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Quantum Materials

Lateral Semiconductor
Nanostructures, Hybrid
Systems and Nanocrystals

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Detlef Heitmann

(Editor)

Quantum Materials

Lateral Semiconductor Nanostructures,
Hybrid Systems and Nanocrystals

With 209 Figures



Springer

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Preface

Introduction

Semiconductor nanostructures are ideal systems to tailor the physical properties via quantum effects, utilizing special growth techniques, self-assembling or lithographic processes in combination with tunable external electric and magnetic fields. We call such systems “Quantum Materials”.

The physical properties of these systems are governed by size quantization effects and discrete energy levels. The charging is controlled by the Coulomb blockade, and one can realize systems with $N = 1, 2, 3 \dots$ electrons, which allows one to study single-particle effects and successively the development of the most elementary many-body effects such as the formation of singlet and triplet states for two electrons, or more complex exchange and correlation effects for more electrons.

An important aspect of these quantum materials is that it is possible to also manipulate the spins of the system, which directly relate the quantum materials to the strongly developing field of spintronic. In quantum materials, not only the electronic properties but also the dispersion of the photons and the phonons will be quantized thus that, respectively, confined electromagnetic optical modes or confined optical and acoustic phonons can be studied. In addition, the high quality of man-made quantum dots also allows one to study the influence of size quantization on the crystal morphology and the formation of bulk, interface, and surface states.

In this book, we cover in different chapters the preparation of quantum materials, a wide variety of experimental techniques for the investigation of these interesting systems, and describe selected experiments which give an overview about the wide field of physics and chemistry that can be studied in these systems. These experiments benefit in an interacting way from sophisticated theoretical concepts that will be addressed in a number of chapters.

Preparation

In several chapters, we describe different methods to fabricate quantum materials. We review the growth of optimized GaAs or InAs quantum wells and heterostructures by molecular beam epitaxy (MBE) with or without modulation doping. Starting from such two-dimensional electron systems (2DES), one-dimensional quantum wires, zero-dimensional quantum dots or antidots can be prepared in a top-down process using etching techniques. We also address MBE based bottom-up approaches for the preparation of self-assembled InAs quantum dots utilizing the Stranski–Krastanov growth mode or droplet epitaxy. Very important is also the preparation of electrical contacts, in particular to control the spin orientation in all-semiconductor devices or in hybrid ferromagnetic/semiconductor systems. The MBE also allows one to grow strained bi-layer system which roll up to microtubes, also called microrolls or microscrolls, if a sacrificial layer is etched away. Another powerful bottom-up process for the fabrication of quantum materials is the wet chemical synthesis of nanocrystals. It is possible to prepare sophisticated core–shell–shell nanocrystals with very narrow size distributions, high stabilities, and photoluminescence yields.

Experimental Techniques

In a number of chapters, we have sections providing introductions into various experimental techniques to study quantum materials. With far-infrared, photoconductivity and Raman spectroscopy, the elementary charge and spin excitations in quantum wells, wires, dots, and antidots can be studied. Photoluminescence in the visible and near-infrared regime gives access to excitonic excitations in the quantum materials. In particular, sophisticated set ups make it possible to perform spectroscopy on a single quantum dot revealing extremely narrow intrinsic line widths. X-ray spectroscopy is an element specific excitation which allows distinguishing between bulk, interface, and surface states in nanocrystals and clusters. X-ray diffraction and near edge X-ray absorption fine-structure spectroscopy give access to the interplay of electronic structure, crystal morphology, and the crystal's phase.

Cantilever magnetometry, capacitance-voltage, and deep-level-transient-spectroscopy measure the ground state properties and density of states in the quantum structures. They are closely related and complementary to transport experiments on the same structures. A very powerful method for quantum materials is the scanning tunneling spectroscopy. On surfaces, step edges, quantum dots or chemically prepared nanocrystals, one can study the local density of states of electrons and holes in different dimensions and directly map the electron's wave functions.

Experiments and Theory

The focus in most of the chapters in this book lies on selected striking experiments and sophisticated theories of these quantum materials, as listed in the Content Section. Self-assembled InAs quantum dots, embedded in gated structures, can be successively charged with $N = 1, 2, 3, \dots$ electrons. This charging is governed by the Coulomb blockade and can be studied by capacitance-voltage spectroscopy. With resonant Raman spectroscopy, one observes for $N = 1$ electron directly the quantized energy levels of the systems. The spectra for $N = 2$ electrons, the so called quantum-dot Helium, one finds, besides singlet-singlet transition, the dipole-forbidden spin-density excitation into the triplet state. The latter resembles the ortho-Helium state of the natural He atom. Far-infrared spectroscopy and photoconductivity give access to a wide variety of charge- and spin-density excitation in quantum dots, antidot arrays and electron systems with internal density modulation arising from many-body effects. Other approaches with complementary information are based on magnetization experiments and deep-level-transient-spectroscopy. A complementary approach to the energy levels of artificial few-electron atoms comes from scanning electron tunneling spectroscopy which, as an ultimate limit, allows a direct mapping of the individual electronic wave functions in the quantum materials.

In two chapters of our book, we review experiments on semiconductor microtubes, in particular the study of the quantum Hall effect in a curved geometry and the realization of optical microtube resonators where it is possible to confine light in three dimensions.

An interesting feature of the quantum materials is the possibility to control the spin. In several chapters, we will review theory and experiments of different aspects of spin transport, in particular, the controlled spin injection from hybrid ferromagnetic/semiconductor contacts, based on permalloy or on Heusler alloys, or all-semiconductor spin valves utilizing the Rashba effect.

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Much of the work reviewed here has been conducted within the Collaborative Research Center SFB 508 ‘Quantum Materials – Lateral Structures, Hybrid Systems and Nanocrystals’. We are very grateful to the German Science Foundation DFG for the generous support for 12 very successful years. We also thank Mrs. Barbara Truppe and Dr. Helga Gemegah for their great and very skillful commitments in all aspects of the administrative organization of our Collaborative Research Center.

Hamburg,
April 2010

Detlef Heitmann

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Chapter 1

Self-Assembly of Quantum Dots and Rings on Semiconductor Surfaces

Christian Heyn, Andrea Stemmann, and Wolfgang Hansen

Abstract Self-assembled semiconductor quantum dots provide almost ideal zero-dimensional quantum confinement for charge carriers. Employing self-assembly mechanisms during epitaxial growth, we are able to fabricate impurity and defect free barriers in all three spatial dimensions with nanometer precision and without the need of lithographic steps. The homogeneity, composition, and geometry of self-assembled nanostructures crucially depend on details of the epitaxial growth process. We illuminate this dependency on the basis of results of three self-assembly methods, the Stranski–Krastanov growth mode, the droplet epitaxy, and the novel technique of local droplet etching. Central aspects are experimental and theoretical studies on the underlying self-assembling process and its influence on the nanostructures structural, optical, and electronic properties. We also discuss the relevance for device applications.

1.1 Introduction

The famous sentence “God made solids, but surfaces were the work of the Devil” is attributed to Wolfgang Pauli (1900–1958) and illustrates the complex properties of the surfaces of solids. This complexity is also present during the growth of thin crystalline films by gas adsorption on solid surfaces where a variety of different processes play the role. On the other hand, control on the processes ruling crystal growth enables the fabrication of a large variety of very interesting surface morphologies being of great interest for current and future device applications.

Depending on strain and the binding state of the surface atoms (Fig. 1.1a), three classical modes are observed during growth of crystalline material, the Frank–van der Merwe or layer-by-layer growth [1], the Volmer–Weber or island growth [2], and the Stranski–Krastanov or layer plus island growth [3] (Fig. 1.1b). In Frank–van der Merwe growth mode, films with nearly atomically flat surface morphology can be fabricated. By switching the composition of the beam of particles directed toward the substrate surface, deposition of layers with nearly abrupt changes of the material composition becomes possible. With the atomic precision of the molecular beam

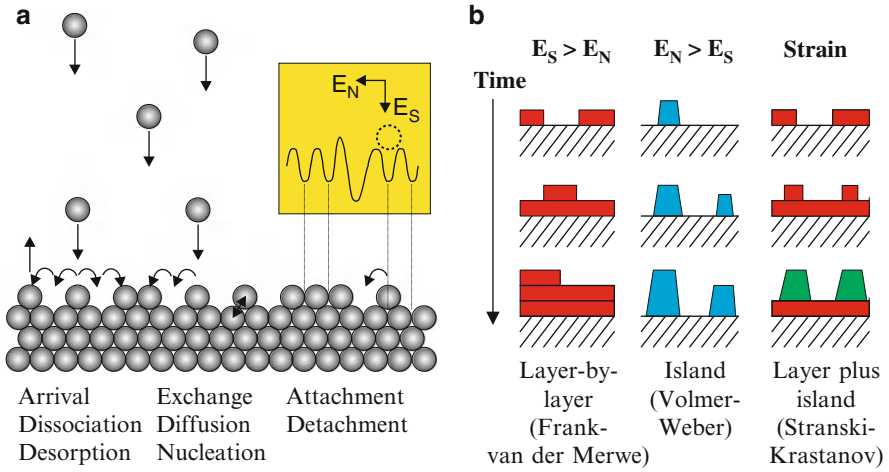


Fig. 1.1 (a) Cross-sectional scheme of the different processes during crystal growth from atomic beams. The insert shows the surface energy landscape illustrating the energy barrier for surface diffusion. The surface diffusion energy barrier has two major contributions: the binding energy E_S to the surface and the lateral binding energy E_N to neighboring atoms. (b) Modes of epitaxial growth dependent on the ratio between E_S and E_N as well as on the influence of strain

epitaxy technique, this leads to the concept of the semiconductor heterostructure¹ which allows control on the local charge and the insertion of barriers for the charge carriers.

The controlled generation of crystalline quantum-size structures employing self-assembly mechanisms represents a fascinating aspect of physics [4]. A very prominent example is the self-assembly of strain-induced InAs quantum dots (QDs) grown on GaAs in the Stranski–Krastanov mode [5–8]. As artificial atomic-like entities in solid-state systems, they intrigue from a fundamental point of view. But self-assembled QDs are also very attractive for device applications where QDs turned out to be superior to bulk material. This has been demonstrated for instance, in 1999, by the first QD-based laser that exhibits a lower threshold current density compared to QW lasers [9]. Further advanced applications for QDs are proposed such as qubits in quantum computing [10] or single-photon sources in quantum cryptography [11, 12]. The structural, electronic, and optical properties of these nanostructures crucially depend on the conditions during the epitaxial growth process. We illuminate this dependency on the basis of three different self-assembly methods, the Stranski–Krastanov growth, the droplet epitaxy in Volmer–Weber mode, and the novel technique local droplet etching (LDE). Examples of nanostructures generated by these methods are shown in Fig. 1.2 and will be discussed in detail in Sects. 1.2–1.4. In the concluding remarks, we comment on the pros and cons of the different routes to self-assembled QDs.

¹ The Nobel Prize in Physics for: Zhores I. Alferov, Herbert Kroemer, and Jack S. Kilby (2000).

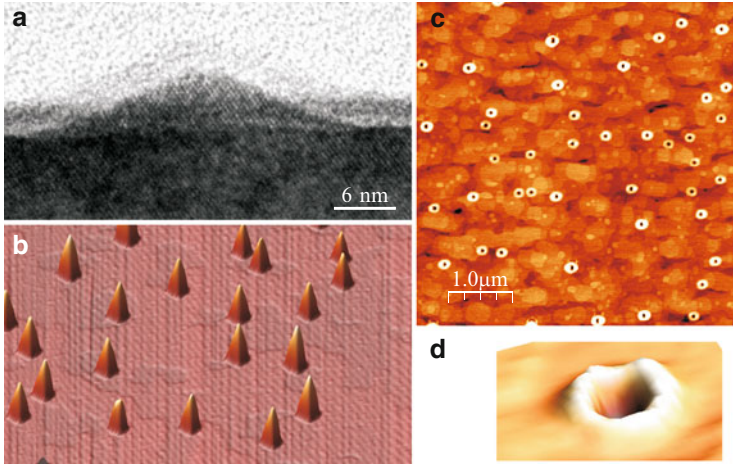


Fig. 1.2 Overview on the various types of nanostructures discussed here: **(a)** TEM cross-section of a strain-induced InAs QD grown in Stranski–Krastanov mode. **(b)** 3D AFM image of an AlGaAs surface with droplet epitaxial GaAs QDs. **(c)** Top view AFM image of an AlGaAs surface with nanoholes and GaAs quantum rings after local droplet etching with Ga. **(d)** 3D AFM image of a nanohole with quantum ring

1.1.1 Molecular Beam Epitaxy

Molecular beam epitaxy (MBE) denotes epitaxial growth of thin semiconductor, metal, or oxide films from atomic or molecular beams and was introduced in the late 1960s by J.R. Arthur and A.Y. Cho for the growth of III/V-semiconductors. Overviews are given, for instance, in [13–16]. The term epitaxy (Greek: “epi” “above” and “taxis” “in ordered manner”) describes crystalline growth with order given by the substrate. The samples studied here were fabricated in a MBE cluster system with two semiconductor growth chambers (Riber 32P and Riber C21). The base pressure inside the MBE chambers is in the low 10^{-11} mbar range in order to avoid unintentional doping with background impurities. The molecular or atomic beams are thermally evaporated from ultra-pure elements in so-called effusion cells. The MBE chambers are equipped with several effusion cells for evaporation of the group III elements Ga, Al, and In, the group V element As, the dopants Si and C, as well as with a Mn cell for the fabrication of diluted magnetic semiconductors. This cell configuration allows the growth of heterostructures composed of the compound semiconductors GaAs, AlAs, InAs, and alloys of these materials. With cell shutters in front of the effusion cells, switching of the respective flux takes less than 0.5 s. In combination with a typical MBE growth speed of about one monolayer (ML) per second, this enables the vertical structuring of semiconductor crystals on the atomic scale. We use 2 in. GaAs, InAs, or InP wafers as substrates for the MBE growth. Most samples discussed here were grown on (001) GaAs substrates.

The time evolution of the surface morphology on the growing crystal was examined in situ using reflection high-energy electron diffraction (RHEED). RHEED is a very powerful method and has been established as a standard technique for instance to study the GaAs surface morphology [17, 18] during MBE, intensity oscillations during GaAs layer-by-layer growth [19–23], and the spontaneous formation of InAs QDs in Stranski–Krastanov mode [24–27]. In our RHEED experiments, we use a 12 keV electron source in combination with a CCD camera and an image processing program on a personal computer for data acquisition. An ex situ analysis of the created nanostructures was performed using atomic force microscopy (AFM) and transmission electron microscopy (TEM).

1.1.2 Kinetics of Crystal Growth

Figure 1.1a gives an overview on the most important processes during crystal growth from molecular or atomic beams. The flat and crystalline substrates are heated to the growth temperature T . Effusion cells provide beams of atoms or molecules that are directed to the substrate surface. The flux of species i to the surface is denoted as F_i and given in units of monolayers per second (ML/s). Molecules impinging on the surface are thermally dissociated into single atoms before incorporation. Dissociation is relevant, e.g., for incorporation of arsenic from As_4 or As_2 beams into GaAs layers [28].

After a surface lifetime, adatoms that are not incorporated into the growing surface by chemical bonding are re-evaporated from the surface by desorption. The ratio between incorporated and impinging atoms is described by the sticking coefficient $\alpha = 1 - R_D/F$, with the desorption rate R_D . Under usual MBE growth conditions, the growth rate determining group III elements completely stick on the surface ($\alpha_{\text{III}} \simeq 1$), whereas As is incorporated only via reaction with a group III element ($\alpha_{\text{As}} \simeq F_{\text{III}}/F_{\text{As}}$) [22]. By applying a slight As overpressure, this allows the fabrication of stoichiometric films [29].

Incorporation of adatoms into the growing film takes place via exchange processes with substrate atoms, attachment to steps on vicinal surfaces, or nucleation of growth islands and subsequent attachment of additional atoms to these islands on flat surfaces. For the latter two processes, the surface mobility of the adatoms is an essential. At sufficiently high temperatures, free adatoms perform a random-walk on the surface, the so-called surface diffusion. In order to jump to a neighboring surface site, free adatoms must thermally overcome the surface diffusion energy barrier E_S , which reflects the binding to the substrate surface. Adatoms that are located at island edges have a higher surface diffusion energy barrier which is $E_S + E_N$, with the lateral binding energy E_N to neighboring atoms. In this picture, collisions between diffusing adatoms on the surface lead to an increase of their surface diffusion energy barrier. As a consequence, the adatoms are nearly immobile and act as nuclei for the formation of growth islands by capturing additional diffusing adatoms. These considerations demonstrate the crucial role of nucleation processes during crystal

growth, which decisively determine the properties of the resulting layers. Classical nucleation theory [30] predicts the density of stable islands as function of growth temperature and speed by a scaling law

$$n = c_n F^p \exp\left(\frac{E_a}{k_B T}\right) \quad (1.1)$$

with the constant c_n , the flux F of the growth rate determining species ($F = F_{\text{III}}$ for growth of III/V-semiconductors under usual growth conditions), and Boltzmann's constant k_B . In the case of complete condensation $E_a = p(E_S + E_i/i)$, with the critical cluster size i , the energy of a critical cluster E_i , and the parameter $p = i/(i + 2.5)$ for three-dimensional (3D) or $p = i/(i + 2)$ for two-dimensional (2D) islands with the height of 1 ML.

Different growth modes are observed dependent on the ratio between E_S and E_N (Fig. 1.1b). Frank–van der Merwe or layer-by-layer growth takes place for materials where $E_S > E_N$. Growth islands in this mode are two-dimensional with the height of one monolayer. For semiconductor homoepitaxy, often a critical nucleus size of one is assumed which simplifies (1.1) to $n = c_n F^{1/3} \exp[E_S/(3k_B T)]$. After nucleation, the islands grow laterally up to coalescence and completion of the layer. In ideal layer-by-layer growth, the second layer starts to grow once the first layer has been completed. Layer-by-layer growth is the preferred growth mode for fabrication of semiconductor heterostructures where abrupt interfaces between heterolayers are desired.

Volmer–Weber or island growth is observed for materials where the neighbor binding energy E_N is higher than E_S . In this case, strong bonds inside the islands lead to the formation of three-dimensional islands on the surface. This growth mode is typical, e.g., for deposition of metals on alkali halogenides and will be discussed in Sect. 1.3 for the self-assembled fabrication of GaAs QDs by applying droplet epitaxy.

In Stranski–Krastanov or layer plus island mode, the first few layers grow flat, i.e., comparable to the layer-by-layer mode. With increasing coverage, the strain-energy induced by the lattice mismatch between substrate material and deposit is relaxed by spontaneous formation of three-dimensional islands. In contrast to the kinetically controlled formation of islands in Volmer–Weber mode, the Stranski–Krastanov islands result from energy minimization and, thus, their size distribution is usually sharper. Prominent examples for Stranski–Krastanov growth in semiconductor systems are Ge islands on Si and InAs islands on GaAs. Such islands grown in Stranski–Krastanov mode are a further very prominent example for self-assembled semiconductor QDs and will be discussed in Sect. 1.2.

A large number of theoretical approaches to model crystal growth from vapor and the influence of the process parameters has been published. Reviews are given, e.g., in [30–33]. In general, a crystallization process is governed by both thermodynamic and kinetic factors. This work concentrates on kinetic growth models, since semiconductor epitaxy usually takes place far from equilibrium. In the following sections, (1.1) will be used as a starting point for the development of

more specific growth models describing, in particular, the formation of InAs QDs under consideration of strain, the generation of droplet epitaxial GaAs QDs by taking Ostwald ripening into account, and the formation of nanoholes by LDE with an InGa alloy, where two different surface diffusion barriers are relevant.

1.2 Strain-Driven InAs QDs in Stranski–Krastanov Mode

The fabrication of coherently strained InAs QDs in Stranski–Krastanov mode has been widely established starting from three pioneering works in 1994 [5–8]. The driving force for the self-assembled QD formation is the strain energy induced by the lattice mismatch of about 7.2% between the GaAs substrate and the InAs deposit. Figure 1.3 shows a phase diagram of the different strain relaxation mechanisms during InGaAs growth on (001) GaAs. We find QD generation in Stranski–Kranstanov mode to be energetically favorable for an In content of at least 40% [27]. As an example, a TEM cross-section of an InAs QD is shown in Fig. 1.3c. Dependent on the growth parameters, InAs QDs have typical densities

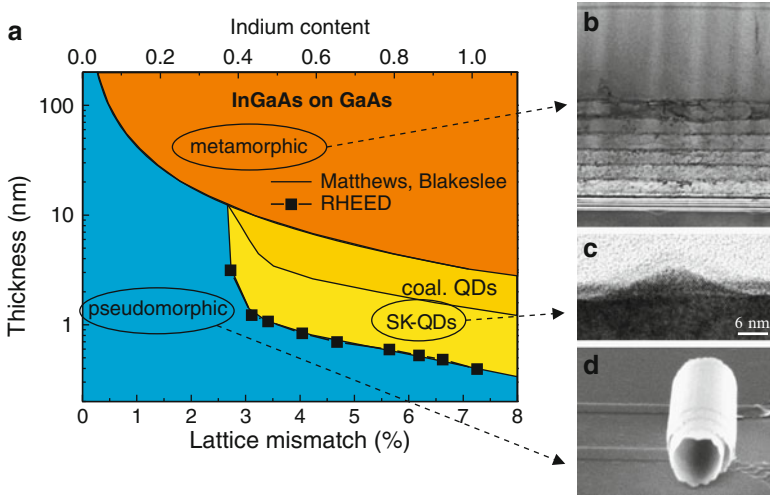


Fig. 1.3 (a) Phase diagram of the strain status of MBE grown $\text{In}_x\text{Ga}_{1-x}\text{As}$ layers on GaAs. At low In content x and for thin InGaAs films the layers are pseudomorphically strained. We use such layers for the fabrication of semiconductor nanotubes utilizing a self-rolling mechanism [34, 35]. For thicker films dislocations are formed at the InGaAs/GaAs interface which we apply in a controlled fashion for the fabrication of metamorphic buffers inside high-mobility InAs HEMTs [36–38]. At high In content, the generation of small islands on the surface is energetically favorable which represents the InAs QD growth in Stranski–Krastanov mode. An increase of the layer thickness in this regime causes coalescence of the QDs. The onset of dislocation formation is calculated according to Matthiessen and Blakeslee [39] and the critical coverage for QD formation is measured by us using RHEED [27]. (b) TEM cross-section of a metamorphic InAs HEMT. (c) TEM cross-section of an InAs QD. (d) SEM image of a semiconductor nanotube

between $1 \times 10^8 \text{ cm}^{-2}$ and $1 \times 10^{11} \text{ cm}^{-2}$, heights between 2 and 12 nm, and diameters of 10–50 nm. The QDs are approximately pyramid-like shaped with an angle of about 25° between the QD side-facets and the substrate surface [40].

From a practical point of view, the QD fabrication process is rather simple and only requires deposition of about 2 ML of InAs on a (001) GaAs substrate. Nevertheless, the growth speed, the III/V flux ratio, InAs coverage, and particularly the growth temperature influence the growth process in a quite complex way. These process parameters control the structural properties of the QDs such as density, size, composition, and, finally, their optical as well as electronic properties [41].

An important quantity, which is very sensitive to the applied process parameters, is the critical coverage $\theta_c = t_c(F_{\text{In}} + F_{\text{Ga}})$ at the instant t_c of the nearly abrupt transition from an initially flat two-dimensional surface morphology to three-dimensional QDs. The critical coverage was precisely determined from the known flux calibration and the critical time t_c taken from *in situ* RHEED experiments [26, 27]. An example for the intensity evolutions of a 2D growth related spot for $\theta < \theta_c$ and a 3D spot for $\theta > \theta_c$ is shown in Fig. 1.4d. The respective RHEED spots marked by arrows are shown in Figs. 1.4a–c.

Most theoretical models of strain-induced QD formation are based on equilibrium arguments [42–45] or consider kinetic effects of the growing surface in terms of kinetic Monte Carlo simulations [46, 47] or mean-field rate-equations [27, 48, 49]. But the very important process of intermixing of the InAs deposit with substrate

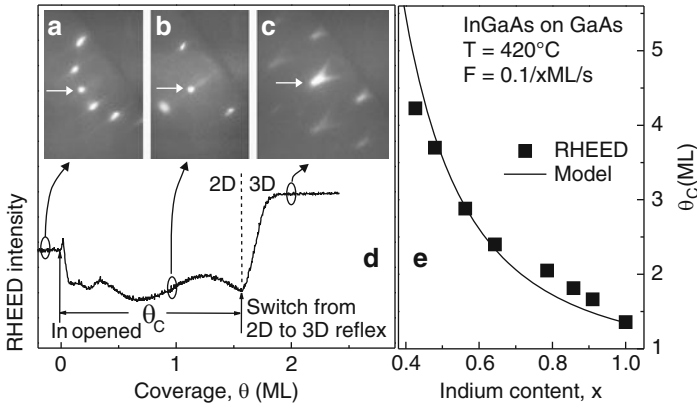


Fig. 1.4 RHEED study of the critical coverage $\theta_c = t_c(F_{\text{In}} + F_{\text{Ga}})$ of the spontaneous $\text{In}_x\text{Ga}_{1-x}\text{As}$ QD formation. Shown are RHEED reflexes from (a) a flat GaAs surface at 420°C in $[1\bar{1}0]$ azimuth, the arrow indicates the 2D-type reflex that is used for the measurement of the time-dependent intensity, (b) after deposition of 1.0 ML InAs, the arrow indicates the 2D reflex, and (c) transmission diffraction and appearance of chevrons [26] after deposition of 2.0 ML InAs, the arrow indicates the 3D reflex used for the time-dependent measurements. The 3D reflex appears at a critical coverage θ_c . (d) Time evolution of the intensity of 2D and 3D growth related reflexes. (e) Critical coverage of $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum dot formation as function of the indium content x . The parameters are $F_{\text{In}} = 0.1 \text{ ML/s}$ and $F_{\text{Ga}} = 0 \dots 0.12 \text{ ML/s}$. The low growth temperature $T = 420^\circ\text{C}$ is chosen so that intermixing is negligible. Symbols denote RHEED data, and the line model results as is described in the text

material is usually neglected. Growth parameter dependent intermixing leads to a high content up to 80% unintentional substrate material in the bottom layer of the QDs [50–53], which significantly modifies the strain status [54] and thus crucially influences the process of QD formation. Furthermore, the high and uncontrolled content of substrate material strongly blue-shifts the optical emission of the InAs QDs which impedes, for instance, the fabrication of QD lasers for the technological relevant wavelengths 1.3 and 1.55 μm .

For a better understanding of the mechanisms behind the strain-induced formation of InAs QDs and, in particular, of the influence of intermixing, we have performed experimental studies accompanied by the development of corresponding growth models [26, 27, 47, 52, 55]. In the following, we will discuss QD formation on basis of a thus developed, simple growth model [55] that allows for calculations without the need of numerical methods. This enables the direct inspection of the influence of the model parameters.

Figure 1.5 shows a sketch of the different layers and growth regimes important for strain-induced InAs QD formation. The growing film is divided into two layers where the initial layer on top of the GaAs substrate is the wetting layer and the second layer on top of the wetting layer we denote as island layer. Due to the strong chemical attraction to GaAs in the substrate [26], migration of In atoms from the wetting layer into the island layer is suppressed. In the island layer, surface diffusion of mobile adatoms leads to the nucleation of monolayer high 2D growth islands. To calculate the average island density n , we refer to the scaling law of (1.1). The total beam flux $F = F_{\text{In}} + F_{\text{Ga}}$ to the surface is the sum of the fluxes from the In and Ga

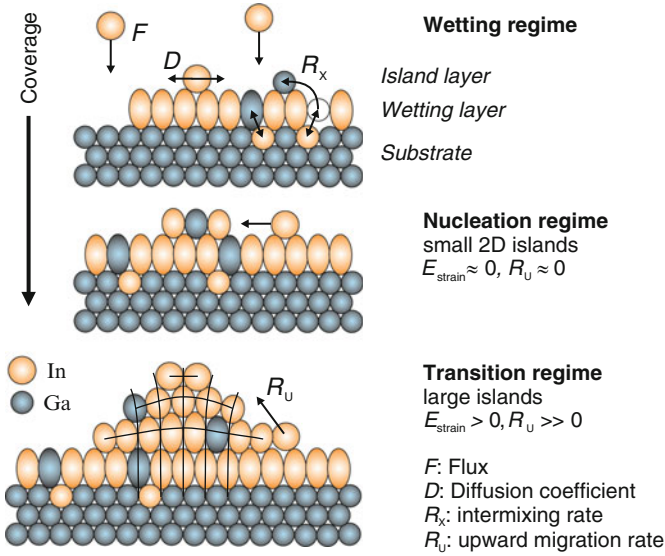


Fig. 1.5 Cross-sectional scheme of the different processes, layers, and regimes considered in the InAs QD growth model

effusion cells. An additional Ga coverage inside the QDs arises from intermixing from substrate material with rate R_x .

With increasing deposition time and thus increasing island size, the strain energy inside the 2D growth islands becomes important and initiates their nearly abrupt transformation into 3D QDs. The strain energy $E_{\text{strain}} = c_s s x^2$ inside a monolayer-high $\text{In}_x\text{Ga}_{1-x}\text{As}$ growth island composed of s atoms can be calculated from Hooke's law, with the average In content x in the islands and the constant c_s [55]. The upwards migration of atoms from the island edges on top of the islands (Fig. 1.5) is the central process for the transition of the initial 2D islands into 3D QDs [27, 52]. The corresponding upwards migration rate is $R_U = s^{1/2} \nu \exp[-E_U / (k_B T)]$, with the energy barrier E_U and the vibrational frequency ν . Following [27], we assume that an increase of the strain energy lowers the upwards migration energy barrier according to $E_U = E_0 - c_u E_{\text{strain}}$, with constants E_0 and c_u .

In order to compare measured values of θ_c with the model calculations, we assume that in the experiments the 2D to 3D transition is observed at the instant at which the upward migration rate becomes significant. In the model, this instant is represented by $R_U(t_c) = 1$. Using this approach, the critical strain energy $E_{\text{strain}}(t_c) = (E_0 - \gamma T) / c_u$ inside the island can be calculated, where the slowly varying $\gamma = -k_B \ln(s^{1/2} \nu)^{-1} \simeq 0.0029 \text{ eV/K}$ is approximately constant for typical values of $s = 1,000$ and $\nu = 10^{13} \text{ s}^{-1}$. The combination of both expressions for the strain energy gives the critical number of atoms inside an island $s_c = s(t_c) = (E_0 - \gamma T) / (c_s c_u x^2)$. This leads to the critical coverage of island material:

$$\theta_{\text{Isl}}(t_c) = s_c n = c F p \frac{E_0 - \gamma T}{x^2} \exp\left(\frac{p E_a}{k_B T}\right) \quad (1.2)$$

with $c = c_n / (c_s c_u)$. For comparison with our RHEED experiments, we now calculate the critical amount of material $\theta_c = F t_c = \theta_{\text{Isl}}(t_c) + \theta_{\text{WL}}(t_c) - \langle R_x \rangle t_c$ at the instant of QD formation:

$$\theta_c = \frac{\theta_{\text{Isl}}(t_c) + \theta_{\text{WL}}(t_c)}{1 + \langle R_x \rangle / F} \quad (1.3)$$

with the average intermixing rate $\langle R_x \rangle$ and the wetting layer coverage $\theta_{\text{WL}} = 1 - \exp(-F t)$. For $\theta > 1.0 \text{ ML}$, the coverage θ_{WL} is only slowly varying and can be approximated by $\theta_{\text{WL}} \simeq 0.8 \text{ ML}$.

Following the study of Joyce et al. [50], we assume that intermixing is negligibly small ($R_x = 0$) at low growth temperatures of $T \leq 420^\circ\text{C}$. In the case of negligible intermixing, (1.3) simplifies to $\theta_c = \theta_{\text{Isl}}(t_c) + \theta_{\text{WL}}(t_c)$ which allows to parameterize the model. The material dependent model parameters are distinguished into nucleation related parameters p and E_a , strain related parameters γ and E_0 , and the constant c . A previous study [27] reveals $E_a = 0.7 \text{ eV}$ and $p = 1/3$ which indicates a critical cluster size of $i = 1$ (see Sect. 1.1.2). The value of $\gamma = 0.0029 \text{ eV/K}$ is given above, and the value $E_0 = 3 \text{ eV}$ is obtained from the condition $(E_0 - \gamma T) > 0$ for the temperatures discussed here. In order to determine the remaining constant c ,

we address earlier RHEED measurements [27], where $\theta_c = 1.36$ ML was found for deposition of pure InAs at $T = 420^\circ\text{C}$ and $F = 0.1$ ML/s. With $x = 1$, we get $c = 0.024$ s/eV.

Furthermore, low temperature growth without significant intermixing allows a controlled adjustment of x by intentional deposition of $\text{In}_x\text{Ga}_{1-x}\text{As}$. Corresponding experimental data are shown in Fig. 1.4 together with values of θ_c calculated using (1.2) and (1.3) with the above parameters. The very good reproduction of the measurements by the calculation results indicates that our simple growth model correctly describes the influence of strain on InAs quantum-dot formation. More elaborate models, which include, for instance, the strain inside the volume and the size distribution of the QDs are described in [27, 52].

In the next step, we have developed a model for the intermixing process by assuming kinetic exchange processes between deposited In atoms and Ga atoms from the substrate (Fig. 1.5) [55]. Figure 1.6a shows values of the Indium content x inside the QDs calculated with this model for $F = 0.01$ ML/s. We find the onset of intermixing at a temperature of about 450°C and nearly completely intermixed layers for $T > 600^\circ\text{C}$. The values of x calculated by the intermixing model are now used as an input parameter for the above QD growth model [(1.2) and (1.3)]. Combining the models for strain-induced QD formation and temperature dependent intermixing, we calculate the critical coverage θ_c of QD formation. In Fig. 1.6b, results are shown as function of T at different values of F . We find a nearly abrupt rise of θ_c at a certain critical temperature. Furthermore, the value of the critical temperature is found to increase nearly linearly with F . The appearance of a critical temperature for QD formation can be explained by intermixing. The intermixing rate and consequently the amount of substrate material inside the QDs increase with T which reduces the strain energy and, thus, the driving force of QD formation. On the other hand, an increase of the In flux reduces this effect and shifts the critical temperature toward higher values. Both, the appearance of a critical temperature

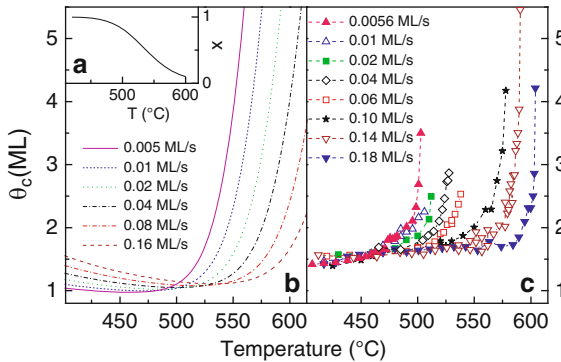


Fig. 1.6 Temperature dependent intermixing during strain-induced QD generation by deposition of pure InAs on GaAs. (a) Calculated Indium content x inside the QDs for an In flux $F = 0.01$ ML/s. (b) Calculated values of θ_c for varied F given in the figure. (c) Critical coverage determined with RHEED for different F

and its flux dependence are found in the experiments, as well (Fig. 1.6c). The close agreement between experimental data and calculation results confirms the assumption that the strain energy reduction due to intermixing causes the experimental critical temperature.

These results illuminate the complex influence of the process parameters and, in particular, of the growth temperature. At $T \leq 420^\circ\text{C}$, where intermixing is negligibly small, a temperature rise causes an increase of both, the island size (1.1), as well as the upward migration rate (1.2). Both effects result in a decrease of θ_c with T (Fig. 1.4e). On the other hand, at higher temperature, intermixing comes into play which reduces the strain-energy inside the QDs and, thus, yields vice versa an increase of θ_c with T (Fig. 1.6).

Based on the results of the above growth studies, we have fabricated optimized heterostructures containing layers with ensembles of highly uniform InAs QDs with controlled structural properties. The quantized electronic states inside the strain-induced InAs QDs have been studied by means of deep level transient spectroscopy (DLTS) [56–59]. We find typical values of the s_1 and s_2 activation energies of 17 meV and the s_2 and p activation energies of 48 meV [56]. Furthermore, the DLTS experiments reveal that apart from pure thermal emission thermally activated tunneling processes are important for an understanding of the spectra, as well. This will be reviewed in the chapter of Schramm et al. in this book.

Garcia et al. [60, 61] have demonstrated that the structural and electronic properties of self-assembled InAs QDs can be modified by overgrowth with GaAs and subsequent growth interruption. In particular, quantum ring-like structures have been obtained. Spectroscopic investigations reveal the quantum ring nature of these systems [62]. Recently, we have shown that the overgrowth with AlAs is also a promising way to form well defined ring-like nanostructures. Photoluminescence (PL) and DLTS studies of their optoelectronic properties have been presented [63, 64]. In addition, we have demonstrated the self-assembled lateral ordering of InAs QDs by an underlying dislocation network [65, 66]. Scanning micro PL allows a spatial mapping of the structures and reveals different emission energies for QDs aligned along [110] and $[-110]$ crystal directions. We attribute this effect to anisotropic surface diffusion during QD formation.

1.3 Droplet Epitaxy in Volmer–Weber Mode

The fabrication of QDs in a self-assembled fashion by applying droplet epitaxy is an interesting alternative to the above discussed technology of strain-driven InAs QD formation in Stranski–Krastanov mode. The method was first demonstrated by Koguchi and Ishige [67] in 1993. In comparison to the Stranski–Krastanov QDs, the method of droplet epitaxy is more flexible regarding the choice of the QD material. For instance, the fabrication of strain-free GaAs QDs [68, 69], InGaAs QDs with controlled In content [70, 71], and InAs QDs [72] has been demonstrated. Furthermore, besides QD like structures, recent experimental droplet epitaxy studies

demonstrate, e.g., the generation of QD molecules [68], quantum rings [73], and concentric double rings [69, 74, 75]. We have developed the first growth model for droplet epitaxy of GaAs QDs [76] and observed an interesting correlation between the QD shape and its volume [77]. In the following, both topics will be discussed.

During QD fabrication [76, 77], first liquid Ga droplets were generated on (001) AlGaAs surfaces in a Volmer–Weber-like growth mode by Ga deposition without As flux. The growth temperature $T = 140\text{--}300^\circ\text{C}$ was kept very low compared to usual MBE growth conditions. Deposition of Ga with flux $F = 0.025\text{--}0.79\text{ ML/s}$ for a time t resulted in a total Ga surface coverage of $\theta = Ft$. We would like to note that the initial AlGaAs substrate surface is As-terminated. Due to the strong binding energy to As in the substrate, the first Ga monolayer is consumed for the formation of a Ga terminated surface and does not contribute to the formation of Ga droplets. That means the coverage of Ga located in the droplets is $Ft - 1$. After Ga droplet formation, 60 s pause was applied for equilibration followed by the crystallization of the droplets and their transformation into GaAs QDs under As pressure. After crystallization, the QDs were annealed for 10 min at $T = 350^\circ\text{C}$.

Figure 1.7a–d shows examples of droplet epitaxial GaAs QDs on AlGaAs. Clearly visible is the strong influence of the growth temperature T on the QD density n . Quantitative results are plotted in Fig. 1.7e for two different values of the growth speed F . The experimental densities n are now discussed on the basis of classical nucleation theory (1.1) as introduced in Sect. 1.1.2. The slope of the temperature dependent data Fig. 1.7e for $T \leq 200^\circ\text{C}$ agrees with a value of E_a of about 0.235 eV . Furthermore, in this regime, the slope of additional flux dependent

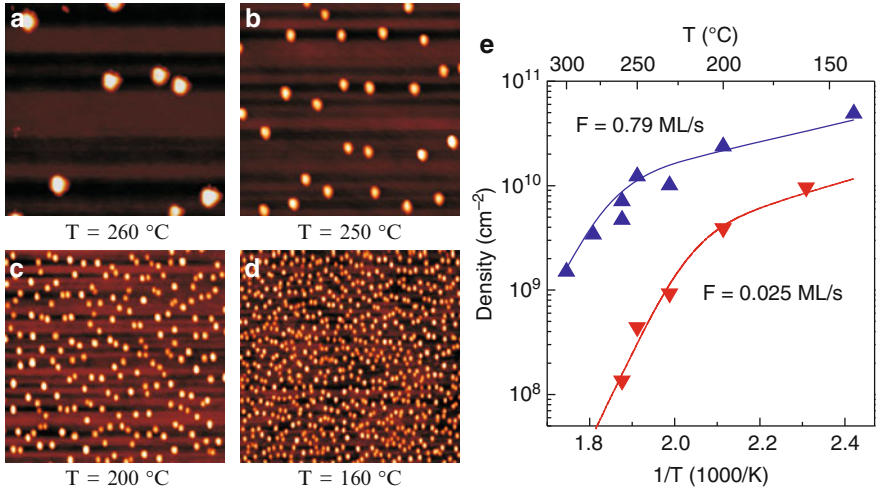


Fig. 1.7 (a)–(d) $2.5 \times 2.5\text{ }\mu\text{m}^2$ AFM images of GaAs QDs grown by droplet epitaxy on (001) AlGaAs at a Ga flux $F = 0.025\text{ ML/s}$, a Ga coverage $\theta = 3.75\text{ ML}$, and indicated growth temperature T . (e) Surface density of GaAs QDs as function of growth temperature T at $\theta = 3.75\text{ ML}$ and indicated F . Symbols reflect results of AFM measurements, and the lines are calculated with the model (1.4)

data [76] fits to $p = 0.50$ and thus $i = 2.5$. This value of i establishes that dimers are unstable and trimers represent the smallest stable island size. On the other hand, in the regime, $T > 200^\circ\text{C}$, the measured values of n are not consistent with classical nucleation theory. We attribute the reduction of the measured QD densities at $T > 200^\circ\text{C}$ to the onset of coarsening by Ostwald ripening [78]. Ostwald ripening means the growth of large clusters on cost of smaller ones and hence causes a decrease of the total cluster density. From mean-field theory [33] of Ostwald ripening under mass conservation, the evolution of the cluster density as function of time t_r is predicted [79]: $n(t_r) = n_0 (1 + t_r/\tau_r)^{-m}$, where n_0 is the droplet density as calculated with (1.1) and m is a scaling exponent that is equal to 1 for three-dimensional islands coupled via adatom diffusion on a two-dimensional surface in the interface-reaction-limited case [79]. We assume an activated temperature dependence $\tau_r = \nu^{-1} \exp[E_r/(k_B T)]$, where E_r is a constant. The ripening time is $t_r = (\theta - 1)/F$. We now expand (1.1) by Ostwald ripening:

$$n = c_n F^p \left[\exp\left(\frac{-E_a}{k_B T}\right) + \nu \frac{\theta - 1}{F} \exp\left(\frac{-E_a - E_r}{k_B T}\right) \right]^{-1} \quad (1.4)$$

As is demonstrated in Fig. 1.7e, results of the extended scaling law of (1.4) using $c_n = 1 \times 10^8$, $p = 0.5$, $E_a = 0.235 \text{ eV}$, and $E_r = 1.5 \text{ eV}$ agree very well with the experimental behavior and hence suggest the validity of our model.

In additional experiments, we have established droplet epitaxy of GaAs QDs on (001), vicinal (001), (110), and (311)A GaAs surfaces [80]. On (311)A GaAs, QDs are formed with higher density and smaller height compared to (001) and (110). A quantitative analysis is performed on basis of (1.4) assuming that the difference in QD density is related mainly to the effect of surface diffusion, whereas the critical cluster size $i = 2.5$ and binding energy $E_i = 0.375 \text{ eV}$ [76] are mostly unchanged. We find a surface diffusion barrier $E_s = 0.32 \text{ eV}$ for the (001) GaAs surface, $E_s = 0.29 \text{ eV}$ for (110), and $E_s = 0.47 \text{ eV}$ for (311)A. Interestingly, on (311)A, QD densities up to 10^{11} cm^{-2} should be realizable, whereas QD densities on (001) and (110) GaAs can be reduced to less than 10^8 cm^{-2} . On vicinal (001) surfaces, step bunches are found to act as preferred nucleation sites for GaAs QDs which opens the possibility for a lateral positioning of the QDs by pre-patterning [80].

Furthermore, our RHEED and AFM experiments establish the existence of two phases for the shape of GaAs QDs grown by droplet epitaxy on (001) AlGaAs [77]. Dependent on the QD volume, the droplets transform either in pyramid-like QDs with $\alpha = 25^\circ$ side-facet angle or truncated pyramids with $\alpha = 55^\circ$. Examples are shown in Fig. 1.8. The angle $\alpha = 25^\circ$ is close to the side-facet angles expected for (113)- or (137)-type side-facets ($\alpha_{113} = 25.2^\circ$ and $\alpha_{137} = 24.3^\circ$, respectively). The considerably higher angle of $\alpha = 55^\circ$ is very close to the angle expected for the (111)-type surface ($\alpha_{111} = 54.7^\circ$). The facet transition is found at a QD volume of about 3×10^5 Ga atoms, where larger QDs form steeper facets. Since the QD volume is correlated to the density, the transition volume corresponds to a QD density of about $6 \times 10^9 \text{ cm}^{-2}$. To our knowledge, a theoretical model that explains the occurrence of these two phases is still missing.

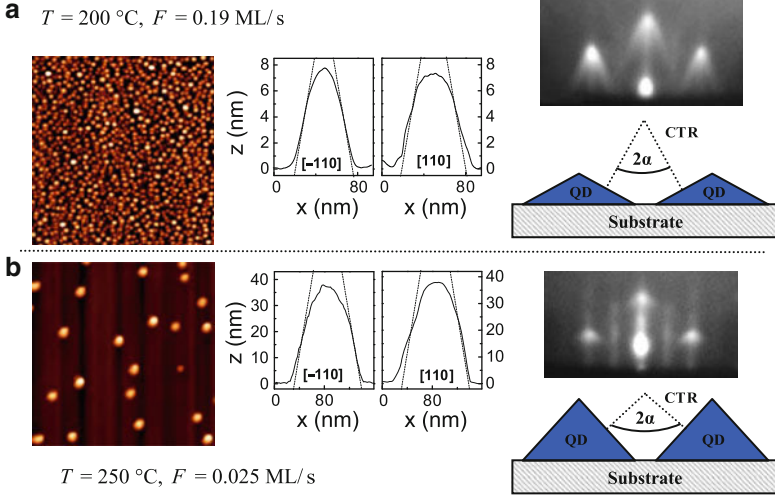


Fig. 1.8 (a) $2.5 \times 2.5\text{ }\mu\text{m}^2$ AFM image, profiles, and RHEED pattern along $[\bar{1}10]$ azimuth from GaAs QDs grown by droplet epitaxy on (001) AlGaAs at $T = 200^{\circ}\text{C}$, $F = 0.19\text{ ML/s}$, and $\theta = 3.75\text{ ML}$. In the RHEED pattern, transmission diffraction spots as well as crystal truncation rods (CTRs) are clearly visible. The sketch illustrates that the angle between the crystal truncation rods is twice the QD side-facet angle α . From the RHEED pattern, we determine $\alpha = 25^{\circ}$ which corresponds to (113)- or (137)-type side-facets. Corresponding facets are plotted as *dotted lines* in the profiles. (b) GaAs QDs grown at $T = 250^{\circ}\text{C}$ and $F = 0.025\text{ ML/s}$. Here $\alpha = 55^{\circ}$ is determined corresponding to (111)-type facets

In order to study the optoelectronic properties of droplet-epitaxial GaAs QDs, we have embedded QD layers in AlGaAs barrier material. Using PL spectroscopy, we find an only very broad and weak optical emission which we attribute to the poor QD size uniformity and to the incorporation of undesired defects or background dopants caused by the low growth temperatures. Similar observations were reported by Mano et al. [81] who have performed additional post-growth rapid thermal annealing steps in order to improve the QD quality. Nevertheless, we have turned to the more promising technique of LDE for the fabrication of strain-free GaAs QDs as is described in Sect. 1.4.

1.4 Local Droplet Etching

The very recent technique of local droplet etching (LDE) provides self-assembled nanoholes by a local removal of material from semiconductor surfaces without the need of any lithographic steps. As an important advantage compared to conventional lithography processes, LDE is fully compatible with usual MBE equipment and can easily be integrated into the MBE growth of heterostructure devices. Examples of LDE nanoholes are shown in Fig. 1.1c and d. The nanoholes are 1–40 nm deep

which can be adjusted by the process conditions. Similar to the droplet epitaxy described above, the LDE process starts with the generation of metallic droplets on the surface. However, here significantly higher temperatures are used. At these temperatures, deep nanoholes are formed at the interface between the liquid droplets and the substrate. The fabrication of such nanoholes was first demonstrated by Wang et al. [82] on GaAs surfaces using gallium as etchant. Later, we have demonstrated LDE on AlGaAs [83, 84] and AlAs [87] surfaces as well as etching with In [83] and Al [87] droplets. In addition, we have observed the formation of walls surrounding the nanohole openings, which serve as quantum rings [83]. In the following, we address the structural properties of the nanoholes and quantum rings together with the influence of the conditions during the fabrication process. Furthermore, we demonstrate the fabrication of highly uniform GaAs QDs by filling of LDE nanoholes.

1.4.1 Structural Properties of LDE Nanoholes and Rings

We fabricate LDE nanoholes on (001) GaAs, AlGaAs, or AlAs substrates. First, the As shutter and valve were closed and droplet formation was initiated at a temperature T_1 by opening the Ga, Al, or In shutter for a time t_1 . During this stage, a strongly reduced arsenic flux is important [88]. The As flux in our experiments was approximately hundred times lower compared to typical GaAs growth conditions. The Ga, Al, or In flux F corresponded to a growth speed of 0.8 ML/s, and droplet material was deposited onto the surface with coverage $\theta = Ft_1$. After droplet deposition, the temperature was set to a value T_2 and a thermal annealing step of time t_2 was applied in order to remove liquid etching residues. For most samples, we have selected $t_1 = 4$ s and $t_2 = 180$ s.

A sketch of the different stages during LDE is shown in Fig. 1.9. The key process for nanohole creation is the diffusion of As from the substrate into the droplet which causes the liquefaction of the substrate below the droplet. From the measured hole volume, we determine a value of 0.03 ± 0.01 for the average As concentration in the droplet material [88]. In [83], we have shown that the walls surrounding the nanohole openings are crystallized from droplet material. This finding is explained by the assumption that As diffuses to the droplet surface and crystallizes during the annealing step with droplet material at the interface to the substrate. Interestingly, the amount of material stored in the walls is equal to the amount of material removed from the holes [88]. This result indicates conservation of arsenic. Nearly all As which has been extracted from the substrate into the droplet will crystallize into wall material.

At present, the mechanism for the removal of the liquid material during the annealing step is not completely clear. Figure 1.10 shows a series of AFM images from AlGaAs surfaces after Ga LDE at $T_1 = T_2 = 570^\circ\text{C}$ and different annealing times t_2 . Directly after droplet formation ($t_2 = 0$ s), only hills are visible on the surface that we identify as the initial droplets. At $t_2 = 120$ s, there is a

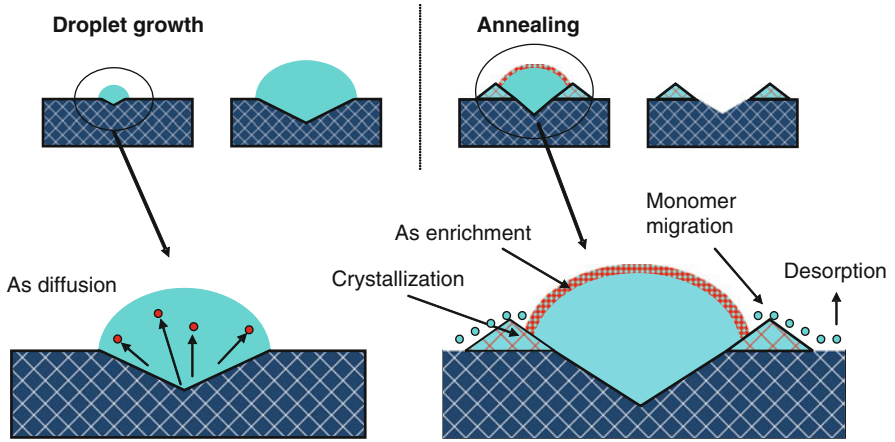


Fig. 1.9 Sketch of the different stages during LDE of nanoholes and wall formation

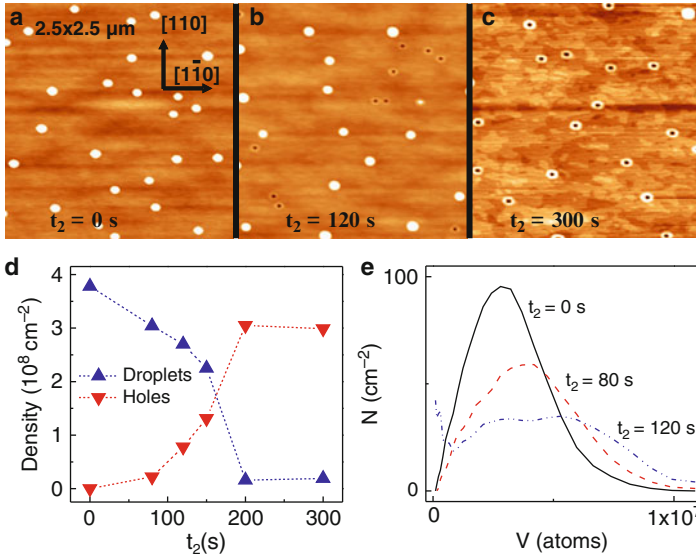


Fig. 1.10 (a)–(c) AFM images from AlGaAs surfaces after Ga LDE at different annealing times t_2 . The temperatures were $T_1 = T_2 = 570^\circ\text{C}$, $t_1 = 4$ s, and $F = 0.8$ ML/s. (d) Density of the droplets and of the nanoholes as function of t_2 . (e) Droplet size distribution at different t_2

co-existence of droplets and nanoholes and at $t_2 = 300$ s nearly all droplets have been removed. A quantitative analysis (Fig. 1.10d) establishes a nearly abrupt transition from droplets to holes. Furthermore, the data show that the hole density is slightly reduced compared to the initial droplet density. Unexpectedly, the center of the droplet size distribution shifts toward higher volumes during the droplet removal

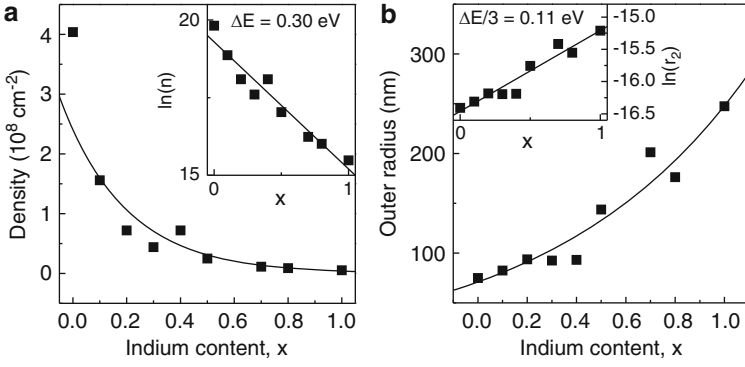


Fig. 1.11 (a) Symbols: measured density n of nanoholes fabricated with $\text{In}_x\text{Ga}_{1-x}$ LDE as function of x . Lines: a characteristic energy $\Delta E = 0.30 \text{ eV}$ is determined from an exponential fit. (b) Symbols: measured outer-radius r_2 of the quantum rings surrounding the nanoholes as function of x . A characteristic energy $\Delta E = 0.33 \text{ eV}$ is determined from an exponential fit indicated by the line

(Fig. 1.10e). This important finding establishes the relevance of droplet coarsening by Ostwald ripening during the annealing step. Ostwald ripening requires a significant exchange of material between the different droplets which can take place only via diffusion on the substrate surface. Therefore, we assume a high density of mobile monomers on the free surface mainly emitted from small droplets. These monomers may attach to larger droplets and increase their volume or re-evaporate, which we consider as central mechanism for the droplet removal (Fig. 1.9).

Additional experiments using a two-temperature process with $T_1 = 570$ and $T_2 = 620^\circ\text{C}$ show a nanohole density which is approximately 10 times lower compared to the density of the initial droplets. Again, this effect might be explained by coarsening of the droplets during the temperature rise from T_1 to T_2 . Importantly, with the two-temperature process, very low nanohole densities down to less than $5 \times 10^7 \text{ cm}^{-2}$ can be achieved which allows one to directly address single nano objects by a focussed laser beam for single QD spectroscopy.

As an additional interesting method to control the nanohole density and size, we have etched GaAs and AlGaAs surfaces with $\text{In}_x\text{Ga}_{1-x}$. By etching with pure In, hole densities as low as $5 \times 10^6 \text{ cm}^{-2}$ have been achieved. To systematically study the influence of the indium content x , a series of 9 samples with varied $x = 0 \dots 1$ was fabricated using $T_1 = 520$ and $T_2 = 580^\circ\text{C}$. The nanohole densities and wall outer radii on these samples are plotted in Figs. 1.11a and b, respectively. Clearly visible is the strongly reduced hole density when x is increased. For a quantitative interpretation of the data, we assume different surface diffusion barriers for In and Ga. Since the critical nucleus size is unknown, we consider here a composition dependent energy $E_a = E_{a,\text{Ga}} - x(E_{a,\text{Ga}} - E_{a,\text{In}}) = E_{a,\text{Ga}} - x\Delta E$. Insertion into (1.1) yields for the nanohole density:

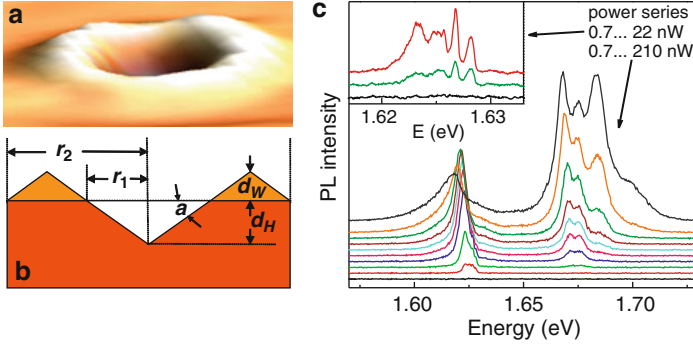


Fig. 1.12 (a) 3D AFM image of a typical nanohole with wall fabricated using LDE with $x = 0$. (b) Schematic profile of a LDE nanohole with wall. r_2 is the wall outer radius, r_1 the wall inner radius which is equal to the hole opening radius, d_H the hole depth, d_W the wall height, and α the angle between the substrate surface and the side wall of the holes. (c) Low temperature PL measurements of a single GaAs quantum ring in AlGaAs. The excitation power was varied from 0.7 up to 210 nW. The inset shows a magnification of the peaks at 1.625 eV at an excitation power varied from 0.7 up to 22 nW

$$n = c_n F^p \exp\left(\frac{E_{a,\text{Ga}} - x\Delta E}{k_B T}\right) = c_n F^p \exp\left(\frac{E_{a,\text{Ga}}}{k_B T}\right) \exp\left(\frac{-x\Delta E}{k_B T}\right). \quad (1.5)$$

From an exponential fit with (1.5) of the measured hole densities vs. x in Fig. 1.11a, a value of $\Delta E = 0.30$ eV was determined. In additional temperature dependent experiments Ga LDE nanoholes were etched on GaAs and AlGaAs surfaces. The measured nanohole densities are analysed using (1.1) and correspond to a value of $E_{a,\text{Ga}} = 0.54$ eV, which allows to estimate $E_{a,\text{In}} = E_{a,\text{Ga}} - \Delta E = 0.24$ eV.

The shape of the nanoholes and walls (Fig. 1.12a) is quantitatively characterized by the nanohole depth d_H , the wall's inner radius r_1 , which is identical to the nanohole opening, and the wall's outer radius r_2 (Fig. 1.12b). As a general trend, for our nanoholes, we find that the wall outer radius is approximately twice the inner radius $r_2 \approx 2r_1$. The continuous decrease of the wall radius with decreasing In content is quantitatively depicted in Fig. 1.11b. Neglecting desorption during the droplet growth regime, the volume of an individual droplet is related to the droplet density via $V = Ft/n$. We assume that the outer radius of the quantum rings represents the radius of the initial droplets [83] which yields $r_2 \propto V^{1/3} \propto \exp[x\Delta E/(3k_B T)]$. Again, the measured values agree well with the exponential law (Fig. 1.11b) and a value of $\Delta E = 0.33$ eV was determined from the r_2 data. The close agreement of both values of ΔE determined independently from the hole density and wall radius indicates the validity of our approach.

The depth of the holes $d_H = r_1 \tan \alpha$ is closely related to the hole radius r_1 via the angle α between the substrate surface and the side wall of the holes (Fig. 1.12b). We find no significant dependence of α on the In content of the etchant, and the average angle is constant $\alpha \simeq 20^\circ$ within the accuracy of the measurements.

We have performed low-temperature PL measurements to study the optical properties of the quantum-ring-like walls around the nanohole openings. For the selection of single rings, a micro-PL setup is used with a focussed laser beam. Figure 1.12b shows a PL power series of a single GaAs quantum ring embedded in AlGaAs. For low excitation power, we find a set of PL peaks at about 1.625 eV. The inset of Fig. 1.12b shows a magnification of these peaks. We attribute the sharp lines to ground-state excitons and the broader ones at lower energy to multiexcitonic transitions. With increasing excitation power, additional peaks occur at higher energies (about 1.67 eV). These we attribute to quantum-ring excited states. Additional investigations are required to clearly confirm the quantum-ring-like confinement potential from the PL data. Alternative techniques for the self-assembled fabrication of semiconductor quantum rings employ, for instance, partial overgrowth of InAs QDs (see Sect. 1.2) or type-II InP/GaAs QDs [85]. For the latter structures, Aharonov–Bohm-type oscillations [86] for neutral excitons have been observed.

1.4.2 Fabrication of QDs by Filling of LDE Nanoholes

In this section, we describe the creation of a novel type of very uniform and strain-free GaAs QDs by filling of LDE nanoholes [87]. Recent concepts for the self-assembled fabrication of strain-free GaAs QDs utilize droplet epitaxy (see Sect. 1.3) or hierarchical self-assembly [89]. However, droplet epitaxy takes place at unfavorable low temperatures and the obtained QDs show a broad size distribution. Hierarchical self-assembly requires in situ etching of buried InAs QDs with AsBr₃ and the samples include a highly strained InAs wetting layer.

For the LDE QD fabrication, nanoholes in AlGaAs and AlAs are filled with GaAs. In a first step, the process conditions were optimized. We used Al droplets for etching in order to avoid an additional charge-carrier confinement caused by the wall. After Al LDE on AlGaAs surfaces, we find a bimodal distribution of the hole depth similar to earlier experiments [84]. We distinguish between the desired deep holes, with depth of more than 8 nm, and shallow holes. From earlier results [84], we know that the formation of flat nanoholes can be suppressed by performing the LDE process at higher temperatures. Due to decomposition of the surface, the maximum temperature for LDE on AlGaAs is about 630°C. Therefore, for QD fabrication, the LDE process was performed on more stable AlAs surfaces with $T_1 = T_2 = 650^\circ\text{C}$. Such etched surfaces show only deep holes with a density of $4 \times 10^8 \text{ cm}^{-2}$, an average hole depth of $d_H = 14 \text{ nm}$, and slightly elliptical hole openings with axis of 39 nm along $[1-10]$ direction and 33 nm along $[110]$.

The nanoholes were filled with GaAs at a substrate temperature of 600°C in a pulsed mode. Very importantly, the holes are only partially filled with a filling level defined by the precise layer thickness control of the MBE technique. The resulting very uniform GaAs QDs are shaped like inverted cones with slightly elliptical base area and heights $h_{\text{QD}} = 4.5$ and 8.0 nm. The height is perfectly controlled by the amount of Ga deposited for filling.

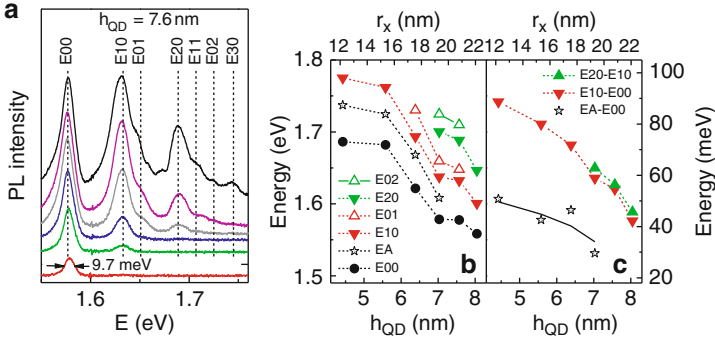


Fig. 1.13 PL measurements of LDE QDs at $T = 3.5$ K. The laser energy is 2.33 eV. **(a)** Power series ($I_e = 8.5 \dots 450$ W/cm²) of a LDE-QD sample with $h_{\text{QD}} = 7.6$ nm. Dashed lines indicate calculated transition energies assuming a parabolic confinement potential. **(b)** Energy of the ground and excited states for LDE QDs as function of h_{QD} . **(c)** Energy separations $E_{10}-E_{00}$, $E_{20}-E_{10}$, and E_A-E_{00} taken from the data of **(b)**. The continuous line is calculated assuming a ratio between hole and electron quantization energy of 0.39

In Fig. 1.13a, PL spectra of a sample with $h_{\text{QD}} = 7.6$ nm are plotted for different excitation intensities I_e . The very small linewidth of the ground-state peak E_{00} with a full width at half maximum of 9.7 meV demonstrates the high homogeneity of the QD ensembles. The number of dots probed in the PL measurement is roughly 4×10^4 . A slight red-shift of 2 meV for the E_{00} peak with increase of I_e is attributed to the occurrence of additional multiexcitonic lines [89]. Additional sharp peaks arise with increasing I_e that are related to excited states. For an understanding of the PL spectra, we approximate the electron and hole energy quantization due to the lateral confinement with an anisotropic parabolic potential model. Optical recombinations between electrons and holes from states with identical quantization numbers n_x, n_y are denoted in the form $E_{n_x n_y} = E_{00} + n_x \hbar \omega_x + n_y \hbar \omega_y$, with the oscillator frequencies ω_x and ω_y . In Fig. 1.13a, the PL data are compared with energy levels calculated using $E_{00} = 1.577$ eV, and equidistant quantization energies $\hbar \omega_x = 56$ meV, and $\hbar \omega_y = 74$ meV. Our approach of a parabolic potential with a slightly anisotropic QD base describes the data very well.

A summary of the PL peak positions is plotted in Fig. 1.13b. We find an increase of all peak energies with decreasing QD height h_{QD} . The energy separations between the E_{00} , E_{10} , E_{20} peaks are plotted in Fig. 1.13c. QDs higher than 6.5 nm show equidistant peaks. This agrees with a parabolic potential. Interestingly, QDs with a height smaller than 7.5 nm show additional peaks E_A marked by open stars in Figs. 1.13b and c. We suggest that these peaks are caused by transitions between ground-state electrons and holes from an excited state.

A very advanced application for semiconductor QDs is the generation of entangled photons for quantum cryptography [91]. As a precondition for entanglement, the QD fine-structure splitting must be smaller than the linewidth. The occurrence of a fine-structure splitting is known for instance for InAs QDs and is related to

strain-induced polarization or a shape anisotropy of the QDs [90]. Studies of the fine structure are only possible on single QDs which requires either masking or a reduction of the QD density combined with QD selection by a focussed laser beam. We have applied the latter method and used the two-temperature LDE process described above for the fabrication of GaAs QDs with a low density of about $5 \times 10^7 \text{ cm}^{-2}$. First results of a sample with $h_{\text{QD}} = 4.4 \text{ nm}$ exhibit sharp excitonic lines with an exciton peak that shows a polarization dependent shift of the emission energy by about $50 \mu\text{eV}$. This shift is related to the corresponding fine-structure splitting. On the other hand, a sample with $h_{\text{QD}} = 7.6 \text{ nm}$ shows no influence of the polarization angle on the exciton peak energy which indicates that the fine-structure splitting is below the resolution of the spectrometer of about $20 \mu\text{eV}$. As a promising result, the very small fine-structure splitting might render this type of QDs suitable for the generation of entangled photons.

1.5 Conclusions

A semiconductor quantum dot can be regarded as the ultimate solid-state nanostructure which features confinement for charge carriers in all three directions. In this context, we discuss here three different types of self-assembled semiconductor QDs all basing on epitaxial growth processes. Most prominent are the strain-induced InAs QDs grown in Stranski–Krastanov mode. We have discussed the influence of the growth parameters on the QD properties with focus on the often neglected unintentional intermixing with substrate material. A model of intermixing is described which quantitatively agrees with experimental data taken with in situ electron diffraction and ex situ X-ray diffraction. This intermixing is one of the weak points of the InAs QDs since it causes a high amount of substrate material inside the QDs with poorly known lateral and vertical material distribution. Furthermore, the InAs QDs are substantially strained which causes a strong fine-structure splitting [90] and impedes their application as emitters for entangled photons.

The fabrication of strain-free QDs without intermixing is possible using the droplet epitaxy. We have developed a growth model that quantitatively reproduces experimental QD densities as function of growth temperature and speed. However, the homogeneity of droplet epitaxial QDs is rather poor and the low process temperatures cause the incorporation of undesired defects and background impurities.

Therefore, we have introduced a novel method for the fabrication of unstrained and very uniform GaAs QDs and rings by applying LDE. In comparison to droplet epitaxy, the LDE process takes place at usual MBE growth temperatures. The QDs produced by filling of the nanoholes are very uniform and have a high optical quality. In comparison to InAs QDs, the unstrained LDE QDs are suggested as emitters for entangled photons in quantum cryptography. At present, we are investigating, for instance, filling of the LDE nanoholes with InGaAs in order to provide InGaAs QDs with independently tunable size and composition. We expect that the optical

emission of such InGaAs QDs can be adjusted over a wide range and in particular to the technologically relevant wavelengths of 1.3 and 1.55 μm .

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Chapter 2

Curved Two-Dimensional Electron Systems in Semiconductor Nanoscrolls

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Abstract The perfect control of strain and layer thickness in epitaxial semiconductor bilayers is employed to fabricate semiconductor nanoscrolls with precisely adjusted scroll diameter ranging between a few nanometers and several tens of microns. Furthermore, semiconductor heteroepitaxy allows us to incorporate quantum objects such as quantum wells, quantum dots, or modulation doped low-dimensional carrier systems into the nanoscrolls. In this review, we summarize techniques that we have developed to fabricate semiconductor nanoscrolls with well-defined location, orientation, geometry, and winding number. We focus on magneto-transport studies of curved two-dimensional electron systems in such nanoscrolls. An externally applied magnetic field results in a strongly modulated normal-to-surface component leading to magnetic barriers, reflection of edge channels, and local spin currents. The observations are compared to finite-element calculations and discussed on the basis of simple models taking into account the influence of a locally modulated state density on the conductivity. In particular, it is shown that the observations in high magnetic fields can be well described considering the transport in edge channels according to the Landauer–Büttiker model if additional magnetic field induced channels aligned along magnetic barriers are accounted for.

2.1 Introduction

In everyday household, we find numerous useful devices and gadgets with functionality based on strain in bimetallic layers as, for instance, the bimetallic strip in thermometers in which the indicator is turned by the strain arising from the different thermal expansion coefficients of the strip layers. More recently, it occurred to experimentalists that strain engineering can also be employed for a bottom-up approach toward novel epitaxial semiconductor micro- or nanostructures. In contrast to conventional top-down approaches based on lithography at the surface of layered semiconductor or metal oxide semiconductor systems, here the spontaneous formation of nanostructures is utilized. Strain is the driving force in the Stranski

Krastanov [1] mode that leads to the formation of self-assembled quantum dots during the epitaxial growth of semiconductor heterolayers with a mismatch of the lattice constant on semiconductor surfaces. This is reviewed in chapter 1 of Heyn, Stemmann, and Hansen. Strain stored in pseudomorphic layers can be utilized to initiate a rolling-up process resulting in objects with a rolled-up carpet like shape. These so-called nanoscrolls will be the central subject of this article. Up to a critical thickness [2, 3], layers of lattice mismatched semiconductor compounds can be epitaxially grown in the Frank–van der Merwe mode without incorporation of lattice defects. Below the critical film thickness, the strain built in the film does not suffice for the formation of misfit dislocations. However, if the film consists of two pseudomorphic layers with different strain, it will roll up once it is released from the substrate. This technique was first introduced by Prinz et al. on InGaAs–GaAs [4] or Si–SiGe [5, 6] bilayer systems. Note that, depending on the length of the rolled-up film, a curved lamella or a nanoscroll with a certain number of turns and a spiral cross section will be obtained. This is in contrast to tubular systems forming cylinder barrels like, e.g., carbon nanotubes. The diameter of the thus formed scrolls is precisely determined by thickness and composition of the layers in the film. This bottom-up approach of nano fabrication of evenly curved films can be combined with top-down techniques that lithographically define the length of the rolled-up film, i.e., the number of windings in the scroll, and the way the scroll remains attached to the substrate, in particular its orientation and location.

A vast number of applications of curved semiconductor films has been envisioned by different authors and already realized in part. Free-standing Si–SiGe micro- and nano-objects like helical coils and vertical rings were fabricated [7]. In electromechanical devices, curved lamellae act as strain-engineered cantilevers for force measurements and displacement sensors [8]. Strained semiconductor films act as flexible hinges in dynamic mirror devices [9, 10]. Aside from mirrors, stress-actuated folding of hybrid material layers is suggested for the fabrication of folded electronic structures such as capacitors [11], compact induction coils, and optical resonators [12]. Particularly, appealing possibilities are offered by the coupling of optical or electrical properties to mechanical deformation of curved semiconductor lamellae or nanoscrolls. Strain in the wall of a semiconductor nanoscroll will modify optical as well as electric properties. To enhance sensitivity, a quantum well [13] or even quantum dots [14] can be implemented in the wall of the nanoscroll. The optical properties of the quantum well or the quantum dots will be controlled by mechanical deformation of nanobridges [15, 16] or microscroll optical resonators [17]. Furthermore, semiconductor microscrolls can operate as high-quality optical resonators with tailorable three-dimensional light confinement [18]. Hybrid permalloy/semiconductor microscrolls have been shown to confine spin waves [19], and multi-rotated Ag/semiconductor microscrolls are promising candidates for magnifying sub-wavelength lenses working in the visible [20]. Also, we note that nanofluidic applications of nanoscrolls have been established by Deneke et al. [21–23]. Nanopipelines, minuscule rockets [24], and syringe tips [25, 26] composed of semiconductor nanoscrolls have been proposed. A comprehensive review focusing on strained Si–SiGe films has been published by Scott and Lagally, recently [27].

Not only undoped quantum wells for optical experiments but also modulation doped wells containing a high-mobility electron system have been successfully fabricated. This offers the appealing possibility to study charge-carrier transport in a two-dimensional manifold curved in the three-dimensional space. Planar two-dimensional electron systems have been previously very successfully generated at the surface of liquid Helium, the interfaces in semiconductor heterostructures, or metal oxide semiconductor devices [28]. Such systems are intensively investigated for a long time and many intriguing properties such as the Quantum Hall [29] and the Fractional Quantum Hall effect (for a review see [30]) have been observed. In contrast to the mature methods developed for planar systems, it is experimentally much more difficult to fabricate a curved charge carrier film. Routes toward the creation of undulated two-dimensional electron systems are given, e.g., by waves on the liquid helium surface or overgrowth of patterned semiconductor surfaces [31, 32]. Soon after, it was shown that thin semiconductor films containing high-mobility electron systems can be fabricated [33]. A method to lift off such thin films from the substrate and transfer them to glass tubes with a few millimeter diameter was reported by Lorke et al. [34]. First evenly curved high-mobility electron systems with bending diameters of a few microns could be realized with the method of self-rolling strained layers [35–37].

Intriguing properties are predicted for charge carrier systems on curved manifolds by many theoretical works. A curvature dependent confinement potential of pure geometric origin is predicted by seminal works on quantum mechanics in constrained geometries ([38–41] and references therein). An additional potential term arises if at the curved interface, where the charge carrier resides, the dielectric constant changes appreciably [42]. However, in present nanoscrolls of several 100 nm curvature radii, these potentials are not significant, yet. Furthermore, in several publications, the role of spin-orbit interaction in curved two-dimensional carrier systems has been discussed. It has been pointed out that, unlike in a planar system, in ballistic systems of finite curvature the sign of the spin–orbit coupling constant can be determined experimentally [43–45]. Entin and Magarill [46] as well as Trushin et al. [47, 48] establish that the curvature introduces a further degree of freedom to manipulate the spin orientation in addition to the electric field control offered by the Rashba spin-orbit interaction with evident implications for spintronic applications.

In this chapter, we focus on charge carrier transport in curved two-dimensional systems exposed to a magnetic field. The effect of a magnetic field on the curved two-dimensional electron systems has been considered theoretically in many publications [42–45, 49–52]. In case the magnetic field is applied perpendicular to the axis of the cylinder, the most obvious effect is the modulation of the field component perpendicular to the electron system. Since the kinetics of a two-dimensional carrier system only reacts on the perpendicular magnetic field component, nanoscrolls with high-mobility two-dimensional electron systems are perfectly suited to study transport in strongly modulated magnetic fields. We can independently control the strength of the magnetic-field gradients as well as the spatial location of the gradient maxima during the experiment just by changing the magnetic field value and orientation. Strong spatial modulation of the magnetic field applied to

a two-dimensional electron system is expected to result in the formation of magnetic barriers [31, 53–55] deflecting the edge states that are responsible for current transport in the quantum Hall regime into the interior of the Hall bar. The thus created novel current carrying states are predicted to feature intriguing properties. The current running parallel to the axis of the nanoscroll is localized to stripes on its perimeter, and the current carrying states have a group velocity that depends on the location on the perimeter where they are localized [49, 56, 57]. In particular, at locations, where the perpendicular magnetic field component changes sign, in addition to the cycloid-like orbits, which are known from edge states in planar systems, so-called snake orbits with opposite drift velocity are expected to exist in the interior of the Hall bar [59–61]. Kleiner has even predicted localization of spin currents along the perimeter of the nanoscrolls which would be of high interest for spintronic applications [57]. A recent review on electron systems in inhomogeneous magnetic fields has been published by Nogaret [58].

With the present curvature radii of nanoscrolls [35–37, 62–64] containing two-dimensional electron systems, the effect of magnetic barriers in magnetic-field gradients seems to be the predominant feature observed in the experiments. We review some of corresponding experiments discussing the transition from the classical regime to the quantum Hall regime [37, 64, 67]. It turns out that the experimental data so far can be well understood within a modified Landauer–Büttiker model taking into account states moving along magnetic barriers. In Sect. 2.2, we will introduce the experimental techniques employed for the fabrication of two-dimensional charge carrier systems in nanoscrolls. The first evidence of rolled-up 2DES in freestanding curved lamellae will be presented in Sect. 2.3. Subsequently, results concerning rolled-up Hall bars in the low and high magnetic field regime will be shown in Sect. 2.4. In this context, the static skin effect and reflected edge channels will be discussed, followed by a conclusion in Sect. 2.5.

2.2 The Basic Principle Behind “Rolled-Up Nanotech”

The principle of rolling-up semiconductor heterostructures was first presented by Prinz et al. for Si-SiGe systems [5, 6] and InGaAs-GaAs heterojunctions [4]. Figure 2.1 shows schematically the basic principle for an InGaAs-GaAs heterostructure. On a GaAs substrate with a lattice constant a_1 a sacrificial layer of AlAs with nearly the same lattice constant a_1 and a pseudomorphically strained InGaAs layer with a lattice constant $a_2 > a_1$ are grown. Above this structure a GaAs layer with lattice constant a_1 is deposited. Selective etching of the AlAs sacrificial layer detaches the strained bilayer system from the substrate. As sketched in Fig. 2.1, the strain results in a torque that bends the film with a radius of curvature r . The In content as well as the thickness of the InGaAs layer are chosen to lie below the critical values for strain relaxation caused by the generation of misfit dislocations [2]. Another constriction for the choice of the In content is the formation of 3D islands, which occur beyond a critical thickness when the lattice mismatch exceeds about

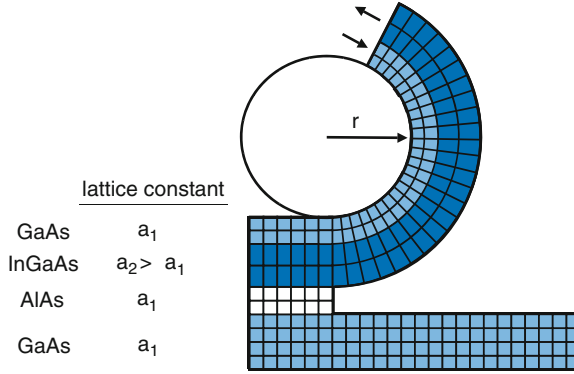


Fig. 2.1 Rolling-up mechanism. An AlAs sacrificial layer (white) with lattice constant a_1 and a pseudomorphically strained InGaAs layer (dark blue) with a larger lattice constant $a_2 > a_1$ are grown on a GaAs substrate (light blue) with lattice constant a_1 . Above this structure, there is another GaAs layer with the same lattice constant a_1 . Due to the torque, the film bends with a bending radius r once it is detached from the substrate by selectively etching the AlAs sacrificial layer

2.7% [3]. The topmost GaAs layer, which is lattice-matched to the substrate can be replaced by a more complex heterostructure layer sequence containing, e.g., a modulation-doped AlGaAs-GaAs quantum well with a 2DES.

The radius of nano- and microscrolls depends on the thicknesses of the different layers in the wall of the scrolls as well as their respective modules of elasticity and lattice constants. For a quantitative prediction, two slightly different models are used. Many authors apply a model of Tsui and Clyne developed for the prediction of the bending of an amorphous metal bilayer [68]. Tsui and Clyne consider both the strain caused by different thermal contraction of the multilayer materials and the intrinsic stresses caused by the deposition process. If we apply this analytical model to the strain caused by the different lattice constants of the InGaAs stressor layer and the AlGaAs-GaAs layers, we are able to derive an equation for the radius of a scroll that depends on the Poisson ratio ν , the lattice mismatch $\Delta a/a$, the thicknesses $d_{1,2}$, and the Young moduli $E_{1,2}$ of these different layers:

$$\frac{1}{r} = \frac{6(1-\nu)E_2E_1d_2d_1(d_2+d_1)(\Delta a/a)}{E_2^2d_2^4 + 4E_2E_1d_2^3d_1 + 6E_2E_1d_2^2d_1^2 + 4E_2E_1d_2d_1^3 + E_1^2d_1^4}$$

For a rough estimate assuming equal thicknesses d of both layers and a relative lattice mismatch $\Delta a/a$, one gets $r \sim d/(\Delta a/a)$. While the model of Tsui and Clyne yields already quite good quantitative estimates of the experimental radii in semiconductor nanoscrolls [69, 70], it does not provide a statement about the preferred rolling directions of epitaxial layer systems. Grundmann calculates the scroll radius by determining the minimum of the strain energy in single-crystalline semiconductor bilayer systems with respect to the bending radius r [71]. He takes into account the cubic symmetry of zinc-blende semiconductors and predicts that

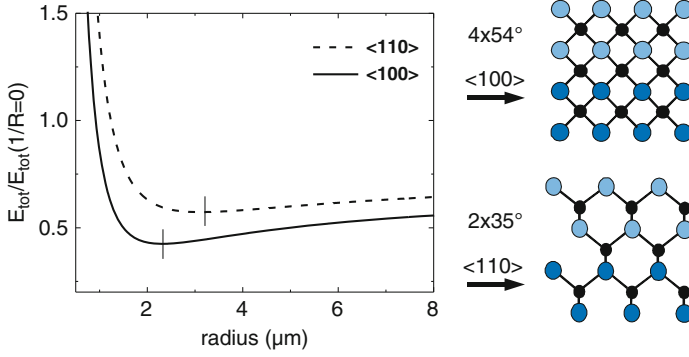


Fig. 2.2 Strain energy normalized on the strain energy of the flat structure plotted against the radius of a scroll for two crystal directions. The minima of the strain energies indicate the radius for the respective rolling direction and are marked by vertical lines. On the right, a strained InAs-GaAs structure is shown for the $\langle 100 \rangle$ and the $\langle 110 \rangle$ direction. Due to the covalent bonding, the $\langle 100 \rangle$ direction is the hardest crystal direction and the strain energy decreases the most

the scroll radius is dependent on the rolling direction. To determine the preferred rolling direction, we calculate the strain energy depending on the radius of a scroll like described by Grundmann. In Fig. 2.2, the strain energy normalized to the strain energy of the flat structure is plotted against the radius for the $\langle 100 \rangle$ and $\langle 110 \rangle$ rolling directions for a bilayer system consisting of a 30 nm $\text{In}_{18}\text{Ga}_{82}\text{As}$ layer and a 30 nm GaAs layer. Obviously, the strain energy and the radius are smallest for the $\langle 100 \rangle$ rolling directions, i.e., the $\langle 100 \rangle$ directions are the preferred rolling directions. This can be explained by the number of covalent bondings acting in the corresponding crystal directions. As sketched in Fig. 2.2, in zinc-blende type semiconductors, each crystal atom has four bonds, which are aligned along the $\langle 111 \rangle$ directions. Correspondingly, only two bonds act along a $\langle 110 \rangle$ type crystal direction with a $\cos 35^\circ$ component. In contrast, in the $\langle 100 \rangle$ directions four bonds are acting at an angle of 54° . For this reason the crystals tend to cleave along the weak $\langle 110 \rangle$ directions. On the other hand, strain relaxation along the hardest $\langle 100 \rangle$ directions reduces the strain energy the most, which makes plausible that the preferred rolling directions of zinc-blende type semiconductors are the $\langle 100 \rangle$ directions.

The preparation of semiconductor nano- and microscrolls comprises alternating lithography, wet etching, and metallization steps [35, 76]. It enables us, e.g., to fabricate rolled-up Hall bars [37] and other structures for magneto-transport measurements. A basic sample structure grown via molecular beam epitaxy (MBE) is shown in Fig. 2.3a. It consists of a GaAs substrate, an AlAs sacrificial layer, a pseudomorphically strained InGaAs layer, and a GaAs top layer. The process starts with a shallow wet etching step using a solution of phosphoric acid, hydrogen peroxide, and ultra-pure water ($\text{H}_3\text{PO}_4:\text{H}_2\text{O}_2:\text{H}_2\text{O}$, 1:10:500) in order to define a mesa that in the end will be rolled up to form the scroll (Fig. 2.3b). For a precise definition of the scroll geometry, it is important that the wet etching stops within the InGaAs layer to

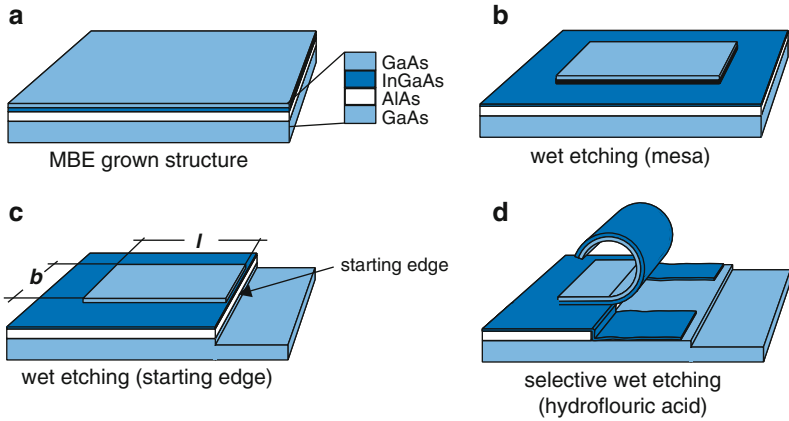


Fig. 2.3 Two-step lithography for the fabrication of well-defined rolled-up 3D objects. (a) Sample structure consisting of a GaAs substrate, an AlAs sacrificial layer, a pseudomorphically strained InGaAs layer, and a GaAs top layer, (b) step 1: shallow wet etching down to the center of the InGaAs layer, (c) step 2: deep wet etching of the starting edge, which selects one of the preferred rolling directions, (d) step 3: selective wet etching of the AlAs sacrificial layer with hydrofluoric acid resulting in a bending of the structure [72]

avoid that the hydrofluoric acid (HF) accesses the AlAs layer through unintentional holes in the InGaAs layer during the last preparation step. On the other hand, the InGaAs layer has to be thin enough to rip when the mesa rolls up. Furthermore, a starting edge for the rolling-up process has to be defined. For this, deep trenches are etched at the starting edge of the mesa by a further wet etching process as sketched in Fig. 2.3c. The starting edge determines precisely the beginning of the rolling-up and selects one of the four preferred $\langle 100 \rangle$ rolling directions. In the last step (Fig. 2.3d), the AlAs sacrificial layer is selectively etched with diluted HF (5%) and the mesa starts to bend. HF has a high selectivity ($>10^6$) for materials with an aluminum content higher than 40% like the AlAs layer used in our sample structure [77, 78]. The number of windings depends on the time the sample is exposed to the etchant and on the length of the mesa.

A virtually unlimited number of curved 3D structures exists that can be realized with the rolling-up principle in combination with lithographic methods. Figure 2.4a shows InGaAs-GaAs scrolls with exactly predetermined length, number of rotations and location on the sample surface fabricated using the two-step lithography described above. On basis of this two-step lithography, even more complex 3D objects as shown in Fig. 2.4b–f were developed by us. The helical solenoid in Fig. 2.4b was fabricated by rolling-up a rectangular mesa which was on purpose misaligned with respect to the preferred $\langle 100 \rangle$ rolling direction. The distance between two windings can exactly be tailored by the dimensions of the mesa and the angle between the mesa edges and the preferred rolling direction. Thus with starting edges misaligned with respect to the $\langle 100 \rangle$ directions the preferred $\langle 100 \rangle$ rolling directions can be proven experimentally [79]. Figure 2.4c shows a single-walled tube

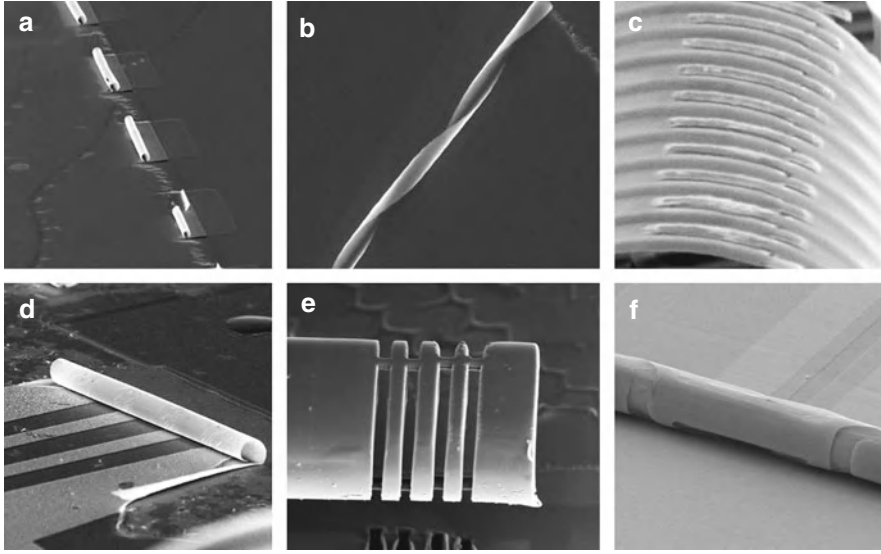


Fig. 2.4 Various curved 3D nanostructures: (a) InGaAs-GaAs scrolls prepared with two-step lithography [72], (b) solenoid with predetermined distance of windings and a radius of $2.2\ \mu\text{m}$ [72], (c) single-walled microtube closed with a tine system similar to an insect-eating plant [73], (d) suspended microscroll with four contact leads visible as light grey stripes on the planar substrate [35], (e) suspended scroll with focused ion beam (FIB) milled rings, (f) curved 2DES in van der Pauw geometry rolled-up with a template scroll [75]

closed by a system of tines which was prepared into two opposite starting edges. During underetching, this structure rolled up from these opposite starting edges and was closed similar to an insect-eating plant [73]. Figure 2.4d shows a tube, which is suspended on four metallized bearings. The fabrication of such suspended tubes was developed by us for the first measurements on rolled-up two-dimensional systems [35] and was also a key prerequisite for the first realization of rolled-up optical resonators [12]. On the other hand, rolling-up metal/semiconductor layers as in the metallized bearings of the suspended tube shown in Fig. 2.4d is a promising method to fabricate tubular shaped metamaterial lenses with sub-wavelength resolution, so called hyperlenses [20]. Figure 2.4e shows a tube which is patterned into three suspended rings by focused ion beams (FIB) at one end. These rings might act, e.g., as coupled optical ring resonators provided ion damaging during FIB preparation can be kept at a minimum to preserve good optical properties. The most sophisticated structure is shown in Fig. 2.4f: A van der Pauw rectangle containing a high-quality, two-dimensional electron system and four metallized contacts was rolled-up into a curved shape with the help of a semiconductor template tube similar to aluminum foil which is rolled-up on a cardboard roll. This template tube concept enabled us, e.g., to fabricate rolled-up gated Hall bars [37].

2.3 First Evidence of Rolled-up 2DES in Freestanding Curved Lamellae

To prepare rolled-up 2DES, specific InGaAs-GaAs heterostructures are grown using MBE. The layer sequence of a typical sample is presented in Fig. 2.5. On a GaAs substrate with an AlAs sacrificial layer, the actual heterostructure which constitutes the wall of the scroll is grown. It consists of a pseudomorphically strained $\text{In}_{18}\text{Ga}_{82}\text{As}$ stressor layer, $\text{Al}_{33}\text{Ga}_{67}\text{As}$ barrier layers, a GaAs quantum well and a GaAs cap layer. The $\text{Al}_{33}\text{Ga}_{67}\text{As}$ layers contain high Si delta dopings which provide the electrons confined in the quantum well and saturate surface states at the front and back side of the lamella. An In content of 18% and a thickness of 20 nm have been chosen for the stressor layer. These values guarantee that the strained film is not relaxed by generation of misfit dislocations.

First, evenly curved semiconductor lamellae containing two-dimensional electron systems were prepared by Lorke et al. [34] with an epitaxial lift-off process and subsequent lamella deposition on a fine glass rod. While this method is restricted to bending radii of the order of 1 mm and larger, rolled-up nanotech enabled the fabrication of 2DES with bending radii of a few microns. The first rolled-up structures containing a 2DES were suspended lamellae with a simple contact geometry and current direction along the axis of the lamella, i.e., perpendicular to the modulation of the magnetic field [35].

In Fig. 2.6, scanning electron microscopy images and transport data of two nano-scroll samples with 8 μm radius are depicted. The lamellae were fabricated from a heterostructure shown in Fig. 2.5 in a preparation process described in Sect. 2.2. In the sample of Fig. 2.6a and b, the curved two-dimensional electron system under investigation covers $0.14 \cdot 2\pi$, i.e., 14% of the scroll circumference. The lamella,

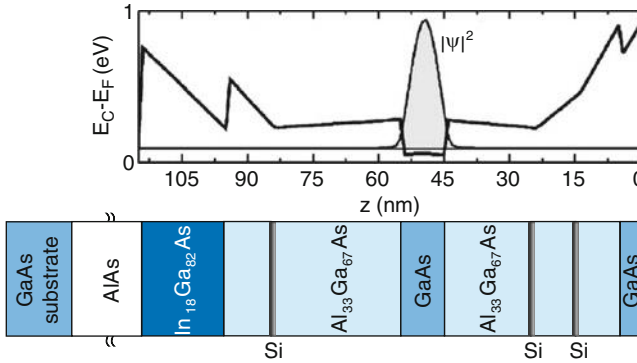


Fig. 2.5 Typical sample structure (*bottom*) and respective potential distribution (*top*). The heterostructure forming the wall of the scroll is grown on a GaAs substrate with an AlAs sacrificial layer. It consists of the pseudomorphically strained $\text{In}_{18}\text{Ga}_{82}\text{As}$ layer, $\text{Al}_{33}\text{Ga}_{67}\text{As}$ layers containing Si delta dopings, the GaAs quantum well, and a GaAs cap layer. The potential distribution for the detached structure and the associated free electron density are presented above the sample structure

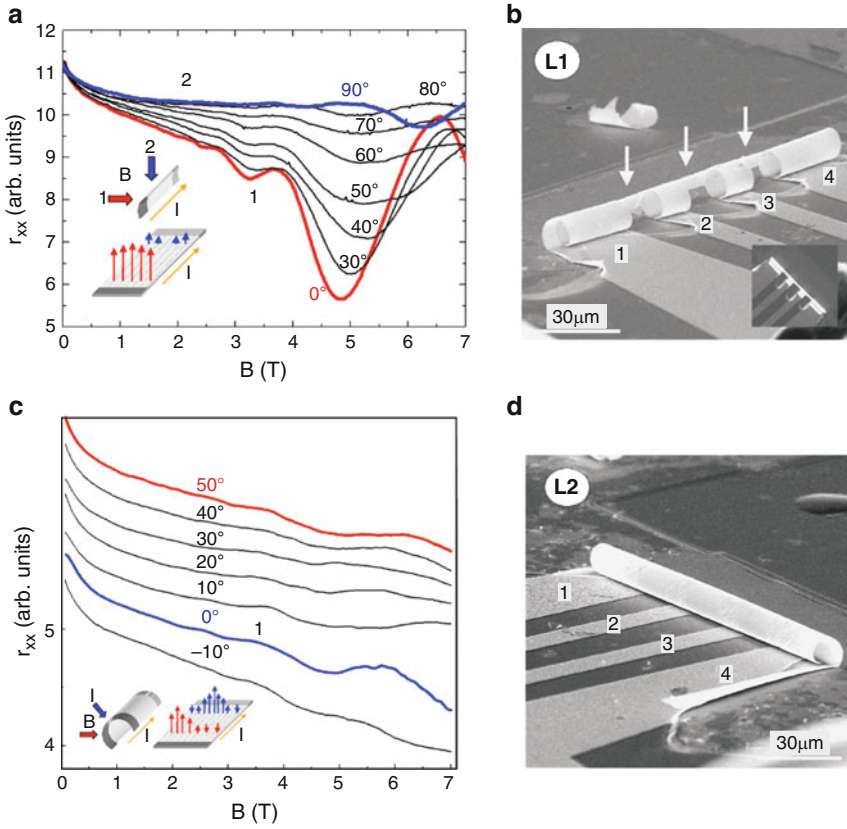


Fig. 2.6 Scanning electron microscopy images and magnetoresistance of two samples for four-probe measurements on evenly curved two-dimensional systems spanning an arc of $0.14 \cdot 2\pi$ (a,b) and $0.6 \cdot 2\pi$ (c,d), i.e., the electron system under investigation covers 14% and 60% of the scroll circumference, respectively. The scroll radius is $8\mu\text{m}$ in both samples. The white arrows in (b) indicate the position of the lamella. The numbers denote the four Gold leads connecting the annealed AuGe/Ni/AuGe contacts on the lamella with the outside world. (a), (c) Measurements of the lamellae for different angles of rotation. The insets show the respective orientation of the lamella relative to the perpendicular component of the external magnetic field. In (c), curves are offset by a constant value for visibility

indicated in Fig. 2.6b by white arrows, is suspended between four arcs, which span a complete scroll revolution and serve as contacts. The lamella is oriented perpendicular to the substrate implying that the signals of the lamella have a maximum when the substrate is parallel to the external magnetic field, i.e., when the maximum of the modulated magnetic field is perpendicular to the center of the lamella which can be described by $B = B_0 \cdot \cos(y/r)$ with $y = 0$ at the symmetry axis of the lamella. Ohmic contacts to the 2DES are provided by four annealed AuGe/Ni/AuGe contacts located on the lamella which are connected to bond pads on the substrate via gold leads evaporated on the sample surface and denoted in Fig. 2.6b and d by numbers. By positioning the annealed contacts on the lamella,

we avoid that resistance changes in the two-dimensional electron system of the contact arcs influence the data. Additionally, in this way, we prevent parallel conduction through another 2DES located at the InGaAs-AlGaAs interface in the undetached part of the heterostructure. While for two-point contact measurements these precautions are obviously indispensable to avoid misinterpretations [36], Vorob'ev et al. argue that four-point measurements can be conducted without rolled-in metal contacts [62]. In general, they anneal ohmic contacts in the flat, undetached areas of the heterostructures. The 2DES in this area, in turn, provides contacts to the curved 2DES in the lamella. Vorob'ev et al. assume that curved 2DES can be used for the current and voltage leads to the Hall bar on the scroll, because even in the curved part of the leads at least one edge channel will maintain connection between Hall bar and ohmic contact and scattering processes among the edge channels as well as passages through zero normal-field regions would effectively bring all edge channels to an equilibrium chemical potential [62].

The four-point magneto-transport measurements depicted in Fig. 2.6 were performed with standard lock-in technique by feeding an AC-current of 10 nA along the axis of the curved lamellae through lead 1 and 4 and measuring the voltage between contact 2 and 3 in a standard liquid Helium bath cryostat at $T = 4.2$ K and at magnetic fields up to $B = 7$ T. The sample was mounted on a rotatable holder allowing for different orientations of the magnetic field as sketched in the insets of Fig. 2.6a and c. In Fig. 2.6a, lamella 1 shows clear Shubnikov–de Haas oscillations, i.e., oscillations in the longitudinal resistance with a periodicity in $1/B$ [80], when the maximum of the perpendicular component of the modulated magnetic field $B_{\max}$ is located at the center of the lamella as sketched in the inset in Fig. 2.6a. Presuming that the resistance minima are shifted insignificantly against the minima in a flat 2DES, the charge carrier density can be estimated to $n = 4.5 \cdot 10^{11} \text{ cm}^{-2}$ with an electron mobility of $\mu = 7,000 \text{ cm}^2 (\text{V s})^{-1}$. On the other hand, lamella 1 exhibits just weak Shubnikov–de Haas oscillations when the modulated magnetic field is parallel to the surface of the lamella. The amplitude strongly decreases and the minima of the Shubnikov–de Haas oscillations shift to higher magnetic fields when the lamella is tilted out of the symmetric orientation. A different result is presented in Fig. 2.6c. Lamella 2 covering $0.6 \cdot 2\pi$, i.e., 60% of a complete revolution, shows only weak Shubnikov–de Haas oscillations in all applied angles of rotation with no clear periodicity in $1/B$. We assume that the charge-carrier density and the mobility are similar to the values estimated for lamella 1. The different behavior of lamella 1 and 2 is attributed to the different widths b of the lamellae compared to the circumference U of the scroll which corresponds to different sections of the sinusoidally modulated perpendicular component of the magnetic field. Lamella 1 covers only 14% of a complete tube and resembles an almost flat suspended stripe with nearly constant magnetic field (see inset in Fig. 2.6a). Therefore, Shubnikov–de Haas oscillations are clearly visible when the perpendicular component of the magnetic field is at maximum at the center of the lamella and decreases when the sample is tilted away from this configuration. The shift of the minima that occurs when the lamella is rotated in the magnetic field, however, cannot be explained by a simple sinus law like in the case of planar 2DES. In lamella 2 which covers 60% of a complete tube,

the perpendicular component of the magnetic field is strongly modulated and the oscillations wash out. These observations, which represent indeed the first conclusive proof for rolled-up 2DES [35], can be explained using a model for the averaged density of states at the Fermi energy which will be discussed in the following.

In a planar 2DES subjected to a homogenous magnetic field, the electron states condense on Landau levels with energy separation $\hbar\omega_c$. Taking into account the broadening Γ of Landau levels caused by scattering processes, the density of states can be approximated by Gaussians [28]

$$D_{B \neq 0}(E) = N_L \sum_n \frac{1}{\sqrt{2\pi}\Gamma^2} \cdot \exp\left[-\frac{1}{2} \left(\frac{E - E_n}{\Gamma}\right)^2\right],$$

where N_L is the Landau level degeneracy. The dashed curve in Fig. 2.7d represents the averaged density of states $D_{B \neq 0}(E)$ for a planar 2DES plotted over the energy in units of $E_F(B=0)$ for a magnetic field with $\frac{\hbar\omega_c}{E_F} = 0.25$. Figure 2.7a shows the oscillations of the Fermi energy E_F , also presented by a dashed line, due to the condensation of the states on Landau levels with increasing magnetic field. The Shubnikov–de Haas oscillations in the longitudinal conductivity σ_{xx} (Fig. 2.7 (c)) are obtained from the following relation between the longitudinal conductivity and the density of states at the Fermi energy

$$\sigma_{xx} = e^2 D_{B \neq 0}(E_F) D_D$$

where $D_D = \frac{1}{2}v_F^2\tau$ is the diffusion constant comprising the averaged scattering time τ and the Fermi velocity v_F at $B = 0$. To transfer these results from planar to curved 2DES, we have to take the impact of the modulated magnetic field into account. Assuming a purely two-dimensional electron system, we neglect the magnetic-field component oriented parallel to the electron plane and model our system with a planar two-dimensional electron system in a perpendicular magnetic field with sinusoidal modulation. The energy spectrum of such a system was calculated in [34, 43] and exhibits in the limit $l_B \ll r$ a sinusoidal modulation of the energy separation as well as the degeneracy of the Landau levels. A corresponding density plot is shown in Fig. 2.7b. To determine the Fermi energy as a function of the magnetic field, we have to summarize the varying number of states that can be occupied and equalize this result with the number of available charge carriers. In thermodynamical equilibrium, i.e., with a Fermi energy being constant over the whole sample, we obtain $E_F(B)$ from

$$\int_0^{E_F} D^*(B, E) dE = N_S$$

where the averaged density of states $D^*(B, E)$ is calculated from the local density of states $D(B, E, x)$

$$D^*(B, E) = \frac{1}{b} \int_{-\frac{b}{2}}^{\frac{b}{2}} D(B, E, x) dx.$$

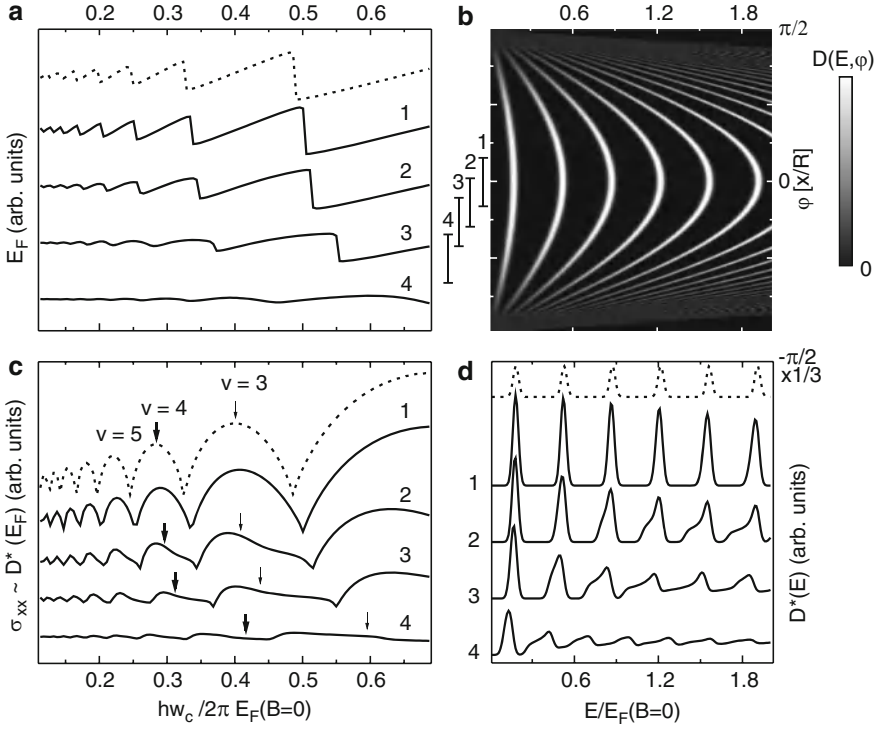


Fig. 2.7 (a) Calculated Fermi energy E_F , (b) local density of states $D(E, \varphi, B)$, (c) longitudinal conductivity σ_{xx} , and (d) averaged density of states $D^*(E, B)$. The broadening of the Landau levels caused by scattering and the magnetic field in (b) and (d) are assumed to be $\Gamma = 0.02 E_F$ and $\frac{\hbar\omega_c}{E_F} = 0.25$, respectively. The *dashed lines* in (a), (c), and (d) represent a flat 2DES and all *curves* are offset by a constant value for visibility. The graphs 1 to 4 belong to a lamella with a constant width $b = 0.08 U$ and different orientations indicated in (b). The *dashed line* in (d) is $D^*(E, B)$ divided by 3 [72]

Here, we assume that the charge carrier density N_S does not change across the lamella, which is expected to be a good approximation, if the width of the Landau levels Γ is not much smaller than the Landau energy $\hbar\omega_c$. A numerical model has been presented recently in [64]. For samples with a very small width b in comparison to the circumference U of a scroll, i.e., $b \ll U$, the lamella has an averaged density of states, Fermi energy, and conductivity similar to a planar 2DES. For $b \sim U$, however, $D^*(B, E)$ is considerably different from the graph of a planar 2DES (cf. Fig. 2.8). In Fig. 2.7d, the averaged density of states is presented for a sample with $b/r = 0.08 \cdot 2\pi$ and four different orientations of the sample in the magnetic field, i.e., different phases of the modulated perpendicular component of the magnetic field. The orientations 1–4 are indicated by lines marking the width of the lamella in Fig. 2.7b. Figure 2.7b shows the local density of states which depends on the absolute value and the phase of the magnetic field in a grey-scale plot. For

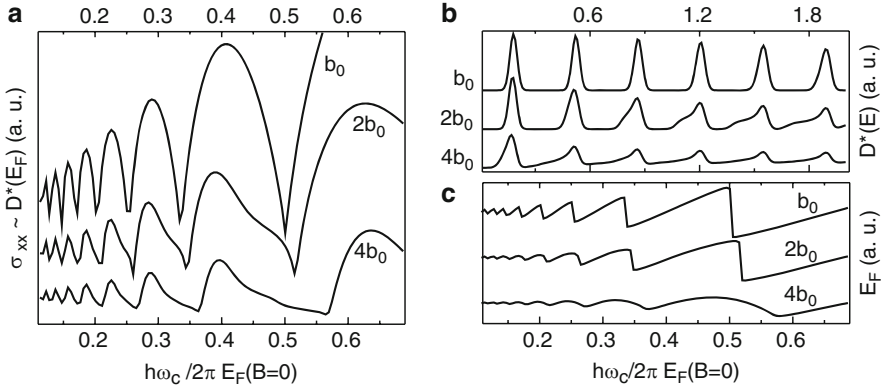


Fig. 2.8 (a) Calculated longitudinal conductivity σ_{xx} , (b) averaged density of states $D^*(E, B)$, and (c) Fermi energy E_F . The graphs belong to lamellae with different widths $b = b_0 = 0.08 U$, $b = 2b_0$ and $b = 4b_0$ and constant symmetric orientation (cf. orientation 1 in Fig. 2.7b). The curves are offset by a constant value for visibility [72]

the symmetric orientation 1, the peaks of the according curve of the averaged density of states $D^*(B, E)$ broaden with increasing energy, because the curvature of the Landau levels increases with the quantum number of the Landau levels. The figure makes clear that the attenuation of the Shubnikov–de Haas oscillations as well as the shift of the minima to higher magnetic fields are attributed to the fact that for a curved 2DES the integral over the broadened peaks of $D^*(B, E)$ is smaller than the according integral of a planar 2DES caused by the lower averaged degeneracy of the curved Landau levels. Tilting the sample out of the symmetric position amplifies this effect (cf. orientation 2–4 in Fig. 2.7b). Furthermore, the broadening of the peaks in the averaged density of states is reflected in the deformation of the curves for the longitudinal conductivity. Both the damping of the Shubnikov–de Haas oscillations and the shift to higher magnetic fields are clearly observed in the experimental curves shown in Fig. 2.6a. Note that the shift to higher magnetic fields is larger than expected for a flat 2DES which is tilted in a magnetic field.

The difference between the magneto-transport measurements in Fig. 2.6a and c can be qualitatively explained by the calculations presented in Fig. 2.8 showing the longitudinal conductivity, the averaged density of states, and the Fermi energy for lamellae with different widths $b = b_0 = 0.08 \cdot 2\pi r$, $b = 2b_0$, and $b = 4b_0$. The orientation is fixed with maximum of the perpendicular magnetic field component at the center of the lamella. Again, the lower averaged degeneracy of the Landau levels and the according smaller integral over $D^*(B, E)$ are reflected in the shift of the minima and the attenuation of the Shubnikov–de Haas oscillations. This effect becomes stronger with increasing width of the lamellae. The weak dependence on the magnetic field orientation of very broad lamellae as lamella 2 is also in accordance with our model (calculations not shown).

2.4 2DES in Rolled-Up Hall Bars: Static Skin Effect, Magnetic Barriers, and Reflected Edge Channels

To further investigate the magneto-transport in evenly curved 2DES, we developed a method to prepare rolled-up Hall bars [37]. Obviously, the Hall bar geometry provides some advantages compared to the simple geometry described in Sect. 2.3, e.g., the exclusion of possible artefacts in the signal caused by current driven through the voltage probes or the comparability with measurements in which the modulation of the magnetic field is realized by other methods [31–34]. In particular, this geometry enables us to measure the Hall resistance in a curved 2DES. Furthermore, the current direction will not be restricted to the axis of a scroll anymore.

This chapter focuses on magneto-transport measurements on evenly curved Hall bars running along the curvature of the microscroll, i.e., the current is driven along the modulation of the perpendicular magnetic field component. The corresponding setup is sketched in Fig. 2.9a. The phase δ of the perpendicular magnetic field component can be tuned in the experiment simply by rotating the rolled-up Hall bar around its axis. For the preparation of such systems, we again adopted the two-step lithography described in Sect. 2.2: In a first step, a gated Hall bar geometry as sketched in Fig. 2.9b is defined by shallow etching. For the reasons discussed in the previous section AuGe/Ni/AuGe contacts are prepared in the direct vicinity of the Hall bar and connected to the outside world by gold leads. Furthermore, the contacts overlap the mesa to avoid Corbino-related effects as illustrated by the circular zoom-in in Fig. 2.9b. In a second deep mesa etching step, the starting edge is defined at the position indicated by the dashed line in Fig. 2.9b. To avoid cracking of the delicate Hall-bar structure, the first winding of the scroll, corresponding to a

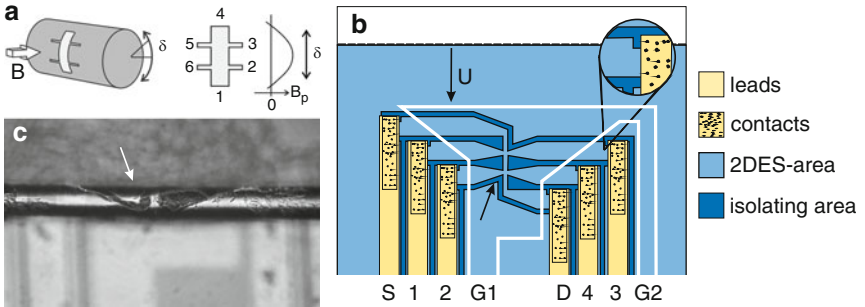


Fig. 2.9 (a) Sketch of an evenly curved Hall bar (ECHB) oriented along the curvature of a scroll in a homogenous magnetic field. Accordingly, the modulation of the external magnetic field is parallel to the current direction in the Hall bar. (b) Diagram of the ECHB in the planar state. The etched trenches (dark blue) define the mesa (blue) consisting of the Hall bar, the annealed contacts (dotted area) and the leads (yellow). The inset in (b) shows that the contacts overlap the mesa in order to avoid Corbino-related effects. The structure begins to roll up at the starting edge (dashed line) in direction of the arrow. (c) Microscopic image of the ECHB

rolled-up length U , remains unstructured and acts as a rolling template in the final selective etching step. As mentioned before, this rolling template concept resembles a cardboard tube which is used to roll up aluminum foil. In Fig. 2.9c, a microscopic image of the ECHB is presented. The white arrow indicates the position of a part of the mesa in the rolled-up structure which is marked in Fig. 2.9b by a black arrow.

2.4.1 Low Magnetic Field Regime: Static Skin Effect and Magnetic Barriers

The solid curves in Fig. 2.10b show magneto-transport measurements on a typical evenly curved Hall bar running along the curvature of the microscroll with a bending radius of $8\text{ }\mu\text{m}$, a Hall bar width of $6\text{ }\mu\text{m}$, and a voltage-probe distance of $12\text{ }\mu\text{m}$. Measurements were performed at 4.2 K with standard lock-in technique driving an AC current of 10 nA . In the symmetric configuration (s) with rotation angle $\delta = 0^\circ$, the maximum of the perpendicular field component modulation is located exactly between the voltage probes as shown in Fig. 2.9a. Due to the fact that magnetic field inversion corresponds to a permutation of source, drain and voltage probes ($1 \leftrightarrow 4$, $2 \leftrightarrow 6$, $3 \leftrightarrow 5$, cf. Fig. 2.9a) [65,66] we expect symmetric magnetoresistance curves

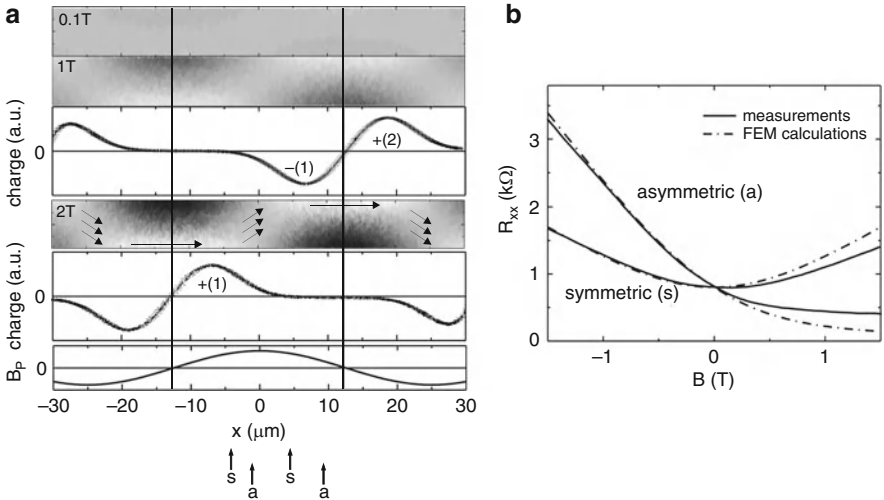


Fig. 2.10 (a) Grey-scale plots of the normalized current density J/J_0 for $B = 0$ calculated by FEM for a Hall bar subjected to a sinusoidally modulated magnetic field with an amplitude of 0.1 T , 1 T , and 2 T . The according charge distributions are presented on the *top* and *bottom* of the 2 T current-density plot, respectively. The *arrows* below the diagram indicate the voltage probe position for symmetric (s) and asymmetric (a) probe configurations. (b) Magnetoresistance measurements of a rolled-up Hall bar (*solid curve*) and FEM calculations (*dashed-dotted curve*) for configuration s and a

for configurations which are not affected by this permutation. Indeed, as shown in Fig. 2.10b, in the symmetric configuration (s) with $\delta = 0^\circ$ the magnetoresistance curve is almost symmetric with respect to $B = 0$. Consistently, we obtain asymmetric curves as soon as the maximum of the perpendicular magnetic field component is shifted from the center between the voltage probes, i.e., the Hall bar is rotated from the symmetric configuration and $\delta \neq 0^\circ$. As an example for such an asymmetric configuration (a), we plotted a curve for $\delta = 40^\circ$, i.e., with modulation maximum shifted by $\frac{2\pi r}{360} \cdot \delta = 5.6 \mu\text{m}$ from the center of the voltage probes. We note that in accordance with the Onsager–Casimir relations [65,66], we also find that the voltage probe permutation $2 \leftrightarrow 6, 3 \leftrightarrow 5$ corresponds to a magnetic field inversion (data not shown).

To understand not only the magnetic field inversion symmetry but also the shape of our curves, we described the magneto-transport in our ECHBs in a first approximation with a classical model [81, 82]: The finite-element method (FEM) is employed to locally solve a Laplace type equation and obtain the spatial distribution of electrostatic potential and current. The dashed-dotted lines in Fig. 2.10b correspond to calculations for an ECHB with the geometric parameters given above, a carrier density of $n = 7.0 \times 10^{11} \text{ cm}^{-2}$ and a mobility of $70,000 \text{ cm}^2 (\text{V s})^{-1}$. These values for carrier density and mobility are typical for our structures.

The underlying physical mechanism which dominates transport in the classical low-field regime is revealed in the corresponding density plots of the normalized current density J/J_0 in Fig. 2.10a. The black arrows indicate the local current direction. The positions of the voltage probes in the symmetric and asymmetric configuration are indicated in the bottom of Fig. 2.10a with s and a for the symmetric and antisymmetric situation, respectively. With magnetic field increasing from 0.1 to 2 T, a meander-shaped pattern evolves, which strongly confines the current to the Hall-bar edge at the zero crossings of the perpendicular magnetic field component. On the other hand, the current path crosses the sample when the modulated magnetic field goes through maxima. This somewhat surprising behavior can be understood if the steady-state carrier distribution at the edges of the Hall bar is taken into account. Charge-carrier densities at the top and bottom edge of the Hall bar are indicated on top and bottom of the 2 T current–density plot in Fig. 2.10a, respectively. In a classical picture, the motion of the electrons is determined by the sum of Lorentz force due to the modulated magnetic field and the electrostatic force due to the carriers at the edges of the Hall bar. While Lorentz force and electrostatic force exactly cancel each other in the steady state for a flat Hall bar the situation is different here. In the areas of high magnetic fields, the Lorentz force dominates and pushes electrons to the upper or lower Hall bar edge depending on the respective sign of the magnetic field. The electrostatic force due to this carrier accumulation at the edges (cf. $+(1)$ and $-(1)$ in Fig. 2.10a) counteracts the Lorentz force, but does not cancel it resulting in a steady state current which runs across the Hall bar. In contrast to this, at the zero crossings of the magnetic field modulation electrons are driven along the Hall bar edges solely by the electrostatic force induced by the carrier accumulations of the last and the next field extremum, e.g., $-(1)$ and $+(2)$. As a result, a meander-shaped

pattern evolves because electrons are driven to opposite Hall-bar edges by magnetic field extrema with opposite sign.

The exponential drop of current density from the Hall-bar edge into the bulk at the zero crossings, demonstrated by numerical calculations and experimentally here [37], was predicted in an analytical model by Chaplik et al. [67]. Since the exponentially decreasing current densities at the Hall-bar edges resemble the skin effect known for AC electric fields at metal surfaces, it has been named the ‘static skin effect’. The crossing of current at the modulation maxima from one edge of the Hall bar to the other has been observed before, e.g., by Leadbeater et al. and is referred to as ‘magnetic barriers’ [31, 32]. While the above classical model well describes the magneto-resistive behavior of our ECHB for the low magnetic field regime, qualitative deviations appear for higher magnetic fields, which are discussed below.

2.4.2 High Magnetic Field Regime: Reflected Edge Channels

The solid lines in Fig. 2.11a–c show the longitudinal magnetoresistance measured in the high magnetic field regime on an ECHB oriented along the curvature of a micro-scroll. The Hall bar width (6 μm) as well as the voltage-probe distance (12 μm) are identical with those of the ECHB discussed above. The bending radius of 9.5 μm is slightly larger here. As above, the Hall bar orientation in the magnetic field is described by the parameter δ , with $\delta = 0^\circ$ (Fig. 2.11a) corresponding to the symmetric configuration, i.e., magnetic field maximum exactly between the voltage probes. At $\delta \approx \pm 33^\circ$ (Fig. 2.11b, c), the situation is asymmetric with field maximum shifted by $\frac{2\pi r}{360} \cdot \delta = \pm 5.6 \mu\text{m}$ from the center of the voltage probes. Also for high magnetic fields, where deviations from the classical behavior appear, the Onsager–Casimir symmetry considerations discussed above are met. Interestingly, the asymmetric longitudinal resistance curves in Figs. 2.11b, c exhibit a shape resembling Shubnikov-de Haas (SdH) oscillations [80] for one magnetic field orientation and Quantum Hall resistance curves [29] for the other field orientation. In the symmetric case ($\delta = 0^\circ$), the longitudinal magnetoresistance curves seem to resemble SdH oscillations with a pronounced positive magneto-resistive background for both magnetic field orientations. To elucidate the physical background of these remarkable observations we adopted the Landauer–Büttiker-formalism (LBF) [88–91] for our curved structures.

In general, the LBF describes dissipationless magneto-transport by one-dimensional current channels at the edges of flat Hall bars. These so-called edge channels form at the intersection of the Fermi level with the filled Landau levels, which reside at energies lower than the Fermi level in the interior of the Hall bar and are lifted by the edge potential. The bending-up of the Landau levels simply describes the potential barrier which is necessary to keep charge carriers inside the Hall bar [83, 84]. For a flat two-dimensional electron system, the LBF well describes the plateau values of the quantized Hall resistance accompanied by zero longitudinal

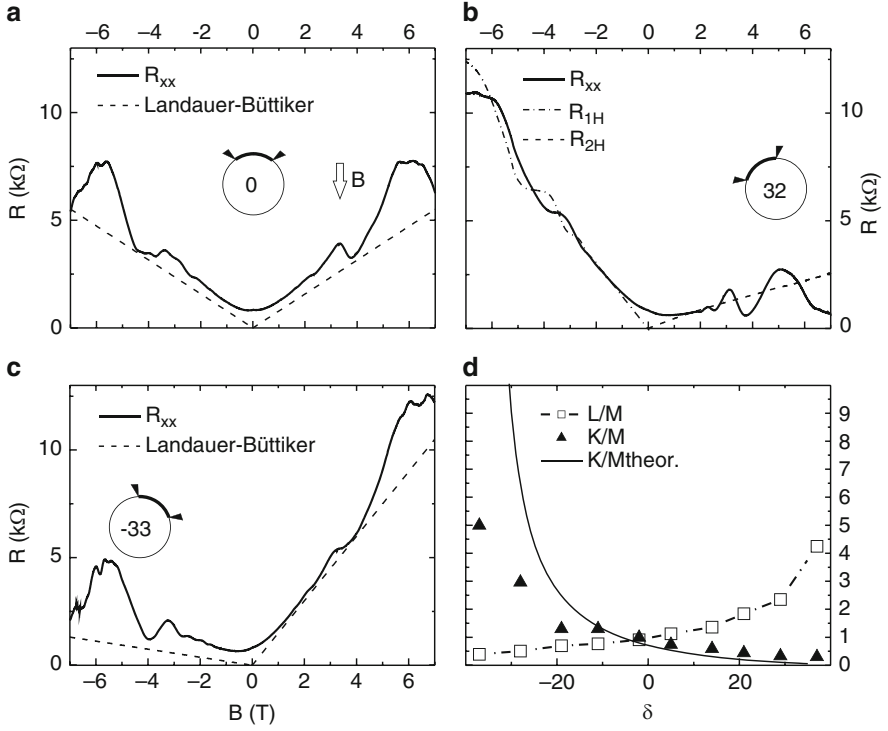


Fig. 2.11 (a–c) Measurements of the longitudinal and Hall resistance in a Hall bar along the curvature of a microscroll and calculations according to the Landauer–Büttiker-formalism. The insets show the respective orientation of the Hall bar in the external magnetic field. The radius of the scroll is $9.5 \mu\text{m}$, the distance between the voltage probes is $12 \mu\text{m}$ and the width of the Hall bar is $6 \mu\text{m}$. The charge carrier density is $n = 3.9 \times 10^{11} \text{ cm}^{-2}$ and the mobility is $34,000 \text{ cm}^2 (\text{V s})^{-1}$. (d) Ratio of bending to passing edge channels determined by $R_{xx}/R_{1H} = K/M$ and $R_{xx}/R_{2H} = L/M$, respectively, and calculated graph assuming continuous filling factors. δ represents the phase shift with respect to the symmetric orientation

resistance as discovered by von Klitzing [29]:

$$R_H = \frac{h}{e^2} \frac{1}{M} \quad (2.1)$$

$$R_{xx} = 0 \quad (2.2)$$

The number of edge channels M corresponds to the number of filled Landau levels $\nu = M = \text{Int} \left[\frac{h N_s}{e B} \right]$, with carrier density N_s and magnetic field B .

Figure 2.12a illustrates the corresponding scenario for a curved system. As discussed in Chap. 2.3, a curved 2DES in a homogeneous magnetic field exhibits Landau levels with a sinusoidal modulation of the energetic separation and density of states [34, 43]. The sinusoidal modulation of the Landau level energy leads to additional intersections with the Fermi level, i.e., additional transport

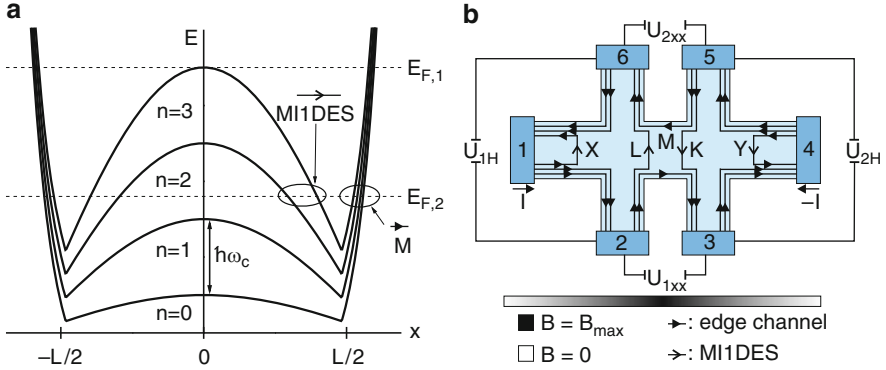


Fig. 2.12 (a) Diagram of Landau levels in a sinusoidally modulated magnetic field. The sample is assumed to have a finite length L . In addition to edge channels (M) crossings of Landau levels with the Fermi energy cause magnetically induced one-dimensional systems (MI1DES). (b) ECHB along the curvature of a scroll with the maximum of the modulated perpendicular component of the magnetic field $B_{\max}$ located at the center of the Hall bar. The color gradient below the Hall bar indicates the strength of $B_{\max}$. M , (K, L), (X, Y) represent edge channels running through all contacts, being reflected into the voltage probes and being reflected into source or drain, respectively

channels in the interior of the Hall bar. Due to their physical origin, we term these additional channels ‘magnetically induced one-dimensional electron system’ (MI1DES). Figure 2.12b shows an LBF scheme of a Hall bar oriented along the curvature with magnetic field modulation maximum located at the center between the voltage probes [34, 72]. The current direction of edge channels and the MI1DES is given by the velocity of states $v_{nk} = \frac{1}{\hbar} \frac{dE_{n,k}}{dk}$ and indicated by closed and open arrows, respectively. Three different types of edge channels can be distinguished: channels running through all contacts (M), channels reflected back into the source or drain contact (X , Y), and channels reflected into one of the voltage probes (K , L). It is obvious that for the classes X , Y , K , and L edge channels at opposite sides of the Hall bar are connected via the MI1DES in the interior. Intriguingly, the position of the MI1DES, i.e., the location where the respective edge channels are reflected, is not fixed and can be tuned over the Hall bar by changing the magnetic field, the carrier density, or simply by rotating the Hall bar in the magnetic field. This is a unique property of MI1DES in nanoscrolls.

To model Hall resistances and longitudinal resistances for the curved system shown in Fig. 2.12b with the LBF, the current balance for all six contacts is calculated. We assume that no dissipation is present in the one-dimensional channels and that no reflection occurs at the contacts. Without dissipation and reflection, the current driven through a one-dimensional edge channel which connects two contacts with chemical potential difference $\Delta\mu$ has the simple form $I_{\text{ID}} = \frac{e}{h} \Delta\mu$ [91]. In the case of flat two-dimensional systems, the condition of dissipationless current carried only in edge channels is not met and the LBF thus is not applicable when the Fermi level coincides with a Landau level and edge channels at opposite sides

of the Hall bar can strongly couple via the conducting bulk. This case cannot occur in the curved system shown in Fig. 2.12b due to the curvature of the Landau levels, i.e., the LBF is applicable for all situations in this simple picture. Under the above assumptions, the current balance for contact i reads:

$$I_i = \frac{e}{h} \left(C_i \mu_i - \sum_j t_{ij} C_{ji} \mu_j \right),$$

C_i is the number of channels leaving contact i . C_{ji} is the number of channels leaving contact j and approaching contact i . The transmission coefficient t_{ij} is one if contact i and contact j are connected and zero otherwise. μ_i and μ_j are the chemical potentials of contact i and contact j , respectively. The current balance for all six contacts for the configuration shown in Fig. 2.12b can be described by

$$\begin{pmatrix} I \\ 0 \\ 0 \\ -I \\ 0 \\ 0 \end{pmatrix} = \frac{e}{h} \begin{pmatrix} -M-L & 0 & 0 & 0 & 0 & M+L \\ M+L & -M-L & 0 & 0 & 0 & 0 \\ 0 & M & -M-K & 0 & K & 0 \\ 0 & 0 & M+K & -M-K & 0 & 0 \\ 0 & 0 & 0 & M+K & -M-K & 0 \\ 0 & L & 0 & 0 & M & -M-L \end{pmatrix} \begin{pmatrix} \mu_1 \\ \mu_2 \\ \mu_3 \\ \mu_4 \\ \mu_5 \\ \mu_6 \end{pmatrix},$$

The solution of this equation gives the Hall resistances R_{1H} and R_{2H} as well as the longitudinal resistances R_{1xx} and R_{2xx} :

$$R_{1H} = \frac{U_{26}}{I} = \frac{h}{e^2} \frac{1}{L+M} = -R_{1H}(B \rightarrow -B) \quad (2.3)$$

$$R_{2H} = \frac{U_{35}}{I} = \frac{h}{e^2} \frac{1}{K+M} = -R_{2H}(B \rightarrow -B) \quad (2.4)$$

$$R_{1xx} = \frac{U_{23}}{I} = \frac{h}{e^2} \frac{K}{(K+M)M} = |R_{2H}| \frac{K}{M} = R_{2xx}(B \rightarrow -B) \quad (2.5)$$

$$R_{2xx} = \frac{U_{65}}{I} = \frac{h}{e^2} \frac{L}{(L+M)M} = |R_{1H}| \frac{L}{M} = R_{1xx}(B \rightarrow -B) \quad (2.6)$$

On the base of these equations, we can explain all main features of the experimental observations in Fig. 2.11a–c. An inversion of the magnetic field is considered in the LBF by reversing all current channel directions and reproduces the magnetic field inversion symmetry found in the measurements. Furthermore, we indeed find that the Hall resistances appear as a magneto-resistive background in the longitudinal resistances as suspected above.

For a quantitative comparison between the LBF results and measurements we, first of all, have to measure the carrier density of the ECHB. For this purpose, Hall curves are taken with two Hall probes, e.g., probes 2 and 6, in a modulation maximum. Considering this situation, no current channels are reflected into

probe 2 and probe 6 ($L = 0$), and (2.3) reproduces the Hall resistance $\frac{h}{e^2} \frac{1}{M}$ for a flat system. Carrier density and mobility can then readily be obtained from the measured Hall resistance slope and the longitudinal resistance at zero magnetic field. We here obtain a carrier density of $n = 3.9 \times 10^{11} \text{ cm}^{-2}$ and a mobility of $34,000 \text{ cm}^2 (\text{V s})^{-1}$. The dashed lines in Fig. 2.11a and c were calculated with (2.3)–(2.6) for this carrier density assuming a continuous change of the filling factors. They well reproduce the magneto-resistive background of the longitudinal SdH oscillations.

In Fig. 2.11b, we plot the measured Hall resistances R_{1H} and R_{2H} , which are responsible for the magneto-resistive background for the respective field orientations together with the corresponding longitudinal magnetoresistance R_{1xx} . Dividing linear fits of these curves gives direct access to the ratio of reflected current channels and current channels running through all contacts, i.e., L/M and K/M . In Fig. 2.11d, experimental values of these quantities are plotted as symbols together with a theoretical curve calculated with the LBF showing a good agreement. Deviations might be attributed to the pronounced SdH oscillations present in our experiment which are not included in the Landauer–Büttiker model. Vorob'ev and coworkers investigated in detail the situation when the magnetic field modulation maximum has left the area between the voltage probes [62] of an ECHB, very similar to the one discussed here. In this case, the field inversion asymmetry becomes even more pronounced and SdH oscillations are only very weak. They found a good agreement with characteristic features of the measured curves if the LBF model introduced here is adopted to their situation and quantized filling factors are used.

It is obvious that more sophisticated models are necessary to get a deeper insight into the exciting physics of features that are unique for curved 2DES in a magnetic field, e.g., the magnetically induced 1D channels. Important facts like the final width of the edge channels [84] and magnetically induced channels, the varying density of states, or coupling effects between current paths are ignored in the LBF based model presented here. An open question is the transition from the classical regime with meander-shaped current paths as calculated in Fig. 2.10a to the quantum Hall regime with current channels as schematically shown in Fig. 2.12. The minimum around 1 T in the longitudinal magnetoresistance plotted in Fig. 2.11 shows that the classical description holds for $B \ll 1$ T and that our LBF based model holds above 1 T. A microscopic picture for this transition does, however, not exist, yet. A first step in this direction was recently published by Friedland et al. [64], who observe deviations from the LBF model at filling factors slightly below integer values. They ascribe these to a reorientation of the current in compressible regions at the field maximum similar to the static skin effect at low fields.

2.5 Conclusions

In conclusion, rolled-up two-dimensional electron systems in a magnetic field represent a fascinating new research field which was made possible by the combination of conventional top-down preparation methods with the bottom-up self rolling effect

of strained heterolayers introduced by Prinz et al. [4]. We gave a brief overview of our pioneering experiments in this field focusing on two key examples, i.e., the first proof of rolled-up 2DES in simple four-finger geometry [35] as well as the experimental proof of the static skin effect [37] and magnetically induced current channels in rolled-up Hall bars [73]. While many features of these measurements can be described with the simple models presented in this review one of the major challenges in this field is the development of more sophisticated models, e.g., to obtain a microscopic picture of the current paths in our samples. On the other hand, the ongoing optimization of rolled-up 2DES with mobilities of up to $900,000 \text{ cm}^2 (\text{V s})^{-1}$ has recently enabled measurements in the ballistic regime [63] where intriguing electron states with opposite direction of momentum and velocity are predicted by theory [56]. Furthermore, in Hall bars on semiconductor nanoscrolls, we can guide one-dimensional Landau states by magnetic barriers, and we are able to change with the orientation and strength of the magnetic field both, their internal properties, as well as their location. The investigation of such effects in rolled-up systems with customized contact geometry [92, 93] might be the next step in the field of rolled-up 2DES.

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Chapter 3

Capacitance Spectroscopy on Self-Assembled Quantum Dots

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Abstract We review studies on energy levels and charge carrier transfer processes in self-assembled InAs quantum dots (QDs). The central aspect of our work is the use of the deep level transient spectroscopy (DLTS) and its related methods, which have been proven to be powerful tools to assess quantization and interaction effects in low-dimensional quantum structures. For a detailed understanding of the experimental results, involved emission and capture processes of carriers via different paths must be considered. We find that the main process of carrier traffic takes place by thermally-assisted tunneling. Furthermore, pure thermal emission or capture and tunneling processes are possible. The competition among these emission (capture) processes depends on the temperature, the electric field across the diode, as well as the charge state of the QDs. To gain further insight in the complex carrier exchange mechanism, we use, e.g., high magnetic fields to suppress competing tunneling processes or low temperatures in order to avoid multiple emission paths. We further discuss related DLTS methods, such as Tunneling-DLTS and Reverse-DLTS in order to complete our understanding of the charge carrier transfer mechanism of QDs in biased diode-like devices. Finally, our review of transient capacitance experiments yields a consistent understanding of the relevant physics behind the charge carrier emission rates on the basis of simple models.

3.1 Introduction

The complex interplay of quantization and interaction in semiconductor nanostructures is found to result in a wealth of intriguing properties that can be employed in applications such as single-photon emitters, detectors, or even quantum computing or cryptographic devices. In self-assembled InAs quantum dots (QDs), both effects are of the same order [1–4]. The evaluation of quantization energies is essential for the understanding of, e.g., Raman and photoluminescence experiments [5, 6] and for the development of applications in optoelectronic devices like quantum-dot photodiodes for coherent optoelectronics [7, 8], photodetectors [9, 10], and basic memory devices [11, 12]. In the latter cases, the QDs are embedded in diode-like

devices, and the QDs are exposed to strong electric fields. Furthermore, for room-temperature operation of QD devices thermal effects are important. Despite its importance for applications, the field dependent emission and capture of charge carriers in QDs are only barely studied so far [13–18]. It has been pointed out that the charge carrier exchange between QDs and their environment, i.e., emission and capture of carriers, can be quite complex since several emission (capture) paths can be involved [18–20]; and it is controversially discussed which process would dominate. Generally, for the escape of electrons from QDs embedded in a Schottky diode thermal, tunneling, and thermally assisted tunneling (TAT) processes may be distinguished [14, 16, 21–23]. Furthermore, the competition among these emission processes strongly depends on the applied electric field at the diode, the temperature, magnetic fields, as well as the charge state of the QDs [20]. In TAT processes, the electron tunnels from an intermediate state that is energetically elevated with respect to the ground state. Activated tunneling from the excited state is favorable with respect to the pure tunneling process because the tunneling rate strongly depends on the barrier height and width. Moreover, it has been pointed out that not only the resonant quantum-dot states [16, 24] can act as intermediate states but also the continuum of evanescent states [14, 20, 21, 25] that arises from the conduction band penetrating the barrier in an electric field. In close analogy to the thermionic tunneling process at Schottky barriers [26], this will lead to a lowering of the apparent activation energy of the emission process that is strongly dependent on the electric field [14, 27].

In the focus of our review, we will discuss transient capacitance spectroscopy data in which the charge state of the self-assembled InAs QDs can be clearly resolved, i.e., we are able to distinguish not only between emission of the s and p shell but also between the singly and doubly occupied s shell and even resolve the number of carriers in the p shell. The data can be quantitatively understood with a simple model that considers the charge-state dependence of the emission rates on the footing of the electric-field dependence of the emission paths in TAT processes.

3.2 Experimental Techniques

In the following, we briefly introduce experimental details of our experiments, such as the experimental techniques, the samples, and the analysis of the data.

3.2.1 Deep Level Transient Spectroscopy

Time-resolved capacitance spectroscopy or deep level transient spectroscopy (DLTS) is a powerful method to identify defect levels in semiconductors [28, 29]. DLTS relies on the evaluation of temperature-dependent emission rates of carriers emitted from localized states that may be defect states, for which the DLTS

technique was originally developed, or the bound charge-carrier states in semiconductor QDs, as discussed in this review. An activation energy E_a and a capture cross section σ_a are determined from the temperature dependency of the emission rate [28, 30]. In case of defect states, both values are characteristic for given defects and can be used for identification. More recently, it was demonstrated that DLTS is also an excellent tool for studying carrier-exchange processes from states associated with low-dimensional quantum structures, such as quantum wells [31–33] or QDs [14, 24, 34–40]. A diversity of different QD material systems (InAs/GaAs [14, 20, 24, 38, 41], GaSb/GaAs [37, 39], InP/GaInP [35], Ge/Si [42]) has been investigated, and a wide spectrum of activation energies and capture cross sections has been reported. Even reports on QD samples of the same composition, e.g., self-assembled InAs QDs in a GaAs matrix, quote values that are significantly different. This indicates that the data are quite sensitive on properties like geometry and composition of the QD system as well as the material they are embedded in.

For DLTS measurements, the QDs are generally embedded in slightly doped Schottky or *pn* diodes. In our experiments, the Schottky diodes and QDs are grown by molecular beam epitaxy (MBE) as sketched in Fig. 3.1. Starting with a highly doped GaAs back-contact layer, the InAs QDs are introduced into *n*-GaAs ($N_D \approx 4 \times 10^{15} \text{ cm}^{-3}$). We usually clad the QDs by few nm thick undoped GaAs layer in order to prevent a direct doping of the dots. On top of the samples metallic Schottky contacts (Cr) are deposited.

In Fig. 3.2, we briefly introduce the principle of DLTS. Voltage pulses V_p of duration t_p are periodically applied at the Schottky gate in order to charge the InAs QDs with electrons ($t < 0$). During the time interval $t_p < t < 0$, at which the pulse voltage V_p is applied, the extent of the depletion region is smaller than the distance of the

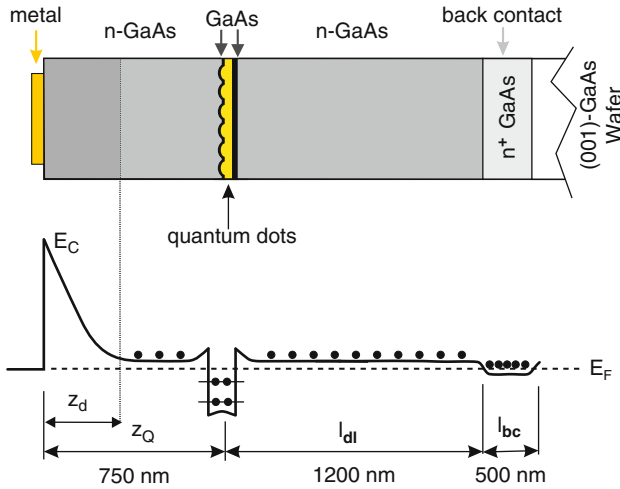


Fig. 3.1 Sketches of layer sequence (*top*) and corresponding conduction-band edge at zero bias (*bottom*) of a Schottky diode with embedded InAs QDs

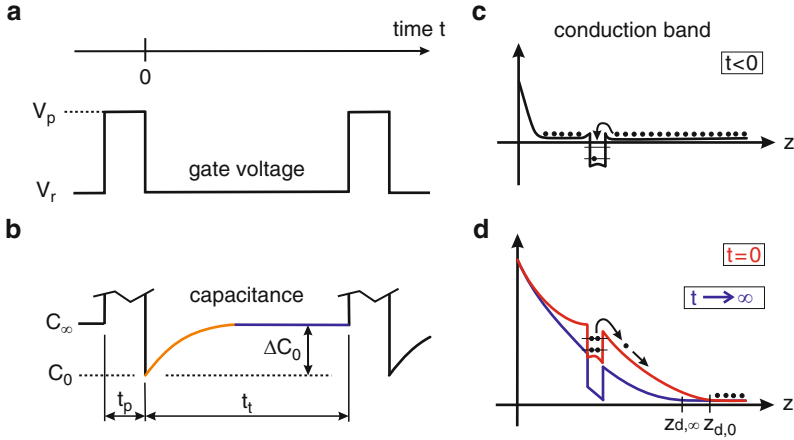


Fig. 3.2 (a) Voltage-pulse sequence and (b) the corresponding capacitance transient in the periodic DLTS cycle. (c) Conduction-band edge at pulse voltage $V = V_p$ and (d) at reverse voltage $V = V_r$. During the time $-t_p < t < 0$, the boundary of the depletion zone lies between the gate of the Schottky diode and the QD layer as indicated in (c). In general, the length of t_p is chosen such that all QD levels below the Fermi energy E_F are occupied. In (d) the band profiles at the beginning and at the end of the transient are shown in red and blue colors, respectively

QD layer from the Schottky contact ($z_d < z_Q$) as illustrated in Fig. 3.2c. At $t = 0$ the bias of the Schottky diode switches from the pulse voltage V_p to a reverse voltage V_r , at which the QD levels are lifted above the Fermi energy E_F . The QD layer now lies within the depletion region ($z_d > z_Q$). Furthermore, the extend z_d of the depletion zone and thus the depletion capacitance C depends on the charge state of the QDs. As sketched in Fig. 3.2b, the capacitance increases from the value C_0 at $t = 0$ where the charge in the QD layer is at maximum to C_∞ at $t \gg 0$ where the QDs are unoccupied. Thus, the measured capacitance change $\Delta C(t) = C_\infty - \Delta C_0 \exp(-e_n t)$ reflects the time evolution of the dot occupation driven by the carrier emission rate:

$$e_n(T) = \sigma_a \gamma_n T^2 \exp\left(-\frac{E_a}{kT}\right), \quad (3.1)$$

where γ_n is a material constant and k the Boltzmann factor.

From the capacitance transients, now we have to extract the emission rates $e_n = 1/\tau_n$, which can be obtained by a single exponential fit in the simplest case. However, a monoexponential fit is usually not satisfying due to multiexponential behavior of the transients caused by inhomogeneous broadening or possible multi-occupancies of the QD shells. In order to tackle this difficulty, several filter methods can be applied onto the transient to extract emission rates. Apart from the often used double-boxcar (DB) filter [30], more sophisticated evaluation methods have been applied as, e.g., lock-in techniques [43], Laplace [44, 45] or Fourier Transformations [46, 47]. In the following, we will discuss results obtained with the

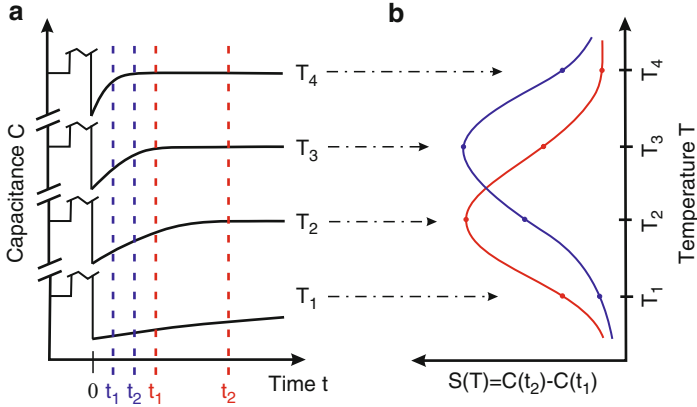


Fig. 3.3 Evaluation of capacitance transients with a double-boxcar (DB) filter: (a) capacitance transients sketched for various temperatures with $T_1 < T_2 < T_3 < T_4$. The blue and red dashed vertical lines denote the times t_1 and t_2 of the DB filter for two different rate windows, respectively. The DLTS signals $S(T)$ obtained for both rate windows at the corresponding temperatures are depicted in (b)

simple rate-window approach using a DB filter [30]. In Fig. 3.3, the principle of the DB filter is illustrated. At times t_1 and t_2 , capacitance values are recorded for the temperature-dependent DLTS spectrum $S(T) = C_2(t_2, T) - C_1(t_1, T)$. It is easily shown that a large DLTS signal $C_2(t_2) - C_1(t_1)$ is observed if a time constant τ_n in the capacitance transients becomes equal to the reference time constant $\tau_{\text{ref}} = (t_2 - t_1)/\ln(t_2/t_1)$ set by the times t_1 and t_2 of the DB filter. Thus, in DLTS spectra a maximum occurs at the temperature T_m at which the relaxation time τ_n in the capacitance transient becomes equal to the reference time: $e_n^{-1} = \tau_n = \tau_{\text{ref}}$. Setting different values of t_1 and t_2 (Fig. 3.3) and thus of τ_{ref} , one obtains DLTS maxima at different temperatures T_m . Assuming the exponential dependence of the emission rate in (3.1), the reference-time dependence of T_m is used in a conventional Arrhenius analysis to obtain values for the activation energy and the capture cross section:

$$\ln(\tau_{\text{ref}} T_m^2) = -\ln(\sigma_a \gamma_n) + \frac{E_a}{k T_m}. \quad (3.2)$$

From the slope in the trap signature, i.e., the logarithmic depiction of $\ln(\tau_{\text{ref}} T_m^2)$ vs. the inverse temperatures of the DLTS maxima $1/T_m$, and its intersection with the ordinate at $1/T_m = 0$, we obtain E_a and σ_a , respectively. A linear behavior is expected if the emission is dominated by a single, purely thermally activated process. In case of several emission paths, such as competing tunneling processes [20, 27] or emission via excited QD states [19], $\ln(\tau_{\text{ref}} T_m^2)$ vs. $(1/T_m)$ shows a nonlinear behavior. We note that the nonlinearity might not be obvious from the experimental data if the temperature window probed in the experiment is too narrow. A conventional Arrhenius analysis might then yield erroneous values.

3.2.2 Capacitance Voltage Spectroscopy on Schottky Diodes

Capacitance voltage (CV) spectroscopy on Schottky diodes is a very powerful tool to gain information about the extent of the depletion region in the semiconductor and the material properties at its boundary. The capacitance between the front contact, that is often called gate, and the back contact is measured at a fixed frequency as function of the dc bias. For the measurement, an ac voltage is superimposed to the dc bias and the ac current is measured with lock-in technique. In CV spectroscopy on Schottky diodes, the bias dependence of the depletion-region extent is probed. For the case of a diode doped with a single, shallow impurity type, an impurity profile of the diode can be obtained from a CV spectrum according to:

$$N_D(V_g) = -\frac{2}{A^2 \epsilon \epsilon_0 e} \left(\frac{d(C^{-2})}{dV_g} \right)^{-1}, \quad (3.3)$$

where N_D is the doping density at the border of the depletion region and A is the gate area. Furthermore, e is the electron charge, ϵ and ϵ_0 are the dielectric constants of GaAs and vacuum, respectively. If there exist additional deep impurities, the distance of the depletion zone boundary from the front electrode is larger than the distance of the deep impurities probed with the ac signal. The situation is very similar, if the depletion zone boundary approaches a layer of self-assembled QDs [48–50]. In particular, the capacitance signal becomes strongly frequency dependent at low temperature, since the carriers have to surpass a barrier between the depletion zone boundary and the QD layer. The relaxation time for the carrier exchange between the bound levels in the QD and the depletion zone boundary results in a phase shift of the ac signal, i.e., the ac current, which is purely out of phase to the ac voltage in the case of a pure capacitor, now contains an in-phase component reflecting a finite admittance of the device.

This fact is employed in the admittance spectroscopy [51], where the temperature and frequency dependence of the ac current is measured phase sensitive. The real part of the complex admittance, i.e., the conductance measured in-phase with the excitation, shows a maximum when the emission rate e_n of the localized state corresponds to $\omega = 2e_n$ with $\omega = 2\pi f$, where f is the lock-in frequency of the superimposed ac voltage [52]. In case of QDs, conductance maxima are observed at certain gate voltages and temperatures T_m . Measuring the conductance at different frequencies f enables us to evaluate an activation energy from an Arrhenius analysis of $\ln(\omega T_m^2)$ vs. $1/T_m$ [52].

CV spectroscopy can also be applied on metal-insulator-semiconductor (MIS) devices with embedded QDs [1, 2, 4]. Here, the semiconductor material between the metallic front electrode and the back contact, in which the dot layer is embedded, is nominally undoped. Between the back contact and the QDs charge exchange is possible. The frequency of the ac excitation is chosen such that even at very low temperatures carrier exchange is possible via tunneling processes. If no charge transfer between the dot layer and the electrode takes place, an almost dc-voltage independent capacitance is observed due to the high doping of the back contact. The

capacitance signal significantly increases from this background value whenever the gate voltage is adjusted to a value at which charge exchange is energetically possible [1,2,4]. Thus, the capacitance spectra of MIS diodes differ significantly with respect to CV experiments on Schottky diodes. Both the constant background capacitance as well as the low temperature allow to much clearer resolve quantum levels of QDs in capacitance spectra of MIS diodes. In Schottky diodes, the voltage dependence of the depletion capacitance and the high temperatures needed for the charge injection from the depletion region boundary into the dot levels tend to smear structure related to QD states in the CV spectra [40]. At the end of this review, we will compare values for the energy separation of the dot levels obtained from CV spectroscopy on MIS diodes with the results obtained from DLTS spectroscopy.

3.3 Experimental Results

In this chapter, we first illustrate CV spectra on three different Schottky diodes containing self-assembled QDs. While the epitaxial layer sequence of the diodes is nominally identical, the dots have different size and density. This leads to characteristic changes in the spectra. CV spectra are invaluable for the performance and the interpretation of DLTS experiments, since they yield the values of the voltages at which the QDs are charged. Afterwards, we will summarize our DLTS experiments. From the DLTS spectra, we obtain the level structure with respect to the continuum energy of the barrier material. Here we will discuss the observation of several maxima in one spectrum, which we associate with the shell structure of the QDs. In particular, we will focus on the electric and magnetic field dependence of the energies. We will demonstrate that the experimental observations can be well understood on the footing of a model assuming thermally activated escape of the charge from the QDs. The consequence is that the activation energies obtained from a conventional Arrhenius analysis of the DLTS data are different from the energetic distance between the bound carrier states in the dot and the continuum of the barrier. This difference becomes increasingly important with the occupation of the QDs. Thus, we are able to resolve in the DLTS spectra not only the shell but also the charge state of the level from which the electron escapes. In the last two subchapters, we will briefly discuss Tunneling- and Reverse-DLTS as well as admittance spectroscopy and, finally, compare the results of CV spectroscopy on MIS diodes with similar QDs.

3.3.1 *Capacitance Spectroscopy on Quantum-Dot Schottky Diodes*

In Fig. 3.4a, we compare CV spectra of three different Schottky diodes measured at $T = 100$ K. At this temperature, the QD states are in equilibrium with states in the surrounding GaAs host. This ensures sufficiently fast charge exchange between all

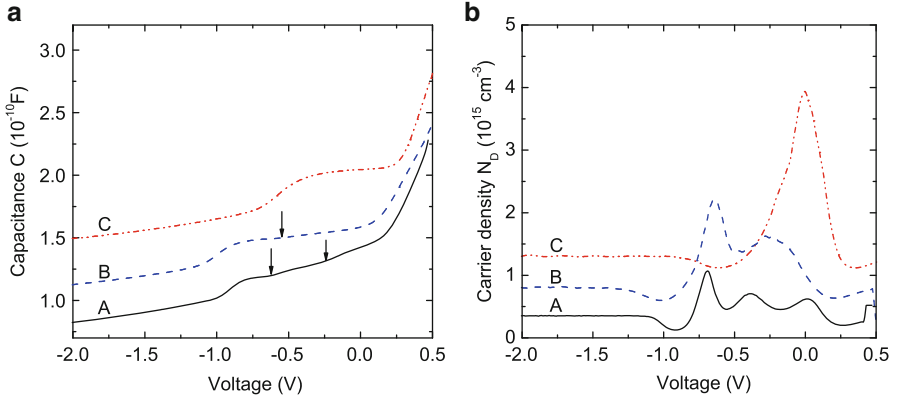


Fig. 3.4 (a) CV spectra of Schottky diodes with embedded QDs. The QDs were grown at different temperatures resulting in different structural properties. The temperatures, the signal frequencies and amplitudes of the lock-in amplifier were $T = 100$ K, $f_{\text{lock-in}} = 258$ Hz, and $V_{\text{lock-in}} = 10$ mV, respectively. (b) Carrier-density profiles of the sample obtained from the CV measurements using (3.3) shown in (a). The traces in (a) and (b) are offset for clarity

QD states and the electron reservoir at the signal frequency of $f_{\text{lock-in}} = 258$ Hz. Despite the different QD growth parameters of the samples, the spectra show similar qualitative behavior. At low gate voltages $V_g < 1.5$ V, a slight increase of the capacitance is observed reflecting the n -type doping level according to (3.3). Below $V_g < 1.5$ V, the border of the depletion zone moves from the highly doped back contact toward the QD layer with increasing voltage V_g . The energies of the QD states are higher than the Fermi energy E_F so that they remain unoccupied. Above this regime, the voltage passes a threshold value V_g^{th} at which the lowest lying QD state crosses E_F and electrons are filled into the QDs. At this threshold voltage V_g^{th} , which is different for each sample, the capacitance rises abruptly and a plateau sets in. We determine V_g^{th} to be 0.92, 1.02, and 0.62 V for sample A, B, and C, respectively. The capacitance rises abruptly because the QD layer is still closer to the front electrode than the depletion-zone boundary. From the distance between the boundary of the depletion region and the dot layer, values for the ground-state energy of the QDs have been estimated [53]. While the QD layer is charged, the boundary of the space charge region remains almost fixed below the QD layer due to the screening effect of the electrons occupying the QD layer. Thus, the capacitance signal remains nearly constant, which is observed in a plateau-like structure. Arrows in Fig. 3.4 mark faint indentations in the plateaus of sample A and B, which are more clearly resolved as minima in the density profiles of Fig. 3.4b. These indentations are associated with charging of different QD shells as will be shown later in this review [14, 40]. No substructures are observed in the CV trace of sample C. At the end of the plateau-like structure, the dots can accommodate no more electrons and the boundary of the space-charge region sweeps across the QD layer. This leads to a nearly abrupt increase of the capacitance. Beyond this point, the capacitance

traces feature the same behavior as expected for a Schottky diode without QD layer.

Whereas V_g^{th} is similar in sample A and B, the threshold voltage in sample C is larger, although the nominal layer thicknesses are identical in all three samples. This can be explained by a reduced energy separation between the ground state in the QDs and the barrier continuum in sample C in comparison with samples A and B.

The doping profiles in Fig. 3.4b are calculated from the CV spectra according to (3.3). For clarity they are offset; the constant background doping below the threshold voltages is identical within 5% for all three samples. At a voltage $V_g = V_g^{\text{th}}$, a strong charge-carrier depletion is observed reflecting that the space-charge region around the QD layer remains partly depleted while charge exchange with the dots takes place. With increasing V_g , we observe several peaks in the apparent doping profiles attributed to charge accumulation in the QD plane. Interestingly, they obviously depend on the QD properties in the different samples. We observe for sample A three maxima at -0.69 , -0.39 , and 0.02 V, which we attribute to charging of the s shell, the p shell, and states in the wetting layer, respectively. These assignments are supported by admittance spectroscopy experiments as discussed in Chap. 3.3.6.

From atomic-force microscopy (AFM) images of the surface dot layers of the samples, we imply that the dot density of sample B is larger than the one of sample A and that the density in sample C is by far the largest. This is in accordance with the larger apparent carrier densities in the profiles of samples B and C. Furthermore, the number of peaks decreases to two and one for samples B and C, respectively. This observation can be understood in view of the smaller dot sizes determined in the AFM images. Due to the smaller size, the shell energies are closer to the barrier continuum. Electron occupation further lifts the levels with respect to the continuum so that in sample B the fully occupied p shell is not stable any more and in sample C the p shell cannot be occupied at all.

3.3.2 Deep Level Transient Spectroscopy on Quantum-Dot Schottky Diodes

The assertions derived above from the CV spectra are confirmed by the DLTS spectra of the corresponding samples depicted in Fig. 3.5. The pulse voltage V_p and its duration t_p ensure a complete filling of the QDs. After the filling pulse, the applied voltage is reduced to the reverse bias V_r , which lifts the QD levels above the Fermi energy ($V_r \leq V_g^{\text{th}}$) in all samples as can be inferred from the CV spectra in Fig. 3.4. In sample A, two pronounced DLTS maxima are observed at $T = 80$ and $T = 40$ K that we associate with electron emission from the QD s and p shell, respectively. The splitting of the s -state maximum is associated with different emission rates of s electrons in singly and doubly occupied QDs. This point will be discussed in the further text. Similarly, in sample A, a fine structure is resolved in the p maximum. It is accordingly associated with emission from QDs occupied by one to four electrons in the p shell [40]. At $T < 20$ K, a temperature-independent DLTS signal

Fig. 3.5 DLTS spectra of the quantum-dot Schottky diodes A, B, and C. The pulse and reverse bias are $V_p = +0.7$ V and $V_r = -1.0$ V, respectively. The pulse-bias duration is $t_p = 1$ ms and the reference time of the DB filter is $\tau_{\text{ref}} = 4$ ms with $t_2/t_1 = 8$

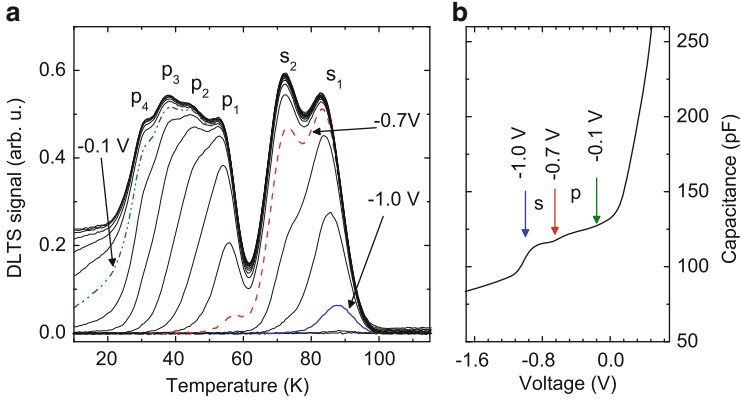
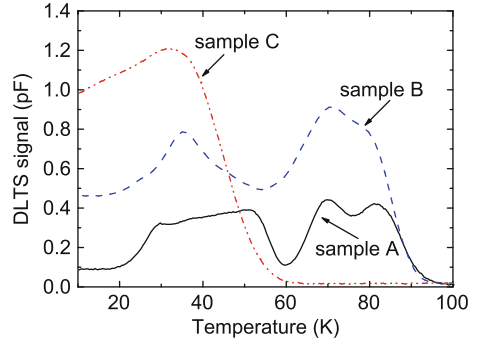


Fig. 3.6 (a) DLTS spectra of a Schottky diode (sample D) with InAs QDs at pulse bias voltages ranging from $V_p = -1.3$ V to $V_p = +0.7$ V and a fixed reverse bias of $V_r = -1.4$ V. The rate window is $\tau_{\text{ref}} = 4$ ms. Traces recorded at $V_p = -1.0$, -0.7 , and -0.1 V are highlighted by a fat, dashed, and dotted line, respectively. (b) CV measurement recorded on the same diode at $T = 100$ K and frequency $f_{\text{lock-in}} = 258$ Hz. The thick blue full line, the dashed red, and the dotted green line in (a) indicate the DLTS spectra taken at the pulse voltages marked by arrows in (b), respectively

is observed which we assign to tunneling processes [15, 23, 24, 43, 54, 55]. In the DLTS spectrum of sample B, the splitting of the s maximum at $T = 70$ K is less well resolved. A second maximum is observed at $T = 37$ K which we associate with the emission of electrons from the incompletely filled p shell. No further substructure is observed in this maximum. Sample C only shows one broad maximum at $T = 33$ K.

In the following, we will further illuminate the substructure of the s and p maxima with the aid of data obtained with a QD Schottky diode in which the structure is more clearly resolved (sample D). In Fig. 3.6a, we present DLTS spectra recorded with different pulse biases starting from $V_p = -1.3$ V in 0.1 V steps. A CV trace recorded on the same Schottky diode at $T = 100$ K is presented in Fig. 3.6b. The

reverse bias $V_g = V_r = -1.4$ V is chosen so that the QDs can relax into the empty state. With the pulse voltage V_p , the charge state at the beginning of the transient is chosen. The charge states inferred from the CV trace in Fig. 3.6b clearly confirm the identification of the DLTS maxima in Fig. 3.6a. Within the voltage range -1.0 V $< V_g < -0.7$ V, we expect the s shell of the QDs to be charged. Indeed, in the DLTS spectra, the two maxima between 60 K $< T < 90$ K gradually arise starting with the one located at higher temperature that, accordingly, is assigned to emission from the singly occupied s shell. At $V_g = -0.7$ V, where from Fig. 3.6b the s shell is expected to be completely filled, the DLTS maximum assigned to the doubly occupied s level starts to saturate and at higher voltages emission from the p states cause the appearance of the p maxima at temperatures below $T < 60$ K. Four peaks labeled with p_1 – p_4 appear one after the other until at $V_p > -0.1$ V saturation of the p related maximum starts. The DLTS signal at lower temperatures $T < 20$ K is related to wetting-layer states and tunneling from the p shell [15, 20, 23, 43].

The reverse voltage controls the location of the QD states with respect to the Fermi level and the average electric field F_{ave} at the QD layer when the transients are recorded. From the behavior of the spectra with this field, we derive further information about the nature of the emission processes. In Fig. 3.7a, the reverse voltage V_r lies within the capacitance plateau, $V_r > V_g^{\text{th}}$. Here, with increasing V_r , the QD levels are swept across E_F . Correspondingly, we clearly observe suppressions of the DLTS peaks for those QD levels that remain below E_F . With increasing V_r , the high temperature parts of the DLTS maxima are quenched while the remaining signal at low temperature even increases in strength. The increase is mainly due to a geometric effect arising from the smaller extend of the depletion region at higher V_r . In addition, at low electric fields, the competition of tunneling emission is reduced [16, 20, 23, 43]. The competition between tunneling and thermal signal can be clearly observed in the color-scale plots depicted in Fig. 3.7c and d at $T < 30$ K. The DLTS signal below $T = 20$ K, which at reverse bias below $V_r < 1.4$ V is associated to tunneling processes, increases with decreasing reverse bias while the thermal DLTS signal above 20 K decreases. We note that, on the other hand, at $V_r > -1$ V a thermal signal arises with a maximum at $T \approx 17$ K, which we tentatively associate with wetting-layer emission. This signal is completely quenched at smaller reverse voltages where due to the higher electric field the corresponding states are already depleted by tunneling processes in the time window used in the experiment.

Figure 3.7b illustrates the development of the DLTS spectra if the reverse bias falls below the threshold voltage. In this voltage range, the QDs always relax into the empty state. With decreasing reverse voltage V_r , the electric field F_{ave} increases and causes the s -state maxima to shift to lower temperatures as observed in Fig. 3.7b as well as in the color-scale images Fig. 3.7c and d taken at reference times $\tau_{\text{ref}} = 3.5$ ms and $\tau_{\text{ref}} = 95$ ms, respectively. Furthermore, with increasing electric field, the p maximum shrinks to a temperature-independent DLTS signal arising from pure tunneling processes [23]. The temperature range in which tunneling processes compete with the thermal emission increases with higher electric fields [20, 23]. In particular, from the color-scale images, it becomes clear that at

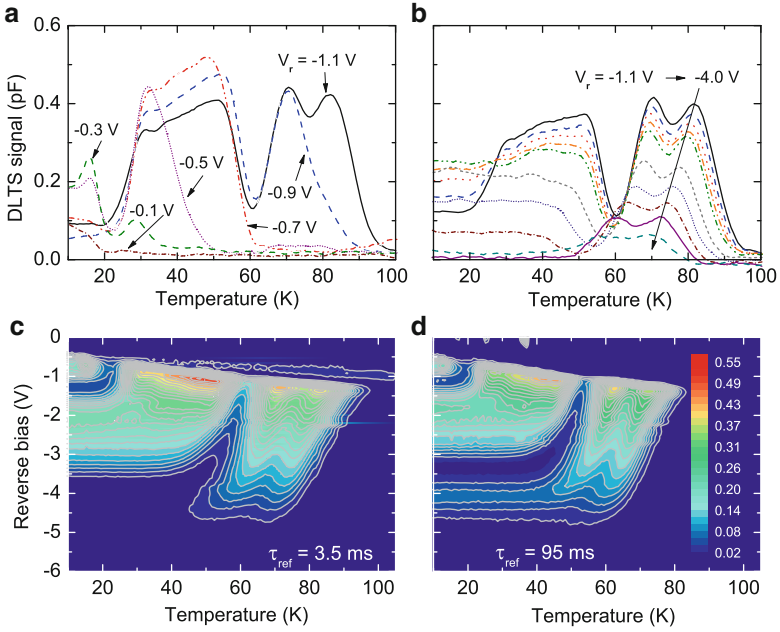


Fig. 3.7 DLTS spectra of sample A in Fig. 3.5 for reverse voltages (a) $-1.1 \text{ V} < V_r < -0.1 \text{ V}$ and (b) $-4.0 \text{ V} < V_r < -1.1 \text{ V}$. The pulse bias is $V_p = +0.7 \text{ V}$ and the rate window is $\tau_{\text{ref}} = 4 \text{ ms}$ with $t_2/t_1 = 8$. The color-scale maps of the DLTS signal vs. V_r and T are taken at reference times (c) $\tau_{\text{ref}} = 3.5 \text{ ms}$ and (d) $\tau_{\text{ref}} = 95 \text{ ms}$, respectively. The unit of the color scale indicated in (d) is pF

the experimental reference times the temperature independent, pure tunneling signal stems from tunneling from the p shell at voltages $V_r > -3.5 \text{ V}$ and from the s shell at $V_r < -3.5 \text{ V}$. Again, the reduction of the signal height with increasing F_{ave} results from a geometric effect and from the competition between thermal and tunneling emission.

3.3.3 Evaluation of Quantum-Dot Shell Energies in the Thermally Assisted Tunneling Model

DLTS data are conventionally evaluated with an Arrhenius analysis. In the following, we will discuss the field dependence of thus derived activation energies for the emission from the QDs. It will reveal that the emission is a TAT process [14, 20, 27, 56]. A simple model will be applied in order to estimate the discrepancy between the apparent activation energies derived from the Arrhenius analysis and the field-independent energy separations E_s and E_p between the QD levels and the conduction band edge of the barrier close to the QD. The model will be

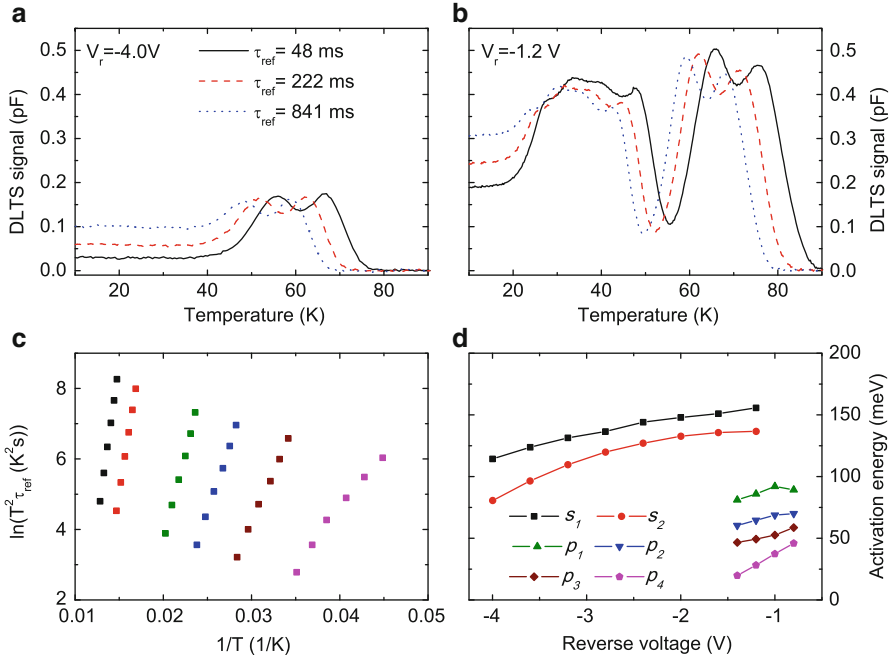


Fig. 3.8 (a) and (b) show DLTS spectra of sample D for three different rate windows (48, 222, 841 ms) at $V_r = -4.0$ V and $V_r = -1.2$ V, respectively. The pulse bias is $V_p = +0.7$ V and $t_2/t_1 = 8$. (c) depicts the trap signatures of the QD electrons at $V_r = -1.2$ V. (d) Activation energies E_a and (e) capture cross sections σ_a at various V_r

corroborated with experiments in magnetic fields and alternative methods discussed in the following two chapters.

We start with the conventional Arrhenius analysis of the DLTS maxima positions in the spectra recorded at different reference times τ_{ref} . This is illustrated in Fig. 3.8a and b for reverse voltages $V_r = -4.0$ V and -1.2 V, respectively. The temperature shift of the maxima with τ_{ref} is clearly observed indicating a dominantly thermally activated process. Figure 3.8c shows the so-called trap signatures of the emission processes obtained from the Arrhenius analysis. The V_r dependence of the apparent activation energies E_a of the s and p maxima is depicted in Fig. 3.8d. The reverse-voltage range in which the apparent activation energies E_a of p states could be determined is much smaller since for the p shell the tunneling signal starts to dominate the DLTS spectra at considerably smaller fields than for the s states.

It is well known that an electric field lowers the barrier in traps with Coulomb-confinement potential as first discussed by Poole and Frenkel [57]. A similar effect can be expected in confinement potentials of different shapes. In particular, for the case of a rectangular quantum well with width $2z_0$ the barrier lowering can be easily estimated to be eFz_0 [58]. The experiments reveal that the field dispersion of the activation energies is much stronger than would be expected from this effect and points to the importance of tunneling processes [14]. Tunneling rates

increase exponentially with the electric field, since the effective length of the tunneling barrier decreases. We could show [14, 20] that the behavior of the DLTS signal associated with the s -shell emission can be explained with a TAT model that previously had been developed to explain the electric field-dependent emission rates from deep impurity states in semiconductors [21, 58–60]. In the model, the emission from the QD due to purely thermal emission as well as competing TAT was considered in a simple one-dimensional approximation with a triangular tunneling barrier. With a refined model considering the tunneling through the Coulomb barrier of charged QDs, the field dependence of the emission from the p shell could be explained as well [56].

The tunneling emission rate e_{tu} is approximated within a one-dimensional, semi-classical Wentzel–Kramers–Brillouin (WKB) approach [20, 23, 61]

$$e_{tu} = e_{tu,0}(F) \exp \left(-\frac{\sqrt{8m^*}}{\hbar} \int_0^{z_1} \sqrt{V_B(z)} dz \right). \quad (3.4)$$

The pre-exponential factor $e_{tu,0}$ is assumed to be only moderately field-dependent as in the case of a Dirac well, where it is linear [21]. Furthermore, $m^* = 0.069m_e$ is the effective mass in the barrier material, $\hbar$ is Planck's constant, and $V_B(z)$ is the barrier potential along the (reverse) growth direction. The barrier potential depends on the energy E of the tunneling electron, which is assumed to remain constant during the tunneling process. The integration spans the nonclassical region between the QD and the point z_1 , where the barrier-band edge meets the energy of the tunneling electron. We note that in order to keep the model simple, resonant and non-resonant QD states are considered on an equal footing. The results indicate that non-resonant states close to the conduction band edge of the barrier dominate the behavior [14, 23]. The tunneling potential was assumed to be approximately linear in [23] (linear TAT model) where the emission from the s shell was analyzed. Later the Coulomb potential of the charges in the dot was included in order to also quantitatively understand the emission rates from the p shell (Coulomb TAT model) [56]:

$$V_B(z) = E - eF_{ave}z - \frac{ie^2}{4\pi\epsilon\epsilon_0} \left[\frac{1}{z_0} - \frac{1}{z_0 + z} \right]. \quad (3.5)$$

The last term is the Coulomb potential of the charge in the QDs, which are modeled by metallic spheres with center at $z = -z_0$ and radius z_0 . The integer i denotes the dot electron occupation after the emission process. The linear part $eF_{ave}z$ arises from the space charge in the Schottky diode and can be controlled by the diode bias:

$$F_{ave} = \frac{eN_D}{\epsilon\epsilon_0} \left(\sqrt{\frac{2}{eN_D} [\epsilon\epsilon_0(V_{bi} - V_r) + ien_Qz_Q]} - z_Q \right) \quad (3.6)$$

where n_Q is the areal quantum-dot density, V_r is the bias, and V_{bi} the built-in voltage of the Schottky diode. We note that the linear TAT model is obtained with $i = 0$ in (3.5). The Coulomb contribution in (3.5) approximates the field of the charge in the

QD from which the considered emission takes place. In contrast, the second term in the square root of (3.6) takes into account the average field of the charges in all other QDs.

The total emission rate is obtained by summing up all contributions for electron energies E between the top of the quantum-dot potential, i.e., the band-edge energy of the barrier at the location of the QD, and the energy of the s or p state at which the TAT process starts. In the TAT model, the enhancement e_{meas}/e_n with respect to the purely thermal emission can be written as [58]

$$\frac{e_{\text{meas}}}{e_n} = 1 + \int_0^{E_B} \Gamma(E) dE, \quad (3.7)$$

where E_B is the barrier height with respect to the energy of the state from which the electron escapes and $\Gamma(E)$ is given by

$$\Gamma(E) = \frac{1}{kT} \exp \left(\frac{E}{kT} - \frac{\sqrt{8m^*}}{\hbar} \int_0^{z_1} \sqrt{V_B(z)} dz \right). \quad (3.8)$$

As will be seen in the following, the most important contributions $\Gamma(E)$ to the integral in equation (3.7) will be at energies close to the band-edge energy of the barrier. We thus may expect that, for the calculation of the emission rate from a highly charged QD with a TAT model, the Coulomb part in (3.5) describing the potential in close proximity to the QD is very important.

In Fig. 3.9, apparent activation energies determined from an Arrhenius analysis of the trap signatures are compared with the energies E_B derived from the Coulomb

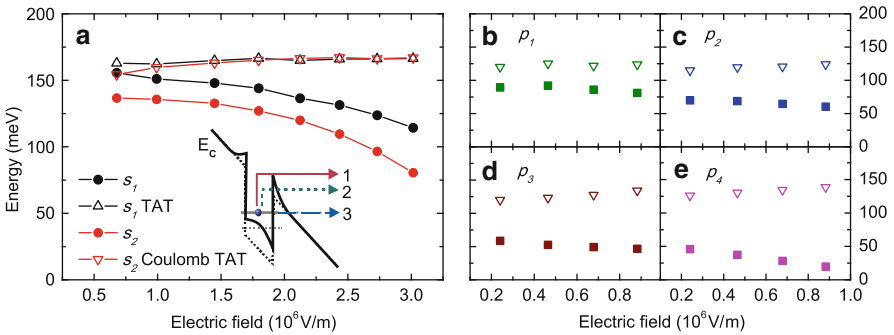


Fig. 3.9 Filled circles in (a) denote apparent activation energies E_a of the s_1 and the s_2 electrons obtained with a conventional Arrhenius analysis from the experimental data at different electric field strengths. The open triangles denote the energy barriers of the s_1 electron and the s_2 electrons derived from the Coulomb TAT model. (b-e) Activation energies E_a (filled squares) and barrier energies (open triangles) obtained from the Coulomb TAT model of p -state electrons as indicated in the figure. The inset in (a) sketches the QD potential in the Schottky diode. The purely thermal, the thermally assisted tunneling (TAT), and the tunneling escape paths are denoted by numbers 1, 2, and 3, respectively

TAT model. Filled circles denote apparent activation energies determined from the conventional Arrhenius analysis. They strongly depend on the occupation state of the QDs as well as on the electric field at the QDs. The open triangles are evaluated with the Coulomb TAT model from the experimental data as follows [56]. First, from the experimental rates and the apparent activation energy, a rate enhancement according to (3.7) is calculated solving numerically the integral equation (3.7). The calculated emission rates e_n are used for a second Arrhenius analysis in order to determine new activation energies. These energies are taken for a next iteration until the energies used in (3.7) and determined from the Arrhenius analysis of the calculated emission rates e_n are consistent. Finally, the thus determined activation energy should resolve the true barrier height E_B as measured from the quantum-dot state, from which the electron escapes. The thus determined barrier heights E_B are depicted in Fig. 3.9a for the s_1 - and s_2 -state electrons. Unlike the apparent activation energies E_a , the barrier height is independent on the occupation state and the electric field F_{ave} . Both facts are expected if the multi-electron states in the QDs can be described by multiply occupied single-particle states shifted by the Coulomb-charging energy and the potential step defining the dot boundary is localized to a length scale much smaller than the scale on which the Coulomb potential decreases. In particular, in this approach, the shift by the Coulomb-charging energy does not alter the relative energy distances between the single-particle levels and the height of the barrier at the dot boundary. For instance, in this model, the height of the barrier with respect to the s and p levels does not depend on the occupation of the level. The repelling potential arising from the charge in the QDs lifts the whole conduction band including the barrier band edge. Correspondingly, assuming a purely thermal emission process, Engström *et al.* have pointed out that the Coulomb-charging energy is not expected to influence the activation energy in DLTS measurements [38]. The open triangles in Fig. 3.9 refer to the height of the barrier with respect to the s or p levels and would be equal to the experimental activation energies if tunneling would be absent. While due to the tunneling contribution, the apparent activation energies depend on the electric field and the dot occupation, the barrier heights calculated with the Coulomb TAT model do not.

In Fig. 3.9b–e, the corresponding apparent activation energies and barrier heights calculated with the Coulomb TAT model are presented for emission from the p shell. The much stronger field dispersion of the apparent activation energies in the p shell as compared to the s shell, which is also obvious from the data in Fig. 3.7d, already indicates the importance of tunneling processes. Also, from the above discussion, we expect that the rate for emission from the p shell is drastically enhanced because of the lower effective barrier and the repulsive Coulomb field of the larger dot charge. Indeed, the barrier heights E_B , denoted by open triangles in Fig. 3.9b–e, are considerably higher than the apparent activation energies and the difference increases with the charge state of the p shell. As in the case of the s -shell emission, the barrier heights determined with the Coulomb TAT model for the p_1 electron are nearly independent of the electric field. The barrier heights average to a value of 121 meV. The barrier heights of the p_1 – p_4 electrons are almost independent on the electric field. The difference between the barrier heights of the p_1 state and

$E_B = E_s = 163$ meV determined for the s shell amounts to $E_s - E_p = 42$ meV. This value is in good correspondence with values determined on similar QDs with different techniques [1–3].

3.3.4 DLTS Experiments in Magnetic Fields

We have performed DLTS experiments in magnetic fields applied parallel as well as perpendicular to the QD layer. In a parallel magnetic field, tunneling paths oriented normally to the QD layer will be quenched [20, 61, 62]. On the other hand, perpendicular magnetic fields can be employed in order to verify the assignment of the low-temperature DLTS peaks to emission from the p states [40].

The conjecture of competing tunneling processes in the thermally assisted emission suggests the use of strong magnetic fields applied parallel to the QD plane in order to suppress the tunneling emission. The results clearly demonstrate that indeed tunneling processes play a crucial role in the carrier transfer processes even at elevated temperatures. Figure 3.10a shows DLTS spectra in parallel magnetic fields up to 7 T. The temperature-independent DLTS signals below $T < 20$ K strongly decreases in a parallel magnetic field providing further evidence that the signals indeed are caused by tunneling processes [20, 61–64]. The ratio of the emission rates measured at finite magnetic fields and zero field can be described quantitatively with a one-dimensional WKB approach [20]. To the potential, the term $\frac{e^2 B^2}{2m^*} (z - z_1)^2$ was added to the ansatz (3.5). Assuming that at $T = 10$ K the DLTS signal results from

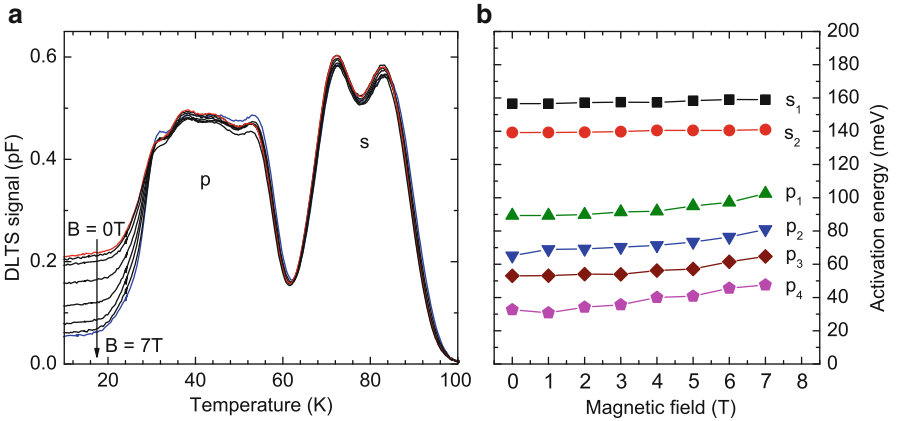


Fig. 3.10 (a) DLTS spectra recorded in magnetic fields $0 \leq B \leq 7$ T directed parallel to the QD layer. The field is increased in steps of 1 T as indicated. Between adjacent traces, the field was increased from 0 to 7 T in steps of 1 T as indicated. The reverse and pulse bias are $V_r = -1.4$ V and $V_p = 0.7$ V, respectively. The rate window of the DB filter is $\tau_{\text{ref}} = 4$ ms with $t_2/t_1 = 8$. (b) Activation energies derived from an Arrhenius analysis of the DLTS maxima in (a). The assignment to the shells and their occupation is indicated. Lines between the data points are guides to the eye

pure tunneling processes, we determine a value of about 80 meV for the barrier height that is in reasonable agreement with the results obtained from the thermal emission rates.

The thermal emission from the QDs, which is responsible for the maxima at $T > 20$ K in Fig. 3.10a, is much less affected by parallel magnetic fields. Nevertheless, the activation energies derived from the trap signatures do depend on the magnetic field as illustrated in Fig. 3.10b. The activation energies obtained for the different occupation states of the s and p shell are marked by different symbols in Fig. 3.10b. While the activation energies for emission from the s shell are only slightly affected by the magnetic field, the energies for emission from the p shell strongly shift to higher values. This observation is in accordance with the Coulomb TAT model discussed in the previous chapter. As a result of the lower binding energy and the higher occupation number, tunneling processes play a much more important role for emission from the p shell than for the s shell. Indeed, at very high fields $B \gg 7$ T, we would expect the activation energies to approach values in accordance with the barrier heights derived from the Coulomb TAT model from the trap signatures. This remains to be checked.

DLTS spectra recorded in perpendicular magnetic field are shown in Fig. 3.11a [40]. The s_1 and s_2 peaks slightly shift to higher temperatures with increasing magnetic field B . A clearly stronger influence of the magnetic field is found for the emission peaks allocated to the p shell. The p_1 and the p_2 peaks shift to higher temperatures and their heights increase approximately linearly with magnetic field. On the other hand, the peaks associated to the emission from the p_3 and the p_4 states shift to lower temperatures and their heights decrease. Figure 3.11b shows the corresponding magnetic field dependence of the activation energies E_a derived

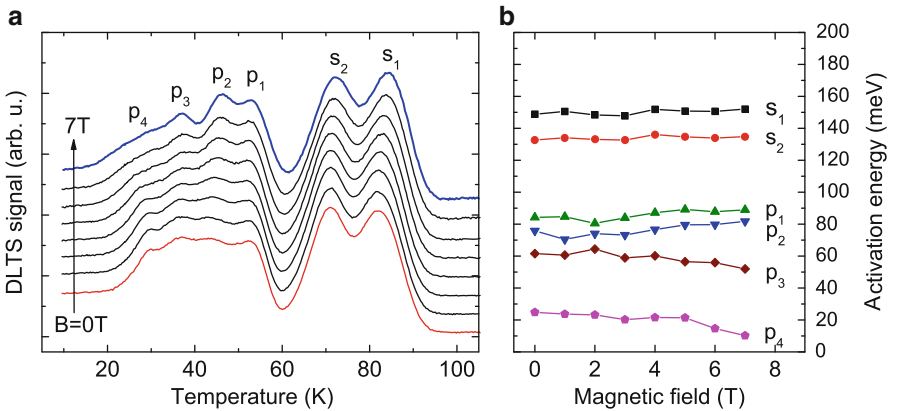


Fig. 3.11 (a) DLTS spectra of the QD sample measured in perpendicular magnetic fields between $B = 0$ T and $B = 7$ T. The field values are increased in 1 T steps between adjacent traces. Data are recorded at a reverse bias $V_r = -1.4$ V and a pulse bias $V_p = +0.4$ V. The rate window is $\tau_{\text{ref}} = 4$ ms. The spectra taken at $B > 0$ are offset for clarity. (b) Activation energies derived from the DLTS spectra in a perpendicular magnetic field. Lines between the data points are guides to the eye [40]

from an Arrhenius analysis of the reference-time dependence of the maxima. The activation energies of the s maxima very slightly increase which we attribute to different shifts of the ground state in the QD and the free-electron continuum in the barrier material. In contrast, the energies associated to p states shift at $B > 2$ T indicating an increasing energy separation between the $p_{1,2}$ and the $p_{3,4}$ branches. The behavior can qualitatively be understood with a model assuming a parabolic confinement potential for the QD states [3, 65]. This observation is in agreement with results of capacitance spectroscopy on similar QDs embedded in MIS diodes [2]. From the magnetic-field dispersion of the p emission energies, an effective mass m^* can be derived within the parabolic model [40, 65]. The obtained value $m^* = 0.03m_e$ is larger than the value $m_{\text{InAs}}^* = 0.023m_e$ for pure InAs bulk material but smaller than values published previously by Miller *et al.* [2], who derived values for the effective mass of $m^* = 0.06m_e$ from capacitance measurements and $m^* = 0.08m_e$ from far-infrared spectroscopy measurements on MIS diodes with similar QDs. Values between those of bulk InAs and GaAs ($m_{\text{GaAs}}^* = 0.067m_e$) may be expected due to wave-function penetration into the barrier material and band non-parabolicity in the dot material.

3.3.5 Advanced Time-Resolved Capacitance Spectroscopy Methods: Tunneling-DLTS, Constant-Capacitance DLTS and Reverse-DLTS

In this section, we discuss three modified transient capacitance techniques that have been applied to InAs QDs. In essence, the results of these methods confirm our TAT model developed in the previous sections. The complexity of the multiple-path scenario for carrier emission from QDs can be avoided with the Tunneling-DLTS method [15, 23, 43, 66]. At elevated temperatures, many emission paths from QDs are possible involving excited levels as intermediate states as pointed out, e.g., by Engström *et al.* [19]. In contrast, at sufficiently low temperature, only one path remains, in which the charge carrier escapes by a pure tunneling process only. As already discussed in connection with Figs. 3.7 and 3.10, we expect the DLTS signal to be dominated by pure tunneling. From a vertical cut through the color-scale map of the DLTS signal in Figs. 3.7c and d, it is obvious that the tunneling signal is strongly electric-field dependent. This field dependence can be employed to derive independent values for the barrier heights with respect to the QD states. In Fig. 3.12, we present a spectrum obtained on sample A of Fig. 3.5 by such a cut, i.e., the temperature was kept constant at $T = 10$ K and the electric field was changed by the reverse bias. We clearly observe two distinct maxima in Fig. 3.12 that we attribute to emission from the s shell at higher and from the p shell at lower electric fields. Similar to the Arrhenius analysis of conventional DLTS spectra, here the rate-window dependence of the electric fields at the maxima can be used for an analysis. Using a simple model for the tunneling rate [23]

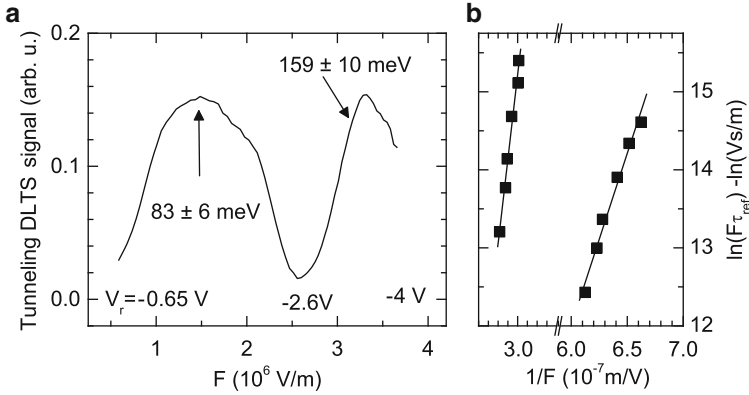


Fig. 3.12 (a) Tunneling-DLTS spectrum showing the capacitance change associated with tunneling emission from the QDs measured at a reference time $\tau_{\text{ref}} = 1,467$ ms using $t_2/t_1 = 2$ and temperature $T = 10$ K. The capacitance signal is normalized accounting for the dependence of the signal on the reverse voltage. The reverse voltages V_r used at the beginning, the minimum, and the end of the trace are denoted. Figure (b) presents logarithms of the reference times vs the inverse fields at the peak positions. Note that the x scale is broken. From the slope of the linear fits (*full lines*), the energies allocated to the peak positions in (a) are determined

$$e_{\text{tu}} = \frac{eF}{4\sqrt{2m^*E_B}} \exp\left(-\frac{4}{3} \frac{\sqrt{2m^*E_B}^{3/2}}{e\hbar F}\right), \quad (3.9)$$

we determine the barrier heights indicated in Fig. 3.12a. In (3.9), a triangular tunneling potential has been assumed for simplicity. Due to the relatively long tunneling path of pure tunneling processes, the Coulomb barrier does not change the field dependence of the tunneling rates, as has been confirmed by numerical calculation [23]. The barrier height, however, is reduced by an amount close to the Coulomb-charging energy. If this is considered, the obtained values are in very good correspondence to results from thermal data evaluated with our TAT model and thus support our understanding of the carrier emission from QDs [23].

Considering the strong electric-field dependence of the emission rates, one might be concerned that the change of the electric field that occurs during the recording of a DLTS transient might influence the data. This question has been tackled experimentally by application of the so-called Constant-Capacitance DLTS [28,67]. In this method, a feedback circuit is used in order to keep the capacitance of the diode and thus F_{ave} constant [68]. The results obtained with this method are almost identical to those obtained with the conventional method [67]. This confirms our assertion that the strong occupation dependence of the emission rates results from the Coulomb barriers at close distance to the charged dots.

Finally, we briefly discuss so-called Reverse-DLTS (R-DLTS) experiments, in which capture processes are studied instead of emission processes [69]. Measurements of capture rates on defects in DLTS experiments are generally performed by the filling pulse method [70] down to the nanosecond scale. But, in case of

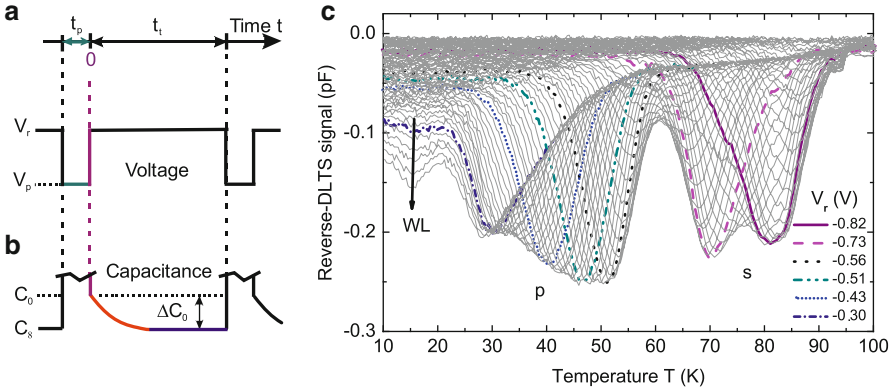


Fig. 3.13 Pulse sequence (a) and capacitance transient (b) in Reverse-DLTS. Gray lines in (c) denote Reverse-DLTS spectra recorded at different reverse voltages. The voltage was changed in steps of 0.01 V between $-1.15 \text{ V} < V_r < -0.15 \text{ V}$. Six of the traces, which are highlighted by color, are obtained with reverse voltages V_r as labeled in the figure. The reference time is $\tau_{\text{ref}} = 3.5 \text{ ms}$ with $t_2/t_1 = 8$

QDs, the capture times are in the range of few picoseconds, rendering the evaluation of capture processes in QDs with common space-charge techniques impossible. Nevertheless, in recent studies, it has been shown that capture of electrons [71] and holes [72] can be assessed by the so-called Reverse-DLTS (R-DLTS) method. In Fig. 3.13a, the pulse sequence used in R-DLTS is sketched. During the voltage pulse, the QDs are in the depletion region unlike in conventional DLTS experiments. Thus, at the beginning of the capacitance transients, the QD levels under consideration are unoccupied. The bias V_r applied during the transient recording is chosen within the plateau region of the CV spectra so that charges are injected into the QDs. Thus at $t > 0$, the space-charge boundary is repelled from the QD layer and the capacitance decreases with time. As a consequence, the capture times are extremely short if the dots are far away from the equilibrium situation. Close to equilibrium the capture time becomes similar to the emission time measured with swapped pulse and reverse voltages in conventional DLTS. In Fig. 3.13c, a number of R-DLTS spectra is presented. A pulse bias $V_p = -1.2 \text{ V}$ is used to empty the QD levels before transient recording. A spectrum recorded at a certain V_r consists of a single peak reflecting the charge injection into the subset of QDs with empty states close to the Fermi energy E_F . States above E_F remain uncharged while states far below E_F are charged already at t_1 for above reason. If spectra recorded at different V_r are plotted together as in Fig. 3.13c, the envelope indeed shows remarkable similarities with the corresponding DLTS spectra with $\tau_{\text{ref}} = 3.5 \text{ ms}$ depicted in Fig. 3.7. We clearly observe capturing of the s and p electrons and even into the wetting layer. These experiments are still ongoing. Although the activation energies derived with this method are similar to those discussed in Chap. 3.3.2, there are open questions that remain to be resolved. Among those is an unexpectedly strong dependence of the signal strength on the reference time and the pulse voltage.

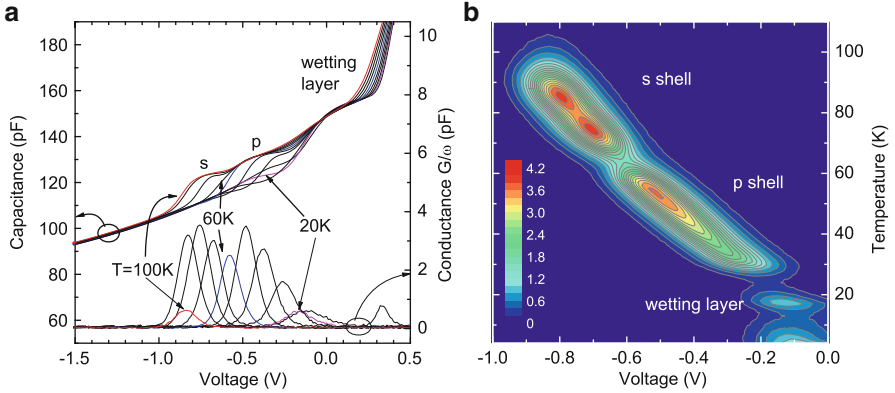


Fig. 3.14 (a) Admittance spectra of a Schottky diode (sample A) with QDs recorded with a frequency $f = 258$ Hz and at temperatures between $T = 10$ K and $T = 100$ K in 10 K steps. The capacitance and the conductance signals are proportional to the out-of-phase and the in-phase ac current components, respectively. (b) Map of the conductance vs. gate voltage and temperature for $f = 258$ Hz. The color scale in (b) is given in pF

3.3.6 Alternative Capacitance Spectroscopy Methods

The admittance spectroscopy method [51] as introduced in Sect. 3.2.2 is an alternative method to probe rates for charge transfer between the depletion zone boundary and QDs [13, 25, 27, 52, 73]. Figure 3.14a presents admittance spectroscopy spectra of a Schottky diode containing QDs recorded at different temperatures. The conductance features a single temperature dependent maximum. At the maximum position, the frequency used in the experiment can be related to the charge exchange rates [51]. This can be understood qualitatively with the aid of the temperature and voltage dependence of the maximum positions depicted in the color-scale plot in Fig. 3.14b. At temperatures above the *s* shell and the *p* shell related maxima, the charge occupation of the corresponding QD shells readily follows the excitation. At temperatures below the maxima position, the charge exchange is suppressed and, therefore, in the maxima the experimental frequency is essentially equal to the emission and capture rates. The temperature and frequency dependence of the maxima positions can be used to evaluate thermal activation energies for the charge exchange process similar to the analysis of DLTS data. For the sample in Fig. 3.14, we determine activation energies of the s_1 , s_2 , and the *p* electrons to be 156, 131, and 110 meV, respectively. As expected, these values are very close to activation energies measured at lowest possible electric fields in DLTS experiments of the sample [27].

Finally, we briefly refer to capacitance spectroscopy experiments performed on MIS diodes with self-assembled QDs. Since the seminal work of Drexler *et al.* [1], this technique has been widely used in order to gain information about the level structure of charge carriers in self-assembled QDs [1, 4, 62, 74–78]. As a typical

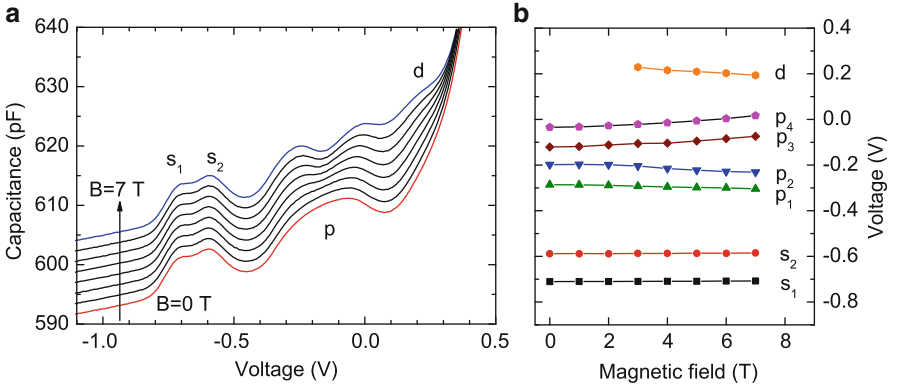


Fig. 3.15 (a) CV spectra of a metal-insulator-semiconductor (MIS) diode with QDs. The perpendicular magnetic fields in which the traces have been recorded are increased in 1 T steps between zero field and 7 T as indicated. The ac signal frequency and amplitude are $f_{\text{lock-in}} = 938$ Hz and $V_{\text{oc}} = 5$ meV, respectively. The traces obtained for $B > 0$ T have been offset by 2 pF for clarity. (b) Magnetic-field dispersion of the QD s and p shell

example, capacitance spectra of a MIS diode with self-assembled InAs QDs embedded in nominally undoped GaAs are presented in Fig. 3.15a. In the structures, a highly doped back electrode (Si, $2 \times 10^{18} \text{ cm}^{-3}$) is separated from the dot layer by a 20 nm GaAs tunneling barrier. The distance to the front electrode is 117 nm. The QD growth conditions were similar to the ones used for the Schottky diodes discussed above. A magnetic field has been applied perpendicular to the QD layer. With the magnetic field dependence, the assignment of the s and p shell to the capacitance maxima is verified. The s maximum is clearly split as a result of the Coulomb-charging energy between the singly and the doubly occupied s shell. Within a simple model [1], a Coulomb-charging energy of 18 meV is derived from the voltage separation of the two s maxima and a single-particle s - p level separation of 39.5 meV is obtained from the voltage separation between the s and the p maxima. The level separation obtained in capacitance spectra is thus in very good correspondence to the difference of the barrier energies derived from DLTS experiments for the s and the p shells. The splitting of the p maxima has been used to determine within a parabolic confinement model an effective mass of the QD p state. The value $m^* = 0.058m_e$ is in close correspondence to values reported elsewhere [2].

3.4 Conclusion and Outlook

In this review, we present an overview of transient capacitance experiments performed on self-assembled InAs QDs embedded in n -doped GaAs Schottky diodes. The DLTS and related techniques, which are well-established for deep carrier traps in semiconductor materials, can also be used to study the relaxation of electrons

out of the QDs as function of temperature, electric and magnetic field. The DLTS spectra contain well-resolved maxima that reflect the emission processes from the QD s shell, the QD p shell and the wetting layer. This identification is verified in DLTS experiments performed in magnetic fields applied perpendicular to the QD layer. Substructure in the DLTS spectra reflects the occupation of the final states. The observations can be well understood with a model considering TAT processes. The model can be verified by DLTS experiments in which the electric field is varied or a parallel magnetic field is applied, and in Tunneling- DLTS experiments at very low temperatures. Furthermore, we have briefly introduced two derived techniques, the Reverse-DLTS as well as the Constant-Capacitance DLTS, which have been applied on Schottky diodes with QDs. The results of both support the previous findings. In addition, it is demonstrated that the values obtained for the QD level structure is in very good agreement with values obtained by alternative techniques such as admittance spectroscopy and CV spectroscopy in MIS diodes.

Ongoing work is devoted to the development of improved techniques derived from the transient-capacitance technique. The by far largest number of the transient capacitance studies on QDs employed the DB technique for an extraction of relaxation times. The large number of quantum states in QDs with nearby energies suggests to compare the DB with different filter techniques such as the Fourier- or Laplace-Transformation technique in order to optimize resolution. Furthermore, experiments are of special interest in which during the charging pulse the sample is illuminated simultaneously. This optical DLTS [79, 80] technique enables us to probe electron and hole emission of the same QD ensemble under conditions of relevance for optical applications. Moreover, experiments are performed, in which the impact of nearby QDs on the conductivity of a low-dimensional electron system is probed [81, 82]. In essence, the detection layer may be replaced by a resistor that depends on the charge state of the QDs. This will enable transient conductance experiments on much smaller systems, even on single QDs [83], than the rather large capacitors of the DLTS technique, in which always large QD ensembles are probed. Also, the potential for memory device applications has been pointed out [41]. Finally, we mention that a systematic study of capture and emission cross sections is missing, so far. It seems that for an understanding simulations will be helpful [84].

Acknowledgements

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Chapter 4

The Different Faces of Coulomb Interaction in Transport Through Quantum Dot Systems

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Abstract Transport through quantum dot systems covers a broad range of phenomena ranging from Coulomb blockade oscillations to the Kondo effect. The role of Coulomb interaction in transport processes has many facets. It influences the electronic structure of quantum dot systems, it introduces a strong dependence on the number of charge carriers in the confined system, and, last but not least, it enhances the appearance of spin effects. In this chapter, we review the different faces of Coulomb interaction on the electronic structure of few-particle quantum dot systems emphasizing the mutual interplay between quantum confinement, dimensionality, and charge interaction.

4.1 Introduction

Electronic transport experiments and theory form a vital and rapidly developing field of condensed matter physics. This is not only because they provide a way to easily investigate the properties of the quantum system of interest and allow to study fundamental quantum mechanics by means of conventional (semiconductor) electronics. They also may guide the way both toward a new kind of (spintronic) devices, in which the electron spin plays the role of the charge in ‘ordinary’ electronics, and a possible realization of quantum bits (see, e.g., [1–8]). Control and variability make quantum dots an ideal system to study the fundamental mechanisms which may be exploited in novel devices. They can be viewed as artificial atoms which allow for a control of the number of carriers, their spin, and the effects of quantum confinement [9].

The impact of competing interactions on the magnetic properties of quantum confined structures lies in the focus of this review, where we will discuss geometry induced spin transitions as well as magnetization transitions in magnetically doped semiconductors. Particularly at weak tunnel coupling of the quantum dot structure to macroscopic charge reservoirs, transport across the system reveals detailed insight into the correlated electronic structure. Transport spectroscopy opens access to excited states which might be invisible to optical investigations due to selection

rules. And – similar to optical oscillator strengths – the differential conductance is a measure for spin and orbital many-body correlations. On the other hand, the non-equilibrium condition imposed on the system by applying a bias between the reservoirs strongly alters the occupation of levels. In this situation, negative differential conductances and level blocking witness spin and orbital Coulomb correlations.

New phenomena occur when the coupling between dot and reservoir becomes stronger. Cotunneling opens transport channels which were energetically forbidden to lowest order tunneling, level renormalization becomes observable due to the formation of hybrid transport channels. Higher order non-equilibrium transport based on the Keldysh formalism will be reported upon in the last part of this chapter.

4.2 Transport Through Quantum Dot Systems

All systems, we consider here, have the same general structure (see Fig. 4.1a). The central region CR, which is of nanoscale proportions, is tunnel coupled to two (or more) macroscopically large parts, the leads L and R, which serve as energy and particle reservoirs. In particular, we focus on single and double quantum dots as central regions. Applying a voltage between the leads will result in a charge and/or spin current, where the charge carriers may be electrons or holes. Additional gate electrodes allow the tuning of the quantum dot potential. Since the current has to pass the central region, the transport characteristics will strongly depend on its internal (quantum

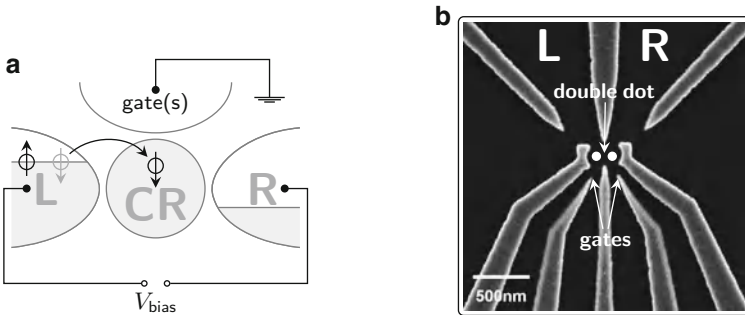


Fig. 4.1 (a) General scheme of the model structure for studying transport through a nanoscale central region (CR). Two metallic, macroscopic leads (L and R) are tunnel coupled to CR. With one (or more) gate(s), the electrostatic potential can be adjusted. An applied bias voltage V_{bias} between L and R, symbolized by different electrochemical potentials (*lines that separate shaded and non-shaded parts of the reservoirs*), leads to a current through the central region, indicated by the tunneling of a single electron (*small circle with arrow*) from L to CR. (b) AFM micrograph of a double quantum dot² (*two white dots*). Shown are the surface electrodes, which deplete the underlying two-dimensional electron gas and thereby define the confinement geometry. Two gates allow to adjust the dot potentials individually. A bias voltage between the source and drain leads (L and R) drives a current through the double dot

mechanical) structure. Even in the simplest case of only sequential, incoherent tunneling of single electrons or holes, the resulting current can show non-linearities, oscillations or blockade effects due to the single-particle spectrum of the central area [1, 10, 11]. There are many different ways to realize such a setup experimentally. One example is shown in Fig. 4.1b. In this case, the quantum dot geometry is ‘impressed’ into a two-dimensional electron gas by electrostatic forces of negatively charged metallic electrodes. The electron gas forms in one of the thin layers of a semiconductor hetero-structure (in this case, GaAs sandwiched between AlGaAs) and the surface electrodes are separated from the gas by another layer of the bulk material. For a survey of other possible realizations, see, e.g., [12, 13].

Electron transport through quantum dot systems inevitably involves states with a different number of particles in the confined structure. Coulomb interaction between the charge carriers affects these states in two different ways: adding particles to a quantum dot increases its *charging energy* U which can be approximated by the classical energy involved in charging a capacitance, $U \approx \frac{e^2}{2C}$, where C denotes the capacitance of the quantum dot associated with its size. On the other hand, the electronic structure of the quantum dot is influenced by quantum mechanical many-body correlations. A generic Hamiltonian $\hat{H}$ describing the tunnel coupled system consists of the dot part $\hat{H}_{\text{dot}}$, a part for the leads $\hat{H}_{\text{leads}}$, and the tunneling or hybridization operator $\hat{H}_{\text{T}}$. The different parts of the Hamiltonian are then:

$$\begin{aligned}\hat{H}_{\text{dot}} &= \sum_i E_i \hat{n}_i + \sum_{i,j,r,s} V_{ijrs} \hat{d}_i^\dagger \hat{d}_j^\dagger \hat{d}_r \hat{d}_s \\ \hat{H}_{\text{leads}} &= \sum_{pk\sigma} \epsilon_{pk} \hat{n}_{pk\sigma} \\ \hat{H}_{\text{T}} &= \sum_{pk\sigma} \gamma_{p,k,\sigma}^i \hat{d}_i^\dagger \hat{a}_{pk\sigma} + H.c.,\end{aligned}\tag{4.1}$$

where $p \in \{\text{L}, \text{R}\}$, $\mathbf{k}$, and σ are the lead index, wave-vector, and spin of a lead electron. E and ϵ denote the dot and lead energies, respectively, $\hat{n}_i = \hat{d}_i^\dagger \hat{d}_i$ and $\hat{n}_{pk\sigma} = \hat{a}_{pk\sigma}^\dagger \hat{a}_{pk\sigma}$ are the corresponding particle number operators, where the dot index i comprises the single-particle energy levels and the spin of a particle in the quantum dot. The second term in the dot Hamiltonian describes the Coulomb interaction between particles in the quantum dot, characterized by matrix elements V_{ijrs} in the single-particle basis of the quantum dot. The coupling strength between dot and leads is given by the complex tunneling amplitude $\gamma_{p,k,\sigma}^i$.

In small quantum dots, a strong confinement of charge carriers may result in a considerable Coulomb energy. But even if it is of the same order of magnitude as the dot energies E_i , the single-particle spectrum can be unaffected (or weakly affected) by Coulomb interaction. In this case, a quantum dot system is a candidate to be described by the *constant interaction model* (see Sect. 4.3 and also [1, 4, 10]). The interaction effects will be, however, observable in transport in form of a classical charging energy U .

A prominent example for a strongly non-linear transport effect in small quantum dots is the so called *Coulomb blockade* [14] of sequential (stationary) transport [10, 15, 16]. It occurs for temperatures and tunnel couplings much smaller than the so called *addition energy* $E_{\text{add}}(N) = \mu(N+1) - \mu(N)$, with the chemical potential $\mu(N) = E(N+1) - E(N)$ between quantum dot states differing by one in particle number. $E(N)$ denotes the energy of the N -particle ground state. In the constant interaction model, the addition energy is given by $2U + E_{N+1} - E_N$. In the low-bias regime, single-electron tunneling can only lead to a current, if at least one transport channel $\mu(N)$ lies in the energy range between the chemical potentials of the leads (bias window). This is illustrated in Fig. 4.2a. Otherwise, due to the Coulomb repulsion and despite the finite bias voltage, sequential tunneling is energetically forbidden. The resulting suppression of the tunneling current is called Coulomb blockade (Fig. 4.2b). But even if a transport channel lies in the bias window, it will only contribute to stationary transport, if there is a finite probability to find the system in either one of the states the channel connects. An example for a situation, in which tunneling through a channel in the bias window is prohibited, is the *Spin-Blockade* of transport [17–20]. The electron tunneling is prevented by (iso-)spin selection rules (see Fig. 4.2c).

At elevated bias voltage, additional transport channels involving excited quantum dot states will eventually fall into the bias window. They give rise to additional transport resonances whenever $E(N+1, \alpha) - E(N, \beta) = \mu_p$, provided the ground state transition lies in the bias window, too. Here, α, β denote many-particle energy levels in the quantum dot, and μ_p is the chemical potential of one lead or the other. This way, the differential conductance gives way for transport spectroscopy of quantum dot structures.

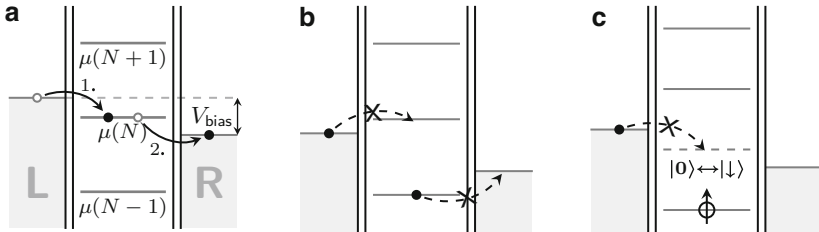


Fig. 4.2 Transport schemes of the electrochemical lead and dot potentials for a small bias voltage V_{bias} . Shaded regions L and R indicate states occupied with electrons, black circles symbolize one of the electrons in the dot and at the Fermi level in L and R, respectively. Thick double lines represent the tunneling barriers. (a) Illustration of sequential transport via channel $\mu(N)$ in the bias window. Hopping lines (1.) and (2.) represent sequential, incoherent single-electron processes, which lead to a current. (b) Coulomb blockade in sequential transport. In this configuration, the Coulomb repulsion of dot electrons prohibits single-electron tunneling. (c) Spin-Blockade of sequential transport. Since the stationary state has spin-up (depicted by circle with arrow on lowest channel) and the dashed channel connects a spin-down ($|\downarrow\rangle$) with a singlet state ($|0\rangle$), transport is blocked

4.3 Electronic Structure of Quantum Dots

Various parameters determine the electronic structure of quantum dots. The ratio between orbital energy and Coulomb interaction is controlled by the strength of the confinement potential. An applied magnetic field or the shape of the confinement potential can lift the orbital degeneracy of highly symmetric quantum dots and can destroy a corresponding shell structure. Transitions in the electronic configuration can be induced by tuning these parameters.

4.3.1 Circular Quantum Dots

Usually, the precise form of the lateral confinement potential in quantum dots is not known. In the limit of small particle numbers, however, it was found that an isotropic harmonic oscillator serves as a very successful model describing the outstanding properties of quantum dots. Assuming a two-dimensional movement of the electrons, the eigenenergies of an electron in a lateral confinement $V(x, y) = \frac{1}{2}m^*\omega_0^2(x^2 + y^2)$ are [21, 22]

$$E_{n,m} = \hbar\omega_{\text{eff}}(2n + |m| + 1) - \frac{\hbar\omega_c}{2}m, \quad (4.2)$$

where $n = 0, 1, 2, \dots$ and $m = 0, \pm 1, \pm 2, \dots$ are the radial and angular momentum quantum numbers, respectively. The effective confinement frequency $\omega_{\text{eff}} = \sqrt{\omega_0^2 + \omega_c^2/4}$ depends on the curvature of the confinement potential ω_0 and the cyclotron frequency $\omega_c = eB/m^*$. Here, electrons in the conduction band of the semiconductor are described in a single-band effective mass approximation with m^* the effective mass of the electron. For simplicity, a possible Zeeman energy is neglected. Figure 4.3 shows the Fock–Darwin spectrum in dependence of the magnetic field strength.

Regarding the zero-field case ($\omega_c = 0$), the eigenenergies of the system reduce to

$$E_{n,m} = \hbar\omega_0(N_0 + 1), \quad (4.3)$$

where $N_0 = 2n + |m|$. By taking spin degeneracy into account, each energy level is $2(N_0 + 1)$ -fold degenerate. For non-interacting electrons, the degenerate states are successively filled, respecting the Pauli principle. This yields to closed-shell configurations with a number of electrons $N_e = 2, 6, 12, 20, \dots$ (for $N_0 = 0, 1, 2, 3, \dots$). Regarding Coulomb interaction, it is energetically unfavorable for the particles to occupy the same orbitals. However, the shell structure is maintained if the confinement energy $\hbar\omega_0$ is larger than the difference in Coulomb energy associated with adjacent orbitals. For partially filled shells, the Coulomb repulsion causes the electrons to maximize the total spin due to exchange-energy saving in accordance to Hund’s rule. This leads to half-filled subshells with $N_e = 4, 9, 16, \dots$. In

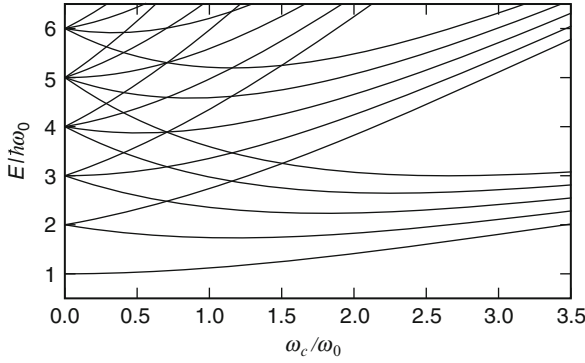


Fig. 4.3 Single-particle spectrum of a circular quantum dot as a function of the magnetic field

experiments [23], the shell structure is revealed by determining the addition energy spectrum, i.e., the difference in the chemical potential $\mu(N_e + 1) - \mu(N_e)$ for different number of electrons N_e . For magic numbers N_e of electrons, the difference in the chemical potential exhibits pronounced peaks implying particular stability for these configurations.

In order to theoretically predict the addition energy spectrum, the energies of N_e interacting electrons have to be calculated. If the confinement energy is of the order of the interaction energy, particular electron correlations become important [24]. There are different methods to calculate the few-particle spectrum. A common way to tackle the problem of correlated electrons is to apply the configuration-interaction methods [25–27]. A disadvantage of these methods is the exponential increase of the computational effort with the particle number and with the reduction of density. However, these exact diagonalization methods generate numerically exact results for the ground state energy and the low-lying excitation spectrum.

Quantum Monte Carlo techniques are another class of methods which are applied to correlated quantum dot systems [28–30]. While these methods suffer from the fermionic sign problem for large numbers of electrons, they are extremely powerful in computing properties of few electrons in the low-density regime and predicting the formation of a Wigner molecule in the quantum dot [31, 32]. They often serve as a reference for effective theories such as the density functional approach [9].

These methods reveal that strong Coulomb interaction may cause a polarized ground state of the quantum dot [31, 33]. In this limit, the single-particle energy-level spacing is much smaller than the Coulomb energy. Different orbitals are occupied by the electrons reducing the direct Coulomb interaction and a parallel spin alignment is favored by exchange energy.

Introducing a small magnetic field first reduces and then even breaks the shell structure [23]. In this case, the single-particle energies (4.2) can be approximated by

$$E_{n,m} = \hbar\omega_0(N_0 + 1) - \frac{\hbar\omega_c}{2}m, \quad (4.4)$$

including only terms linear in ω_c . The degeneracy with respect to m is lifted and the energy of levels with $m < 0$ is increased by the magnetic field while the energy of the $m > 0$ levels is reduced. However, accidental degeneracies occur for certain values of ω_c . The *two-electron* case provides an instructive example of ground state transitions induced by adjusting external parameters, such as the magnetic field. Without a magnetic field ($\omega_c = 0$), the electrons form a spin-singlet configuration occupying the lowest single-particle orbital. As ω_c increases, the spacing between energy levels with $m = 0$ and $m > 0$ reduces. If the level energy difference between consecutive levels is smaller than the difference in their Coulomb energy, the electrons will occupy orbitals of larger angular momentum, reducing the interaction. (Be aware that due to an increase of ω_{eff} with increasing magnetic field no crossing of single-particle levels with the ground state occurs (Fig. 4.3)). In fact, the total angular momentum increases with the magnetic field. This leads to spin-singlet-spin-triplet transitions as the total spin of the electrons is linked to the angular momentum by particle-exchange symmetry [34]. Spin-singlet states and spin-triplet states can only possess an even or odd angular momentum number, respectively. For three and four electrons, it is observed that the angular moment increases monotonically as a function of the field, which is not true for the spin [25, 35]. Again, only certain spin values are linked to different angular momenta which can be proven by symmetry considerations [25, 36]. At high magnetic fields ($\omega_c \gg 4\omega_0$), (4.2) yields the energies

$$E_{n,m} = \hbar\omega_c \left(M_0 + \frac{1}{2} \right), \quad (4.5)$$

where $M_0 = n + (|m| - m)/2$. The initial energy levels condensate into so called Landau bands of infinite m degeneracy.

4.3.2 Elliptical Quantum Dots

Experiments on rectangular quantum dots [37, 38] suggest an anisotropic confinement which can be modeled by an elliptical harmonic oscillator potential $V(x, y) = \frac{1}{2}m^*(\omega_x^2 x^2 + \omega_y^2 y^2)$. The eigenenergies of an electron moving in this potential with an applied magnetic field are given by [39]

$$E_{n_+, n_-} = \hbar\omega_+ \left(n_+ + \frac{1}{2} \right) + \hbar\omega_- \left(n_- + \frac{1}{2} \right), \quad (4.6)$$

where

$$\omega_{\pm} = \frac{1}{\sqrt{2}} \sqrt{\omega_x^2 + \omega_y^2 + \omega_c^2 \pm \text{sgn}(\omega_x^2 - \omega_y^2) \sqrt{(\omega_x^2 + \omega_y^2 + \omega_c^2)^2 - 4\omega_x^2 \omega_y^2}}. \quad (4.7)$$

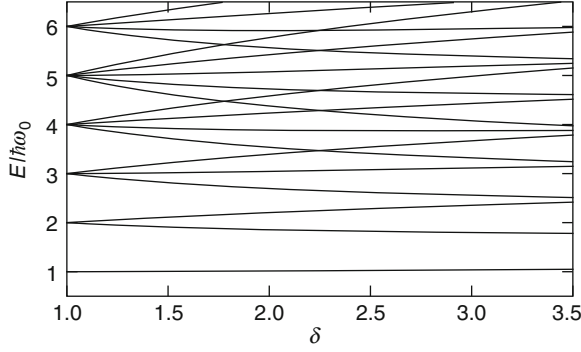


Fig. 4.4 Single-particle spectrum as a function of deformation $\delta = \omega_x/\omega_y$ for $B = 0$

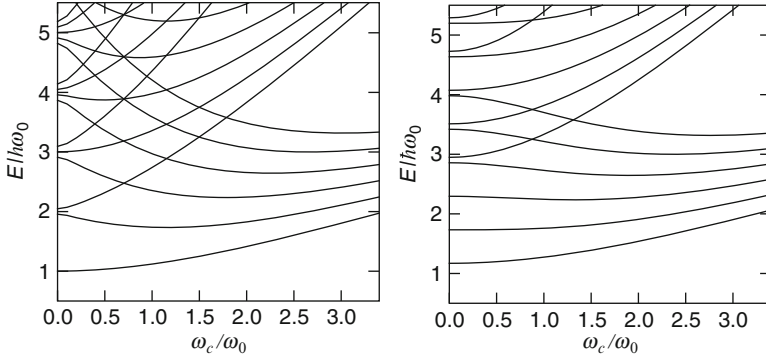


Fig. 4.5 Single-particle spectrum as a function of the magnetic field for $\delta = 1.2$ (left) and $\delta = 10.0$ (right)

Defining a deformation δ such that $\omega_x = \omega_0\sqrt{\delta}$ and $\omega_y = \omega_0/\sqrt{\delta}$ yields the constraint $\omega_0^2 = \omega_x\omega_y$, i.e., the area of the quantum dot is preserved during the deformation. Figure 4.4 shows the single-particle energies as a function of deformation in the case $\omega_c = 0$. The single-particle spectrum as a function of magnetic field for different deformations $\delta = \omega_x/\omega_y$ is shown in Fig. 4.5.

Without magnetic field ($\omega_c = 0$), the system represents two harmonic oscillators in x and y direction, respectively. In the special case, when $\omega_x = \omega_y$, the isotropic harmonic oscillator (4.2) is recovered with $n_x = n + 1/2|m| - 1/2m$ and $n_y = n + 1/2|m| + 1/2m$.

A slightly anisotropic potential lifts the degeneracy at $B = 0$. However, the energy levels are still very similar to the case of circular symmetry and a shell structure can be observed [40]. For stronger deformations, the shell structure is completely destroyed except for special deformations where accidental degeneracies occur [38, 41]. If the degeneracies are removed, the orbitals are successively filled by a spin-up and a spin-down electron. This leads to an antiferromagnetic

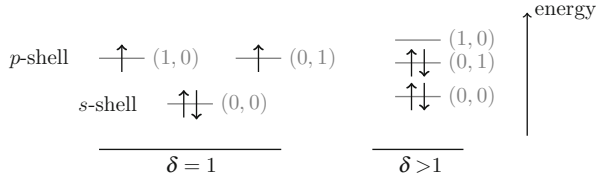


Fig. 4.6 Schematic energy levels denoted by (n_x, n_y) . In case of a circular dot, a Hund's rule ground state is found with a configuration of a doubly occupied s -shell and both p -orbitals singly occupied reducing the exchange interaction (*left figure*). A slight deformation of the dot (*right figure*) removes the degeneracy of the p -shell. The ground-state spin turns to $S = 0$ if the splitting is sufficient to overcome the exchange-energy saving associated with the $S = 1$ state

ground state configurations for even particle numbers. Especially in the case of four electrons, the splitting between initially degenerate states can be sufficient to overcome the exchange-energy saving associated with the spin-triplet state (Fig. 4.6). Anisotropy induces in this case a transition from a spin-triplet to a spin-singlet ground state [37].

The $N = 6$ electron ground state is predicted to switch from total spin $S = 0$ to $S = 1$ at a certain deformation indicating a 'piezo-magnetic' behavior [38]. For special deformations, a shell structure with a different sequence of electron numbers can be found [42]. In the limit of $\omega_x \gg \omega_y$, the single-particle energies (4.6) are

$$E_{n_+, n_-} = \hbar\omega_y \left(n_- + \frac{1}{2} \right), \quad (4.8)$$

that is, the system becomes quasi-one dimensional resembling a quantum wire. In such systems, an antiferromagnetic ground state and the appearance of spin-density-wave states are predicted [43, 44].

4.3.3 Quantum Rings

The topology of quantum rings [45–47] makes them suitable to observe the Aharonov–Bohm effect. A fractional dependency of the Aharonov–Bohm effect on the flux quanta was found recently experimentally and theoretically as a consequence of interparticle interaction [48, 49]. Different radii and ring widths can be realized depending on the process of manufacture. This variety of features attracted a strong interest in the theoretical description of the physical properties of quantum rings.

Numerical investigations have revealed that the energy spectrum and pair-correlation functions have different characters depending on the size of the ring. The occurrence of a transition between spin order and disorder has been examined using quantum Monte Carlo methods [50]. Increasing the ring radius, the interaction energy between electrons dominates the Hamiltonian leading to a strongly

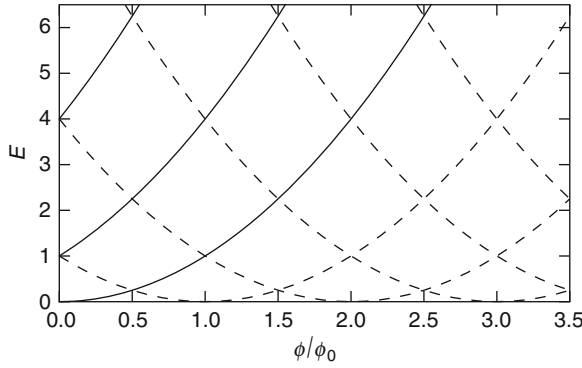


Fig. 4.7 Single-particle spectrum of a quantum ring as a function of the magnetic field (energy in units $\hbar^2/2m^*r_0^2$)

correlated system. Accordingly, the formation of a rotating Wigner molecule was suggested, where the electrons are localized on an equilateral polygon in a rotating frame. In this regime, the energy levels can be classified as rotational and vibrational states [51].

The simplest approach to an understanding of the properties of ring-shaped quantum dots is the model of an ideal one-dimensional ring penetrated by a magnetic flux Φ . The energy levels of this system are given by

$$E_\ell = \frac{\hbar^2}{2m^*r_0^2} \left(\ell + \frac{\phi}{\phi_0} \right)^2, \quad (4.9)$$

where $\ell = 0, \pm 1, \pm 2, \dots$, r_0 is the radius of the ring and $\phi_0 = hc/e$ the flux quantum. Without magnetic field, all energy levels are two-fold degenerate with respect to the direction of angular momentum, except of the first (Fig. 4.7). This leads to a shell structure as it is the case for a quantum dot, but with different magic numbers [52]. With increasing magnetic field, the energy levels start to oscillate which leads to recurring degeneracies. As a consequence of these degeneracies and Coulomb interaction, the total spin of the ground state oscillates with the magnetic field, which can be observed for different numbers of electrons [53].

Insight into the effect of dimensionality can be obtained from modeling the confinement potential of the quantum rings by a shifted parabola, $V(x, y) = 1/2m^*\omega^2(r - r_0)^2$ [48, 54]. This model allows a finite ring width determined by $w = 2\sqrt{\hbar/m^*\omega_0}$. If the ring is narrow ($w \ll r_0$), the spectrum resembles the one dimensional model. Lately, theoretical investigations on this model revealed transitions in the ground state of interacting electrons solely induced by geometrical changes. An analysis using numerical methods implies that in a quantum ring containing three interacting electrons a transition of the ground state from $S = 1/2$ to $S = 3/2$ occurs while increasing the radius [55, 56]. However, such a transition is found to be absent in a four-electron ring retaining the ground state to be $S = 1$ as given by Hund's rule.

4.3.4 Magnetically Doped Quantum Dots

Faultless manipulation of the spin in quantum dot structures is of vital importance both for applications in quantum computing as well as in spintronics. Recent progress in the fabrication of dilute magnetic semiconducts (DMS), as, e.g., manganese-doped GaAs, [57–63] has stimulated a broad interest in the exploration of their physical properties [64–66] and applications, e.g., as spin aligners, spin memories, and spin qubits [80, 81]. Control of the magnetic properties can be achieved via electric fields [57, 58, 67–69] and with the help of light [70–76].

Driving the study of quantum dots made from DMS is the possibility to obtain a nanomagnet that can be controlled by external parameters. The findings of Fernández-Rossier and Brey [77] that magnetization in quantum dots doped with Mn can be controlled by single electrons motivate the analysis given at the beginning of this section. It is meant to illustrate the evolution and properties of carrier mediated ferromagnetism in a finite model system. A more realistic model which takes into account band structure effects occurring in Mn doped GaAs quantum dots will be discussed at the end of this section.

The electrons in the half filled d -shell of a manganese atom generate a magnetic moment due to their total spin of $5/2$. The interaction between the itinerant electrons in the quantum dot and this localized Mn spins can be modeled by a contact interaction term

$$H_J = J_c \sum_{i,R} \mathbf{S}_R \cdot \hat{\mathbf{s}}_i \delta(\mathbf{R} - \hat{\mathbf{r}}_i),$$

where $\mathbf{S}_R$ denotes the impurity spin at position $\mathbf{R}$, $\hat{\mathbf{s}}_i$ is the spin of the i th electron and J_c is an effective coupling constant characterising the s – d -exchange between conduction band electrons and the magnetic Mn- d electrons [75, 76]. In (II,Mn)VI compounds, the Mn impurity is electrically neutral, whereas in (III,Mn)V compounds the Mn ions act as effective mass acceptors[66]. The coupling J_c is usually taken to be antiferromagnetic for electrons in (II,Mn)VI compounds [75–79], the local moments are thereby aligned antiparallel to the itinerant carriers. One Mn spin after the other is aligned according to the interaction with the environmental carrier spin moment. This indirect interaction between Mn ions, called *Zeners kinetic-exchange interaction* [65], induces *carrier-mediated ferromagnetism* in DMS.

Quantum dots doped with a single magnetic impurity have been exhaustively studied [57, 58, 72, 73, 75, 76]. Investigation of carrier induced magnetic correlations between the Mn spins already start to emerge when two [75, 76] or more impurities are embedded in the quantum dot. As pointed out in [78], competition between intercarrier Coulomb correlations and spin–spin interaction modifies the simple picture which suggests a maximum of the Mn magnetization at half filling of carrier-energy shells (maximum carrier spin according to Hund’s rule filling) and a vanishing magnetization at completely filled shells. For a quantum dot containing three Mn impurities and two electrons Fig. 4.8 reflects the complex interplay between the competing energy scales. The three impurity spins are not placed on

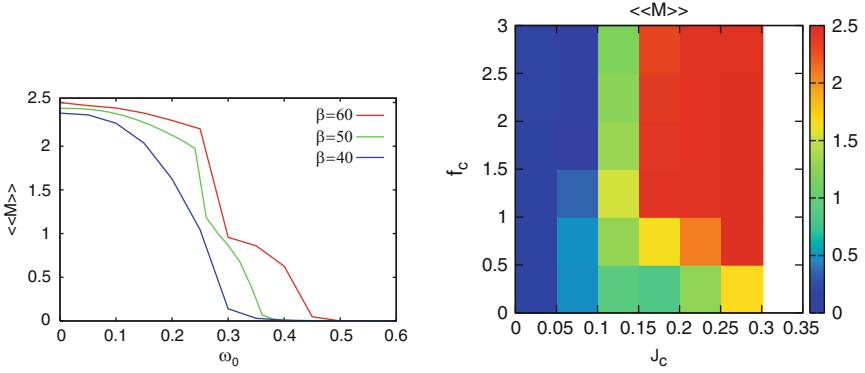


Fig. 4.8 Expectation value of the magnetization $\langle M \rangle$ (left) as a function of the dot confinement at different temperatures $\beta = 40, 50, 60$ corresponding to $T = 3.71, 2.97, 2.48$ K in (Cd,Mn)Te ($B = 0$, $N_c = 2$); Phase diagram (right) of the magnetization in dependence of the Coulomb coupling f_c and the exchange coupling J_c at a constant confinement ($\hbar\omega_0 = 0.5$ Ryd) of the dot. All values are in units of the effective Ryberg

a straight line. To compute their total magnetization, we use the self-consistent approach introduced in [77].

At low temperature, Fig. 4.8 reveals different features in the magnetization at increasing confinement strength $\hbar\omega_0$ of the dot. Up to a confinement strength of $\hbar\omega_0 \approx 0.3$ Ryd*, the magnetization is nearly saturated, dropping thereafter to a value which indicates that on average only one Mn spin is aligned with the electron spin. This is a result of a triplet-singlet transition in the electron state when the confinement energy overcomes the exchange interaction of the electrons. Note, that despite a vanishing spin polarization of the electrons, a residual Mn magnetization can be observed which results from the odd number of Mn atoms. The continuous decrease of the magnetization beyond this transition point can be attributed to thermal effects [77].

On the right hand side of Fig. 4.8, we see that at strong confinement ($\hbar\omega_0 = 0.5$ Ryd*) and without Coulomb interaction ($f_c = 0$, where f_c has been introduced to scale the strength of the Coulomb interaction. It can be viewed as the inverse of an effective dielectric constant) the magnetization does not attain its maximum value. Only the combination of Coulomb and spin-spin-interaction enables a triplet state of electrons in mutual accordance with a saturation magnetization of the Mn spins.

When we introduce a manganese impurity into *GaAs*, it will act as an acceptor. The hole thereby introduced into the valence band has a bounding energy of 112.4 meV. While electrons at the bottom of the conduction band of III-V-semiconductors are well described by an effective mass approach in single-band envelope function approximation, spin-orbit coupling and band structure effects have to be taken more seriously when treating holes at the top of the valence band. Following Kohn and Luttinger [82, 83], we can describe the wave function of the hole by taking only the topmost four valence bands into account and treating the

influence of all the others as a perturbation. The total angular momentum of the hole is a good quantum number to describe the Bloch bands in a crystal. It is the sum of the hole spin and the (atomic) angular momentum of the band which is $l = 1$ for GaAs. The topmost valence bands consist of states with total angular momentum $J = 3/2$ which split into four magnetic subbands labeled by $J_z \in \{\pm 3/2, \pm 1/2\}$. The effective mass of the holes is anisotropic. Owing to their larger mass in z -direction, $J_z = \pm 3/2$ states are called heavy holes. Accordingly, $J_z = \pm 1/2$ bands are named light hole bands. In the plane perpendicular to the z -axis, the order of the masses is inverted. The heavy holes are light and vice versa. As in the case of conduction band electrons confining potentials as well as long-range interactions are treated within the envelope function approximation. Typically, the confinement in z -direction is the strongest causing the z -mass to determine the energetic order of the bands. In this case, the ground state of the hole is mainly composed of states from $J_z = \pm 3/2$ bands. With strong z -confinement and weak confinement in the lateral (xy -)plane, the coupling between the four bands is weak. With stronger lateral confinement, the mixing between heavy and light hole bands is enhanced, leading to an increase of the hole mass in the xy -direction. Due to time inversion symmetry at vanishing magnetic field, the ground state degenerates with respect to reversal of the total angular momentum. In GaAs, the Zeeman energy of the hole $2\mu_B\kappa BJ_z$ determines the order of the energetically lowest states in weak magnetic fields. Here, κ is a material-dependent parameter. In stronger fields, only the $J_z = +3/2$ state efficiently couples to the light hole bands. The other bands become decoupled. This behavior leads to a crossing of the lowest states and for sufficiently strong lateral confining potentials an ordering opposite to the Zeeman term at all magnetic field values occurs [84]. In InAs, in contrast, the Zeeman energy of the hole is much larger and the Zeeman term determines the ordering of states in the whole magnetic field range.

If due to the geometry of the confining potentials, one pair of bands becomes dominant, then the lowest levels of the hole can be described by a single-band theory with a single effective mass similarly to an electron in the conduction band. Only then, the spectrum for a parabolically confined hole becomes approximately equidistant. The magnetic field, however, changes the band coupling and, therefore, alters this effective mass. In small dots, such as self assembled quantum dots, the ground state consists almost totally of $J_z = \pm 3/2$ bands.

In the envelope function approximation, the basis set of hole wave functions is formed from products of an envelope function F_m and a Bloch function u_{J_z} . For circular symmetric confining potentials $M = J_z + m$, i.e., the sum of the z -component of the total angular momentum of the bulk hole J_z and of the orbital angular momentum of the envelope function m , is a good quantum number. Accordingly, the energy eigenstates of the holes are superpositions of the form

$$\Psi_{M,J_z} = \sum_{J_z} F_{M-J_z}(\mathbf{r}) u_{J_z}(\mathbf{r}).$$

Mixing of band states due to the confining potential in a quantum dot is therefore accompanied by mixing of envelope functions with different orbital angular momentum.

Several approaches have been taken to model the interaction between the magnetic manganese acceptor and the valence band hole [85–87]. Within the envelope function approximation, the attractive Coulomb interaction between the negatively charged manganese ion and the positive hole can be modeled by a $1/r$ potential with a correction accounting for a distance dependent effective dielectric constant. This correction usually affects only the ground state.

Additionally, the p – d exchange coupling between the localized Mn- d -electrons and the valence band hole can be modeled by a Heisenberg type interaction between the manganese spin $\mathbf{S}$ and the total spin $\mathbf{J}$ of the hole. Due to the short range of the p – d -exchange interaction, $J_{pd}(\mathbf{R} - \mathbf{r})$, the effective interaction potential in the envelope function approximation is similar to (4.3.4), where the pointlike disturbance acts on the envelope function. Its expectation value in a specific hole state with angular momentum J_z is given by [88]

$$\langle \Psi_{J_z} | J_{pd}(\mathbf{R}_I - \mathbf{r}) \mathbf{S} \cdot \mathbf{J} | \Psi_{J_z} \rangle = \overline{|f_m(\mathbf{R}_I)|^2} \frac{J_{pd}}{3} \langle S_z | \langle J_z | \mathbf{S} \cdot \mathbf{J} | J_z \rangle | S_z \rangle.$$

$\overline{|f_m(\mathbf{R}_I)|^2}$ is the averaged value of the envelope function over the unit cell containing the impurity. The effective exchange coupling J_{pd} amounts to 40 meV (nm)³ [89]. For the following considerations, a single manganese atom is assumed to be located at the center of the dot. Due to the large extend of the hole wave function, its overlap with the manganese atom is small. Therefore, the energy contribution due to the spin–spin interaction is typically much smaller than the splitting of the hole states in a pristine dot. Therefore, only the ground state of the hole will participate in the low-energy sector of the Mn-doped quantum dot (Fig. 4.9).

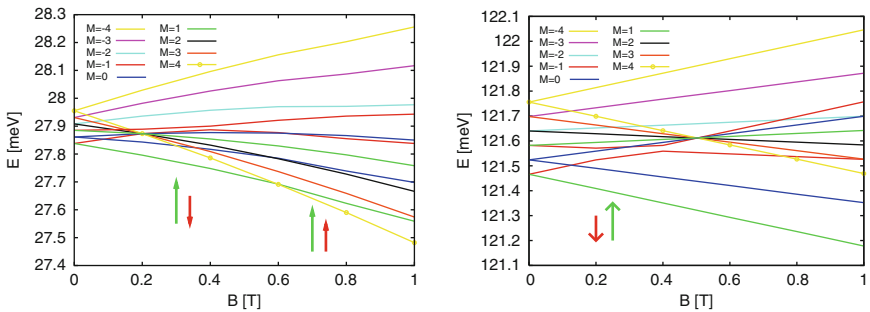


Fig. 4.9 Hole spectrum of a *GaAs* quantum dot with a manganese impurity at the center for different confinement. The confining potentials for the dot on the *right* are approximately five times stronger than for the dot on the *left*. The *arrows* show the manganese (*green*) and hole (*red*) spins of the lowest state. The magnetic field points in ($-z$)-direction. Due to a much higher localization of the hole at the manganese site in the right figure the antiferromagnetic p – d -coupling plays the dominant role

For vanishing magnetic field, the ground state is doubly degenerate due to time inversion symmetry of the Hamiltonian [90]. In this state, the hole and manganese spin are aligned maximally antiparallel, i.e., the degenerate spin states are

$$|J_z = +3/2\rangle |S_z = -5/2\rangle \text{ and } |J_z = -3/2\rangle |S_z = +5/2\rangle.$$

The magnetic field lifts the degeneracy. For non-vanishing magnetic fields, the alignment of the manganese spin in *GaAs* follows the Zeeman term. The alignment of the hole depends on the confining potentials. Strong lateral confinements aligns the hole spin opposite to the Zeeman term. This alignment of the hole is also favored by the antiferromagnetic *p-d* exchange coupling, which is enhanced by the stronger concentration of the hole wave function at the manganese site. Therefore, in weak magnetic fields in (+*z*-direction) and strong lateral confinement, the ground state will be $|J_z = +3/2\rangle |S_z = -5/2\rangle$ while for strong magnetic fields and weak band coupling the Zeeman energy will dominate and $|J_z = -3/2\rangle |S_z = -5/2\rangle$ becomes the ground state. In each case, the alignment of the manganese is independent of the hole state and follows always the external magnetic field. Thus, in *GaAs*, band mixing and *p-d* exchange simultaneously enhance the tendency of an antiparallel alignment of the hole and Mn spins. This constructive interplay is material dependent and cannot, e.g., be observed in II-VI semiconductors where the manganese impurity is charge neutral.

For impurities not in the center of the dot, the circular symmetry is broken and the total angular momentum ceases to be a good quantum number. Despite this, the eigenstates retain a dominating component with the aforementioned spin alignments.

4.3.5 Correlations Beyond Hund's Rule

The competition between Coulomb interaction and single-particle excitations in quantum dots does not only give rise to a reordering of levels. A genuinely quantum effect is the formation of many-body correlations when several single-particle configurations become quasi-degenerate with regard to their respective interaction energy. Entanglement of different configurations, technically speaking a superposition of several Slater-Determinants, is the consequence. In quantum dots, these many-body correlations gain importance when the total spin of the quantum dot state does not reach its maximum value, i.e., when a parallel spin alignment of all carriers in the dot cannot be achieved and, thus, exchange interaction partly vanishes [27]. Transport spectroscopy offers a unique tool to unravel these many-body correlations.

Similar to the optical oscillator strengths which govern the intensity of optical spectra, the spectral weights

$$S_{\alpha\beta} = \sum_i \left| \langle \alpha | d_i^\dagger | \beta \rangle \right|^2$$

determine the amplitude of transport resonances [17, 91–99]. Moreover, like the optical oscillator strengths, they provide selection rules for the availability of various transport channels. Measuring the overlap of the product wave function – built from the incoming electron wave function and the N -particle state $|\beta\rangle$ of electrons in the dot – with a $(N + 1)$ -particle state $|\alpha\rangle$ of the quantum dot, the spectral weights sensitively distinguish between correlated and uncorrelated few-particle states.

The most obvious selection rules result from spin quantization. Since an electron tunneling into the dot has total spin $\sigma = 1/2$ and might come in two different polarizations, the initial and final dot state must differ in both, their total spin and its z -component by one half, $\Delta S = 1/2 = \Delta S_z$. Therefore, transitions between spin-forbidden multiplets will not appear in the excitation spectrum [17, 100]. These spin-selection rules are lifted when the spin no longer is a good quantum number for carriers in the dot, as it is, e.g., the case for dots with magnetic impurities.

The strong variation of the ground state magnetoconductance shown in Fig. 4.10, however, does not result from spin-correlations, but rather reflects strong orbital correlations in the wave functions of a parabolic quantum dot containing two/three electrons [92, 97, 98, 101]. In a correlated electron state, many single-particle orbitals are partially occupied, and transitions between states with different particle numbers usually require a severe reorganization of the electron configuration. This leads to a drastic reduction of the spectral weight (4.3.5) and correspondingly to a small differential conductance accompanied with this transition. This is the reason for the extremely low conductance which can be observed in Fig. 4.10 at high magnetic fields ($B > 5T$). The large angular momentum ground states ($M(N_e = 2) = -3$, $M(N_e = 3) = -9$) can be viewed as finite size precursors of a strongly correlated bulk fractional quantum Hall state. In contrast, the high magnetoconductance in the

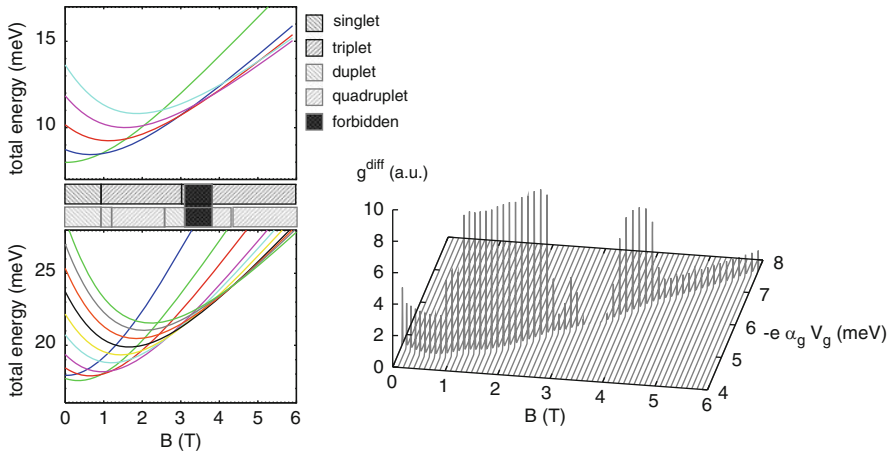


Fig. 4.10 Differential conductance $g = \partial I / \partial V_{\text{bias}}$ as a function of a magnetic field vertically applied to a parabolic quantum dot at the transition between two and three particles in the dot. $\hbar\omega_0 = 2$ meV, GaAs parameters

magnetic field range $1.2\text{T} < B < 2.5\text{T}$ indicates a regime where both the two-particle and the three-particle ground states are only weakly correlated. Indeed, the two-particle triplet state at $M(N_2 = 2) = -1$ has an overlap of 95% with a single Slater-determinant, the overlap of the three-particle quadruplet state at $M(N_e = 3) = -3$ amounts to 84%. The transition between both states can be accomplished by merely adding the third electron. Note that the transition between the low-spin states at even smaller magnetic fields again indicates strong orbital correlations. A spin-forbidden transition regime is also visible in Fig. 4.10.

4.4 Transport Beyond Spectroscopy

So far we have considered transport as a spectroscopic tool which allows to study the electronic structure of quantum dot systems. However, coupling a quantum system to reservoirs inherently alters its eigenmodes. If the tunnel coupling is weak, this effect may be negligible. Up to now, we only considered stationary, incoherent, sequential transport, which is conveniently described by a Markovian rate or master equation

$$\begin{aligned} \frac{dP_i}{dt} &= - \sum_{j \neq i} \Gamma_{i \leftarrow j} P_j = 0 \\ \sum_i P_i &= 1, \end{aligned} \quad (4.10)$$

where i numbers the single-particle states, the P_i are stationary probabilities for occupation of state i , and the rates $\Gamma_{i \leftarrow j}$ for transitions from state j to i can be obtained with Fermi's Golden Rule (see, e.g., [102]) and by summing over all states of the equilibrium leads. Whenever (a) sequential tunneling is not sufficient, (b) a dynamical generation of coherent superposition states of the unperturbed dot has to be allowed for, or (c) the real-time dynamics of the system are to be investigated, however, a more sophisticated approach is required.

Many non-equilibrium transport theories, suitable to be applied to these kinds of problems, base on the *Keldysh formalism* [103, 104]. Among these, the real-time diagrammatic technique by Schoeller et al. [15] is well-established to investigate stationary transport with weak dot-lead coupling. It allows to construct a systematic perturbation expansion in orders of the tunnel coupling for transition rates and observables (such as tunneling current) and to treat coherent superpositions via a master equation for the *reduced density matrix* $\hat{\rho}_{\text{dot}}(t) := \text{Tr}_{\text{leads}}[\hat{\rho}(t)]$ of the central region. For the stationary state (denoted by superscript), this approach yields the *kinetic equation*

$$0 = \frac{d\hat{\rho}^{\text{st}}}{dt} = -\frac{i}{\hbar}[\hat{H}_{\text{dot}}, \hat{\rho}^{\text{st}}] + \hat{\Sigma}^{\text{st}}\hat{\rho}^{\text{st}}, \quad (4.11)$$

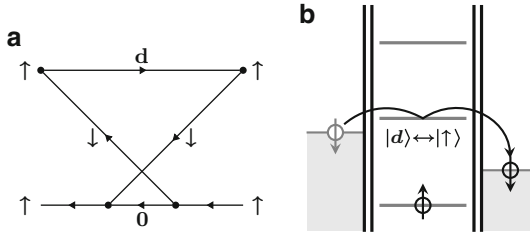


Fig. 4.11 (a) Irreducible Keldysh diagram, contributing to tensor $\hat{\Sigma}^{\text{st}}$ in (4.11) of an SLQD system. The horizontal directed lines (Keldysh branches) symbolize free time propagation of dot states (empty dot: 0, spin-up (down) electron: *uparrow* (*downarrow*), double occupation: d), the lines connecting vertices on the Keldysh branches denote tunneling of electrons between leads and dot. This diagram can be associated with a coherent two-electron process (cotunneling). (b) In the same configuration as Fig. 4.2b, coherent two-electron processes can lead to a finite cotunneling current. Shown is an elastic process of one spin-down electron tunneling twice through a virtual state. The dot is in a spin-up stationary state

where the elements of the 4th-rank tensor $\hat{\Sigma}^{\text{st}}$ are given by the sum over all *irreducible* Keldysh diagrams.³ Figure 4.11 shows an example for such a Keldysh diagram for a single-level quantum dot system and the kind of process which it can be associated with. If we take into account second-order tunneling (or cotunneling), we expect a finite current through the dot, which scales as $|\gamma|^4$ in the small coupling γ (Fig. 4.11b) even when sequential transport is Coulomb- or spin-blocked as in Fig. (4.2b, c).

But even in the sequential regime, dynamical generation of coherent superposition states can considerably affect the transport behavior. An example of this situation was presented by Wunsch et al. [20, 106]. It was shown that in sequential transport through a double quantum dot, the dot energies are effectively renormalized due to the coupling to the leads. The resulting deviations of the tunneling current from the bare current are of the same order of magnitude as the latter, which is calculated for a *diagonal* reduced density matrix $\hat{\rho}^{\text{st}}$. This is illustrated by Fig. 4.12.

As we saw above, however, it is not always sufficient to account for single-particle tunneling only. This is the case for transport in the deep Coulomb-blockade regime, where the sequential current is exponentially suppressed. Since the lowest order perturbation vanishes in this regime, at least cotunneling has to be taken into account to obtain a physically sound result for the stationary state of the system.

An example of a very simple model with cotunneling as the dominant transport process is given in [107–109]. The SLQD's stationary state and transport are studied in the one-particle Coulomb-blockade valley with non-degenerate single-electron levels. As shown in Fig. 4.13a, b, the consideration of elastic and inelastic cotunneling processes reveals a rich internal structure in the otherwise completely uniform

³ Kinetic equations can, however, often be rewritten as effective rate equations [105].

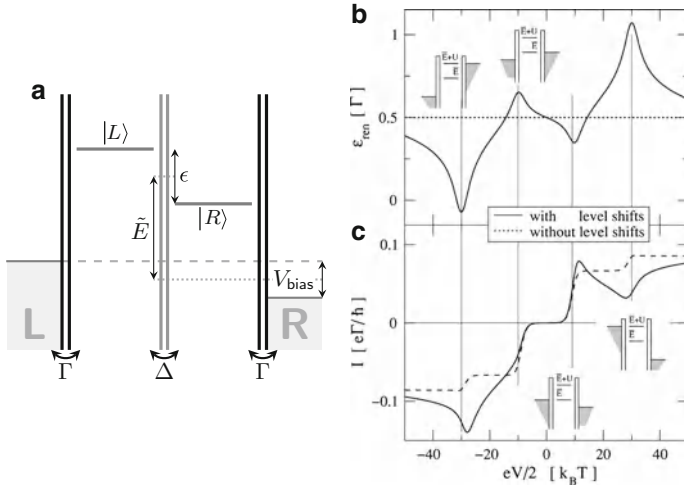


Fig. 4.12 (a) Energy scheme of a double quantum dot. Transport channels in *left* and *right* dot connect empty dot states with the respective single-particle ground states $|L\rangle$ and $|R\rangle$. The tunneling barriers between double dot and reservoirs are characterized by a scalar parameter $\Gamma \propto |\gamma|^2$, the intra-dot coupling by Δ . Parameters $\tilde{E}$ and ϵ denote mean value of and relative distance between the dot states, respectively. In the sequential transport regime, the bare distance ϵ is renormalized to ϵ_{ren} due to the dot-lead-coupling: (b) Shown is ϵ_{ren} against the bias voltage. Renormalization is maximal whenever the chemical potential of a lead is in resonance with a transport channel for either single ($\tilde{E}$) or double occupation ($\tilde{E} + U$, see insets). (c) Current I versus bias voltage for dot renormalized level spacing (*solid*) and without renormalization (*dashed*). The lower (*higher*) ϵ_{ren} is compared to ϵ , the more (*less*) current flows compared to the bare value. Parameters are $\epsilon = \Delta = \Gamma/2$. All three taken from [106]

Coulomb diamond. Not only is cotunneling necessary to uniquely determine the spin-up and spin-down occupations, it also leads to signatures of the single-particle spectrum and a sequential current in face of Coulomb blockade. As soon as eV_{bias} exceeds the excitation energy $\delta = E_{\uparrow} - E_{\downarrow}$, inelastic processes (Fig. 4.13c) dynamically populate state $|\uparrow\rangle$. This population leads to a sequential current, when one of the electrochemical lead potentials is in resonance or below the channel $|0\rangle \leftrightarrow |\uparrow\rangle$ (Fig. 4.13d).

4.5 Outlook

These few examples show that even small conceptional steps, which go beyond incoherent, sequential transport, may yield new physics and strongly non-linear transport effects. In recent years, several methods and theories were developed, which aim to further expand the range of accessible parameters, reproducible physical effects, and treatable systems. For the stationary case, the Bethe ansatz [110–112] is suitable to treat quantum dots with strong (non-perturbative) tunnel

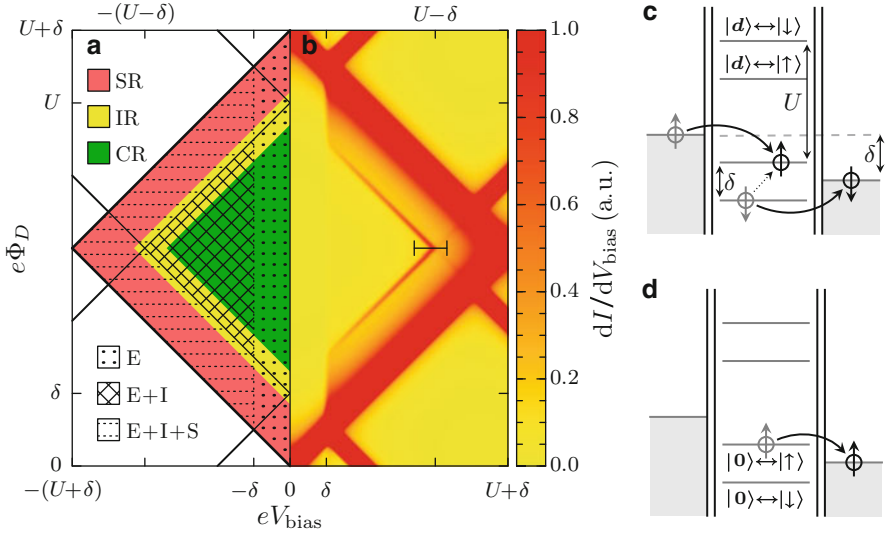


Fig. 4.13 (a) Schematic picture of the *diamond-shaped* (one-particle ground state) cotunneling regime for an SLQD showing its partition into areas with different possible tunneling processes (*hatched areas*) as well as into core (CR), shell (SR), and intermediate region (IR), in which the occupation of $|\uparrow\rangle$ and $|\downarrow\rangle$ are determined either by cotunneling, sequential tunneling, or both (*shaded areas*). E is for elastic (Fig. 4.11b), I for inelastic cotunneling (see below), and S is for sequential tunneling (Fig. 4.2a). (b) Differential conductance dI/dV_{bias} (arbitrary units) against eV_{bias} with parameters $\delta = 45k_B T$, $U = 225k_B T$, and $\Gamma = 4.5 \times 10^{-3}k_B T$. The *sharp line* in the part of IR, in which $eV_{\text{bias}} > \delta$, is the signature of cotunneling mediated sequential transport. Both (a) and (b) can be extended to regions with opposite sign of V_{bias} . Taken from [107]. (c) Inelastic cotunneling that excites the SLQD from state $|\downarrow\rangle$ to $|\uparrow\rangle$ (*dotted arrow*). Via the coherent two-electron process, the excitation energy δ is transferred to the dot and a dynamical population of state $|\uparrow\rangle$ is generated. (d) The population of $|\uparrow\rangle$ due to inelastic cotunneling is reduced to almost zero by sequential tunneling, once the chemical potential of a lead aligns with channel $|0\rangle \leftrightarrow |\uparrow\rangle$

couplings. Various renormalization group approaches [113–119] as well as real-time quantum Monte-Carlo (RTQM) [120–122], and the flow-equations method [123] go also beyond the weak coupling regime and allow to study real-time dynamics. With a similar scope of application, although restricted to very small quantum dot systems, the numerically exact iterative summation of the fermionic path integral (ISPI) [124] permits to simulate long propagation times and avoids the fermionic sign problem of RTQM.

The results that are obtained by these methods are not only interesting in their own right, for they convey insight into the physical structure of the quantum system at hand. They also encourage to ask the question, whether and how quantum dot systems may be deployed as novel nanoscale devices that exhibit different physical effects and/or superior (spin-)electronic properties compared to conventional (classical) components.

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Chapter 5

Far-Infrared Spectroscopy of Low-Dimensional Electron Systems

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Abstract In this chapter, we review far-infrared (FIR) transmission and photoconductivity spectroscopy on quantum materials which are fabricated with sophisticated micro and nano lithographic techniques from modulation-doped InAs and GaAs heterostructures. We will show that it is possible to measure the FIR response of quantum dot arrays where each quantum dot contains just one single electron. We will demonstrate that with increasing electron number and tailored geometrical shape, the charge density excitations are dominated by a complex interplay of one-electron and many-body effects. We will address circular and elliptically shaped quantum dots, the manifestation and the breaking of the Kohn theorem, the interaction with Bernstein modes, anticyclotron motion in antidot arrays, and other examples. Photoconductivity measurements are extremely sensitive. They allow the observation of a quantized plasmon dispersion in the edge regime of two-dimensional electron systems (2DES) under the condition of the quantum Hall effect (QHE) and the excitation of collective spin excitations which becomes possible in materials with strong spin-orbit interaction.

5.1 Introduction

The Hydrogen and the Helium atoms are the most prominent textbook examples to explain quantum mechanics and to understand energy quantization in confining potentials, the formation of wave functions, spin-orbit effects, Coulomb, exchange and correlation effects, and the Pauli principle. Historically, understanding of these fundamental systems came dominantly from spectroscopy investigations. So it was quite natural to also study quantum materials like quantum dots, quantum wires, antidot arrays, or structured two-dimensional electron systems (2DES) with spectroscopic methods. Since typical confinement energies are in the range of 1–10 meV, corresponding to about $10\text{--}100\text{ cm}^{-1}$ on the wave number scale, and to wavelengths of $1,000\text{--}100\text{ }\mu\text{m}$, far-infrared (FIR) spectroscopy is a very powerful tool to study the dipole excitation in these systems. The quantum materials do not just mimic the systems in the real world. Due to the possibility to vary easily the number of

the electrons, the confining potential through the lithographically defined shape or through gate voltages, and with the application of external magnetic fields, it is possible to realize unique conditions and to study fundamental interactions that are not present in real nature. This aspect of fundamental physics is the strong motivation and the beauty of the research on quantum materials. An alternative and complementary approach for the investigation of fundamental excitations in low-dimensional quantum systems is the resonant electronic Raman spectroscopy, which will be reviewed in the Chapter by Kipp, Schüller and Heitmann.

In this chapter, we will first give a brief introduction into the experimental techniques and the preparation of sophisticated samples. We introduce theoretical models to describe the dynamic excitation in 2DES, in quantum wires, quantum dots, and antidot arrays. We will then review selected experiments to give examples of the unique excitations that can be studied in these tailored man-made systems.

5.2 Experimental FIR Spectroscopic Techniques

A typical experimental set up is shown in Fig. 5.1. FIR radiation from a Fourier transform spectrometer (FTS) is guided through an oversized brass wave guide (diameter 10–15 mm) into the center of an axial superconducting magnet in a He cryostat. The radiation is transmitted through the sample and detected by a sensitive Si bolometer (operated at 4 or 2.2 K) that is mounted far enough to minimize the effects of the stray fields from the magnet. The sample is typically wedged by 3 degrees to avoid interference effects in the samples and related features in the spectra. In our lab, we can tune the magnetic field B up to 16 T. The sample temperature is usually 2.2 or 4 K. Using variable temperature inserts (VTI), it is also possible to vary the temperature from 1.7 K to above room temperature. For some experiments, in particular, the photoconductivity measurements, it is extremely helpful to have even lower sample temperatures, for example, 0.3 K in a ^3He insert. However, it is a real challenge to construct a ^3He insert with waveguides that fit into the standard 52-mm diameter bore of a magnet. Thus one has to reduce the size of the waveguide, which means less intensity, in particular at long wavelengths. It is very convenient to use a broad-band FTS. A mercury lamp or a glow bar serves as the light source. Depending on the available beam splitters, the sensitivity of the detector and the size of the waveguides, one can cover the frequency regime starting at about 5 cm^{-1} up to 500 cm^{-1} in our set up that is optimized for the FIR regime. In a scanning FTS, the signal from the bolometer is sampled along the path of the moving mirror which produces an interferogram. This is numerically Fourier transformed into the wave number depending spectrum. The spectrum depends on the spectral efficiency of the bolometer, the mirrors, the beamsplitter, the waveguide, and so on.

In our experiments, we are interested in the relative change in transmission

$$\frac{\Delta T}{T} = \frac{T(V_g) - T(V_t)}{T(V_t)} \quad (5.1)$$

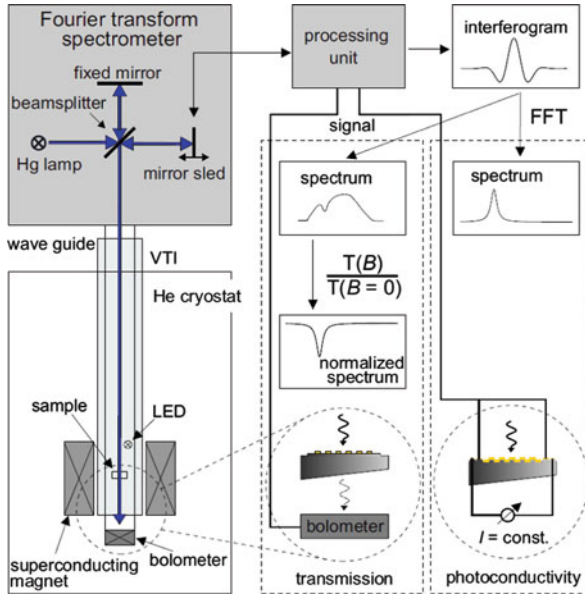


Fig. 5.1 Scanning Fourier transform spectrometer and wave guide systems for FIR transmission and photoconductivity measurements in an axial magnetic field (adopted from [4])

which can be directly and absolutely related to the dynamic conduction of the electron system (see below). Here it is extremely convenient if one can switch the electron in the system on and off by a gate voltage V_g and threshold voltage V_t , respectively. With special tricks, coadding, say alternatively 10 interferograms with gate voltage on and off, over several hours of measurements, one can eliminate long-term drifts of the setup and measure relative changes in transmission as small as 0.1%. This enables one to resolve also the very tiny signal on quantum dot arrays where each quantum dot contains only one or two electrons. Such techniques are described in detail by Batke and Heitmann [1] or Meurer et al. [2]. If it is not possible to change the number of electrons, however, the investigated resonances have a strong enough dispersion in a magnetic field, one can also use different magnetic fields to normalize the spectra. Here it is not so easy to avoid long term drifts. Also, a residual impact of the magnet's stray fields slightly changes the sensitivity of the detector, resulting in small modifications of the base line in the normalized spectra. Nevertheless, resonances with $\Delta T/T$ larger than about 1% can be clearly resolved.

For the photoconductivity measurement, the sample is used as its own detector. A current is fed into the contacted sample and the change in the conductivity of the sample is Fourier transformed. More details on these techniques are described later in Sect. 5.6.

5.3 Preparation of Arrays of Quantum Materials

Due to the long wavelength of the FIR radiation, and in a waveguide setup under cryogenic conditions, it is not possible to focus the radiation on to a small spot. Usually, a brass Winston cone is used in front and behind the sample with a focus spot size of 2–5 mm in diameter. Thus, to achieve large signals, one needs samples with a large active area, 2 mm or even up to 5 mm in diameter. If one wants to prepare a quantum dot array, let us say with a period of 330 nm, one has 10^8 dots in total on an active area of 3 mm^2 . It is impractical to write so many dots with electron-beam lithography. Therefore, interference (also called holographic) lithography is a very powerful tool to prepare such structures. The photoresist layer, which is spincoated onto the semiconductor wafer, is exposed to the interference pattern of two coherent expanded laser beams of wave length λ_L . This results in a periodic line grating where the grating period is determined by $a = \lambda_L / 2 \sin(\theta)$ and 2θ is the angle between the two laser beams. For large angles and using the blue 457 or 384 nm UV lines of an Argon laser, one can reach grating periods as small as 200 nm. One can then rotate the sample, for example, by 90 degrees around its normal, and, if desired, change the angle between the laser beams, and expose a second grating. Controlling the two exposure times and the subsequent development process, one can create dot arrays, antidot arrays, width-modulated quantum wires, as shown, for example, in Fig. 5.2b, or elliptically shaped dots. These photoresist patterns are the starting point for further processing. With these techniques, we can fabricate different types of quantum structures: (a) gated structures, or (b) etched structures, (c) a combination of both.

In Fig. 5.2a, we sketch a field-effect confined quantum dot array. A holographic photoresist dot array is prepared on top of a modulation-doped heterostructure. A thin Ni or Ti gate, which is semitransparent for FIR radiation, is evaporated onto the sample. With a negative voltage applied to the gate, the electrons are depleted in the regions between the photoresist dots and are thus laterally confined. A semitransparent Si-doped backgate, at a distance of about 200 nm from the 2DES, is inserted during the MBE growth and allows one to charge the dots. The typical period of the array is $a = 200\text{--}1,000 \text{ nm}$ and the electronic diameter, i.e., the spread of the electronic wave function in the dot, can be made as small as 50 nm. In such a device, as demonstrated by Meurer et al. [2] it is possible to charge each dot of the array simultaneously with $N = 1, 2, 3, \dots$ electrons. These well-defined electron numbers are controlled by the large Coulomb charging energy in the dot, which requires an increase of the gate voltage by typically 10 mV to transfer the next electron into the individual dots.

The advantage of such gated structures is the tuneability of the electron numbers. The challenge is, however, that it is not easily possible to avoid leakage currents due to some residual defects over such a large gate area. Another point is that the potential in gated structures is usually very shallow, resulting in low confinement and excitation energies. Here it is desirable to prepare samples with a very small array period. This would also increase the total number of dots, the total number of electrons at the same occupancy, and thus the signal. However, with a smaller

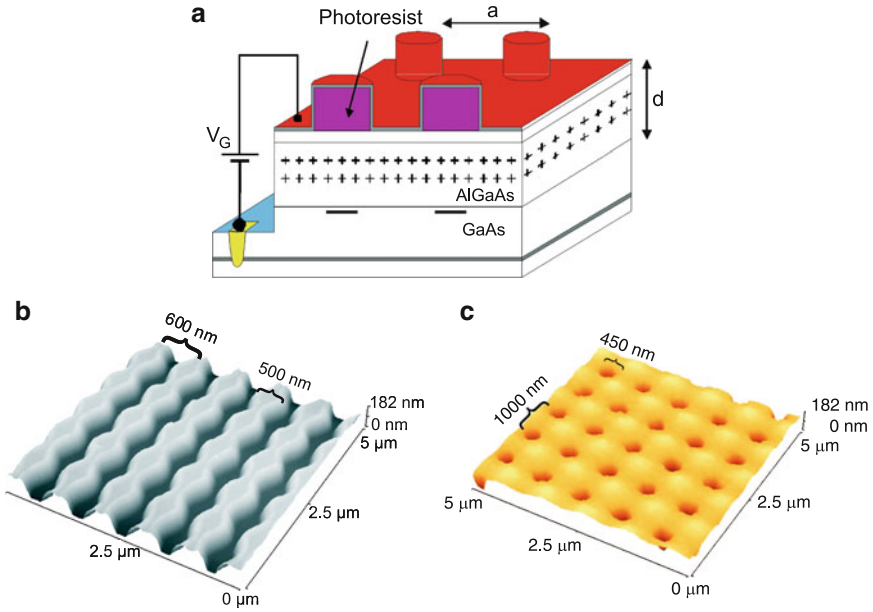


Fig. 5.2 (a) Sketch of a field-effect confined quantum-dot array. With a negative gate voltage electrons in a modulation-doped AlGaAs/GaAs heterostructure are depleted except under the photoresist dot and are thus laterally confined. (b) AFM image of a quantum-wire array after dry etching of a modulation-doped AlGaAs/GaAs heterostructure and evaporating of a Ti gate. The period is $a = 1,000$ nm. In the narrow regions of the lithographically defined laterally modulated wires the electrons can be completely depleted thus that elliptical dots are formed. The lithographic width of the wire, at maximum, is $w_x = 500$ nm. (adopted from [3]) (c) AFM image of an antidot array after dry etching. The period is $a = 1,000$ nm, the geometrical diameter of the holes is $2R = 450$ nm

period, the spatial Fourier component of the confining electric field will also decay more strongly in the growth direction. So the electron layer has to be brought in close distance to the modulated gate. As a rule of thumb, the distance between the modulated gate and the electron system should be less than $1/10$ of the period. This means one has to use the so-called shallow HEMTs which are not easily prepared with high quality and are unfortunately often hampered by leakage currents.

So another approach for the fabrication of quantum structures is etching. The structured photoresist is used as an etching mask. Usually, reactive ion beam etching with optimized low acceleration voltage and additional annealing is applied to minimize and heal damages from the etching process. One distinguishes so-called ‘deep’ or ‘shallow’ mesa etching. In the former case, the etching is executed all the way through the active electron layer; in the later case, only the modulation-doped AlGaAs is etched, depleting the system under this regime. These etching processes for quantum materials are discussed in more detail by Grambow et al. [5].

In Fig. 5.2b, we show an AFM image of a quantum wire array that has been fabricated by deep mesa etching. In addition, a thin metal layer, evaporated on the

top and side walls of the wires, serves as a gate and allows one to tune the electron density in the wire and eventually deplete the system at the narrow constrictions; thus elliptically shaped dots are formed (for more details, see [3]). Figure 5.2c shows an AFM image of an antidot array, where an array of geometrical holes is etched into a heterostructure.

GaAs has the intrinsic property to form inherently negatively charged states at the surface. This is extremely helpful for deep mesa etched samples since some of the electrons from the doped regime are transferred into these states on the sidewalls of the etched structures and help to confine the electron in the active channel of the wire or dot. So the actual ‘electronic’ size of the system, the spread of the wave functions, is smaller than the geometrical size, separated by the so-called ‘lateral depletion length’ of typically 100 nm from the geometrical edge. This lateral depletion has been carefully studied by Riege et al. [6]. The nice thing about the lateral depletion is that, with the confined electron being far away from the geometrical edge, any small-scale fluctuation or roughness of the etched profile will not have a large impact on the smoothness of the confining potential, since their corresponding Fourier field component have died off. So, although etched quantum wires or dots sometimes look quite rough in REM or AFM pictures, the same samples show beautiful sharp resonances in the spectra or well-resolved anticrossing in the dispersions, indicating the smoothness and the large-area homogeneity of the actual confining potential.

5.4 Theoretical Models

In our FIR transmission experiments, we measure the relative change $\Delta T/T$ which for an infinite homogeneous 2DES can be calculated in a straightforward way from electrodynamics. Assuming normally incident FIR radiation, it is

$$\frac{\Delta T}{T} = \frac{-2\text{Re}(\sigma(\omega))}{\sqrt{\epsilon_0/\mu_0}(1 + \sqrt{\epsilon^*})^2 + \sigma_g} \quad (5.2)$$

Here, ϵ^* is the effective dielectric function of the surrounding media and σ_g the conductivity of the gate, if it applies. This result is correct for small values of N_s and a not too high mobility. If this is not the case, the so-called ‘signal saturation’ occurs, which can also be treated by straightforward linear electrodynamics, but it is not relevant here [7].

The dynamic conductivity in a magnetic field B can be described by a Drude ansatz for left (–) and right (+) circularly polarized radiation:

$$\sigma_{\pm} = \frac{N_s e^2 \tau}{m^*} \frac{1}{1 + i(\omega \pm \omega_c)\tau}. \quad (5.3)$$

Here, N_s is the carrier density, m^* the effective mass, τ the Drude relaxation time, which is related to the mobility $\mu = e\tau/m^*$, and $\omega_c = eB/m^*$, the cyclotron

frequency. In a magnetic field B , the denominator in (5.3) determines the well-known cyclotron resonance at frequency ω_c . Note that we measure $\Delta T/T$ directly in %. So we can determine, combining (5.2) and (5.3), absolutely the density N_s from the amplitude, the scattering time τ , and thus the mobility μ from the width of the cyclotron resonance, and m^* from the resonance frequency. In the following, we assume in the theory and have in all the experiments discussed here the so-called Faraday configuration, i.e., the magnetic field is perpendicular to plane of the sample and parallel to the incident radiation.

Besides the cyclotron resonances, the 2D plasmons are characteristic excitations in an infinite 2DES. They have the dispersion [8, 9]

$$\omega_p^2 = \frac{N_s e^2}{2\epsilon^* \epsilon_0 m^*} \cdot q \quad (5.4)$$

and in a magnetic field, we have magnetoplasmons:

$$\omega_{mp}^2 = \omega_p^2 + \omega_c^2. \quad (5.5)$$

Here, q is the wave vector and ϵ^* the effective dielectric function of the surrounding media, for example, $\epsilon^* = 0.5(\epsilon_{\text{Se}} + 1)$ if the 2DES is located close to the boundary of the semiconductor (Se) and vacuum. At larger q also, so-called non-local effects become important and one has to add a term $(3/4)(v_F q)^2$, where v_F is the Fermi velocity, to (5.4). [8]. Similar effects lead in magnetic fields to an anticrossing of the plasmon dispersion with Bernstein modes [10, 11] at $2\omega_c, 3\omega_c, \dots$. The relative splitting of the disperions in the anticrossing regime is, for a 2DES, $(\Delta\omega/\omega) = (qv_F/\omega_c)^2 = (1/3)a^*q$, where $a^* = a_0\epsilon^*m_0/m^*$ is the effective Bohr radius [12–14].

2D plasmons are charge density oscillations that propagate with wave vector q along the plane of the 2DES. They are accompanied by transverse electromagnetic fields which decay exponentially perpendicular to the plane of the 2DES. In this sense, they are directly related to surface plasmons in a metallic slap (and not the volume plasmons in a bulk metal); in particular, they present the limit of an infinitely thin metallic slab as noticed by Ritchie [15] when he first derived the 2D plasmon dispersion.

Although 2D plasmons have transverse components of the electromagnetic field, they do not couple directly to FIR radiation. Rather, grating couplers are necessary to couple the FIR radiation to the surface plasmons and provide the necessary wavevector. For example, a periodic array of metallic stripes of period a spatially modulates the incident FIR radiation and excites plasmons at wave vector $q_n = n(2\pi)/a$ with $n = \pm 1, \pm 2, \dots$. In principle, any periodic modulation near the electron system, in particular, a periodic modulation of the electron density itself, will lead to a grating coupler effect and will couple 2D plasmons with radiation. An extended abstract on 2D plasmons was given by Heitmann [16].

A first example for a confined system is an infinitely long quantum wire in y direction, where electrons are confined in x direction. In a simple plasmon-in-the-box

picture, one finds [17]

$$\omega_{\text{cp}}^2 = \frac{N_s e^2}{2\epsilon\epsilon_0 m^*} \cdot \sqrt{\left(\frac{\pi n}{w}\right)^2 + q_y^2}, \quad (5.6)$$

where w is the width of the wire and $n = 1, 2, 3, \dots$ is an index counting the number of plasmon modes due to the confinement in x direction. This model assumes a constant 2D density in the wire over the whole width and ‘ideal’ boundary conditions, resulting in a set of standing wave plasmon modes for the confinement in x direction and freely propagating modes in y direction. It works surprisingly well, as demonstrated, for example, in the experiments of Demel et al. [18] for deep mesa etched quantum wires. For realistic quantum wires, one has to include the real density profile starting from the external potential. Usually, one then has to apply self-consistent Hartree-Fock methods to calculate the screened one-particle potential and the one-particle wave functions of the 1D subbands, and then apply RPA theories to evaluate the dynamic response, for example [19, 20]. It should also be noted that additional logarithmic corrections are necessary if one approaches the ideal 1D case for small values of $w \cdot q$, see e.g., Kukushkin et al. and references therein [21]. In a magnetic field, one finds for the confined modes

$$\omega_{\text{cmp}}^2 = \omega_{\text{cp}}^2 + \omega_c^2. \quad (5.7)$$

An interesting aspect of the finite size of the quantum wire, and thus the presence of edges, is that, in addition to the confined mode discussed above, which has a positive B dispersion, there are edge magnetoplasmon modes, which have a negative B dispersion. For a quantum wire, they cannot be described by a simple analytic expression. We will discuss these modes for quantum dots and antidot arrays below.

Very similar approaches can also be applied for quantum dots. Fetter et al. discussed in several papers circularly shaped dots, assuming different types of density profiles in the dots [22]. He found the following resonance frequencies:

$$\omega_{i\pm} = \sqrt{a_{i1}\omega_{0i}^2 + a_{i2}\left(\frac{\omega_c}{2}\right)^2} \pm a_{i3}\frac{\omega_c}{2}, \quad (5.8)$$

where

$$\omega_{0i}^2 = 0.81 \frac{N_s e^2}{2\epsilon\epsilon_0 m^*} \cdot \frac{i}{r}. \quad (5.9)$$

r is the radius of the quantum dot and a_{nm} are numerical factors close to 1, which depends on the exact shape of the density profile [22].

For all mode indexes i , one finds two modes, one approaches with increasing magnetic field the cyclotron resonance, representing at large B , a cyclotron type motion. The other mode decreases in a magnetic field, representing with increasing B edge magnetoplasmon modes.

Another approach for the calculation of the dynamic excitations in a quantum dot is an atomic-type picture. This is analytically possible if we assume that the external confining potential has a parabolic shape $V(x, y) = 1/2m^*\omega_0^2(x^2 + y^2)$. Since the confinement arises from the electrostatics of the charged donor and surface states, this is in many cases a very good approximation (see for example [6]). We first evaluate the energy levels of a one-electron parabolic quantum dot in a magnetic field, as first calculated by Fock and Darwin [23, 24] and find:

$$E_{n,m} = (2n + |m| + 1)\sqrt{(\hbar\omega_0)^2 + (\hbar\omega_c/2)^2} + m\hbar\omega_c/2 \quad (5.10)$$

For the dipole-allowed transitions $\Delta m = \pm 1$, we find

$$\hbar\omega_{\pm} = \sqrt{(\hbar\omega_0)^2 + (\hbar\omega_c/2)^2} \pm \hbar\omega_c/2 \quad (5.11)$$

The interesting point of a parabolic external potential is that one can show rigorously that the one-electron (5.11) also holds for any number of electrons in the quantum dot. This is the manifestation of the Generalized Kohn Theorem [25, 26]. The original Kohn theorem was formulated for the dipole excitation of the cyclotron resonance in a translationally invariant 2D or 3D system [27]. The dipole excitation in a quantum dot with an arbitrary number of electrons in a parabolic external potential represents a rigid center-of-mass motion of all electrons (and is in this sense a collective excitation!), which is totally decoupled from all internal relative motions. Note that in a real sample it is not easy to change the number of electrons without changing at the same time the external potential. So, in general, changing the gate voltage and thus the number of electrons will nevertheless lead to a shift in the resonance frequency. From (5.11), we find also that for a parabolic potential, cyclotron-like and edge magnetoplasmon type modes with, respectively, positive and negative B dispersion. In contrast to the Fetter approach for an arbitrary density profile, (5.8), for this parabolic profile only one set of modes is dipole allowed. For a parabolic quantum dot, it is also easy to calculate the transition elements and thus determine from an experimentally measured $\Delta T/T$ the number of electrons per dot. The total intensity of both branches is exactly the same intensity as for a cyclotron resonance with the same number of electrons per unit cell area of the quantum dot array. The intensity ratio between the high- and low-frequency resonances is $(I_+/I_-) = (\omega_+/\omega_-)$ [28]. Thus, at high B , most of the intensity is in the high frequency branch ω_+ , whereas the low-frequency edge-magnetoplasmon type branch decreases in intensity.

For circularly shaped dots, the two types of branches are degenerate at $B = 0$. This degeneracy is lifted in quantum dots with elliptical shape. The two resonances at $B = 0$ then represent standing wave confined plasmons, with the higher and lower resonance frequency determined by the length (or potential along) of the longer and shorter axis of the ellipse, respectively. Analytical dispersions are given, for example, in [29, 30].

Another type of quantum material is an antidot array as shown in Fig. 5.2c. Kern et al. [31] were the first to observe a characteristic two-mode behavior, which

we will discuss in more detail below. Following these experiments, Mikhailov and Volkov [32] developed an effective medium theory which gives an implicit expression for the resonance frequencies.

$$1 - \frac{1-f}{\frac{\omega}{\omega_0}(\frac{\omega}{\omega_0} \pm \frac{\omega_c}{\omega_0})} - \frac{f}{\frac{\omega}{\omega_0}(\frac{\omega}{\omega_0} \mp \frac{\omega_c}{\omega_0})} = 0. \quad (5.12)$$

The antidot array is characterized by an areal filling factor $f = \pi R^2/a^2$, where R is the radius of the depleted area under the geometrical hole and ω_0 is the energy of the $\omega_{1,0}^\pm$ modes at $B = 0$. This equation can be solved easily, and allows one to calculate the dispersion that reproduces the characteristic features, for example, in the experiments of Kern et al. [31] or Hofgräfe et al. [33] quite nicely. More sophisticated theories [34–36] require a detailed knowledge of the shape of the antidot potential.

5.5 Far-infrared Transmission Experiments

In Fig. 5.3, we show measurements on an array of quantum dots with a period of $a = 330$ nm. The number of electrons per dot can be tuned by a gate voltage. This characteristic two-mode behavior of quantum dots filled with a small number of

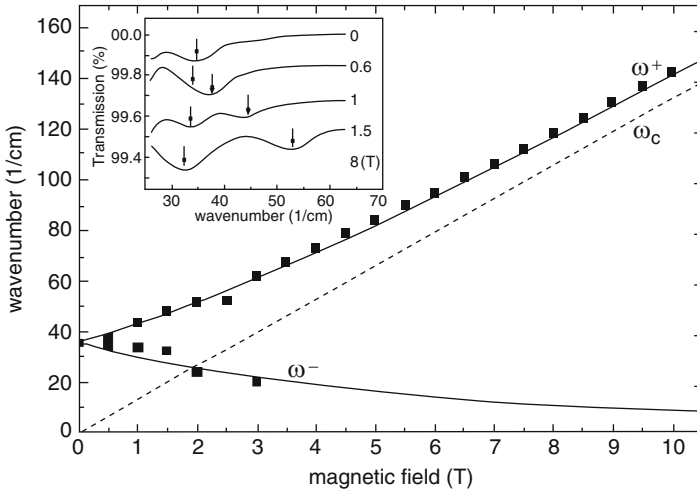


Fig. 5.3 Transmission spectra (*inset*) and dispersion for a 330-nm field-effect induced array of quantum dots for a gate voltage, where each dot is charged with one electron. The two characteristic Kohn modes are observed. The cyclotron resonance frequency ω_c of a 2D system is shown for comparison. It is not an eigenmode of a quantum dot and is not observed in the experiment. (Unpublished results in cooperation with R. Krahne and M. Hofgräfe.)

electrons was first observed by Sikorski and Merkt [37]. In the experiment shown in Fig. 5.3, each quantum dot is filled by exactly one electron. We find the two branches expected from (5.11) with a high-frequency cyclotron-like resonance with positive B dispersion and the low-frequency branches with negative dispersion. The upper and lower branch show the reversed circular polarization as expected from theory and proven experimentally, e.g., in [43]. In our experiments we measure the relative change in transmission $\Delta T/T$. With only one electron per dot, we have for a period $a = 330 \text{ nm}$ only $10^9 \text{ electrons/cm}^2$, about 100 times less than that in typical measurements of a cyclotron resonance in a 2DES. Accordingly, the signal, shown in the inset, is very weak, $\Delta T/T < 0.1\%$. Nevertheless the resonances can be clearly resolved and can be followed over a wide B regime.

For an active sample area of 2 mm^2 , we have about 10^8 electrons in total. It is surprising that for such a large number of electrons all dots are charged simultaneously with the same number of electrons. As was demonstrated by Meurer et al. [2], the reason is the high Coulomb charging energy in the quantum dot. They found a stepwise increase in the integrated intensity of the resonance with increasing gate voltage, reflecting directly the incremental charging of the dots with additional electrons. Note that the transition matrix element is proportional to the number of electrons. From $\Delta V_g = 10 \text{ mV}$ on the voltage axis in their experiments, one can estimate a capacity $C = e/\Delta V_g = 5 \times 10^{-18} \text{ F}$ and a Coulomb charging energy $E_c = e^2/2C = 15 \text{ meV}$. This value is significantly larger than $k_B T$ and stabilizes the number of electrons per dot.

Breaking Kohn's Theorem

In a first step, it was quite nice to confirm the Kohn theorem experimentally, demonstrating that independently on the number of electrons one always saw only the two branches in a magnetic field. Compared to the rich spectra that we know from atomic spectroscopy, the question is, is this also possible for our man-made quantum dots? The answer is 'yes'. We have to fabricate quantum dots with strongly nonparabolic external potential.

Figure 5.4a shows spectra measured on a gated deep mesa etched sample as sketched in Fig. 5.2b. With a gate voltage, quantum dots with elliptical shape are formed. The spectra are extremely rich with up to 5 resonances, which all strongly depend on the magnetic field. The corresponding B dispersions are shown in Fig. 5.4b. The dominant intensity is in the branches labeled ω_{+1} and ω_{-1} . In contrast to the circularly shaped dots in Fig. 5.3, there is a splitting of the two resonances at $B = 0$. These two resonances correspond to confined plasmon oscillations along the long and short axes of the ellipse, which can be experimentally confirmed from the measured linear polarization along the respective axis. The low-frequency resonance decreases in intensity with increasing B and can, due to the decreasing sensitivity of our experimental set up at low frequencies, no longer be resolved at large B . The high-frequency resonance increases in intensity with increasing magnetic field. This is expected from the transition matrix elements. Note that the cyclotron resonance

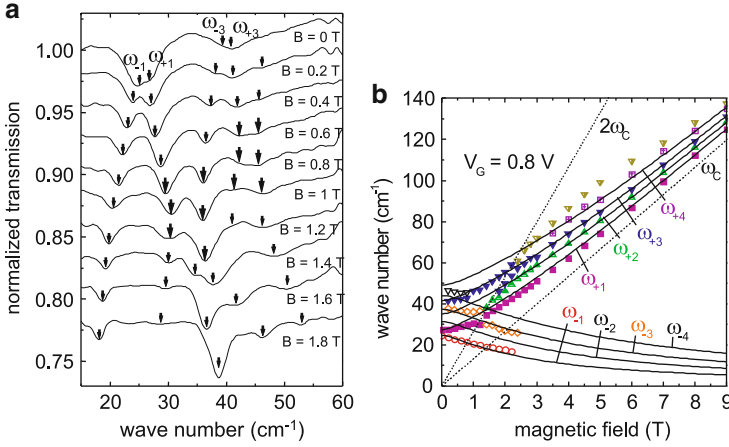


Fig. 5.4 (a) Spectra of elliptically shaped quantum dots (Fig. 5.4b at $V_G = 0.8$ V corresponding to $N = 320$ electrons per dot) from $B = 0$ T to $B = 1.8$ T incremented by 0.2 T. Spectra are shifted vertically for $B > 0$ for clarity. The resonance positions are marked by arrows. The regime where an anticrossing of the modes occurs is marked by *thick arrows*. (b) Experimental dispersions of elliptically shaped dots for $N = 320$ electrons per dot. Modes which have a linear polarization along the short (long) axis at small B are marked by full (open) symbols. (Adopted from [46])

itself is not observed. It is no eigenmode of the quantum dot and only occurs in a system where the electrons fill an infinitely large area completely.

The two dominant modes can be perfectly described by the lowest mode in the Fetter type plasmon-in-a-box model (5.8) if expanded to the elliptical case [29, 30]. According to the Kohn theorem, these two modes would be the only modes if the external potential would be parabolic in both x and y directions. In Fig. 5.4, we observe not only the two fundamental Kohn modes but sets of modes which undergo different types of anticrossings in a magnetic field. This directly indicates that the external potential is not parabolic.

The higher frequency modes $i = 2, 3, \dots$ correspond to higher confined plasmon modes. The ω_{-i} modes represent confined edge magnetoplasmon modes, which exhibit a characteristic negative B dispersion. In a hard wall potential, these modes can be described microscopically by a collective electron motion, where the individual electrons perform skipping orbits along the circumference of the dot.

Another interesting finding is that the dispersions shown in Fig. 5.4b exhibit two different types of anticrossings. One type occurs close to $2\omega_c$ and resembles the interaction of plasmons in a 2DES with the Bernstein modes. In this interaction regime, we have a complex internal excitation that has been discussed in detail, using Hatree-Fock and RPA type of theories, in [20] and [38]. Another type of anticrossing occurs if a higher order ω_{-i} mode intersects with an ω_{+j} mode ($i > j$). Model calculations for few-electron systems [39] have shown that such an anticrossing does not occur for circularly shaped dots, even if the potential is not parabolic, rather it was shown that a noncircular, for example, a quadratic shape, is required to

break this degeneracy. It was discussed in [39] that besides the symmetry and geometry, the electron–electron interaction determines the strength of the interaction and the resulting splitting in the anticrossing regime. We believe that the same parameters, the geometrical shape and the electron–electron interaction, are responsible for the anticrossing at $B > 0$ that we observe for our elliptically shaped dots.

We like to note that the observation of higher order modes, which we have discussed here for elliptically shaped quantum dots, is also observed in circularly shaped dots, provided that the external potential is nonparabolic, see for example [40]. This ‘breaking of the Kohn Theorem’ can also be used to study the filling-factor dependent formation of compressible and incompressible stripes in quantum dots and antidot arrays under conditions of the quantum Hall effect (QHE) [41] or observe excitations below the Kohn mode in field-effect induced quantum dot arrays with flat potentials [42]. Other examples of strong deviations from a nonparabolic behavior are discussed in [45, 46]. Strong modification of the two mode behavior also arises if quantum dots are close enough thus coupling between them occurs. This was demonstrated in FIR experiments by Lorke et al. [48].

Transition from Zero to Two Dimensions

In Fig. 5.5, we would like to demonstrate the transition from zero to two dimensions. The sample is an array of 100-nm high Ni dots of 200-nm diameter which was prepared on top of a shallow HEMT structure. The period of the array is $a = 400$ nm. (The ferromagnetic behavior of these Ni dots has no influence on the spectra here.) Due to the combination of stress and surface effects, a potential landscape is induced which can be filled successively with electrons utilizing the persistent photo effect. With this raising of the Fermi level, we successively have first isolated quantum dots, then an antidot array, and in a last step, a continuous but strongly density-modulated 2D electron system.

For the dot case Fig. 5.5a, we observe the two Kohn modes. (The lower frequency branch cannot be resolved in this sample, due to the decreased sensitivity of the FTS at low frequencies. In the antidot case, (b) and (c), we find again a two-mode behavior which, however, is quite different for the experiments on quantum dots. At high B , the dispersion of both branches resembles the excitation spectrum of quantum dots which we have discussed above. With decreasing B , the resonances of the low-frequency branch ω_{EMP}^- first increase in frequency, but then, in contrast to dots, these resonances decrease in frequency at a certain magnetic field and approach the cyclotron frequency ω_c . This was first observed by Kern et al. [31]. To explain this behavior, we use, as sketched in Fig. 5.6, an intuitive one-particle picture, being aware that the excitation has in reality a strongly collective character. The ω_{EMP}^- mode is at high B , the edge magnetoplasmon mode where the individual electrons perform skipping orbits around the outer orbit of the geometrical hole. With decreasing B , the electron orbits of the ω_{EMP}^- mode become larger, and eventually the electrons can perform classical cyclotron orbits $r_c = \sqrt{2\pi N_s \hbar / eB}$ around a hole. Then the collective edge magnetoplasmon excitation gradually changes into a classical CR

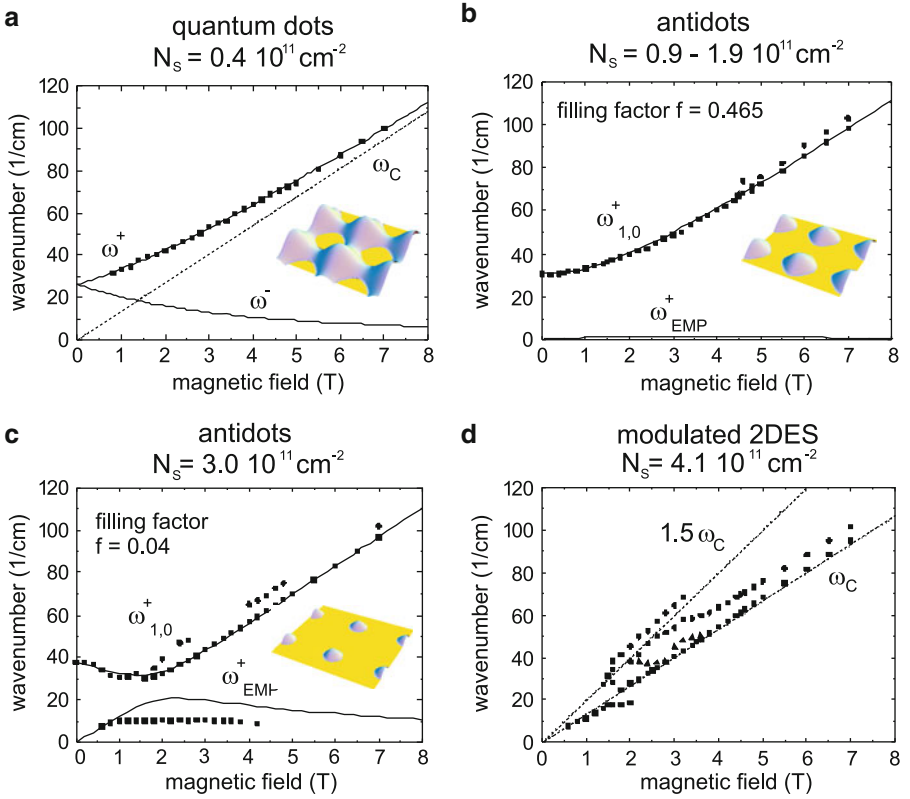


Fig. 5.5 Array of 100-nm high Ni dots of 200-nm diameter prepared on top of a shallow AlGaAs/GaAs HEMT structure. By raising the Fermi level, we successively have first isolated quantum dots (a), then an antidot array, (b) and (c), and in a last step, a continuous but strongly density-modulated 2D electron system. N_s gives the averaged charged density under these conditions. The experimentally observed dispersions are very different in the three cases and explained in detail in the text. The experimental dispersion in (a) is compared with the Kohn modes from (5.11). The experimental dispersions in (b) and (c) are compared with the theoretical dispersion (5.12) assuming area filling factors $f = 0.465$ and $f = 0.04$, respectively. (Unpublished results in cooperation with R. Krahne and M. Hochgräfe.)

excitation. One can estimate the value of B , where this transition occurs, by evaluating the condition that the classical cyclotron radius r_c becomes equal to the radius of the holes r_g . One finds indeed good agreement in experiments on samples with different geometrical hole sizes [31]. The high-frequency mode, which increases in intensity with increasing B , represents at small B a plasmon type of collective excitation of all electrons. A unique behavior is that at small B this resonance shows a weak, but distinct negative B dispersion, which was observed on all our samples where we were able to evaluate the resonance down to $B = 0$. In dots, a positive B dispersion is found (see Figs. 5.3 and 5.5a) and confined local plasmon oscillations in wire structures start with a constant and then increasing B dispersion

[9]. The negative B dispersion of the $\omega_{1,0}^+$ mode at small B in Fig. 5.5c indicates that it represents a kind of 1D magnetoplasmon that propagates with a wavevector $q = 2\pi/a$ along the charged stripes between the geometrical holes. A similar negative b dispersion has been observed for 1D plasmons by Demel et al. [18]. At higher fields, the ω_+ branch approaches ω_c and represents, as denoted in Fig. 5.5a by ω_c , a cyclotron-type excitation in the region between the holes. From this explanation, we conclude that in an antidot array both the high-frequency and the low-frequency branches have the same circular polarization, in contrast to the behavior of quantum dots. This has been confirmed experimentally [43].

Anticyclotron Motion in Antidot Arrays

Looking at Fig. 5.6 and using the same type of picture as just discussed, one could also imagine that there should be an excitation as sketched for the pillow-shaped trajectory labeled $\omega_{1,0}^-$. We see that this antidot mode has an anticyclotron polarization. This mode has actually been predicted by theory [35]; however, it was a challenge to observe this mode, because of its intrinsically weak oscillator strength at $B = 0$, which decreases even further with increasing B .

We have prepared antidot arrays with optimized potentials and were able to detect this mode [33]. The experimental dispersion is plotted in Fig. 5.7 and compared with theory. The mode $\omega_{1,0}^-$ is the new mode with the anticyclotron polarization. To confirm this in detail, we show in Fig. 5.7 the dispersion and the intensity of the two modes. I^+ and I^- are, respectively, the intensities of the cyclotron and the anticyclotron resonance. In Fig. 5.7, the $\omega_{1,0}^+$ mode dominates the spectrum at large B and decreases in intensity with decreasing B to a finite value $I^+(B = 0)$. The $\omega_{1,0}^-$ mode starts at the same value $I^- = I^+$ at $B = 0$ and then decreases in intensity. Theory says that the ratio $I_{1,0}^+(B = 0)/I_S^+ = I_{1,0}^-(B = 0)/I_S^+$, where I_S^+ is the saturated value at large B , depends on the shape of the antidot potential. An explicit expression for the normalized oscillator strengths $S_{m,n}^{+(-)}(B)/S_{m,n}(B = 0)$ for the two modes is given in (17) of [35]. We compare in Fig. 5.7 our experimental

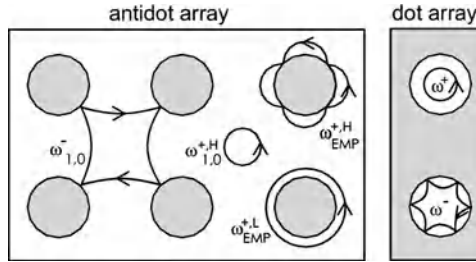


Fig. 5.6 Sketch to visualize in a one-particle picture the cyclotron motion of the $\omega_{1,0}^{+H}$ mode at large B , the skipping orbit motion of the $\omega_{EMP}^{+H(L)}$ mode, and the pillow-shape trace of an anticyclotron motion, $\omega_{1,0}^-$. (Adopted from [49])

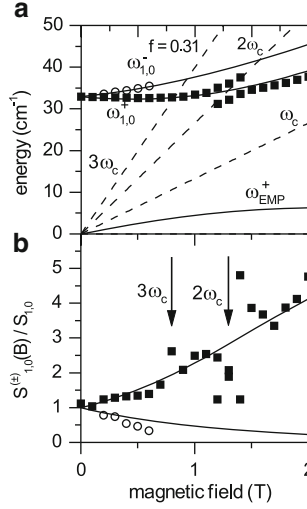


Fig. 5.7 Experimental dispersion (*top*) and normalized oscillator strength $S_{1,0}^{\pm}(B)/S_{1,0}(B=0)$ (*bottom*) of an antidot array, similar to (Fig. 5.2c). The grating period is $a = 1,000$ nm, the diameter of the geometrical holes $d = 450$ nm. The full lines in (a) are the calculated dispersions according to the theory of [35] for an areal filling factor $f = 0.31$, the full lines in (b) are the calculated normalized oscillator strengths. At $B \approx 1.4$ T, interaction with Bernstein modes near $2\omega_c$ occurs, which is not included in the theory for the dispersion and oscillator strength. At $B \approx 0.8$ T, an interaction with $3\omega_c$ is observed in the oscillator strength. (Adopted from [33])

intensities with this theoretical expression and find, within the experimental accuracy, a reasonable agreement. In the original paper, we discuss also the experimentally measured polarization. Although the scatter of the data (within an accuracy of 20%) is relatively large, due to the difficult determination and the interaction with the Bernstein modes, the experimental data clearly demonstrate the anticyclotron polarization.

Strongly Modulated Two-Dimensional Systems

Coming back to Fig. 5.5d, we look more closely into the case where the charge density has been raised so much that the area is completely filled with electrons, with the 2D density being strongly modulated. In this case, we directly observe the cyclotron resonance ω_c . In addition, we find modes above the cyclotron resonance frequency which represent 2D plasmons, as described in (5.4) and (5.5). They are excited via the grating coupler effect of the periodically modulated density. These 2D plasmons exhibit a strong interaction with Bernstein modes at $1.5\omega_c$. For a 2DES with constant density, interaction with Bernstein modes occurs at $2\omega_c$ (and higher harmonics); in systems with modulated densities or in quantum dots and wires with nonparabolic external potential, this interaction can occur at lower frequencies, as was observed in many experiments [44, 47] and explained by theory [20].

We like to mention that there are several more very interesting review articles on FIR transmission experiments which cover different aspects of the extremely wide field of physics that can be studied in these low-dimensional systems and contain many more references which could not be given here within the allotted space, for example, by Kotthaus [50], Merkt [51], or Heitmann and Kotthaus [52].

5.6 FIR Photoconductivity Spectroscopy

In this section, we review experimental results obtained by using the highly sensitive far-infrared photoconductivity spectroscopy (FIR-PC), which involves (a) detecting electron cyclotron resonance in the optical phonon regime [53], (b) measuring the spin-orbit coupling strength via the spin-flip excitation [54], (c) discovering a quantized dispersion of magnetoplasmons in the quantum Hall regime [55], and (d) studying far-infrared radiation induced magneto-resistance oscillations [56].

The Bolometric Model

The bolometric effect plays a major role in the development of the FIR-PC spectroscopy. Via this effect, dipole excitation of electrons effectively heats the 2DES and changes its resistance. Neppl et al. [57] demonstrated that in the case of a weak FIR illumination and by applying a small bias current I , so that non-resonant heating of both the 2DES and the lattice can be neglected, the amplitude of the photo voltage is determined by the steady state of the hot electron gas formed under the condition that its energy loss rate is equal to its power absorption. It is expressed as

$$|\Delta V_{xx}| = I \cdot |\Delta R_{xx}| = I \cdot \left| \frac{\partial R_{xx}}{\partial T} \right| \cdot \frac{A(\omega) \cdot \tau}{C_e}, \quad (5.13)$$

where ΔR_{xx} is the photo-resistance, $A(\omega)$ is the absorbed power of the FIR radiation, C_e is the heat capacity of the 2DES, and τ is the energy relaxation time of the dipole-excited nonequilibrium electrons. Hirakawa et al. [58–60] have shown that by combining the bolometric effect with QHE, the photoresponse of a 2DES can be used for realizing very sensitive, tunable narrow-band FIR detectors. The responsivity and the detectivity of such quantum Hall FIR detectors can reach as high as 1.1×10^7 V/W and 4.0×10^{13} cm Hz^{1/2}/W, respectively, at 4.2 K.

Based on the bolometric effect, FIR-PC of 2DES can be measured with a FTS using the sample itself as the detector (Fig. 5.8). Our experiment is performed by applying a DC current of several μ A to the sample and measuring the changes of the voltage drop caused by FIR radiation. At fixed magnetic fields, the broadband FIR radiation is modulated by the Michelson interferometer of a FTS. The corresponding change in the voltage drop of the sample is AC coupled to a broadband preamplifier and recorded as an interferogram, which is Fourier transformed to get the

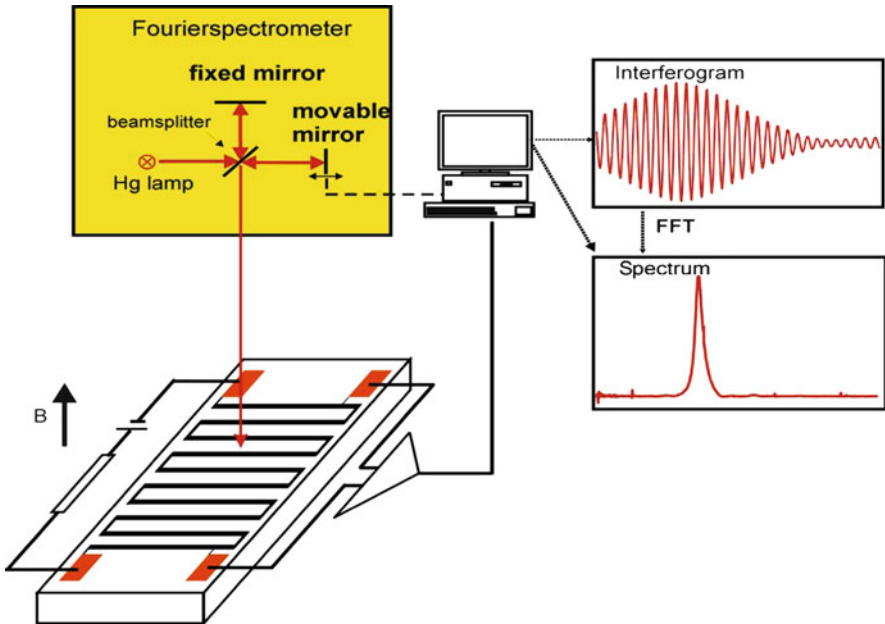


Fig. 5.8 Schematic view of the Fourier transform spectrometer (FTS) and bias circuit to the sample, showing a meandering long Hall bar with ohmic contacts

photo-conductivity spectrum. To increase the photoconductivity signal, the samples are usually fabricated as a meandering long Hall bar by chemical wet etching. The Hall bar has a typical width W of a few tens of micrometer and a total length L of several centimeter. Ohmic contacts are prepared by evaporating AuGe alloys followed by annealing. The sample is mounted in a He cryostat with a superconducting solenoid. A Si bolometer behind the sample allows us to measure the direct absorption and to monitor the phase correction factor sometimes needed if the signal was too weak. All data reviewed here are obtained in Faraday geometry. We begin with cyclotron resonances and spin-flip excitations measured by FIR-PC spectroscopy.

Cyclotron Resonances Within the Reststrahlen Band

The Same Questions Asked in Every 20 Years

Investigation of charge excitations in the regime of optical phonons in semiconductor multilayered structures is the subject of high interest with controversial results. As early as in 1968, Dickey and Larsen [61] published a paper entitled “*Evidence for Electron-TO-Phonon interaction in InSb*”. The idea of Electron-TO-Phonon interaction was introduced to explain the energy discontinuity they observed in the combined resonance of localized electrons. This might be the first time that the question

was brought up whether electrons may interact with the TO phonon. Only half year later, McCombe and Kaplan [62] published experimental results showing strong evidence against the existence of such an electron-TO-phonon coupling in InSb.

Seventeen years later in 1985, Nicholas et al. [63] published a paper suggesting that resonant polaron coupling exists for both TO and LO phonon in GaInAs heterojunctions. This paper was followed by a comment from Das Sarma [64] published one year later, which argued that the discontinuity in the CR mass at the TO phonon frequency ω_T observed by Nicholas et al. might be due to an interface phonon mode that has a frequency very close to ω_T . Both interpretations were incorrect as we know today.

In 2001, Poulter et al. [65] at the Grenoble High Magnetic Field Laboratory, France, published a paper reporting observing the pinning of the CR energy in a highly doped GaAs quantum well, which occurs at an energy close to that of ω_T . They interpret that the CR couples to a longitudinal collective magnetoplasmon-phonon mode with an energy close to ω_T . Half years later, Zhang, Manger, and Batke [66, 67] showed that such a coupling approach cannot be justified. They pointed out the importance of including influences of optical origin in the analysis, which was used in [68] to explain results observed by Nicholas et al. [63].

The Grenoble group continued studying this subject. In 2004, Faugeras et al. [69] published infrared magnetoabsorption data on similar samples, which showed interesting absorption features near the TO phonons of GaAs and AlAs. They attribute the result to the interaction of electrons with some modes with frequencies close to ω_T based on the deformation potential. They further argue that the concept of the Fröhlich polaron theory, which predicts a resonant magnetopolaron coupling near the longitudinal optical-phonon frequency ω_L , has to be reexamined. In June 2005, a comment written by Klimin and Devreese [70, 71] was published, which concludes that such an anticrossing near ω_T cannot be interpreted in terms of the deformation electron-phonon interaction, and the concept of the Fröhlich polaron theory holds.

All of these papers and comments were published in the Physical Review Letters. Theoretically, according to the Huang Equation for lattice dynamics [72], it is clear that the macroscopic electric field and polarization are only associated with the LO phonon; therefore, TO phonon should not couple to electrons. However, experimental investigation of charge excitations in the TO phonon regime is not trivial, due to the Reststrahlen band defined between ω_T and ω_L . Here, the strong coupling between photons and the TO-phonon causes strong reflections and prohibits measuring transmission spectra within the Reststrahlen band. FIR-PC provides the elegant solution to overcome the obstacle. This technique allows us to measure charge excitations within the Reststrahlen band via the resistance change of the 2DES, which paves the way for the detailed investigation of the electron-phonon interaction.

Cyclotron Resonances at the TO Phonon Regime

Our sample is a parabolic quantum well grown by molecular beam epitaxy on a GaAs substrate. To compensate the lattice mismatch between GaAs and $\text{In}_x\text{Al}_{1-x}\text{As}$, a metamorphic buffer was grown with continuously increasing In content

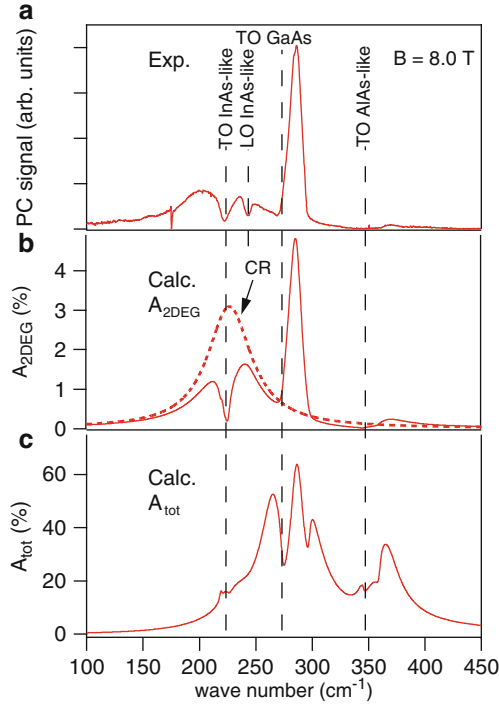


Fig. 5.9 (Adopted from [53]) (a) Experimental photoconductivity spectrum of the parabolic quantum well structure for a magnetic field of 8 T. (b) Calculated spectrum of the absorption of the electron gas. (c) Calculated total absorption spectrum. The vertical lines indicate the positions of the TO phonon modes of $\text{In}_x\text{Al}_{1-x}\text{As}$ and GaAs and the InAs-like LO phonon mode of $\text{In}_x\text{Al}_{1-x}\text{As}$. The dotted curve in (b) shows the absorption of the CR mode at this magnetic field without including the phonon modes

up to $x = 0.75$. The total thickness of this buffer is about $1.2\ \mu\text{m}$. A Si-doped layer and a spacer follow with a thickness of 10 and 20 nm, respectively. The parabolic quantum well is composed by first increasing the In content up to $x = 1$ and second decreasing it down to $x = 0.75$. The total thickness of the quantum well is about 100 nm. The sample is capped with a 20-nm thick $\text{In}_{0.75}\text{Al}_{0.25}\text{As}$ layer. The back side of the sample is wedged by an angle of 3° to suppress interference effects.

Figure 5.9a shows the photoconductivity spectrum of the parabolic $\text{In}_x\text{Al}_{1-x}\text{As}$ quantum well for a magnetic field of $B = 8\ \text{T}$ at a sample temperature of $T = 1.5\ \text{K}$. The spectrum was taken by applying a dc current of 450 nA. The dashed vertical lines indicate the positions of the TO phonon modes of both $\text{In}_x\text{Al}_{1-x}\text{As}$ and GaAs. In addition, the frequency of the InAs-like LO phonon is shown.

To analyze the PC spectra, we have used the Fresnel equations to calculate both the total absorption of the multilayered structure and the absorption of the electron system. The influence of the optical phonons is included in the dielectric function of

the crystal. The absorption of the 2DES is modeled via the high-frequency conductivity of the electrons. The absorption in a single layer of a multilayered structure can be calculated from the amplitudes of the incoming and outgoing electromagnetic waves at both sides of the layer.

In Fig. 5.9b, the absorption of the electron gas confined in the parabolic well is plotted. In (c) the calculated total absorption of the whole multilayered structure is shown. By comparison of the calculated absorption spectra and the experimental PC spectrum, one can see the strong similarity between the absorption of the electron gas and the photoconductivity response. In the calculated spectrum of the electron gas, only the cyclotron resonance is responsible for the absorption. At $B = 8$ T, the CR mode is expected at $\omega_c = 226 \text{ cm}^{-1}$, indicated by arrow in Fig. 5.9b. The absorption of the electron gas without including the phonon modes is shown in Fig. 5.9b as the dotted line. For the result including the phonon modes, one can see the strong deviation from the Lorentzian line shape, which indicates the interference feedback of the multilayered structure with optical phonon modes. The strongest response in both the calculated absorption and the PC measurement can be found at $\omega = 285 \text{ cm}^{-1}$ inside the Reststrahlen band of the GaAs substrate. Such a strong photoconductivity response inside the Reststrahlen band of the GaAs substrate can be observed for a large magnetic field regime. In the regime of the InAs-like TO phonons, a strong splitting can be found, which can be well described by the dielectric calculations. Note that in the theory only the absorption of the cyclotron resonance is calculated. The CR leads to a single absorption process whose energy is proportional to the magnetic field. The strong modulation of the dielectric properties and the interference effects of the multilayered structure yield the multi-peak behavior with splittings around the TO phonons which require no microscopic electron-phonon-coupling mechanism.

The only deviation between the calculated absorption of the electron gas and the experimental result appears at the InAs-like LO phonon mode of $\text{In}_x\text{Al}_{1-x}\text{As}$. In the experiment, a reduced response is observed which shows an anticrossing-like behavior by switching the B field. This effect reflects the microscopical electron-LO-phonon coupling [73–78] which is not taken into account in our dielectric calculations.

This work, therefore, demonstrates that FIR-PC spectroscopy can be applied to detect excitations within the Reststrahlen band. The results show that since the heterostructure is an optically coherent system, the power absorption spectrum of the 2DES depends non-locally on the optical property of the multilayer structure, which is influenced by the macroscopic dielectric effect of each layer. As the consequence, the high-frequency conductivity and the dielectric constant describing the charge and lattice dynamics, respectively, have to be treated on equal footing by using Maxwell's equations. As the result of such an optical effect, the line shape of the charge excitation differs significantly from the Lorentzian line shape observed outside the Reststrahlen band.

Measuring Spin-Orbit Coupling Via the Spin-Flip Excitation

In the classical picture, conduction electrons of a semiconductor placed in a magnetic field $\mathbf{B}$ feel a Lorentz force that drives the electron moving in the cyclotron orbit, while the magnetic moment of the spin feels a torque that causes the spin to precess. These motions resonantly interact with the electromagnetic radiation, with cyclotron resonance (CR) frequency $\omega_c = eB/m^*$ and electron spin resonance (ESR) frequency $\omega_z = -\nu_0\omega_c$ being typically in the THz and GHz regime, respectively ($\nu_0 = gm^*/2m_e < 0$ for most semiconductors). They provide textbook examples of accurate determination of the electron effective mass m^* and the Landé g factor. It is known that in InGaAs/InAlAs heterojunctions, the structure inversion asymmetry dominates the spin-orbit interaction over the bulk inversion asymmetry, so that the spin-splitting energy is given by [79]

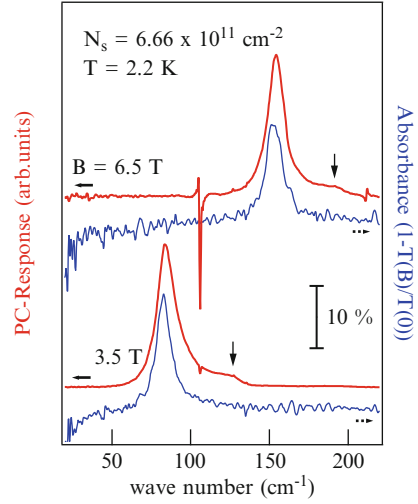
$$|\hbar\omega_s| = \{[\hbar(\omega_c + \omega_z)]^2 + (2\Delta_R)^2\}^{1/2} - \hbar\omega_c, \quad (5.14)$$

which approaches the Zeeman splitting energy $\hbar\omega_z$ only at high B fields when the cyclotron energy $\hbar\omega_c \gg 2|\Delta_R|/(1 - \nu_0)$. Here, the matrix element $\Delta_R = \alpha k_F$ depends on the spin-orbit parameter α and the Fermi wave vector k_F , which can both be controlled via a front gate [80–83]. To determine the spin-orbit coupling strength in the 2DES is of fundamental importance for spintronics, as is best illustrated in the classic paper of Datta and Das for a novel spintronic device [84]. The commonly used method for measuring spin-orbit coupling strength is the magnetotransport technique utilizing either a beating pattern or the weak antilocalization effect. Here, we show that FIR-PC provides an alternative spectroscopic approach by detecting a combined resonance (CBR) with both the Landau and spin quantum numbers changed.

Our sample is an inverted-doped InAs step quantum well with a 40-nm $\text{In}_{0.75}\text{Al}_{0.25}\text{As}$ cap layer. The step quantum well is composed of 13.5-nm $\text{In}_{0.75}\text{Ga}_{0.25}\text{As}$, an inserted 4-nm InAs channel, and a 2.5-nm-thick $\text{In}_{0.75}\text{Ga}_{0.25}\text{As}$ layer. Underneath the quantum well is a 5-nm spacer layer of $\text{In}_{0.75}\text{Al}_{0.25}\text{As}$ on top of a 7-nm-wide Si-doped $\text{In}_{0.75}\text{Al}_{0.25}\text{As}$ layer. The sample is grown by MBE on a buffering multilayer accommodating the lattice mismatch to the semi-insulating GaAs substrate. A self-consistent Schrödinger-Poisson calculation shows that the 2DES is about 55 nm below the surface, mainly confined in the narrow InAs channel [85]. The carrier density N_s and mobility μ at 2.2 K were determined by Shubnikov-de Haas measurement to be $6.66 \times 10^{11} \text{ cm}^{-2}$ and $150,000 \text{ cm}^2/\text{Vs}$, respectively. Ohmic contacts were made to the meandering long 2DES Hall bar ($L = 10 \text{ cm}$, $W = 40 \mu\text{m}$) by depositing AuGe alloy followed by annealing.

Figure 5.10 shows typical FIR-PC spectra (thick lines) measured at two B fields of 3.5 and 6.5 T. A weak resonance is observed at the high-energy side of the dominant peak. For comparison, conventional absorption spectra measured using the Si bolometer under the same experimental conditions are plotted as thin lines. The weak resonance, whose intensity is only about 0.8% of that of the strong one, is only resolved in our highly sensitive photo-conductivity spectrum.

Fig. 5.10 (Adopted from [54]) THz photo-conductivity spectra (*thick lines*) measured at two magnetic fields in comparison with conventional absorption spectra (*thin lines*) under the same experimental conditions using a Si bolometer. In addition to the CR, *thick arrows* indicate the weak CBR, which are only observable using the highly sensitive photo-conductivity technique



In Fig. 5.11a, we plot the B -field dispersion of both resonances. Also shown is the magnetoresistance R_{xx} measured without FIR radiation with the same excitation current of $4.5 \mu\text{A}$. The open circles determined from the strong peaks are easily identified as the CR with $\omega_c = eB/m^*$ that can be described (dashed line) using $m^* = 0.039 m_e$. The solid circles for the weak resonances are fits (dashed curve) to $\omega_c + \omega_s$ using (5.14) with two fitting parameters $\Delta_R = 38 \text{ cm}^{-1}$ and $g = -8.7$. The fairly good fit using a reasonable g factor for InAs encourages us to attribute it to the CBR. Observing the FIR dipole-excited CBR in Faraday configuration requires spin-orbit interaction, in accordance with the obtained zero-field spin splitting $2\Delta_R = 76 \text{ cm}^{-1}$, which gives a spin-orbit parameter $\alpha = 2.38 \times 10^{-11} \text{ eV}\cdot\text{m}$. Using these parameters, we calculate the Landau levels [79] and plot them in Fig. 5.11b together with the dotted lines showing the Fermi level. Thin and thick arrows illustrate the CR and CBR, respectively.

By carefully checking the CBR intensity which are normalized using the CR, we find that the CBR disappears around $\nu = 5$ and 7. This is caused by a many-body effect. Via electron–electron interaction, the CBR is shifted to form collective spin-flip excitation. Theory [86] has predicted that at odd filling factors where the ground state of the 2DES is spin polarized, the collective spin-flip excitation decays into a magnetoplasmon and a spin wave that conserve spin, momentum, and energy. Such a many-body effect allow us to verify the weak resonance as an excitation of the 2DES instead of as impurity transitions that are independent from the filling factors. However, it also indicates that our analysis using the single-particle picture coined in (5.14) is over simplified. We note that it remains a theoretical challenge to determine correctly the g factor and spin-orbit parameter α , by taking into account both electron–electron interaction and spin-orbit coupling.

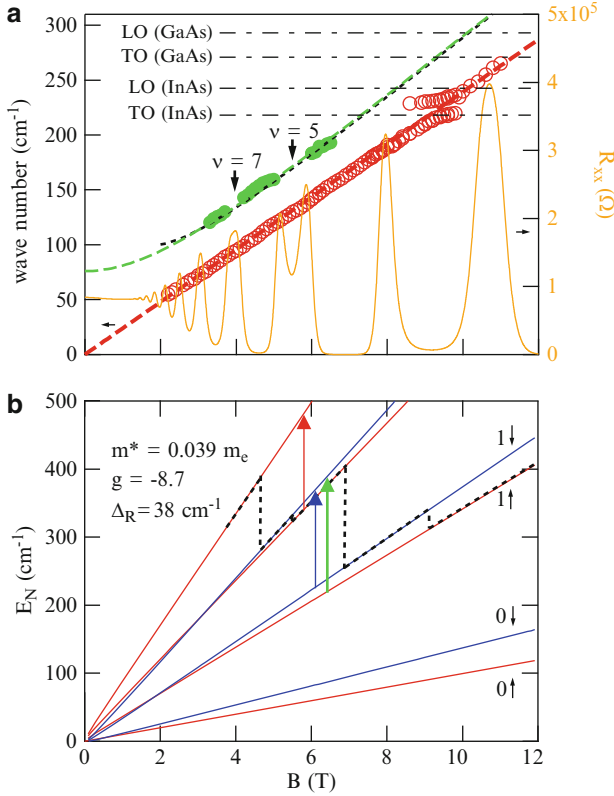


Fig. 5.11 (Adopted from [54]) (a) Resonance dispersions determined from the photo-conductivity spectra and magnetoresistance R_{xx} measured without FIR radiation. The *dashed line* and *curve* are fits for the CR and the CBR using a constant effective mass and (5.14), respectively. *Dash-dotted lines* indicate the optical phonon energies of InAs and GaAs. (b) Landau levels calculated using the band parameters obtained from the fit in (a). *Dotted lines* indicate the Fermi energy. *Thin and thick arrows* illustrate the CR and CBR, respectively

Deviation from the Bolometric Model

In the absorption spectroscopy, one detects the elementary excitations by measuring the transmitted radiation, assuming that absorption of photons does not change the properties of the electronic system. On the contrary, in the PC experiments, elementary excitations are detected by measuring the photo-induced change of the resistance, which monitors exactly the change of the electronic system caused by absorption of photons. The bolometric model explains well the different sensitivities of these two spectroscopic techniques; it also indicates that both techniques detect the same elementary excitations, as in the cases of CR and CBR which we have reviewed. However, it is important to note that the bolometric model applies under the condition that the excited electronic system reaches a steady state characterized

by a slightly raised temperature. This condition may break if the bias current is spatially inhomogeneous as in the QHE regime [55, 58], or if the energy relaxation is spin-dependent as in the spin-polarized electronic system [87], or if intense radiation drives the electronic system far beyond equilibrium [56]). All provide us chances for exploring unique natures of elementary excitations unable to be investigated by conventional absorption spectroscopy. In the following we review the results of quantized dispersion for magnetoplasmon, and FIR-induced magnetoresistance oscillation, both detected by FIR-PC and show deviations from the bolometric model.

Quantized Dispersion for Magnetoplasmons

At first glance, two-dimensional plasmons seem to be unlikely a subject to give us surprises. Its dispersion, given in (5.4) and (5.5) of our Theory Section, was predicted as early as in 1957 by Ritchie [15], 1967 by Stern [8], and 1972 by Chaplik [9]. Both (5.4) and (5.5) have been confirmed by many experiments [16, 89, 90], which makes the plasmon a very well-understood elementary excitation of the 2DES.

By combining (5.4) and (5.5), it is straightforward to define a renormalized magnetoplasmon frequency Ω_{mp} and find

$$\Omega_{\text{mp}} \equiv \frac{\omega_{\text{mp}}^2(B) - \omega_c^2}{\omega_c} = \frac{\hbar \cdot q_{\text{TF}} \cdot q}{2m^*} \nu, \quad (5.15)$$

where we define $q_{\text{TF}} = m^* e^2 / 2\pi\epsilon\epsilon_0 \hbar^2$ as the effective Thomas-Fermi wave vector depending on $\epsilon(q)$. The monotonous linear dependence of Ω_{mp} on the filling factor $\nu = \hbar N_s / eB$ emphasizes the semiclassical nature of the magnetoplasmon, because (5.5) was obtained by analyzing the self-consistent response of the 2DES to a longitudinal electric field in the semiclassical limit, in which the quantum oscillatory part of the polarizability tensor was disregarded [88]. It is therefore astonishing to find deviation of Ω_{mp} from (5.15) for resistively detected magnetoplasmons in high-mobility 2DESs as we review in this section.

To explore a wide range of filling factors, we choose a high-mobility 2DES with high density confined in a GaAs quantum well [91], cf. the sample M1218 in Table 5.1, and we compare it with 2DESs with different mobilities either formed at the interface (HH1295) or in a quantum well (M1266). On top of the

Table 5.1 Parameters of the samples. The two q_{TF} values for sample HH1295 are obtained for the $n = 1$ ($n = 2$) plasmon mode, respectively

Sample	μ ($10^6 \text{ cm}^2/\text{Vs}$)	N_s (10^{11} cm^{-2})	m^* (m_e)	q_{TF} (10^6 cm^{-1})
M1218	1.3	5.58	0.0726	1.83
HH1295	0.5	1.93	0.0695	1.55 (1.86)
M1266	0.3	7.18	0.0730	1.94

Fig. 5.12 (Adopted from [55]) Schematic view of the sample structure and bias circuit showing a meandering long Hall bar with ohmic contacts and a grating coupler

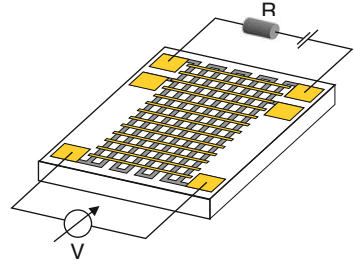
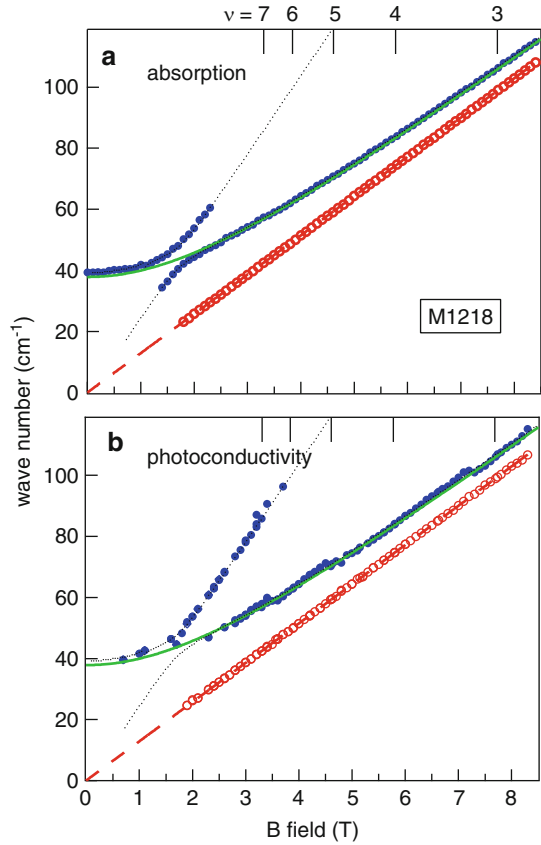


Fig. 5.13 (Adopted from [55]) B -field dispersion of the CR and magnetoplasmons measured in sample M1218 by (a) absorption and (b) FIR-PC spectroscopy. In (a) the CR frequency is fit to the relation $\omega_c = eB/m^*$ (dashed line), and the magnetoplasmon frequency is fit either to (5.5) (solid curve) or by the hydrodynamical model [12] (dotted curve). Theoretical curves in (b) are identical to that plotted in (a)



meandering long 2DES Hall bar ($L = 0.1$ m, $W = 40$ μ m), we have made a gold grating coupler with a period of $a = 1$ μ m (see Fig. 5.12), which allows us to couple the 2D plasmon at $q = 2\pi \cdot n/a$ ($n = 1, 2, \dots$) with FIR radiation. In this frequency regime, the excitations measured by FIR-PC and absorption spectroscopy can be directly compared. CR and magnetoplasmon were measured at 1.8 K in the Faraday geometry by both absorption and FIR-PC spectroscopies.

In Fig. 5.13a, b, we plot the B -field dispersions of the charge excitations determined, respectively, from the absorption and FIR-PC spectra measured on sample

M1218. In both cases, the CR can be well fit (dashed lines) by $\omega_c = eB/m^*$ with $m^* = 0.0726 m_e$. Knowing the effective mass, we fit (solid curve) in Fig. 5.13 the dispersion of the magnetoplasmon using the (5.4) and (5.5), and determine $q_{\text{TF}} = 1.83 \times 10^6 \text{ cm}^{-1}$. Similar fitting procedures are performed for other samples, and the obtained values for m^* and q_{TF} are summarized in Table 5.1. Equations (5.4) and (5.5) capture well the general feature of the magnetoplasmon dispersion except for the nonlocal effect [13], which is responsible for the anticrossing of the magnetoplasmon with the harmonics of CR (the Bernstein modes). Using the hydrodynamical model [12], taking into account the nonlocal effect and with the same parameters of m^* and q_{TF} , the calculated magnetoplasmon dispersions (dotted curves) agree well with that measured by absorption spectroscopy in the whole B -field range, in accordance with the previous studies [13, 14]. In contrast, compared to the theoretical curves and the absorption data, the magnetoplasmon dispersion measured by FIR-PC spectroscopy shows obvious deviations in Fig. 5.13. Plotted in this scale that covers the entire CR frequency range, the deviation looks small. In fact, it is well beyond the experimental accuracy as we plot in Fig. 5.14 which summarizes the filling-factor dependence of Ω_{mp} resistively measured on all our samples.

For comparison, semiclassical predictions for Ω_{mp} calculated by (5.15) using the parameters of m^* and q_{TF} listed in Table 5.1 are plotted in Fig. 5.14 as solid lines. In Fig. 5.14a, Ω_{mp} measured on sample M1218 with the highest mobility deviates clearly from the semiclassical prediction. Very interestingly, the data show plateaus forming around even filling factors of $\nu = 4, 6$ and 8. In Fig. 5.14b, we plot Ω_{mp} obtained on sample HH1295 with a smaller density. The grating coupler of this sample has a higher efficiency, which allows us to measure the magnetoplasmon modes at $q = 2\pi \cdot n/a$ with $n = 1$ and 2. Both show plateaus in the dispersion around even filling factors of $\nu = 2$ and 4. The oscillatory behavior is less obvious in Fig. 5.14c for sample M1266, which has the lowest mobility.

The results shown in Fig. 5.14 are astonishingly reminiscent of the celebrated QHE measured by DC magnetotransport [92], where the Hall conductivity equals its semiclassical prediction $\sigma_H = (e^2/h) \cdot \nu$ at even filling factors (if the spin degeneracy is not lifted), with plateaus forming around them. Currently, two theoretical models have been proposed to explain our experimental data.

Rolf R. Gerhardt developed a screening theory [93] of the integer quantized Hall effect (IQHE) based on the combination of a self-consistent, nonlinear screening theory with a linear, local transport theory. According to this theory, the confinement of the current to incompressible strips may have a total width much smaller than the sample width. Since a dissipative current is used in the FIR-PC experiment to detect the magnetoplasmon, the quantized dispersion of magnetoplasmon is then explained as a consequence of the confinement of the current to strips with integer local filling factor, even if the average filling factor deviates from the integer value.

Toyoda et al. used an alternative approach [94] to explain the filling-factor-dependent plateau-type dispersion, by adopting the electron reservoir hypothesis previously proposed in order to explain the integer QHEs. They notice that following the quantum statistical mechanical derivation of the dispersion relation (5.4), the

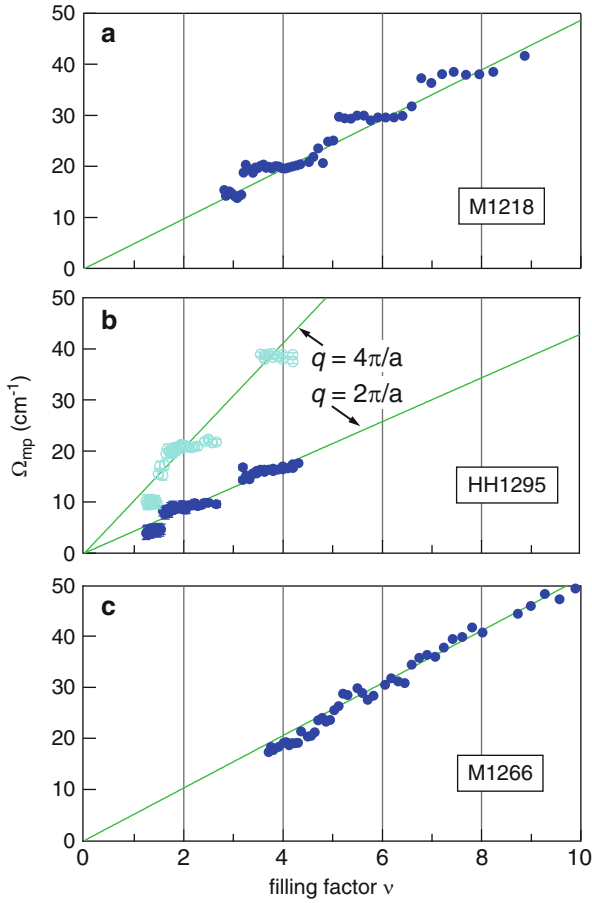


Fig. 5.14 (Adopted from [55]) Filling-factor dependence of Ω_{mp} for the resistively detected magnetoplasmons measured in three different samples. The *solid lines* are the semiclassical predictions calculated using (5.15), which fit exactly to the dispersions measured by the absorption experiments

charge density N_s in the dispersion formula is actually the grand canonical ensemble expectation value for the electron number density in the system. By assuming that the donor impurities in the barrier that confines the 2DES may act as a reservoir, they find an excellent agreement of theoretically derived dispersion curve with the experimental one. However, they also pointed out that the microscopic mechanism that realizes the electron reservoir needs independent experimental verification.

Although the detailed mechanism is still being sought after, the observed feature of quantized dispersion with plateaus forming around even filling factors reveals clearly a previously unknown relation between the magnetoplasmon and the integer QHE. The result is intriguing for investigating the nature of both magnetoplasmon

and QHE, and it shows the importance of going beyond the bolometric model to analyze the results measured by FIR-PC spectroscopy.

Far-Infrared Induced Magnetoresistance Oscillations

Another situation that deviates from the bolometric model is the photoresistance of high mobility 2DES under intense FIR radiation.

In 2001, Zudov et al. [95] reported the observation of Microwave-Induced Resistance Oscillations (MIROs) in a high-mobility ($\mu = 3 \times 10^6 \text{ cm}^2/\text{Vs}$) 2DES. Subsequently, in very high-mobility 2DESs ($\mu > 10 \times 10^6 \text{ cm}^2/\text{Vs}$), MIROs were found to extend all the way to zero resistance, forming microwave-induced Zero Resistance States (ZRSs) [96, 97].

MIROs are determined by $\epsilon = \hbar\omega/\hbar\omega_c$, resulting in a $1/B$ periodicity, where $\omega = 2\pi f$ is the radiation frequency. Following the notation of Zhang et al. [98] the n -th MIRO maxima₍₊₎ and minima₍₋₎ are usually found for $\epsilon_{n\pm} = n \mp \phi_n$. For overlapping LLs $\phi_n \sim 1/4$ [99–101] and in a simplified picture, the field for the n -th MIRO minimum is given by

$$B_{\min} = B_{\text{CR}} \cdot 4/(4n + 1), \quad (5.16)$$

where $n = 1, 2, 3, \dots$ and $B_{\text{CR}} = \omega m^*/e = 2\pi f m^*/e$ is the cyclotron resonance field. For higher fields when LLs get separated, ϕ_n decreases with increasing B [99, 101].

Initially, MIROs and ZRSs were explained in terms of a photon-assisted scattering mechanism, where a transition between LLs is coupled to scattering, resulting in electron displacement in real space [102–108]. Alternatively, it was suggested that a distribution function mechanism, where the microwave radiation leads to nontrivial changes of the Fermi distribution function [109–111], is the dominant contribution. Independent of the microscopic mechanism a negative microscopic resistance leads to macroscopic instabilities [112, 113] and current domain formation, [114, 115] resulting in the observation of zero resistance.

From earlier work, it appeared that MIROs would also be observable at higher radiation frequencies, [96] but experiments show that the MIRO amplitude is diminished with increasing radiation frequency [116–118]. However, studying MIROs at higher frequencies in the FIR regime would provide valuable experimental input to test the frequency dependence of the various theoretical models [108, 110, 119] and identify the contributions of the photon-assisted scattering mechanism and the distribution function mechanism experimentally. Additionally, the interaction of dissipationless states in the QHE at fully separated LLs with ZRSs promises to be interesting with respect to current domain formation [114, 115]. In a FIR-PC experiment, we observe MIRO-analogous oscillations in a moderate mobility 2DES, which shifts the oscillations toward higher fields. We refer such Far-infrared Induced Resistance Oscillations as FIROs.

Our sample is a 10-nm GaAs/AlGaAs quantum well. The mobility and carrier density as determined from magnetotransport are, respectively, $\mu = 1.6 \times 10^6$ cm²/Vs and $N_s = 7.0 \times 10^{11}$ cm⁻² corresponding to a mean free path of $l_e = 22$ μ m. The effective mass is $m^* = 0.0759 m_e$ determined from cyclotron resonance in transmission. On top of an extremely long meandering Hall bar ($L = 10$ cm, $W = 37$ μ m), a grating coupler with a period of $a = 1$ μ m, a width of Au stripes of 0.5 μ m, and a thickness of 15 nm was defined by UV holography. The presence of a grating coupler allows us to check possible coupling between magnetoplasmons and FIROs [120], helping to clarify whether many-particle or single-particle energy levels are relevant to the mechanism underlying the FIROs.

Unlike the results we reviewed in the previous sections, the measurements here were performed in Faraday geometry at $T = 4.2$ K by using a far-infrared laser operating on the strong difluoromethane line at $\lambda = 184.3$ μ m ($f = 1.63$ THz) with a CW output power of $P \sim 60$ mW. Using a light beam chopper and a lock-in technique, ΔR_{xx} was directly measured. The presented ΔR_{xx} signal is the in-phase signal, i.e. the real component of the lock-in measurement.

Figure 5.15 shows two typical traces of the simultaneously measured transmission intensity (a) and the resistance change ΔR_{xx} and longitudinal resistance R_{xx} (b,c) at $I = 0.2$ μ A. Two absorption dips reproduce in all our transmission measurements and are identified as the cyclotron resonance (CR) at $B_{CR} = 4.4$ T and the magnetoplasmon (MP) resonance at $B_{MP} = 2.9$ T. As will be shown, the ΔR_{xx} signal depicted in Fig. 5.15b can be understood as a superposition of a bolometric resistance change and a non-bolometric resistance change caused by the FIROs, which does not follow the $1/B$ periodicity of the bolometric signal.

Strikingly, ΔR_{xx} in Fig. 5.15b shows well-pronounced positive and negative cusps in certain positions. This behavior indicates the existence of certain resonance fields. We focus now on the noteworthy large positive amplitudes of ΔR_{xx} at even filling factors. Within the bolometric model, ΔR_{xx} is well understood to be determined by a convolution of the filling-factor dependent sensitivity due to C_{el} and $\partial R_{xx}/\partial T$ and the resonant field positions. The large positive ΔR_{xx} at $\nu = 10$ and $\nu = 6$ is indicative of an increased 2DES temperature due to the proximity of the magnetoplasmon and cyclotron resonances. Interestingly, ΔR_{xx} is also very large at $\nu = 14$ and $\nu = 8$, indicative of an elevated 2DES temperature close to the positions of the $n = 1$ and $n = 2$ FIRO minima given by the simplified (5.16). In contrast, ΔR_{xx} is much smaller at $\nu = 12$, where no resonance is close by.

In Fig. 5.15c, we focus on the negative components of ΔR_{xx} . The usual behavior of a bolometric signal at partially separated LLs is a smooth oscillation inverted with respect to the SdH oscillations. Strikingly, the negative amplitudes deviate strongly from a bolometric behavior at certain field positions, which are very close to the FIRO minima fields calculated according to the simplified (5.16) and marked by solid vertical lines. In line with the behavior of MIROs, the $n = 1$ minimum is the strongest. We interpret these pronounced minima as being caused by FIROs superimposed on the bolometric 2DES response. We note that the superposition with the bolometric ΔR_{xx} may cause a slight shift of the minima positions. Toward

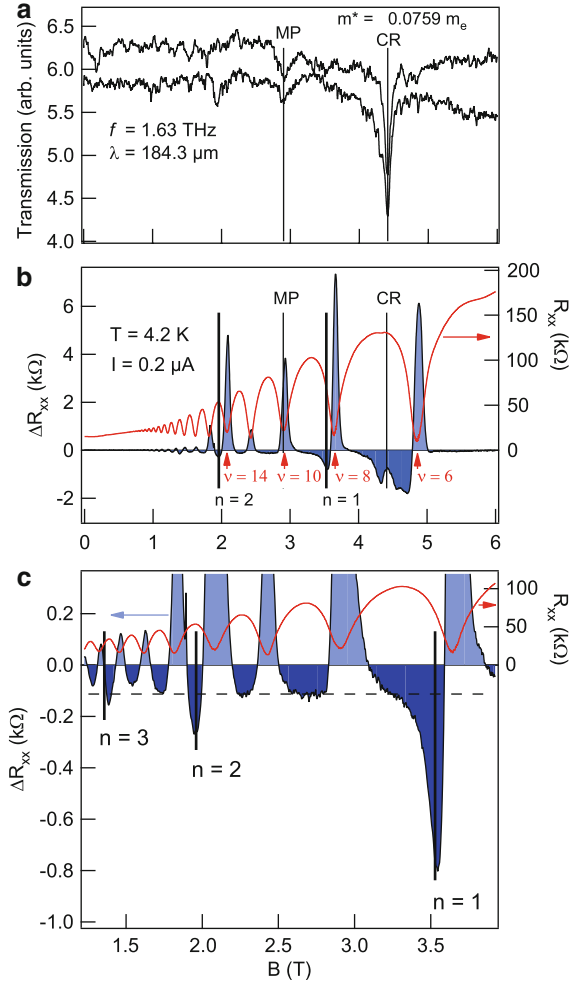


Fig. 5.15 (Adopted from [56]) Simultaneously measured transmission, radiation-induced resistance change ΔR_{xx} and R_{xx} at $\lambda = 184.3 \mu\text{m}$. (a) Two typical transmission curves. The magnetoplasmon resonance (MP) and cyclotron resonance (CR) are marked. (b) ΔR_{xx} and R_{xx} at $I = 0.2 \mu\text{A}$. Additionally, the n -th FIRO minimum according to (5.16) and even filling factors are marked. Note the non-monotonic behavior of the maximum amplitudes of ΔR_{xx} at even filling factors. (c) Magnification of (b) showing the remarkable deviations toward negative ΔR_{xx} around $n = 1, 2, 3$. The dashed horizontal line is a guide to the eye

lower fields, FIRO minima up to $n = 8$ will be shown, measured at increased current levels, which suppresses the SdH-like bolometric ΔR_{xx} .

Figure 5.16a shows ΔR_{xx} in the low-field regime for low and high currents up to $I = 20 \mu\text{A}$. FIROs up to $n = 8$ are directly observed at high currents, where the bolometric ΔR_{xx} is suppressed due to the increased 2DES temperature.

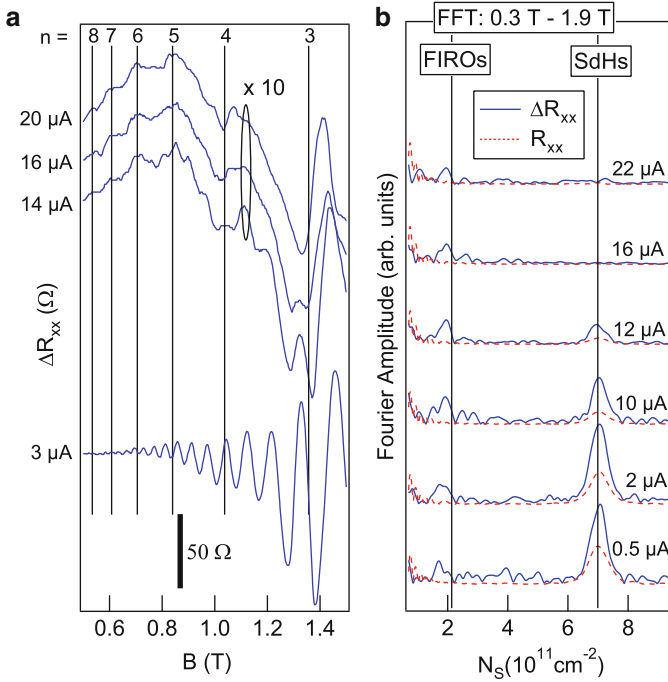


Fig. 5.16 (Adopted from [56]) (a) ΔR_{xx} at low fields for low and high currents. Note how the bolometric SdH-like ΔR_{xx} gets suppressed at higher currents, while the FIROs are more clearly observable. ΔR_{xx} develops a broad maximum around $B = 0.8$ T at the high currents. Additionally, below $B = 0.9$ T, the FIROs show maxima instead of minima at the expected minima positions. (b) Fourier transformations in $1/B$ of ΔR_{xx} and R_{xx} for various currents. The peak around $N_s = 7.0 \cdot 10^{11} \text{ cm}^{-2}$ is caused by the SdH-like bolometric ΔR_{xx} . The structure close to the fictitious carrier density $N_s^* = 2.13 \cdot 10^{11} \text{ cm}^{-2}$ corresponds to FIROs which persist to higher currents. Traces are offset for clarity

Additionally, the existence of FIROs at high and low currents is very clearly revealed in the Fourier transformations in $1/B$ of ΔR_{xx} shown in Fig. 5.16b. Two types of peaks are observed there: One type appears around $N_s = 7 \times 10^{11} \text{ cm}^{-2}$ and corresponds to the SdH oscillations and the bolometric ΔR_{xx} , whose period in $1/B$ is given by $\Delta(1/B) = 2e/hN_s$ (neglecting spin). The other type of peak appearing close to $N_s = 2.13 \times 10^{11} \text{ cm}^{-2}$ corresponds to the FIROs. Following the $1/B$ periodicity of MIROs of $\Delta(1/B) = 1/B_{CR} = e\lambda/2\pi cm^*$, FIROs at $\lambda = 184.3 \mu\text{m}$ are expected to appear at the fictitious carrier density $N_s^* = 4\pi cm^*/h\lambda = 2.13 \times 10^{11} \text{ cm}^{-2}$, which agrees well with our data. The Fourier transformation reveals that FIROs are also present at low currents, being superimposed on the relatively large bolometric ΔR_{xx} . At high currents, both the SdH oscillations and the bolometric ΔR_{xx} vanish, while the FIROs are less sensitive to the increased 2DES temperature, consistent with previous observations of MIROs [95].

Notably, the FIROs for $n \geq 5$ exhibit a sign reversal. A very similar sign reversal has recently also been observed for MIROs [98] in a very high-mobility

($\mu \simeq 1.2 \times 10^7 \text{ cm}^2/\text{Vs}$) 100- μm wide Hall bar. We observe a qualitative agreement with the data of Zhang et al. [98] in that the sign reversal is found to appear at low fields for increased 2D current densities. The broad background signal in ΔR_{xx} , which develops at higher currents centered around $B = 0.8 \text{ T}$, is related to a commensurability effect [121, 122] caused by a stressed lattice due to the grating coupler. Such commensurability oscillations are observed more clearly at fields below $B = 0.4 \text{ T}$ (not shown).

Finally, we like to note that in our sample, we observe magnetoplasmons [8, 9, 55, 89, 90] both in transmission and ΔR_{xx} (not shown), and the application of FIR-PC technique allows us to verify directly that the radiation-induced resistance oscillation is correlated only with CR, whereby the observed magnetoplasmon excitation shows no coupling to the FIROs. This work demonstrates the capability of using the sensitive FIR-PC technique to detect the MIRO analogous oscillations in the THz regime. In contrast to MIROs, such intriguing FIROs are observed even at the relatively large temperature of $T = 4.2 \text{ K}$ and in 2DES with moderate mobility of $\mu = 1.6 \times 10^6 \text{ cm}^2/\text{Vs}$. These experimental results shed new light on the complex photo-electronic processes of low-dimensional electron gases, whereby the striking phenomena of the radiation-induced resistance oscillations have attracted a broad interest in the international community.

5.7 Summary

Far-infrared transmission and photoconductivity spectroscopy are very powerful tools to investigate the elementary excitations in man-made semiconductor quantum materials. They allow us to investigate quantum dots filled successively with one, two, three, . . . electrons. In quantum dot and antidot arrays with tailored shapes and potentials, a rich variety of single-particle and many-body effects can be studied. Photoconductivity measurements are extremely sensitive. They make it possible to observe not only charge density excitations, like the quantized plasmon dispersion in the edge regime under the condition of the QHE, but also spin density excitation, e.g., the excitation of collective spin excitations, which is possible in materials with strong spin-orbit interaction.

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Chapter 6

Electronic Raman Spectroscopy of Quantum Dots

Tobias Kipp, Christian Schüller, and Detlef Heitmann

Abstract We review selected experimental and theoretical results on Raman spectroscopy of electronic excitations in charged quantum dots. Mainly, two different quantum dot systems have been investigated: GaAs–AlGaAs QDs fabricated by a lithographic top-down approach and InGaAs QDs grown by molecular beam epitaxy in a self-assembling bottom-up process. We recapitulate and compare the results of Raman experiments on both systems. We address collective many-particle charge and spin excitations, their magnetic field dispersion and dependence on a wave vector transfer, as well as their particular selection rules. We also review very recent experiments on self-assembled QDs containing exactly two electrons, since they form the simplest systems to study the most fundamental many-particle effects.

6.1 Introduction

During the past 40 years in semiconductor physics, Raman spectroscopy has proven to be a very useful tool in the investigation of free electrons, their interaction with themselves, and their coupling to other elementary excitations such as phonons. These experiments started with the investigation of bulk material, but, with sophisticated growth and structuring techniques, two-dimensional electron systems (2DES) in quantum wells and later 1DES in quantum wires and 0DES in quantum dots (QDs) were also investigated.

In a Raman process, light is inelastically scattered by the creation (Stokes process) or annihilation (Anti-Stokes process) of elementary excitations, which, in semiconductors, typically have energies in the far infrared (FIR) spectral range. The energy difference of the scattered light compared to the incoming light directly gives the excitation energy. Thus, Raman spectroscopy makes it possible to measure excitations with energies in the FIR region using visible light. Due to the two-photon process, Raman transition selection rules principally differ from the selection rules in FIR dipole absorption and thus give a complementary insight into the physics of low-dimensional systems.

This chapter focusses on Raman spectroscopy on electronic excitations in semiconductor QDs, widely called artificial atoms, in which a distinct number of electrons are confined. Since the signals that one deals with are inherently small, all measurements so far have been performed on ensembles of many QDs. In order to achieve a detectable signal strength at all, Raman transitions involving real electronic resonances have to be exploited instead of virtual states. Very recent experimental results, however, raise hope that Raman spectroscopy even on a single QD level might be feasible.

Most of the work of electronic Raman spectroscopy on ODES structures have been performed on two different types of semiconductor QDs: (1) GaAs–AlGaAs QDs fabricated in a top-down approach by laterally structuring modulation-doped semiconductor layer systems grown by molecular beam epitaxy (MBE) (e.g., [1–6]) and (2) In(Ga)As QDs grown in a self-assembled bottom-up process by MBE (e.g., [7–10]). Compared to real atoms, these QDs are lens-shaped, much larger, and of different confinement potentials, exhibiting energy level separations on the order of some millielectron volt. They allow a tailoring of the number of confined electrons. In particular, in self-assembled QDs, a precise tuning of the number of electrons even after the fabrication process and during the actual Raman experiment is possible. In principle, also the electronic structure of chemically synthesized QDs, such as CdSe nanocrystals, should be investigatable by Raman spectroscopy. Nevertheless, here, the comparatively low stability against high laser powers and the problem of a precise tailoring of the number of carriers inside the nanocrystals make these experiments technically highly demanding.

From the experimental point of view, Raman measurements on QDs are challenging not only because of the weakness of the signal but also because of the smallness of the energy of the electronic excitations which requires a very good stray light suppression of the experimental setup. In all experiments, the samples have been cooled down to cryogenic temperatures to avoid thermal population of excited states.

From the theoretical point of view, the electronic structure of charged QDs and also their corresponding Raman spectra can be modeled by different approaches. Concerning the electronic structure of QDs with many electrons approaches, such as Hartree–Fock or local density-functional calculations have been extensively used. For QDs containing only few (up to about 6) electrons, numerically exact calculations, including the full Coulomb interaction together with all exchange and correlation effects, can be done by exact diagonalization of the Hamiltonian usually in the basis of single-particle states. Based on these calculations, the Raman scattering cross sections can also be obtained by evaluating the corresponding matrix elements.

Recently, several reviews about Raman spectroscopy on nanostructures, especially on QDs, have been published. Christian Schüller's book about *Inelastic Light Scattering of Semiconducting Nanostructures* deals in very detail especially with etched GaAs–AlGaAs QDs [11]. Alain Delgado et al. reviewed electronic Raman scattering in QDs from the theoretical point of view, concentrating on relatively large QDs with several tens of electrons, such as experimentally achieved in etched QDs [12].

The scope of this chapter is not to give a complete review on the topic of electronic Raman spectroscopy on QDs. Instead, we select some of the earlier and already reviewed results to link them to very recently published experimental and theoretical work. The structure of this chapter is as follows: In Sect. 6.2, we sketch the two different fabrication processes of laterally etched and self-assembled QDs. Section 6.3 then gives a brief theoretical introduction on their electronic ground and excited states and possible electronic transitions. In Sect. 6.4, we first briefly recapitulate Raman experiments on QDs containing many electrons, since, historically, they form the basis of the later investigated systems with only a few electrons. We then briefly review recent experiments on etched QDs containing only few (≥ 4) electrons. In Sect. 6.5, we present results obtained on InAs QDs containing up to seven electrons. Here, we especially focus on the two-electron case, since this is the most fundamental many-particle system. Results are compared and linked to the previously discussed systems of etched QDs. Finally, in Sect. 6.6 we summarize and give a short outlook on new perspectives of Raman spectroscopy on QDs.

6.2 Fabrication of Charged Quantum Dots

Starting point for the fabrication of laterally etched QDs are MBE grown layer systems consisting typically of a one-sided modulation-doped $\text{GaAs-Al}_x\text{Ga}_{1-x}\text{As}$ quantum well. The number of electrons in the later QDs is predefined by the doping concentration during the growth. The confinement length in growth direction, typically in the range of only some nanometers, is of course also predefined by the layer growth. After growth, QDs are fabricated by lithographic processes. Here, a photoresist pattern is created by holographic laser interference or electron-beam exposure. The photoresist pattern is transferred into the sample by etching processes, most prominently by reactive ion etching. Typical lateral dimensions of the QDs are in the range of several 100 nm. Due to surface charges, the lateral confinement length of electrons inside the QDs is smaller than the geometrical dimensions, however, the lateral dimension of the QDs is still much larger than the confinement in z direction. It has been shown that the positively charged donor ions together with the negative surface charges lead to a more or less parabolic confinement potential for the electrons. Thus, the QDs can be modeled by a quantum well with hard walls in the growth direction and a soft harmonic confinement with quantization energies in the range of some millielectron volts in lateral direction. The QDs investigated in the earlier measurements typically contain many, i.e., several tens of, electrons. Only recently, laterally etched QDs containing down to four electrons have been investigated by Raman spectroscopy.

The fabrication process of InGaAs QDs follows a different approach. Here, the QDs are formed in a self-assembled way during the MBE growth in the so-called Stranski–Krastanov mode. Given the correct growth conditions, on a GaAs substrate, the deposition of InAs leads to the growth of one monolayer InAs, the so called wetting layer. The deposition of further InAs then leads to the formation

of three-dimensional InAs islands, driven by the slightly larger lattice constant of InAs compared to the GaAs substrate. Typical dimensions of self-assembled InAs QDs are 5 nm in height and 25 nm in width. Thus, these self-assembled QDs are also lens-shaped and can be regarded as being two dimensional. After their formation, the QDs are overgrown with GaAs to passivate their surface and ensure their clean electronic properties. Such InAs QDs typically exhibit lateral electronic quantization energies of 30–60 meV.

As in etched QDs, the number of electrons in self-assembled QDs can be tailored by a certain doping level during growth. Nevertheless, the most appealing approach would be to tune the number of electrons after the QD growth, even during the experiment. This can be accomplished by embedding them into a capacitor-like structure consisting of a MBE-grown back gate and a metallic semitransparent front gate evaporated on top of the sample. The back gate consists of either a two-dimensional electron system formed by an inverted heterostructure or a highly n -doped and thus conducting layer. By applying a voltage between the separately contacted back and front gate, electrons can tunnel from the back gate into the QDs. Due to the Coulomb blockade, this charging occurs stepwise on the voltage scale. The number of electrons charged into the QDs can be monitored by capacitance-voltage spectroscopy.

6.3 Electronic States in Quantum Dots

Both, laterally etched and self-assembled QDs can be described with a strong confinement potential in the growth direction and a much weaker confinement potential in the lateral directions. It can be assumed that the quantization energies in growth direction are large enough that only the corresponding ground state is populated. In this sense, one can regard the QDs as two dimensional. For etched QDs, it has been shown that the positively charged ionized dopants together with negative surface charges trapped at the sidewalls of the QDs lead to a harmonic oscillator potential in lateral direction for a test electron inside the QDs. The same kind of potential holds for self-assembled In(Ga)As QDs, although the underlying mechanism is different. Here, a compositional transition from GaAs to InAs material leads to a gradual change in the bandgap and, in first approximation, parabolic lateral confinement.

The single-particle energies of an electron in the conduction band in a QD of circular lateral shape are given by the eigenvalues of a two-dimensional harmonic oscillator

$$E_{nm} = \hbar\omega_0(2n + |m| + 1) = N\hbar\omega_0. \quad (6.1)$$

Here, n and m are the radial and angular quantum numbers, respectively, which can be merged into a single quantum number $N = 2n + |m| + 1$, characterizing the $2N$ -fold degeneracy of the energy levels. The quantization energy $\hbar\omega_0$ of the parabolic potential is in the range of some millielectron volts for etched QDs or some tens of millielectron volts for In(Ga)As QDs. If one applies now a magnetic

field B parallel to the growth direction, i.e., perpendicular to the QD plane, the energy levels split up into the Fock–Darwin single-particle levels

$$E_{nm} = (2n + |m| + 1)\hbar\sqrt{\omega_0^2 + \frac{1}{4}\omega_c^2} + \frac{1}{2}m\hbar\omega_c^2, \quad (6.2)$$

where $\omega_c = \frac{eB}{m^*}$ denotes the cyclotron frequency with the effective mass m^* . Thus, for $B \neq 0$ but negligible Zeeman splitting, each single-particle state is twofold degenerate due to the spin degree of freedom. According to the analogy in atomic physics, states with $|m| = 0, 1, 2, \dots$ are called $s, p, d, \dots$. The description of a QD in the so-far introduced single-particle picture is of course valid only for a single electron, nevertheless it is also extremely helpful for illustrating many-particle effects in QDs.

To describe many electrons of number N_e in a QD correctly, one has to consider their Coulomb interaction together with the Pauli exclusion principle. The model Hamiltonian reads

$$H = \sum_{i=1}^{N_e} \left[\frac{\mathbf{p}_i^2}{2m^*} + \frac{m^*}{2}\omega_0^2\mathbf{r}_i^2 \right] + \frac{e^2}{4\pi\epsilon\epsilon_0} \sum_{i \neq j}^{N_e} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}, \quad (6.3)$$

where the first sum represents the single-particle Hamiltonian and the second sum considers the Coulomb interaction. For two electrons, the problem can be solved more or less analytically, since the respective Hamiltonian separates into a center-of-mass and a relative Hamiltonian, each representing a harmonic oscillator [13]. In full analogy to the situation in a helium atom, the two electrons form either singlet (para helium) or triplet (ortho helium) states. On the one hand, the singlet states exhibit the same energy separation as the corresponding single-particle levels due to the generalized Kohn theorem. On the other hand, the triplet states have, compared to the corresponding singlet states, eigenenergies decreased by the exchange interaction. From the theoretical point of view, a QD containing just two electrons is the most fundamental and simplest system to observe quantum-mechanical many-particle effects. From the experimental point of view, electronic Raman spectroscopy on QDs historically started with structures containing several hundreds of electrons. Over the time, by developing superior fabrication techniques, the number of electrons decreased. Only recently it was possible to resolve singlet-triplet transitions in QDs containing just two electrons by Raman spectroscopy [10].

The electronic energy levels and wavefunctions for QDs containing up to about six electrons can be calculated by an exact numerical diagonalization of the corresponding many-particle Hamiltonian given in (6.3) (see, e.g., [8, 14, 15]). For more electrons, the system cannot be calculated in a numerically exact way. One usually uses self-consistent calculations such as Hartree- or Hartree–Fock-approximations to describe the ground state of the electron system inside the QDs. The calculated self-consistent effective potentials lead to new energy-level structures differing from the one of the external parabolic potential. In this sense, the parabolic potential

acting on one test electron is screened by the other electrons inside the QDs. Excitations of single electrons between energy levels of the effective potential are then called single-particle excitations. This is of course again only a crude approximation since the dynamic response of all other electrons on the excitation of the test electrons is not taken into account. Considering this dynamic response, qualitatively, one expects collective many-particle charge-density and spin-density excitations, CDEs and SDEs, respectively. For CDEs, all electrons in a QD with spin up and spin down oscillate in phase. Their energies are strongly affected by the direct part of the Coulomb interaction, which results in a blueshift compared to the single-particle excitation energy of the effective potential. The lowest-lying CDE is a plasma oscillation where all electrons in a QD oscillate in phase back and forth, thus, it exhibits a large electric dipole-moment and is FIR active. This confined plasmon is also called Kohn mode, since, as stated by the generalized Kohn theorem, its energy equals exactly the quantization energy of the unscreened external parabolic potential, independent of the number of electrons inside the QDs. In contrast to CDEs, for SDEs, the electrons with spin up and spin down oscillate with a phase shift of π against each other. They do not exhibit an electric but a large spin (or magnetic) dipole moment, thus being FIR inactive. Exchange-correlation interaction leads to a redshift of their energies compared to the single-particle energies. To treat the collective excitations in a more quantitative way, one has to consider a time-dependent perturbation and then calculate the excitation spectrum self-consistently. This is usually done in the framework of the random-phase approximation (RPA), which is a time-dependent Hartree approximation. Further improved methods also include exchange and correlation effects, such as the so-called time-dependent local-density approximation (TDLDA). The exact description of these sophisticated modeling methods is far beyond the scope of this article, thus, here, we refer to, e.g., [11, 12] and the references listed therein.

Until now, we briefly explained the calculation of the ground state and the excited states of electrons in QDs. The Raman process leads to transitions between these electronic states. Experimentally, such transitions can be observed only under specific resonance conditions involving intermediate valence-band states. Theoretically, such transitions can be modeled by a second-order perturbation approach, which gives for the transition amplitude A_{fi} ([12] and references therein):

$$A_{fi} \sim \sum_{\text{int}} \frac{\langle f | H_{e-r}^+ | \text{int} \rangle \langle \text{int} | H_{e-r}^- | i \rangle}{\hbar \nu_i - (E_{\text{int}} - E_i) + i \Gamma_{\text{int}}}. \quad (6.4)$$

Here, $|f\rangle$ and $|i\rangle$ represent the final and the initial state of the N_e -electron QD, respectively. E_f and E_i are the corresponding eigenenergies, their difference gives the Raman shift $\Delta E = E_f - E_i$. All possible intermediate states of the electronic system consisting of N_e electrons and an additional electron-hole pair are represented by $|\text{int}\rangle$. The Hamiltonian H_{e-r} describes the interaction between electrons and radiation, $\hbar \nu_i$ is the incident photon energy and Γ_{int} represents a phenomenological damping constant. Descriptively, one may regard the Raman process as a two-step process. In a first step, an intermediate state is populated by exciting an

electron from a valence band state into the conduction band. In a second step, the system goes into the final state by the recombination of a conduction-band electron with the valence-band hole. The involvement of valence-band holes in the intermediate states, which necessarily have to be taken into account if one wants to include resonance behavior, makes the calculations extremely challenging, in particular due to the heavy-hole–light-hole mixing of the valence-band states. Note that, for an even more precise modeling of the Raman process, polaronic effects from the influence of phonons also have to be taken into account.

From the evaluation of (6.4), important polarization selection rules for Raman spectroscopy can be deduced. It can be shown that, for nanostructures in zinc-blende semiconductors, on the one hand, CDEs can be detected in polarized configuration, where the polarization of the detected scattered light is parallel to the polarization of the incident light. On the other hand, SDEs occur in depolarized configuration, in which the polarization of the detected light is perpendicular to the one of the incident light [11, 12]. These polarization selection rules are no longer valid for an externally applied magnetic field [11, 12].

Generally, since the Raman process is a two-photon process, allowed electronic transition in QDs should have even parity. For circular QDs, this means that the total angular momentum of the electronic system should change by $\Delta M = 0, \pm 2$. The excitation of the above mentioned FIR-active Kohn mode comes along with a change of $\Delta M = \pm 1$. Representing the exclusion principle between FIR- and Raman-allowed transitions, this excitation is in first approximation not Raman active. Nevertheless, as has been shown also theoretically from a detailed evaluation of (6.4) [15, 16], virtually forbidden transitions can be observed in Raman experiments by transferring a finite wave vector of the incident photons into the electron system.

Note that from (6.4), which includes all intermediate states, general selection rules and particularly the amplitudes of peaks in resonant Raman spectra can be calculated. However, the positions of the peaks are exclusively given by the energy difference between the final $|f\rangle$ and the initial state $|i\rangle$.

6.4 Raman Experiments on Etched GaAs–AlGaAs QDs

6.4.1 QDs with Many Electrons

From Figs. 6.1 and 6.2, most of the important features of electronic Raman scattering in etched QDs can be deduced. The figures are taken from [3, 4]. The QDs under investigation have been prepared by deep-mesa etching, i.e., etching through the active layer of a 25-nm-wide, one-sided modulation-doped GaAs–Al_{0.3}Ga_{0.7}As single quantum well. The electron density and mobility of the unstructured samples were in the range of $(7\text{--}8)\times 10^{11}\text{ cm}^{-2}$ and $(3\text{--}7)\times 10^5\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$, respectively. The period of the array was $a = 800\text{ nm}$. The geometrical diameter of the QDs was about 240 nm. The number of electrons in the QDs was large such that several

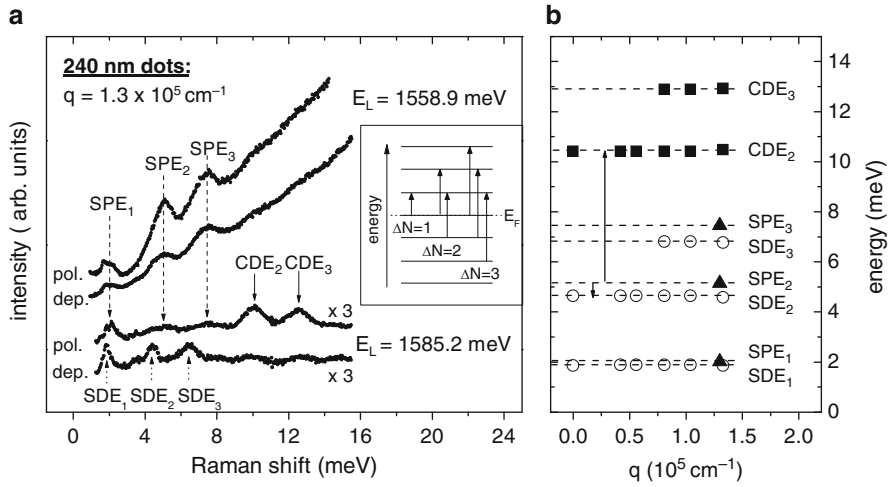


Fig. 6.1 (a) Polarized and depolarized spectra of an ensemble of quantum dots each containing several tens of electrons for different exciting laser energies E_L . The subscripts of the labels give the change ΔN in the quantum number N for the transitions, which contribute predominantly to the observed excitations. The corresponding transitions are sketched in the *inset*. The increasing background is due to luminescence, which is particularly pronounced for the extreme resonance condition at $E_L = 1558.9 \text{ meV}$. (b) Wave-vector dispersions of the experimental excitations in a QD sample. The arrows indicate the energy renormalizations of the $\Delta N = 2$ collective excitations with respect to the corresponding SPE_2 . The horizontal lines are guides to the eyes. (Following [3])

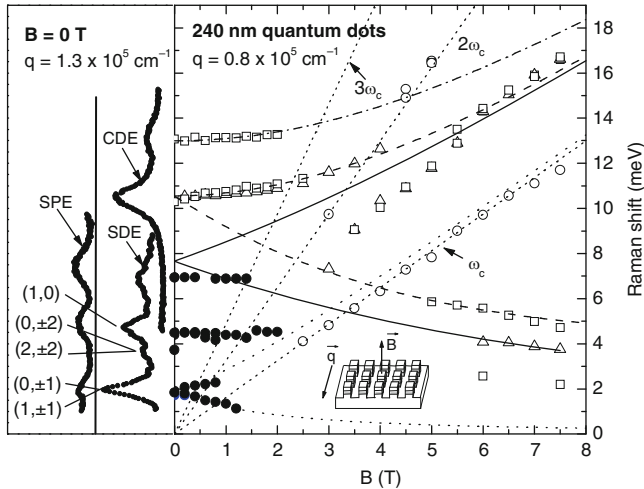


Fig. 6.2 B dispersions of SDEs (solid symbols) and CDEs (open symbols) in a quantum-dot sample. In the left panel, spectra of SDEs and CDEs for $B = 0$ are displayed, which were taken at $E_L = 1,587 \text{ meV}$. The spectrum of SPEs was recorded at a laser energy $E_L = 1,561 \text{ meV}$ under conditions of extreme resonance [Reprinted from [4]. Copyright (1998) by the American Physical Society]

zero-dimensional (0D) levels were occupied. The Raman experiments were performed at $T = 12$ K using a closed-cycle cryostat. The energy of the exciting Ti:sapphire laser was in the range of transitions from various confined hole states to the first excited electron state of the unstructured quantum well. The power densities were below 10 W cm^{-2} . The spectra were analyzed using a triple Raman spectrometer with a liquid nitrogen cooled charge-coupled device camera.

Figure 6.1a shows four Raman spectra: for each of the two different excitation laser energies E_L , two different polarization configurations have been applied. For a polarized (depolarized) spectrum, the polarization of the detected scattered light is chosen to be parallel (perpendicular) to the polarization of the exciting laser. Several lines are observed which can be classified in three categories: (1) peaks that occur predominantly in polarized configuration, (2) peaks that occur predominantly in depolarized configuration, and (3) peaks that appear in both configurations. Following the polarization selection rules given in Sect. 6.3, the peaks are assigned to (1) CDEs and (2) SDEs. Peaks of the third category are assigned to so-called single-particle excitations (SPEs), which are now regarded as superpositions of collective CDEs and SDEs ([14], see below).

The spectra in which either CDEs or SDEs occur have been recorded with a laser energy E_L well above the effective bandgap of the QDs. The peaks are labeled with a related change in quantum number ΔN . In polarized configuration, the CDE_2 is most prominent. The CDE_3 can only be observed for a finite wave vector transfer q , which has a value of $q = 1.3 \times 10^5 \text{ cm}^{-1}$ for the spectra in Fig. 6.1a. The CDE_1 is not resolved in the spectrum. In the depolarized configuration and for the given value of q , the SDE_1 to SDE_3 are observed. Figure 6.1b summarizes the energy positions of the observed peaks for different values of q . In polarized spectra, for $q = 0$, only the CDE_2 occurs. The CDE_3 appears only for larger values of q . In depolarized spectra, the SDE_1 and SDE_2 are visible for $q = 0$, while the SDE_3 occurs only for larger q . The lowest lying CDE_1 is the above mentioned Kohn mode. Its excitation would come along with a change in total angular momentum of $\Delta M = \pm 1$, thus the excitation is of odd parity, which makes it FIR active but Raman forbidden. The CDE_2 can be assigned to the monopole excitation with $\Delta M = 0$, thus being of even parity and Raman active. The CDE_3 can be again assigned to a dipole mode which is Raman forbidden, in first approximation. It cannot be observed until a transfer of wave vector q weakens the parity selection rules. In principle, CDEs and SDEs should behave similarly concerning the parity selection rules. Nevertheless, the violation of the parity selection rules by a wave-vector transfer q seems to be much stronger for SDEs than for the CDEs, since, e.g., the symmetry-forbidden SDE_1 is visible for even small values of q [4].

In Sect. 6.3, it is already mentioned that the energies of collective excitations are affected by the full Coulomb interaction. Compared to the single-particle excitation energies for a single test electron in the effective potential, which is the external parabolic potential screened by all other electrons, the CDEs are blue-shifted, whereas the SDEs are slightly red-shifted. This explains the observed energy differences between the corresponding CDEs and SDEs.

The spectra of Fig. 6.1a, in which the so-called SPEs occur, have been recorded with a laser energy E_L close to the effective bandgap of the QDs. This situation is usually called extreme resonance. The peaks are visible in both two polarization configurations and their energies lie somewhere in-between the energies of the corresponding SDEs and CDEs. They occur close to the energy levels for a single test electron in the self-consistently calculated effective potential. The labeling of these peaks as SPEs results from the analogy to reports on polarization independent and nearly unrenormalized excitations in a 2DES [17]. The occurrence of CDEs and SDEs can be expected from (6.4) even if it is drastically simplified by (1) assuming off-resonance excitation, which cancels the influence of the denominator, and (2) using the completeness relations for the intermediate states [12, 18]. However, to describe the situation in extreme resonance correctly, more elaborated evaluations of (6.4) without further simplifying assumptions have to be performed. These calculations show that for the case of extreme resonance, besides the well known excitations of a CDE and a SDE, further collective excitations with charge and spin character can be excited. In both cases, these excitations occur close to the single-particle energy. It is assumed that these excitations sum up to a rather broad peak, whose fine structure typically cannot be resolved in experiments. Since the broad peak is a superposition of charge and spin density excitations, it appears in, both, polarized and depolarized configuration, and slight differences in the spectra of different polarization, again, cannot be resolved experimentally [12, 14, 16, 18, 19].

The number of nearly unrenormalized excitations increases with the number of shells filled with electrons. For example, for a QD with six electrons, i.e., with two filled shells, one charge- and one spin-density excitation contribute to the SPE_2 . For 12 electrons, i.e., for three filled shells, two charge and two spin density excitations contribute to the SPE_2 peak [11, 14].

Figure 6.2 shows, in its right panel, the dispersion of the observed CDEs (open symbols) and SDEs (solid symbols) for a magnetic field B perpendicular to the QD plane, i.e., parallel to the growth direction of the sample. The left panel displays characteristic polarized and depolarized spectra for $B = 0$ T, showing, respectively, CDEs and SDEs. For comparison, a spectrum obtained under conditions of extreme resonance, which is dominated by the so-called SPE peaks, is also given. Until now, the excitations are characterized by the change in the general quantum number N . The magnetic field dispersion helps to characterize the excitation in more detail by the changes in the radial- and angular-momentum quantum numbers (Δn , Δm) of the dominantly involved single-particle transitions. This can be done in a simple qualitative model which assumes that not only the external potential for the QDs is parabolic but also, in first approximation, the effective screened potential. The single-particle energies in a magnetic field are then given by (6.2). The lowest energy SDE observed in the measurements is attributed to transitions with $\Delta N = 1$. From its splitting in a magnetic field, one can conclude that the SDE_1 consists of transitions with $\Delta m = \pm 1$, corresponding to a spin dipole mode, thus for the radial quantum number holds $\Delta n = 0$ or 1. Since the SDE_2 peak shows neither a strong dispersion nor a splitting in a magnetic field, it is attributed to ($\Delta n = 1$, $\Delta m = 0$)

transitions, which are the Raman-active monopole modes. Similar considerations are valid for the observed CDEs [4, 11].

From Fig. 6.2, another important experimental finding concerning the polarization selection rules can be deduced. The position of CDEs are marked by open symbols. These symbols are either squares, when the CDE is most prominent in polarized configuration, or triangles, when the peaks are most prominent in depolarized configuration. It can be seen that the usual polarization selection rules are weakened for finite magnetic fields $B > 0$ T [4, 11]. This behavior has also been predicted by theoretical calculations [12, 19].

6.4.2 QDs with Only Few Electrons

Recently, Raman experiments were reported on laterally etched GaAs–AlGaAs QDs containing only a few electrons [5, 6, 20]. Here, we briefly review results from César Pascual García and coworkers [5]. The QDs under investigation were fabricated from a 25 nm wide one-sided modulation-doped GaAs–Al_{0.1}Ga_{0.9}As quantum well. The electron density of $1.1 \times 10^{11} \text{ cm}^{-2}$ is considerable lower compared to the above discussed samples. The QDs had a geometrical diameter of 210 nm and were expected to be close to the regime of electron depletion. The number of electrons in the QDs is not known exactly and it is assumed that there is actually a distribution of the electron number in the QDs which leads to a broadening of the observed Raman signals. However, in the low-temperature ($T = 1.8$ K) spectra shown in Fig. 6.3a, the authors observe a peak at an energy shift of about 5.5 meV

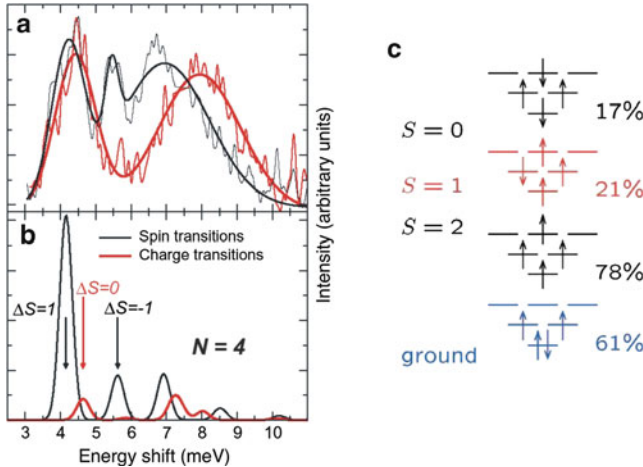


Fig. 6.3 (a) Experimental low-temperature ($T = 1.8$ K) polarized (red) and depolarized (black) Raman spectra. (b) Theoretical spectra for electron number $N_e = 4$. (c) Sketch of the most dominant Slater determinants for the ground state and Raman-accessible excited states, with their corresponding weight percentage [Reprinted with permission from [5]. Copyright (2005) by the American Physical Society]

in depolarized configuration which is extraordinarily sharp compared to the width of other peaks. Detailed calculations of Raman-allowed transitions of even parity (with $\Delta M = 0$) for QDs charged with $N_e = 2 \dots 6$ electrons reveal that such a peak can be expected only for QDs charged with $N_e = 4$ electrons. The peak is attributed to a transition from the triplet ground state (with total spin $S = 1$) to a singlet state (with $S = 0$). Comparison of the calculated spectra with the experiment shows that all other broader peaks can be associated with superpositions of excitations in QDs containing 4–6 electrons. The calculations have been performed by evaluating (6.4) with many-particle initial, intermediate, and final states obtained by the so-called configuration-interaction method, in which the full Hamiltonian of the QD is diagonalized on the basis of single-particle states [5, 21]. Calculated polarized and depolarized Raman spectra for $N_e = 4$ electrons are shown in Fig. 6.3b. The correlated wave functions of the ground and excited states can be written as Slater determinants. The most dominant single-particle configurations for the ground state and for three excited states, which can be accessed by an even-parity transition, are depicted in Fig. 6.3c. All states have an angular momentum of $M = 0$. The total spin of the ground state is $S = 1$. Transitions to excited states with $S = 2$ and $S = 0$ are spin excitations and are thus observed in depolarized configuration. The transition with no change in the total spin is a charge transition occurring in polarized configuration.

Researchers from the same groups have investigated similar samples of QDs containing $N_e = 4$ electrons also at even lower temperature ($T = 200$ mK) in magnetic fields perpendicular to the QD plane [6, 20]. Calculations reveal that at about $B = 0.35$ T, the electronic ground state of the QDs changes from the above described triplet state with $M = 0$ into a singlet state with $M = 2$ [20, 22]. This transition can be identified by Raman experiments, since the prominent $\Delta S = -1$ peak in Fig. 6.3a is peculiar for the triplet ground state. For $B = 0.4$ T, instead, a broad peak at slightly higher energy is observed which is attributed to the superposition of three neighboring collective spin excitations with $\Delta S = +1$ [20]. In [6], the authors discuss roto-vibrational modes of few electrons in QDs. For $N_e = 4$, they observe excitations associated with changes in the relative electronic motion, which are nearly independent of the total angular momentum $M = 0$ or $M = 2$ of the ground state.

6.5 Raman Experiments on Self-Assembled In(Ga)As QDs

6.5.1 QDs with a Fixed Number of Electrons, $N_e \approx 6-7$

The first observation of electronic excitations in self-assembled QDs was reported in the year 2000 by L. Chu and coworkers [7]. The sample under investigation contained 15 layers of InGaAs QDs, each layer having a n -type GaAs doping layer in its vicinity. The number of electrons inside the QDs was estimated to be about $N_e = 6$. The experiments were performed at low temperature $T = 4.2$ K with the

exciting laser at an energy of $E_L = 1.71$ eV exploiting resonances at the $E_0 + \Delta$ gap between conduction band and spin–orbit-split valence band states of the InGaAs QDs, far above the fundamental gap $E_0 \approx 1.07$ eV. Peaks in depolarized Raman spectra at about 50 meV were attributed to interlevel SDEs, between states with the general quantum number $N = 2$ and $N = 3$ (cf. (6.1)). The authors observed very similar spectra in polarized configuration, even though CDEs shifted to slightly higher energy compared to the SDEs were expected. This was explained by the inhomogeneous broadening of the observed peaks.

Multilayers of InGaAs QDs charged with electrons via adjacent doping layers were also investigated by B. Aslan and coworkers. In [9], they reported on the direct observation of polarons in QDs by resonant Raman scattering. The samples under investigation contained 50 layers of InGaAs QDs. The average electron number per dot was estimated to be $N_e \approx 7$. For a set of small pieces from the same part of the MBE grown wafer, a rapid thermal annealing process at different temperatures ranging from 750°C to 940°C was applied to tune the electron level spacing in the QDs from ≈ 50 meV to ≈ 20 meV across the LO-phonon energy of GaAs. The Raman experiments were performed at a low temperature $T = 15$ K with the excitation laser energy slightly above the QD ground state transition energy of the corresponding piece of sample. Peaks from electronic interlevel excitations were observed, showing a large anticrossing with both the InAs and the GaAs-like QD phonon, which characterized the strong coupling of electronic and phonon modes. The question whether the electronic excitations had spin or charge character was not addressed.

6.5.2 QDs with a Tunable Number of Electrons, $N_e = 2 \dots 6$

Self-assembled QDs offer the great possibility to tune the number of electrons N_e by placing them in a capacitor structure and by applying a gate voltage. This opens the possibility to observe electronic excitation by Raman spectroscopy in dependence of N_e on one and the same sample.

The first Raman measurements on QDs with a tunable electron number N_e have been reported by Thomas Brocke and coworkers [8, 23]. The investigated samples were grown by MBE on a GaAs(100) substrate. On top of a GaAs buffer layer and an AlGaAs–GaAs superlattice, a two-dimensional electron system of an inverted modulation-doped heterostructure consisting of 30 nm Si-doped $\text{Al}_{0.33}\text{Ga}_{0.67}\text{As}$, 15 nm AlGaAs, and 40 nm GaAs served as the later back contact. Then, one layer of self-assembled QDs was grown by depositing nominally 2.5 monolayers of InAs. A 33 nm GaAs layer, a superlattice of 16 pairs of AlAs and GaAs (2 nm each), and a 7 nm GaAs cap layer completed the MBE growth. A scheme of the band structure of such a sample is shown in Fig. 6.4a. To complete the capacitor-like structure of the sample, on its top, a 5 nm thick semitransparent titanium gate was deposited and separate alloyed contacts, which connected the 2DES, were fabricated. By applying an external gate voltage between back and front gate, the QDs were charged by

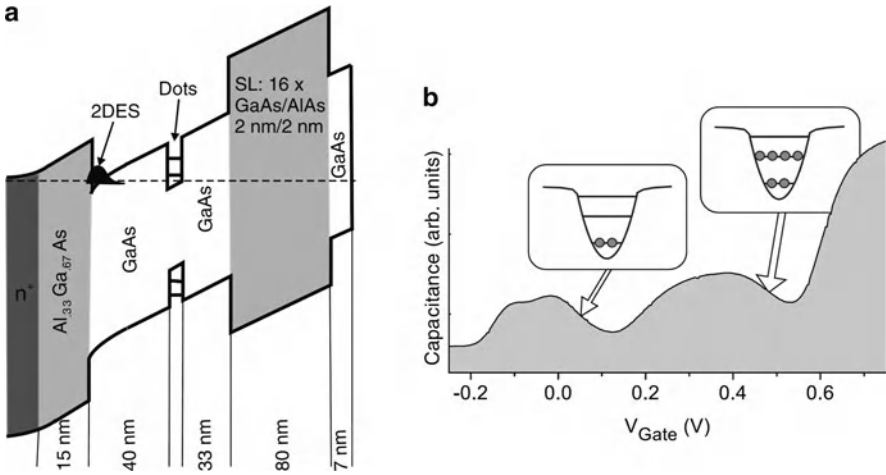


Fig. 6.4 (a) Scheme of the band structure of the QD sample. (b) Capacitance trace of the QD sample [Reprinted from [8]. Copyright (2003) by the American Physical Society]

electrons. The charging was monitored by measuring the capacitance [24]. A capacitance trace of the QD sample is given in Fig. 6.4b. The doublet structure around $V_{\text{Gate}} = -0.05$ V originates from the subsequent charging of the QDs by the first and the second electron. At $V_{\text{Gate}} = 0.12$ V, indicated by the local minimum in the trace, most of the QDs are occupied by exactly two electrons. A further increase of V_{Gate} leads to a further charging of the QDs by electrons occupying the p states. At around $V_{\text{Gate}} = 0.52$ V, the p shell of most of the QDs is completely filled, thus the QDs contain six electrons.

The Raman measurements were performed at low temperature $T = 8$ K with the excitation laser energy, similar to the above mentioned experiments by Chu et al., in the range of the $E_0 + \Delta$ gap of the QDs. Figure 6.5a shows Raman spectra measured in polarized configuration for 11 different gate voltages, i.e., for QDs containing $N_e = 2 \dots 6$ electrons. The corresponding electronic ground state configurations are sketched in a single-particle picture, assuming a parabolic potential of finite height, which is flattened at the edges due to continuum states of the adjacent wetting layer. In each spectrum, at 33.4 and 36.6 meV, two sharp lines occur which are due to the TO- and LO-phonon excitations of the GaAs bulk material. In the energy range of about 43–50 meV, broader bands are visible which are attributed to the electronic excitations inside the QDs. In a simple single-particle picture one can assign these excitations to transitions of electrons from the $N = 1$ shell (s shell, cf. (6.1)) to the $N = 2$ (or p) shell (peak A) or from the $N = 2$ shell to the $N = 3$ shell (peak B). The former transition can only occur when the p shell is not completely filled. The latter transition can only occur, when the p shell is at least partly filled. It has a smaller energy due to the flattened parabolic potential. This assignment is supported by resonance measurements: Peak B resonantly occurs for larger excitation laser energies than peak A [23]. This reflects the larger energy which is necessary to bring

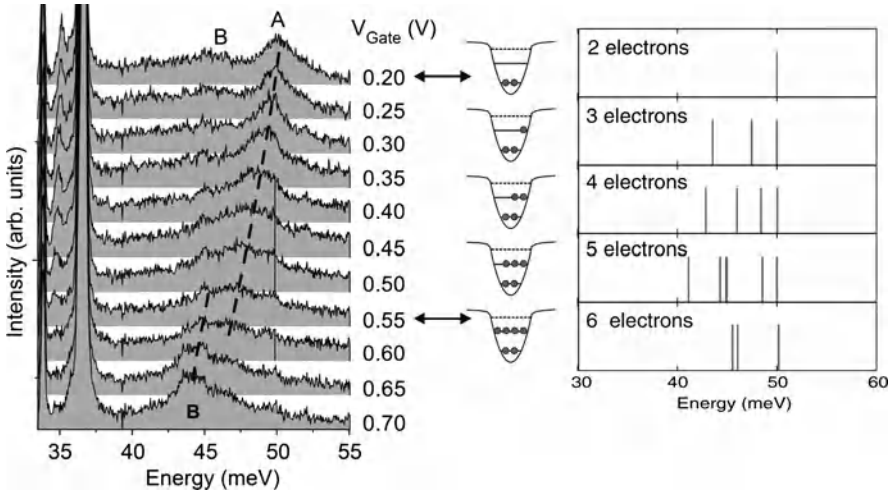


Fig. 6.5 (left) Polarized Raman spectra for different gate voltages V_{Gate} , corresponding to different charging states of the QDs. (right) Calculated energies of low-energy collective excitations of a two-dimensional QD for different electron numbers in the dot [Following [8, 23]]

an electron from a split-off valence-band state into an $N = 3$ state of the conduction band compared to a transition into a $N = 2$ state.

With increasing number of electrons, peak A broadens and its spectral weight shifts to smaller energies. In order to explain this behavior, Coulomb interaction of the electrons has to be considered. In [8], the many-particle Schrödinger equation with the Hamiltonian given in (6.3) has been solved by exact numerical diagonalization for $N_e = 2 \dots 6$. The lateral quantization energy was set to $\hbar\omega_0 = 50$ meV. For the effective mass, the InAs bulk value $m^* = 0.024m_0$ and for the dielectric constant $\epsilon = 15.15$ were used. Figure 6.5b gives the calculated energies of low-lying excitations, for which the total spin is preserved. The spin conservation is justified by the polarized configuration used in the experiments. One observes that, independent of N_e , there is always an excitation at the energy $\hbar\omega_0 = 50$ meV of the external confining potential. This is a consequence of the generalized Kohn theorem, which is already introduced in Sect. 6.3. For $N_e > 2$, additional modes appear below the energy of the Kohn mode. These additional modes cannot be resolved in the experimental spectra, but they might explain the shift and the broadening of peak A observed in the experiments.

The exact numerical diagonalization of the many-body Hamiltonian in the basis of single-particle states allows an additional insight into the microscopic picture of the low energy modes by expanding their corresponding wavefunctions in series of Slater determinants. Instead of going more into detail, we refer to [8], where the excited states of a QD containing $N_e = 3$ electrons are discussed in terms of Slater determinants.

Note that the theoretical modeling of the Raman experiments on QDs with tunable electron number which results in Fig. 6.5b is based just on the calculation of the

energy difference between ground and excited states obtained by diagonalizing the many-body Hamiltonian (6.3). Within this approach, only the position of excitation is calculated, but, of course, no resonance behavior via valence band states is considered, no Raman transition amplitudes are calculated, and thus no parity selection rules are regarded. The only link to the Raman experiments is that only excitations with preserved total spin are calculated, since they are assumed to occur in polarized configuration.

6.5.3 Comparison to Calculated Resonant Raman Spectra for $N_e = 2 \dots 6$

In order to calculate resonant Raman spectra, (6.4) for the transition amplitude has to be evaluated. Here, not only the initial ground state and the final excited state of the N_e -electron system contribute but also all intermediate states of $N_e + 1$ electrons and one hole play a role. Very recently, Alain Delgado and coworkers published a work which deals with exact diagonalization studies of electronic Raman scattering in self-assembled QDs [15]. This theoretical paper was motivated by the above mentioned experiments on self-assembled QDs [7–9]. Here, we briefly mention only some of its interesting results. The initial and the final states entering in (6.4) have been calculated similarly to the above mentioned method, by exact diagonalization of the many-particle Hamiltonian. The intermediate states entering the transition amplitude are interband excitations of the QDs. They are calculated by diagonalizing the appropriate Hamiltonian which includes electron–hole interaction. The initial, intermediate, and final states have then been used to calculate Raman spectra which are essentially proportional to the square of the transition amplitude (given in (6.4)) summed up over all final states. The authors show, amongst others, results for a $N_e = 6$ QD for polarized configuration under backscattering conditions with the incidence angle of 60° and the laser in resonance with the lowest lying absorption energy of the QD. The lateral confinement potential has been assumed to be parabolic with a characteristic energy of $\hbar\omega_0 = 30$ meV. It is shown that monopole excitations with $\Delta M = 0$ occur in an energy range between $2\hbar\omega_0$ and 10 meV below. These excitations are of strongest intensity, which reflects the parity selection rules for Raman processes. However, due to a finite wave vector transfer, also dipole excitations ($\Delta M = \pm 1$) occur, predominantly at $\hbar\omega_0$ (the Kohn mode) and below. Their intensities are calculated to be smaller than 10% of the strongest monopole excitations. A band of dipole excitations is also observed in an energy range slightly below $3\hbar\omega_0$, but with even lower intensity. Quadrupole excitations ($\Delta M = \pm 2$) give negligible contributions to the spectra.

From their calculations, the authors of [15] conclude that in experimental Raman spectra, only monopole excitations should be observable. Therefore they restrict their further theoretical investigations on monopole excitations. Independently on N_e , these modes occur in the energy range from clearly above $\hbar\omega_0$ to slightly above $2\hbar\omega_0$. Collective and single-particle-like charge and spin excitations are discussed

together with the possibility to discriminate between them by polarization selection rules. The theoretical results are compared to experimental results of Chu et al. [7], Brocke et al. [8], and Aslan et al. [9] introduced above in this review. Within these comparisons, the authors assume that only monopole excitations ($\Delta M = 0$) are observed in the experiments and that the lateral confinement energy for the QDs under investigation is $\hbar\omega_0 \approx 30$ meV. Then, the observed peaks in the experiment by Chu et al. [7] which occur around 50 meV with no clear dependency on the polarization configuration can be explained by monopole excitations in QDs with five instead of six electrons. It is also suggested to reinterpret the observed peaks in the experiment by Brocke et al. as monopole transitions. Superpositions of transitions for QDs containing four or five electrons could then explain the observed broadening and shift of the peak at 50 meV (peak A in Fig. 6.5a).

In our opinion, the interpretation of experimental Raman peaks observed in [7–9] as monopole excitation is disputable. Monopole transitions should occur around and slightly below the doubled energy of the external confinement energy, i.e., $2\hbar\omega_0$, as calculated, e.g., by Delgado et al. [15]. The Raman peaks in the experiments of Chu et al. occur at energies below 60 meV, which implies that $\hbar\omega_0 \leq 30$ meV, provided they are a matter of monopole excitations. However, Chu et al. reported on photocurrent measurements on similar samples as the one investigated with Raman spectroscopy [25], where they demonstrate the quantization energy for electrons to be 66% of the *total* confinement energy for electrons *and* holes which can be deduced from photoluminescence (PL) spectra. From the PL spectrum given in [7], one would thus expect the quantization energy to be $\hbar\omega_0 \approx 42$ meV. This is larger than the estimated value for the assignment of the measured peaks to monopole excitations. The same argumentation holds for the experiments of Brocke et al. Here, Raman peaks occur at energies smaller than 50 meV, which, again assuming monopole transitions, would imply that $\hbar\omega_0 = 25$ meV. PL measurements which, because of space limitations, were unfortunately not explicitly reported in [8, 23] reveal a *total* quantization energy for electrons *and* holes of about 69 meV [26]. Assuming again that 66% of this energy can be attributed to the electron quantization, one gets $\hbar\omega_0 \approx 46$ meV. This value is again much larger than estimated, assuming monopole transitions. Actually, this value even more suggests the assignment of the Raman peak at about 50 meV to the Kohn mode, even though it is a dipole transition which should be suppressed in first approximation due to parity. We stress here again that the resonance measurements in [23] strongly supports the assignment to, in a single-particle picture, dipole transitions from $N = 1$ states to $N = 2$ states for peak A and from $N = 2$ to $N = 3$ states for peak B in Fig. 6.5.

Aslan et al. prove a strong coupling of electronic excitations in QDs with phonons [9]. They observe for QDs with a total quantization energy of about 70 meV that coupled electron–phonon excitations occur at about 60 meV, where the energy of this coupled mode should be blue-shifted with respect to the underlying pure electronic excitation. Following our above given argumentation, these values also suggest that the underlying pure electronic excitation is more likely a dipole excitation close to $\hbar\omega_0$ than a monopole excitation close to $2\hbar\omega_0$ as proposed by Delgado et al.

The above given comparison of the so far most sophisticated calculations of resonant Raman spectra of self-assembled InAs QDs to experiments show that further effort has to be made on both, the experimental and theoretical side, to achieve an exact insight into the nature of electronic excitations in charged self-assembled QDs. Limitations of the comparability of the calculations of Delgado et al. to the experiments by Chu et al. and Brocke et al. are probably arising due to the fact that the theory considers resonant excitation at the fundamental E_0 gap whereas the experiments exploit resonances at the $E_0 + \Delta$ gap. The experiments of Aslan et al. evidence that the electron–phonon interaction is also crucial and should be considered in calculations. Further discrepancies definitely occur because of the deviations of the lateral QD shape from the perfect rotational symmetry assumed in calculations. Asymmetries of the QDs might strongly alter the selection rules which strongly favor monopole excitations in circular QDs. For the experiments, it would be favorable to have more homogeneous samples. Until now, all experiments have been performed on ensembles of QDs with inhomogeneities of shape, size, and electron number. Ultimate experiments should be performed on a single QD to overcome inhomogeneities.

6.5.4 QDs with $N_e = 2$ Electrons: Artificial He Atoms

In the last part of this section, we review very recent experiments of Tim Köppen and coworkers on self-assembled InGaAs QDs which could be charged by two or one electron [10]. These systems are highly interesting since two-electron QDs, also called artificial helium atoms, are the simplest systems to observe many-particle effects, i.e., the occurrence of singlet (para helium) and triplet (ortho helium) states.

The investigated samples are very similar to the ones of Brocke et al. which are described above (see Fig. 6.4). The main difference is that the samples have been treated by a rapid thermal annealing process before the fabrication of front gates and contacts to increase the fundamental bandgap of the QDs. This makes it possible to directly excite at the E_0 gap of the QDs. Figure 6.6a shows a low-temperature nonresonant PL spectrum of the sample on which the Raman experiments shown in the following have been performed. The ground-state recombination of electrons and holes occur at 1.308 eV. The sequence of emission peaks out of higher excited states proves the total lateral quantization energy of electrons and holes to be about 33 meV. Figure 6.6b gives a low-temperature capacitance trace of the sample in which the subsequent charging of the QDs with the first and the second electron is resolved. The Raman experiments were performed for a gate voltage of either $V_{\text{Gate}} = 0.30$ V or $V_{\text{Gate}} = 0.16$ V corresponding to $N_e = 2$ or $N_e = 1$. The sample was cooled down to $T = 9$ K in a split-coil cryostat allowing for magnetic fields up to $B = 6.5$ T.

Figure 6.7 shows spectra in Raman depiction at $B = 4.5$ T for varying excitation laser energies E_L . Several sharp peaks of different resonance behaviors are observed. The peaks labeled T_- , S_- , and T_+ get resonant at lowest E_L . With

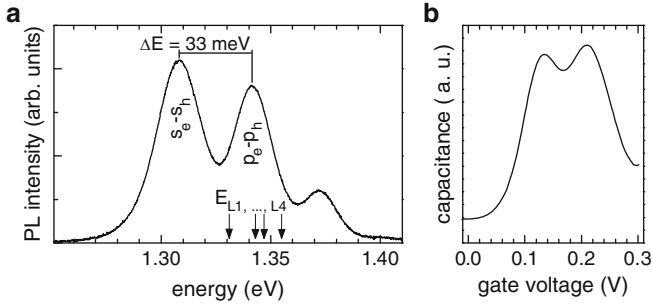


Fig. 6.6 (a) Nonresonant PL spectrum of the QD sample. (b) Capacitance trace of the QD sample [Reprinted from [10]. Copyright (2009) by the American Physical Society]

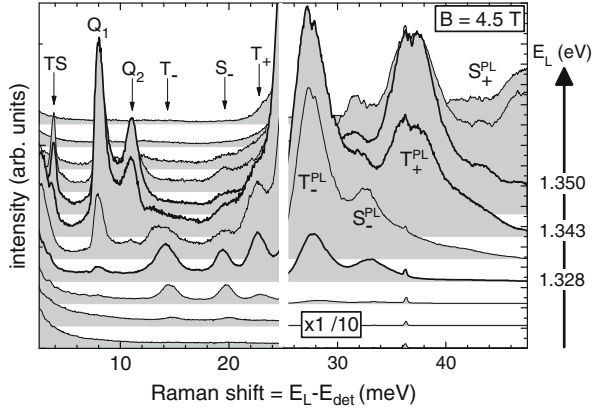


Fig. 6.7 Polarized spectra obtained at $B = 4.5$ T for laser energies E_L varying between 1.306 and 1.380 eV. The spectra are vertically shifted and intensities corresponding to energies above 25 meV have been divided by 10. [Reprinted from [10]. Copyright (2009) by the American Physical Society]

increasing E_L ; the peaks labeled T_-^{PL} , Q_1 , Q_2 , and T_+^{PL} get successively resonant. Figure 6.8a is a compilation of single spectra such as the ones shown in Fig. 6.7 for different magnetic fields B for resonant excitation with four different laser energies E_{L1} to E_{L4} which were chosen close to the resonance of the T and S peaks, the T_-^{PL} peak, the Q peaks, and the T_+^{PL} peak, respectively. The measured intensities are encoded in a gray scale. Regions of different excitation energies are separated by white gaps. The dispersive branches indexed with PL which occur for laser energies around E_{L2} and E_{L4} result from resonantly excited PL. Figure 6.8b sketches the corresponding transition scheme in a single-particle picture for $B > 0$ (see (6.2)) exemplarily for the T_-^{PL} and S_-^{PL} branches which involve $m = -1$ ($\Leftrightarrow p^-$) states for electrons and holes. Analog schemes can be drawn for T_+^{PL} and S_+^{PL} . In a first step, the excitation laser resonantly creates a p_e - p_h electron-hole pair. In a second step, energy dissipation into the lattice occurs when the hole relaxes into the s_h state.

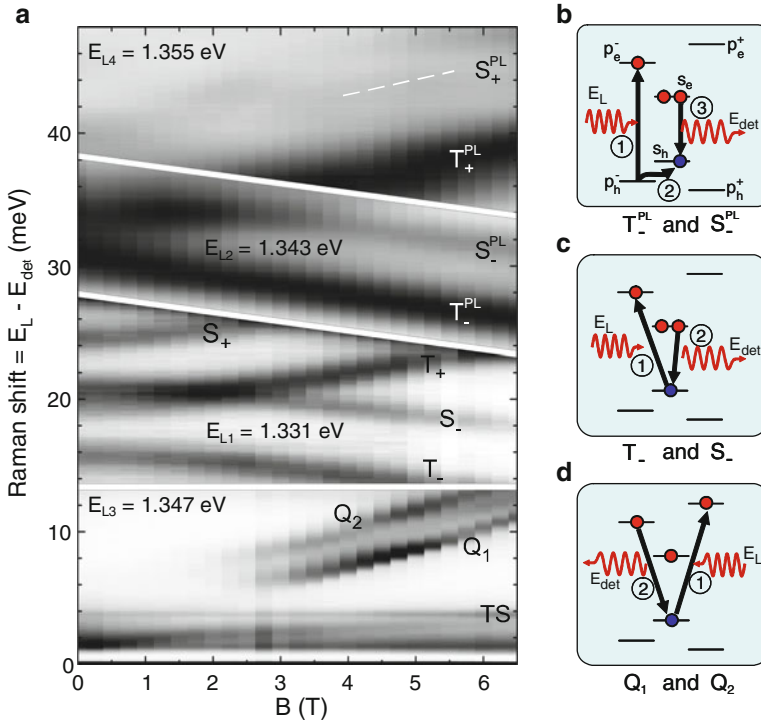


Fig. 6.8 (a) Compilation of spectra of quantum dots in He configuration for resonant excitation with different laser energies E_{L1} to E_{L4} . Intensities are encoded in a gray scale, the horizontal axis gives the magnetic field B , the vertical axis gives the difference between E_{Li} ($i = 1, \dots, 4$) and the detection energy E_{det} . Spectra were taken in polarized configuration. (b–d) Transitions schemes in a single-particle Fock–Darwin picture for (b) resonant PL and (c, d) resonant Raman processes [(a) is reprinted from [10]. Copyright (2009) by the American Physical Society]

The p_e electron cannot relax since the s_e state is initially filled by two electrons. In a third step, a radiative recombination process takes place leaving the QDs behind in a configuration with one electron in the s state and the other electron in the p state. Beyond the single-particle picture, if one regards Coulomb interaction, these two electrons form either a singlet or a triplet [13, 27], in full analogy to the situation in a He atom. The recombination process into different final states leads to different emission lines T_-^{PL} , T_+^{PL} , S_-^{PL} , and S_+^{PL} .

The four dispersive branches labeled T_- , S_- , T_+ , and S_+ in Fig. 6.8a are assigned to transitions from the singlet QD helium ground state to excited triplet and singlet states provoked by resonant Raman scattering. These transitions have their resonances at laser energies around $E_{L1} = 1.331$ eV, clearly below the energy of the dipole-allowed p_e - p_h transition (cf. Fig. 6.6). Compared to resonant PL branches, they occur about 11 meV closer to the energy E_L of the exciting laser and their intensities are more balanced among each other. The underlying Raman scattering process is sketched exemplarily for the T_- or S_- branches in Fig. 6.8c. First, the

laser light resonantly creates a p_e^- - s_h electron-hole pair (p_e^+ - s_h for T_+ or S_+), then, a radiative s_e - s_h transition occurs, leaving behind the QD in an excited either triplet or singlet state. Compared to the resonant PL process, this process fundamentally differs in the absence of the p_h - s_h relaxation, i.e., energy dissipation into the lattice via phonons. Consequently, the excitation step has an energy decreased by the hole quantization energy (11 meV, i.e., $\sim 33\% \cdot \Delta E$, of the total lateral confinement energy). Thus, the Raman process gives directly the excitation energy from the ground state QD He into the excited para- and ortho-He QD states without any ambiguity due to assumptions on the hole confinement energy. Like the T_-^{PL} and T_+^{PL} branches, both the T_- and T_+ as well as the S_- and S_+ branches are not degenerate even for $B = 0$ T, which arises from a slight asymmetry of the lateral potential.

The low energy branches Q_1 , Q_2 , and TS in Fig. 6.8 are assigned to transitions between excited states. The Q_1 and Q_2 branches are attributed to excitations, respectively, from T_- to T_+ and from S_- to S_+ states. The TS branch is tentatively assigned to transitions from the excited triplet to the excited singlet states. All these excitations resonantly occur around laser energies $E_{L3} = 1.347$ close to the p_h - p_e transition. For these excitations, two resonance conditions have to be fulfilled. First, the excited states are resonantly populated within the resonant PL process sketched in Fig. 6.8b. Then, resonant transitions between excited states take place. Exemplarily for the Q branches, this is sketched in Fig. 6.8d. Here, we do not go into further details of these excitation and just refer to Ref. [10].

Coming back to the $T_{+/-}$ and $S_{+/-}$ branches and comparing them to the $T_{+/-}^{\text{PL}}$ and $S_{+/-}^{\text{PL}}$ branches, one finds several important differences. Besides the already mentioned difference in energy shift and resonance position, the Raman peaks are much more balanced in their intensities, which also manifests that the different branches result from a different excitation process. Furthermore, as has been shown in [10], the Raman peaks show a clear polarization configuration dependence and can be enhanced by a lateral wave vector transfer, opposed to the PL peaks. The polarization dependency is such that the T branches occur dominantly in depolarized configuration whereas the S branches occur dominantly in polarized configuration. This is in agreement with the polarization selection rules for SDEs and CDEs, since the T transitions are from singlet into triplet states with a change of the total spin $\Delta S = 1$, whereas the S transitions are between singlet states and affect only the charge and not the spin. In the experiments, these polarization selection rules weaken for finite magnetic field, just like it has been reported before for etched QDs (see Sect. 6.4.1). The underlying single-particle transition for both the T and S excitation is ($\Delta n = 0$, $\Delta m = \pm 1$), as can be seen from Fig. 6.8c. Thus, these excitations are dipole excitations (with a change of the total angular momentum of $\Delta M = \pm 1$), which are in first approximation Raman forbidden because of parity, as discussed above. In [10], a strong enhancement in intensity of the Raman peaks compared to the PL peaks for an increased lateral wave vector transfer q is reported. Indeed, the enhancement of nominally Raman-forbidden transitions by a wave vector transfer has been measured and calculated (see Sects. 6.4.1 and 6.5.3). However, the occurrence of dipole transitions even for negligibly small q proves the parity selection rules in the InGaAs QDs to be inherently weakened. The main

reason for that might be the anisotropy in the lateral potential of the QDs for which, strictly speaking, M is no longer a good quantum number.

As already mentioned in Sect. 6.3, electronic states in QDs containing two electrons described by the Hamiltonian in (6.3) can be calculated more or less straight forward. It is calculated that for $B = 0$, the transition energy from the ground state into the triplet state is about 71% of the singlet-singlet transition energy. This is in good agreement with the value of 78% deduced from the measurements. For a more precise modeling of the QDs, of course, the anisotropy of the lateral potential has to be considered. Even more interesting than the bare electronic states and their eigenenergies would, of course, be the calculation of resonant Raman spectra by evaluation of (6.4) similar to the calculations of Delgado et al. [15] which are reviewed above in Sect. 6.5.3. We suppose that such calculations which would also take into account the asymmetry should explain the experimentally observed rather large intensity for dipole transitions. Even more sophisticated theory should also include heavy- and light-hole mixing for the intermediate states and the interaction with phonons. Strong last-mentioned polaronic effects have been observed in Raman measurements on InAs QDs containing about seven electrons [9]. Interestingly, in above mentioned experiments by Köppen et al., no polaronic effects have been observed for QDs containing two electrons, but strong polaronic effects occur when the same QDs are charged with only one electron. This behavior is still not completely understood.

6.6 Summary

The experiments reviewed in this work show that Raman spectroscopy is indeed a powerful tool to investigate electronic excitations in charged QDs.

The first systems under investigation were laterally etched GaAs-AlGaAs QDs containing many, usually several tens or hundreds of, electrons. In these QDs, one observes charge- and spin-density excitations as well as so-called single-particle excitations. The CDEs and SDEs occur in different polarization configurations, i.e., in polarized and depolarized spectra, respectively. The SPEs occur in both polarization configurations. Raman spectra of QDs with many electrons can be modeled theoretically by self-consistent calculations including time-dependent perturbations. These calculations correctly reflect the polarization selection rules for CDEs and SDEs. Furthermore, they reveal the parity selection rules resulting in the dominance of monopole transitions in Raman scattering. Also, the experimentally observed weakening of the parity selection rules, when a lateral wave vector is transferred, as well as the weakening of the polarization selection rules in the presence of an external magnetic field are modeled by the theory. The SPEs, experimentally observed when exciting in extreme resonance with the QDs, are identified as superpositions of CDEs and SDEs of nearly equal energy.

Later experiments on etched QDs containing only a few, i.e., less than 7, electrons also show charge- and spin-monopole excitations occurring in different polarization configurations. These systems allow a theoretical modeling by the exact numerical

diagonalization of the underlying many-particle Hamiltonian, which leads to a more intuitive insight into the electronic excitations by expanding them with the help of Slater determinants.

In the first Raman experiments on self-assembled QDs, electronic excitations have been observed, but a clear discrimination between charge and spin excitations such as for etched QDs seemed to be difficult. This was most likely because of comparatively large inhomogeneities in size and charging of the QD ensembles under investigation. In more recent investigations of InGaAs QDs exhibiting a higher degree of homogeneity, indeed, charge and spin excitations are observed which follow the polarization selection rules. For self-assembled In(Ga)As QDs, the validity of the parity selection rules obtained from theoretical considerations is still under discussion. In contrast to the theory, electronic excitations observed in different experiments by different groups were in each case attributed to dipole excitations. In our opinion, especially the most recent experiments of Köppen et al. clearly demonstrate the excitation of dipole transitions. We expect this discrepancy to be solved by a theory which includes particularly the experimentally observed lateral anisotropy of the QD potential and also heavy- and light-hole mixing as well as, probably, the interaction with phonons.

The recent measurements on InGaAs QDs which contain two electrons somehow take up a special position in the row of Raman experiments on QDs. Due to the possibility to charge these QDs during the actual Raman experiments by an external gate voltage and to monitor the charging by capacitance voltage spectroscopy, the adjustment of the electron number is much more accurate than in QDs charged by dopants of a predefined concentration. Due to the completely filled s shell in QDs with two electrons, i.e., in artificial QD helium atoms, the charging condition is excellently homogeneous even for a large ensemble of QDs. Compared to QDs containing more electrons, in these artificial QD helium atoms, the many-particle effects are reduced to their simplest and most fundamental representation as singlet and triplet states. The resonant electronic Raman transitions in QD helium exhibit considerably strong intensities. This might even allow for Raman measurements on individual QDs, which actually is an ultimate but not yet reached goal.

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Chapter 7

Light Confinement in Microtubes

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Abstract We review recent developments in the field of light confinement in semiconductor microtube resonators fabricated by utilizing the self-rolling mechanism of strained bilayers. We discuss resonant optical modes in the framework of a waveguide model that naturally explains the occurrence of two-dimensional ring modes by constructive interference of light azimuthally guided by the tube wall. Experiments show that diverse geometries of a microtube have strong impact on the emission properties, including preferential and directional emission, as well as on a three-dimensional light confinement. We show that by lithographically structuring the microtube, it is possible to reach a three-dimensional confinement in a fully controlled way. The evolving confined modes can be described by an intuitive model using an expanded waveguide approach together with an adiabatic separation of the circulating and the axial light propagation.

7.1 Introduction

Semiconductor microcavities are optical devices in which light is spatially confined on a scale of its wavelength. These cavities gained considerable interest in the last years because, on the one hand, they offer the possibility to study fundamental interaction effects between light and matter, and on the other hand, they might be applicable in new and superior optoelectronic devices [1]. Pioneering works, for example, demonstrated the Purcell effect, i.e., the modification of the spontaneous emission rate, of quantum dot (QD) emitters embedded in microcavities [2,3]. Later, it was shown that one can even reach the strong coupling regime between QDs and cavity modes, proven by the so-called vacuum Rabi splitting [4–6]. Prerequisites for such experiments are high quality factors and low mode volumes inside the microcavities. Concerning possible applications, microcavities might lead to superior lasers with low or even no threshold [7–9] or to single-photon sources applicable in quantum cryptography [10, 11]. Furthermore, their use in possible quantum computers are discussed [12].

Three different kinds of semiconductor microcavities have been intensively investigated: (1) Micropillars, (2) two-dimensional photonic-crystal microcavities, and (3) microdisks. Micropillars result from lateral structuring of vertically arranged Bragg reflectors. The periodic modulation of the refractive index inside the Bragg mirrors leads to a strong light confinement between the mirrors in vertical direction. In lateral direction, the confinement is caused by the large difference in refractive index between semiconductor material and air. In two-dimensional photonic-crystal microcavities, the periodic modulation of the refractive index of a thin semiconductor membrane leads to a strong lateral light confinement, whereas the vertical confinement is caused again by the difference in refractive index between semiconductor and air. Microdisks consist of circular semiconductor slabs centered on a thin semiconductor post. Here, light confinement is caused by internal total reflection at the border of the disk.

Optical microtube resonators form a new class of microcavities, firstly demonstrated in the year 2006 [13]. The basis for their fabrication is the self-rolling mechanism of strained layer systems lifted-off from their substrate [14, 15] together with its full lithographic control [16]. For further reading, this book's chapter by Peters, Mendach, and Hansen, especially the section "The Basic Principle Behind 'Rolled-Up Nanotech,'" is recommended. This basis is used to fabricate self-supported microtube bridges in which optical emitters like QDs serve as internal emitters. Typical dimensions of these microtubes are $5\text{ }\mu\text{m}$ for diameter, $100\text{--}200\text{ nm}$ for the wall thickness, and $10\text{--}50\text{ }\mu\text{m}$ for the length. The tubes' walls serve as waveguides for the luminescence light of the internal emitters. The azimuthally guided light interferes after a round trip along the circumference of the tube, which leads to optical modes for constructive interference. Microtubes resonators exhibit the striking features of a nearly perfect overlap between embedded emitters and the intensity maximum of the optical modes as well as low surface scattering rates. The strong evanescent fields of the optical eigenmodes should enable a good coupling to optical networks by waveguides or to emitters brought in the vicinity of the thin walls.

In the last years, optical microtube resonators have been extensively studied [13, 17–27]. These studies deal with different material systems – based on, e.g., InGaAlAs [13, 17, 19, 20, 22, 23, 27] or Si [18, 24–26] – different emitters – like QDs [13, 17, 20, 21, 23] or quantum wells (QWs) [19, 22] – and different possible applications, e.g., as lab-on-chip refractometers [24].

The main topics of our work on microtube resonators so far [13, 19–22] were the demonstration, understanding, modeling, and exact tailoring of three-dimensionally confined optical modes in microtube resonators. The ringlike, cross-sectional shape of a microtube of course has the strongest impact on the optical modes since it ensures confinement of light in azimuthal direction. However, in order to achieve a real three-dimensional confinement of light, confinement mechanisms along the axis of the microtube are of great importance. In this review, we want to carry together selected results concerning the three-dimensional light confinement in microtubes. Our experimental results were obtained on systems based on InGaAlAs microtubes, but they are not restricted to this material system.

We will first give a very brief introduction to the fabrication of optical microtube resonators and to the kind of optical experiments that are reviewed in the following. We will then present selected experiments with which the properties of optical modes in microtubes are discussed.

7.2 Fabrication

Starting point for the fabrication of our microtube cavities are molecular beam epitaxy (MBE) grown samples. A typical layer sequence is sketched in Fig. 7.1a. On top of a GaAs substrate, an AlAs layer serves as a sacrificial layer in the later processing. On top of this, a strained layer system is grown, which will form the actual microtube. The design of this layer system predefines the structural properties like the rolling radius and the wall thickness, as well as the electronic properties by its band structure. Importantly, also optical emitters can be integrated in the structure. In the case of the sample sketched in Fig. 7.1a, which leads to microtubes with QDs embedded as internal emitters, it consists of strained 20 nm $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$ and 30 nm GaAs, centrally containing one layer of self-assembled InAs QDs. We also investigated microtubes with QWs as internal light sources. A typical strained layer system then for example consists of 14 nm $\text{In}_{0.15}\text{Al}_{0.21}\text{Ga}_{0.64}\text{As}$, 6 nm $\text{In}_{0.19}\text{Ga}_{0.81}\text{As}$, 41 nm $\text{Al}_{0.24}\text{Ga}_{0.76}\text{As}$, and 4 nm GaAs. Here, both In-containing layers are pseudomorphically strained grown and the InGaAs layer forms a QW sandwiched between higher bandgap barriers.

Figure 7.1b shows an optical microscope image of the microtube, for which the observation of optical modes was firstly reported [13]. On the basis of this image, we briefly introduce the fabrication process. It starts with the definition of a U-shaped strained mesa by etching into the strained InGaAs layer. In a next step, a starting edge is defined by etching through the AlAs layer. Here, the AlAs is now uncovered

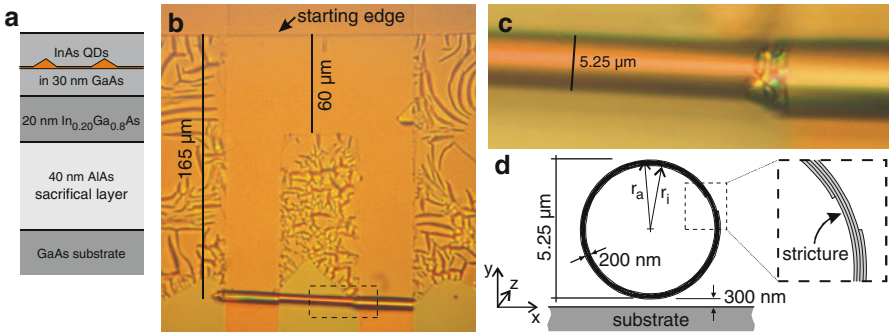


Fig. 7.1 (a) Schematic sample structure for the fabrication of microtubes with QDs embedded. (b) Optical microscope image of a microtube bridge. The U-shaped part has rolled-up beginning at the starting edge. (c) High resolution optical microscope image of the microtube bridge and its bearing. (d) Scaled schematic section of the self-supported part of the microtube. (Following [13])

and, in the last step, a HF solution starts to undercut the strained layer system with a high selectivity. This leads to a bending of the strained mesa over its whole width resulting in the formation of a microtube. After a distinct distance defined by the U-shaped mesa (60 nm for the tube shown in Fig. 7.1b), only the side pieces of the tube continue rolling. This raises the center tube, leading to a self-supporting microtube “bridge,” where in the middle part the tube is separated from the substrate. This preparation process can be improved by etching deeply into the AlAs layer in the region between the legs of the U-shaped mesa and protecting the laid-open AlAs by a photoresist layer during the following selective etching step. This procedure leads to a larger and more controllable lifting of the center part of the microtube from the substrate [21]. Figure 7.1d shows a scaled cross-section of the center part of the tube. The outer diameter is about 5.25 μm . The 3.8 revolutions lead to an overall tube wall thickness of only 200 nm (150 nm in the region of the stricture). Since microtubes have the shape of rolled carpets, they exhibit discontinuities at the inside and outside surface, which we call rolling edges in the following. These rolling edges have a large impact on a microtube resonator concerning its light confinement and emission properties, as will be explained in the following.

7.3 Experimental Setup

The optical modes in our microtubes were probed by the photoluminescence (PL) light emitted from optically active QDs or QWs, which are embedded in the tube walls. The samples were mounted in a cryostat at low temperature ($T = 5 - 7$ K). The excitation laser was focussed onto the sample by a microscope objective. The microtube was imaged by the same objective and further optics on the entrance slit of a grating spectrometer. For detection, we used a cooled charge-coupled device (CCD) camera. One possibility to perform spatially resolved measurements is to scan the sample underneath the fixed excitation laser spot while taking spectra. Another technique makes use of the fact that the grating disperses light from each position along the entrance slit of the spectrometer but conserves the spatial information along the slit. Thus, by evaluating the signal of the two-dimensional CCD chip, one obtains energy-resolved spectra for each spatial position along the entrance slit, on which the sample is imaged. A third technique uses the grating in its zeroth order of diffraction as a mirror, thus allowing a direct two-dimensional imaging of the sample onto the CCD chip.

7.4 Microtubes with Unstructured Rolling Edges

Figure 7.2a shows PL spectra of the microtube presented in Fig. 7.1 in the energy range, in which the QDs emit. One observes a regular sequence of sharp peaks superimposed on the broad QD luminescence. These sharp peaks are optical resonances

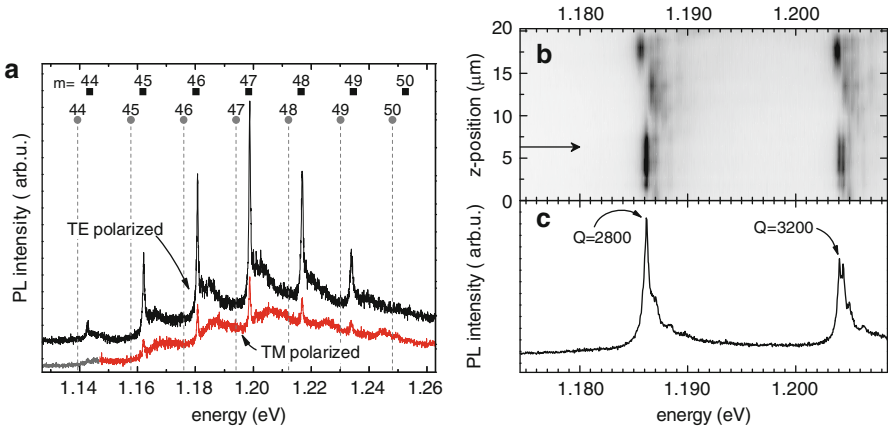


Fig. 7.2 (a) Micro-PL spectra of a microtube bridge measured in TE (*upper graph*) and TM (*lower graph*) polarization configuration. The spectra are vertically shifted for clarity. The *symbols* indicate calculated mode positions (without any fitting) labeled with their azimuthal mode number m . The squares (*circles*) represent the waveguide (exact) approach. (b) Gray scale plot of PL spectra measured at different positions on the microtube along its axis. *Dark regions* represent strong intensities. The spectrum marked with the arrow is depicted in (c). (Following [13])

arising from light that is guided around the tube axis inside the tube wall and that constructively interferes with itself. Two spectra obtained on one and the same position on the center part of the microtube but for different polarization configurations are compared. The upper curve corresponds to the transversal electric (TE) polarization, which we define as having the electric field vector parallel to the tube axis. We prove the optical modes to be TE polarized. Their appearance also in the TM spectrum (which is much less pronounced than in the TE case) is due to a limited polarization selectivity of the setup used. Two different theoretical models explain the experimental results. In the first, the so-called waveguide model, we regard the microtube wall as a dielectric waveguide with a height given by the overall tube wall thickness $h = r_a - r_i$, see Fig. 7.1d. We calculate the modes of a planar waveguide and assign them to an effective refractive index n_{eff} . To ensure phase matching of guided light after one round trip, we apply the periodic boundary condition $n_{\text{eff}}l = \lambda m$ (with the tube circumference $l = 2\pi(r_a - h/2)$, the vacuum wavelength of the propagating light λ and the azimuthal mode number $m \in \mathbb{N}$). It is this model that prompts us to name modes with the electric field vector parallel to the tube axis TE polarized. We assume a radially averaged energy-dependent refractive index of $n(E) = 3.46 + (E[\text{eV}] - 1.1)/2$ for the tube wall. The positions of the lowest lying radial TE modes calculated within this model, using $h = 200 \text{ nm}$ and $2r_a = 5.25 \mu\text{m}$, are depicted as squares in Fig. 7.2a. The exact positions of the calculated modes are afflicted with some uncertainty because they are very sensitive to the assumed radius. Therefore, mode spacing is the important quantity to compare to the experiment. This comparison exhibits striking accordance. In the second, the so-called exact approach, we solve Maxwell's equations for a dielectric disk with a

hole in its center. The dots in Fig. 7.2a represent the results obtained from this solution. The deviation to our first approach is very small. Especially, the mode spacings fit perfectly. This shows that the first approach, which is easier to calculate, delivers sufficiently accurate results. Until now, the resonator has only been regarded as a two-dimensional ring. However, as we will show below, the waveguide approach will prove to be very useful even in a three-dimensional description of confined modes.

Besides the sharp resonances identified as constructively interfering lowest-order TE modes of the waveguiding tube wall, we observe broader signals on the high-energy side of every TE mode, sometimes exhibiting a fine structure. These signals are regular with the TE modes and therefore cannot be attributed to higher order TE modes. Furthermore, we do not observe any distinct modes in TM polarization. The absence of both TM modes and higher order TE modes can be explained with their weaker confinement inside the tube wall and, consequentially, with their higher susceptibility to imperfections of the waveguide surface, especially its discontinuities induced by the rolling edges.

Figure 7.2b shows spatially resolved TE spectra obtained by scanning the sample in steps smaller than $1\text{ }\mu\text{m}$ in direction of the microtube axis underneath the fixed excitation-laser spot. Here, $z = 0$ corresponds to the laser spot being somewhere in the middle of the self-supporting tube. The PL intensity is encoded in a gray scale, where dark means high intensity. In the displayed energy range, each spectrum exhibits two groups of peaks representing two optical modes with neighboring azimuthal mode numbers m . Over a distance of about $20\text{ }\mu\text{m}$ along the microtube, the mode positions shift less than 2 meV . A variation of the microtube radius of just 0.3% would lead to a larger shift. This impressively demonstrates the homogeneity of the microtube and of its underlying self-rolling mechanism. Figure 7.2c shows the spectrum indicated with an arrow in Fig. 7.2b at about $z = 6\text{ }\mu\text{m}$. Here, the fine structure on the high-energy side of the modes is clearly visible. If we fit the peaks by multiple Lorentzians, we receive quality factors defined by $Q = E/\Delta E$ of 2,800 and 3,200 for the modes at 1.186 eV and 1.204 eV , respectively. We now want to address the signals at the high-energy side of the modes. In a perfect homogeneous and infinite long microtube, only light traveling perpendicular to the tube axis, i.e., having no wave vector component k_z , propagates in discrete modes. A nonzero k_z leads to spiral-shaped orbits with continuous mode energy. A finite length of the microtube would allow only discrete values of k_z leading to fully discretized modes. Therefore, we interpret the strong peaks in Fig. 7.2 as modes with $k_z = 0$, whereas signals on their high-energy side represent modes with finite k_z . Following this model, from the fine structure of the broad signal, we can approximately determine the confining length L_z in z direction. For the spectrum depicted in Fig. 7.2c, this leads to $L_z = 10\text{ }\mu\text{m}$, which is much shorter than both the length of the whole tube ($120\text{ }\mu\text{m}$) and the length of its self-supporting part ($50\text{ }\mu\text{m}$, cf. Fig. 7.1b). Indeed, L_z is comparable to the length over which the peak positions are nearly constant, see Fig. 7.2b. This finding strongly suggests that light is confined also along the tube axis on a scale of about $10\text{ }\mu\text{m}$. We will show later in this chapter that this interpretation is consistent with measurements on different microtubes

in which the confinement along the axis is deliberately tailored. Nevertheless, until now, it is an open question, which mechanism exactly causes the axial confinement for the particular tube of Figs. 7.1 and 7.2. SEM pictures do not resolve any inhomogeneities like decreasing radius or deformation of the tube.

7.5 Influence of the Rolling Edges on the Emission Properties

The cross-section of a microtube differs from the ideal ringlike shape by the discontinuous rolling edges inside and outside the ring. These rolling edges surely intrinsically limit the quality factor of our microtube resonators. However, these edges also have a positive impact on the cavity mode properties concerning preferential and directional emission and axial confinement. Here, we first want to address the emission.

Figure 7.3a shows a magnified scanning-electron microscopy (SEM) picture of a part of a self-supporting microtube bridge investigated in [19]. In this case, the starting point for its fabrication was the strained layer system containing a QW described in Sect. 7.2. The tube has rolled-up slightly more than two times. For this particular microtube, the small region where the wall consists of three rolled-up strained layers was orientated on top of the tube. Using a rather high acceleration voltage of 20 kV at the SEM, not only the outside edge of the microtube wall but also the inside edge can be resolved. Since the microtube wall in the region between the inside (left) and outside (right) edges consists of three rolled-up layers, it appears slightly brighter in the SEM picture than the material besides this region. We find that both edges of the microtube tend to randomly fray over some microns instead of forming straight lines. The fraying occurs predominantly along the $\langle 110 \rangle$ direction of the crystal, whereas the rolling direction of the microtube is along $\langle 100 \rangle$. This fraying happened unintentionally during the etching process in this particular sample. Interestingly, these structural inhomogeneities do not destroy the formation of optical modes. In fact, the frayed edges offered the possibility of studying the influence of the rolling edges on the mode confinement and the emission.

Figure 7.3b shows the microtube imaged on the CCD chip of our detector by using the grating of the monochromator in zeroth order of diffraction as a mirror (cf. Sect. 7.3). Here, the microtube was excited by the laser, but in the collected signal, the laser stray light was cut off by an edge filter. Therefore, Fig. 7.3b shows an undispersed PL image of the sample. We observe strong emission at the excitation position centrally on the microtube. We also observe a large corona around this position due to PL emission of the underlying GaAs substrate. The borders of the microtube become apparent by vertical shadows. Furthermore, both the inside and outside rolling edge of the tube are visible in the CCD image. Even though close to the resolution limit, the larger frays especially of the outer rolling edge can be identified in (b) by comparison with the SEM picture in (a). Interestingly, we observe a strong enhancement of PL emission near the inside edge of the microtube. Having aligned the microtube axis in the way that its image is parallel to the entrance slit of

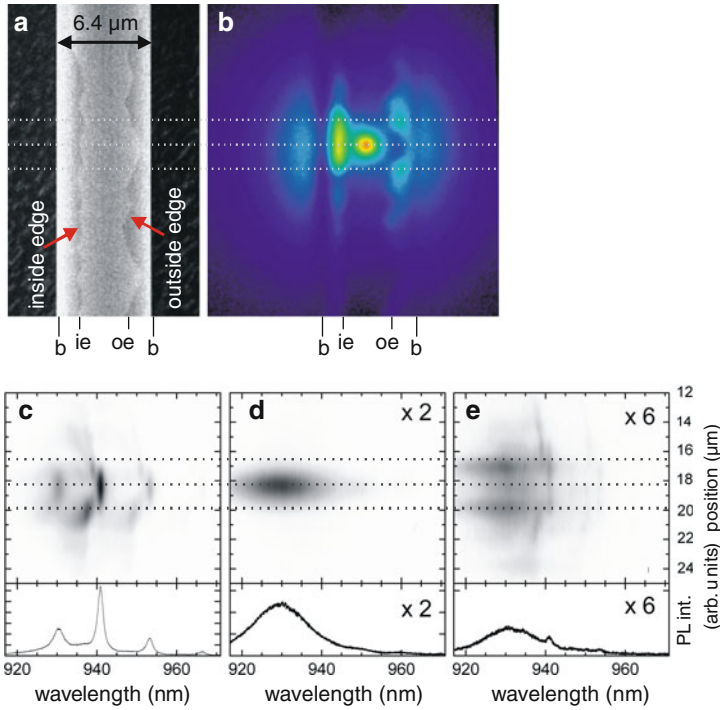


Fig. 7.3 (a) SEM picture of a part of the microtube. Both the inside and outside edges are visible. (b) Undispersed PL image of the sample shown in (a). Both images are equally scaled; the positions of the borders (b), the inside edge (ie), and the outside edge (oe) are marked. (c–e) Spectrally analyzed PL emission at the (c) inside edge, (d) radial position of the laser spot, and (e) outside edge. In the upper panels, the spatial information along the tube z axis is retained. The three *dotted horizontal lines* correlate the axial position to the SEM and PL images of (a) and (b). The lower panels show spectra obtained at the z position on the level of the laser spot (marked by the *central dotted horizontal line*). (Following [19])

the spectrometer, we can spectrally analyze the PL light of different radial positions retaining spatial resolution along the tube axis (cf. Sect. 7.2). In Fig. 7.3c, the signal along the inside edge is analyzed. The vertical axis gives the spatial position along the tube axis (z direction), the horizontal axis gives the spectral position, and the PL intensity is encoded in a gray scale. The lower panel in (c) shows a spectrum obtained at the z position on a level of the laser spot (central dotted horizontal line). The sequence of maxima of different wavelengths shows that the emitted light is indeed dominated by resonant modes. In Fig. 7.3d, the signal emitted underneath the laser spot is analyzed. In contrast to the signal at the inner edge, here, only one peak around 930 nm is observed, which is the emission of the QW into leaky modes, i.e., modes that do not fulfil the condition of total internal reflection. At last, Fig. 7.3e analyzes the emission at the outside edge. Here, we observe both sequences of resonant modes and leaky modes but with a much smaller intensity than in (c) or (d).

Note that the intensity in (e) is multiplied by a factor of 6 compared to (c). Thus, as a summary of Fig. 7.3, preferential emission of modes at the inside edge is proven. This result is valid for every position along the tube and, as many further of our investigations reveal, it seems to be a general result for microtube resonators.

The preferential emission at the inside edge of microtube resonators of course directly raises the question of possible directional emission. We addressed this point in emission angle-dependent measurements reported in [21]. We could prove the emission from the inside edge of the microtube shown in Fig. 7.3a to be strongly directional along an axis forming a 60° angle with the tangent of the tube's ringlike cross-section. This again shows the potential of functionalizing the rolling edges in terms of emission properties of the microtube resonator.

7.6 Controlled Three-Dimensional Confinement by Structured Rolling Edges

A closer look on the spatially resolved emission spectra in Fig. 7.3c already shows that a structuring of the rolling edges can lead to a three-dimensional confinement of light. Here, at the spatial position of the laser spot marked by the central dotted horizontal line, one can deduce that the modes are localized in axial direction on a length of about $2\text{--}3\text{ }\mu\text{m}$. In [19], we investigated the light confinement in the depicted microtube in very detail. Here, we just want to recapitulate our main findings. In scanning micro-PL measurements along the axis of the microtube, we observe for nearly every z position a sequence of optical modes. Their energies are shifting along the axial direction, but interestingly, this shifting is not continuous: The mode energies sometimes seem to be spatially pinned.

The shifting can be explained by expanding the waveguide model already introduced in Sect. 7.4. For each position in the z direction, a cross-section perpendicular to the tube axis is regarded as a circular waveguide with a circumference of πd . The waveguide thickness abruptly changes at the edges, thus also the effective refractive indices n_{eff} change at the edges. From the SEM image of the microtube (see Fig. 7.3a), for each z position, the part of the thicker waveguide related to the total circular waveguide (i.e., essentially the tube circumference) can be deduced. Thus, for each z , an overall effective refractive index $n_{\text{eff}}^{\text{circ}}(z)$ for the whole circular waveguide can be calculated by a weighted averaging of the two different effective refractive indices of the thicker and the thinner parts of the tube wall. The periodic boundary condition for a resonant mode, already introduced in Sect. 7.4, then reads $n_{\text{eff}}^{\text{circ}}(z)\pi d = \lambda m$, with the vacuum wavelength λ and the azimuthal mode number $m \in \mathbb{N}$. The calculations following this waveguide model nicely reproduce the overall z dependency of the measured resonant wavelengths.

The spatial pinning of resonances is a consequence of light confinement in axial direction. In [19], using the above-introduced waveguide model, we showed that such a confinement occurs predominantly in regions of local maxima of $n_{\text{eff}}^{\text{circ}}(z)$. This important result can be qualitatively explained within the waveguide model.

Until now, it was implied in the model that the light has no wave vector component along the tube axis. If one now regards light with a finite but small wave vector component along the axis, this light can experience total internal reflection also in z direction due to the change in $n_{\text{eff}}^{\text{circ}}(z)$. Total internal reflection in this case is quite similar to the situation in a graded-index optical fiber.

The experimental data reviewed above and intensively discussed in [19] show that it is possible to confine light also along the axis of a microtube ring resonator by a slight change of the wall geometry along the axis. These experiments were performed on a microtube, which exhibits unregulated frayed rolling edges. In the following, we show that one can actually tailor the three-dimensional confinement of light in microtube resonators with a very high degree of precision by deliberately structuring the rolling edges, as we have reported in [21].

Figure 7.4a sketches a microtube resonator with its typical multiwalled geometry and the two rolling edges. The most important feature of this microtube resonator is the structured rolling edge, in this case, exemplarily, exhibiting a parabolic lobe. This lobe turns the structure into a bottle resonator, as will be worked out in the following. Figure 7.4b shows a SEM image of a microtube bridge, which resulted from rolling-up an U-shaped strained mesa. In this case, InAs QDs were embedded as optically active material. The underlying MBE grown layer system is described in Sects. 7.2 and 7.4. In the center part of the tube, which is raised from the substrate, the outside edge forms a parabolic lobe, which represents a locally increased winding number. The geometrical parameters such as radius ($R = 2.6$ nm), winding

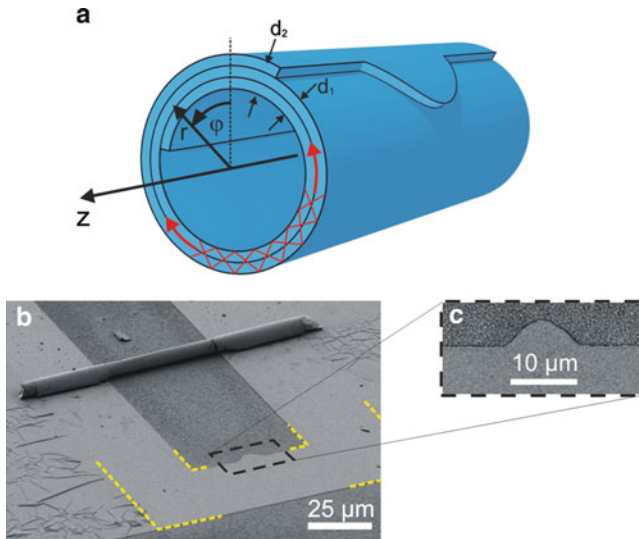


Fig. 7.4 (a) Sketch of a microtube bottle resonator exhibiting a parabolic lobe on its outside rolling edge. Red arrows illustrate the circular light propagation by multiple total internal reflections. (b) SEM image of a microtube bottle resonator. Yellow lines clarify the edges of the U-shaped mesa. (c) Magnified top view on the region marked in (b). (Following [21])

number ($N = 2.3$), wall thickness ($d_1 = 100$ nm, $d_2 = 150$ nm), the specific shape of the lobe (parabolic in Fig. 7.4c), and the distance of the center part of the tube to the substrate are precisely defined by the underlying semiconductor layers, as well as by the optical lithography and the wet etching techniques before rolling-up the U-shaped mesa.

Figure 7.5a shows a spatially integrated spectrum from the central part of the microtube depicted in Fig. 7.4b. The vertical axis gives the photon energy and the horizontal axis gives the PL intensity. The signal from the quantum dot ensemble is dominated by optical eigenmodes of the microresonator: one observes about six groups of at least seven sharp spectral eigenmodes in each group. The groups resemble each other in their spectrally resolved structure. Within one group the peaks are almost equidistant in energy. Figure 7.5b shows the spatially resolved measurement of the microtube. Here, the horizontal axis gives the spatial position along the tube axis measured relatively to the extremum of the lobe. The PL intensity is encoded in a color scale. In this depiction, it becomes obvious that the groups are overlapping in their energies and each group consists of up to 11 peaks. Most interestingly, modes within a group are localized in special regions along the tube axis: there are

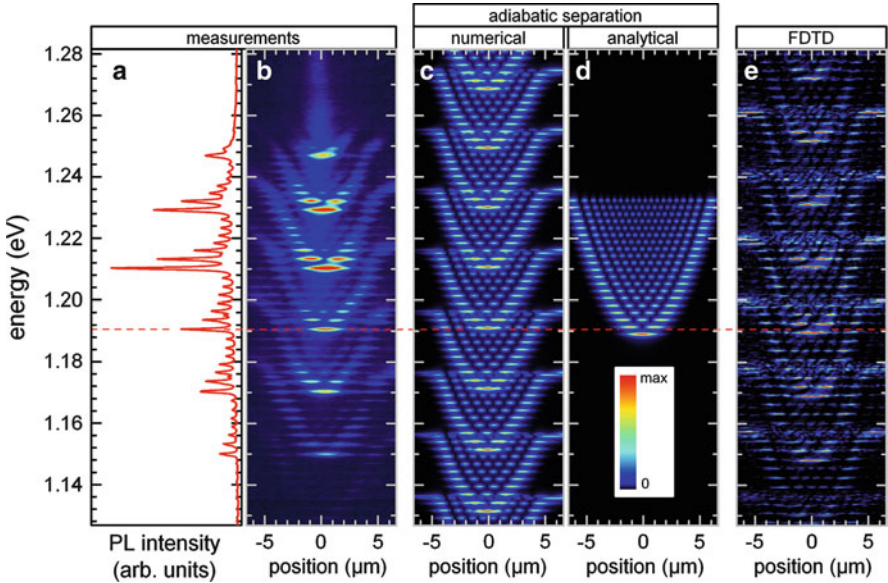


Fig. 7.5 (a) Spatially integrated and (b) spatially resolved PL spectra of the microtube bottle resonator with a parabolic lobe. (c)–(d) Axial field distributions calculated within the adiabatic approximation. (c) Numerical solution for a finite potential obtained from SEM images of the microtube. (d) Analytical solution for an infinite parabolic potential for one group of axial modes. (e) FDTD simulation of the axial field distribution. Intensities in (b)–(e) are encoded in a color scale as depicted in the inset of (d). Horizontal axes in (b)–(e) give the relative position with respect to the extremum of the lobe. The dashed horizontal line at about 1.19 eV serves as a guide for the eye. (Following [21])

nodes and antinodes in the axial intensity distribution, and their numbers increase with increasing energy. This spatial mode distribution demonstrates that the modes are confined at the lobe position and form a system of higher axial harmonics. As already suggested above, the observed sequence of the modes can be explained as follows: Neighboring groups of modes in Fig. 7.5a, b fulfil the periodic boundary condition $n_{\text{eff}}^{\text{circ}}(z)\pi d = \lambda m$ for azimuthal waveguiding but differ from each other by their azimuthal mode number m with $m = \pm 1$. The modes within each group arise from the axial confinement of light, which is induced by the lobe. Thus, one can regard the three-dimensionally confined modes as superpositions of ringlike modes in the circumference of the tube and back and forth reflected modes along the tube axis.

In a previous theoretical work on prolate-shaped dielectric resonators, similar modes have been named “bottle modes” in analogy to magnetic bottles in which charged particles can be trapped [28, 29]. We adopted this term for our microtube resonators, even though the term “empty-bottle modes” would be more precise, since we are dealing with hollow microtubes with thin walls.

The measured axial field distributions in Fig. 7.5b remind one of the probability density of a quantum mechanical particle in a parabolic potential. In the following, we want to elucidate that the actual profile of the lobe can be translated into a photonic quasi potential, which goes into a photonic quasi-Schrödinger equation. The solution of this equation then yields the experimentally observed axial field distributions.

As a first approximation, we assume that we are dealing with electric fields linearly polarized parallel to the tube axis (TE polarization). Indeed, this is what is observed experimentally [13, 18, 19] and in numerical finite-difference time-domain (FDTD) simulations [30].

Maxwell’s equations then result in the scalar wave equation for the z component $E_z(r, \varphi, z)$ of the electric field

$$-\frac{1}{n^2(r, \varphi, z)}\nabla^2 E_z(r, \varphi, z) = k^2 E_z(r, \varphi, z), \quad (7.1)$$

with the absolute value of the wave vector in vacuum k , and the refractive index of the medium n . The cylindrical coordinates (r, φ, z) are defined in Fig. 7.4a. We now apply the adiabatic approximation by separating the differential operator in (7.1) in a circular r – φ part and an axial z part. We also write the electric field as a product of two functions

$$E_z(r, \varphi, z) = \Phi(r, \varphi, z)\Psi(z), \quad (7.2)$$

where $\Phi(r, \varphi, z)$ is the solution of the circulating propagation for a fixed parameter z and $\Psi(z)$, the solution of the axial propagation. With this ansatz, we directly follow the procedure of the adiabatic (or Born–Oppenheimer) approximation given in quantum mechanics textbooks, e.g., in [31], except that we are dealing with a wave equation for electromagnetic fields instead of a Schrödinger equation for quantum mechanical particles. In our ansatz, for each position z along the tube axis, $\Phi(r, \varphi, z)$ has to satisfy the two-dimensional wave equation

$$-\frac{1}{n^2(r, \varphi, z)} \nabla_{r, \varphi}^2 \Phi(r, \varphi, z) = k_{\text{circ}}^2(z) \Phi(r, \varphi, z), \quad (7.3)$$

where k_{circ} is the absolute value of the wave vector component of the circular propagation in the r - φ plane. The solutions of (7.3) characterize the electromagnetic field in the r - φ plane for fixed values of the coordinate z . Then, the axial propagation is described by

$$-\frac{1}{n^2} \frac{\partial^2}{\partial z^2} \Psi(z) + k_{\text{circ}}^2(z) \Psi(z) = k^2 \Psi(z). \quad (7.4)$$

Since this equation is formally similar to the equation for particle waves, we call it photonic quasi-Schrödinger equation. In contrast to the original Schrödinger equation, in (7.4), the eigenenergies k occur squared. The quantity $k_{\text{circ}}(z)$, which also occurs squared in (7.4) and which is defined by the solutions of (7.3), acts as a quasi potential $V_{\text{eff}}(z)$ for the axial propagation. Recapitulating, within the adiabatic approximation, solutions of (7.3) for discrete parameters z define the quasi-potential $V_{\text{eff}}(z) = k_{\text{circ}}^2(z)$ that fully determines the mode structure of the microtube.

In order to solve (7.3), we use the expanded waveguide model briefly introduced in the beginning of this section. But here, we change the notation: Instead of describing the tube as one circular waveguide with an averaged refractive index, we regard the thin microtube wall as two-coupled planar waveguides with different thicknesses d_1 and d_2 , (see Fig. 7.4a) and lengths L_1 and L_2 , with $L_1 + L_2 = 2\pi R$. The ring-shaped cross-section is taken into account by assuming periodic boundary conditions, which ensures phase matching of the light after one round trip. Solving (7.3) within this approximation, we find the very important result that the circular component of the wave vector k_{circ} depends linearly on the quantity $p = L_1/(L_1 + L_2)$ with a deviation from linearity of only 0.1% when it is varied from 0 to 1. This means that the parabolic lobe of the microtube in Fig. 7.4 directly leads to a parabolic energy dependence along the tube axis for the circulating propagation. By measuring the exact geometry of the lobe in SEM images, we are able to determine p and to calculate the quasi potential $V_{\text{eff}}(z) = k_{\text{circ}}^2(z)$. We then solve (7.4) by a spatial discretization followed by the diagonalization of the resulting algebraic equations. With this procedure, arbitrarily shaped quasi potentials can be modeled. Furthermore, also the dispersion of the refractive index of the material can be taken into account.

The numerically calculated mode energies and axial field distributions for the microtube shown in Fig. 7.4 are depicted in Fig. 7.5c. The squared absolute values of the axial eigenfunctions $\Psi(z)$ are encoded in a color scale, spatially resolved in horizontal direction, and energy resolved in vertical direction. The energy width is assumed to lead to quality factors of $Q = E/\Delta E = 2,000$. For the calculations, we took into account the exact geometry of the tube measured from SEM images, including slight asymmetries in the lobe. We observe a very nice agreement with the experimental data in Fig. 7.5b for both the spatial intensity distribution and the mode energies. The calculations yield groups of eigenmodes, which are almost equidistant in energy and exhibit the axial intensity distribution of the measurements. Each group belongs to a discrete solution of (7.3) for the circular field distribution with

the eigenvalue $k_{\text{circ}}^{(m)}(z)$, where m gives the azimuthal mode number. Within a group, the eigenmodes are discrete solutions of (7.4), for which a new axial mode number $h = 0, 1, 2, \dots$ can be introduced. Interestingly, also the depth of the quasi potential in which bound solutions exist is reproduced very well. This directly reflects the validity of the description by the waveguide model. There is a small deviation in the absolute energy between the measured and calculated spectrum, which results from slight uncertainties in the radius of the microtube and in the dispersion of the refractive index of the material.

One can estimate the validity of the adiabatic approximation following quantum mechanics textbooks like [31]. In our approach, we separated the circular propagation in the r - φ plane with the wave vector k_{circ} from the axial propagation with the wave vector $k_z = \sqrt{k^2 - k_{\text{circ}}^2}$. One finds that this adiabatic separation is a good approximation when the sufficient condition $\Delta k_z^2 \ll |(k_{\text{circ}}^{(m)})^2 - (k_{\text{circ}}^{(m+1)})^2|$ is satisfied [31], with $k_z = \sqrt{k^2 - k_{\text{circ}}^2}$ and $\Delta k_z^2 = |(k_z^{(h)})^2 - (k_z^{(h+1)})^2|$. Note that in [31], this estimate is made for the Schrödinger equation with energies instead of squared wave vectors. From experimental data, we estimate $\Delta k_z^2 / |(k_{\text{circ}}^{(m)})^2 - (k_{\text{circ}}^{(m+1)})^2| \approx 10^{-4}$, which justifies our assumption.

We also performed calculations, using the commercial software “Lumerical FDTD Solutions 5.1.” These three-dimensional FDTD calculations take into account the details of the real structure including not only the rolling edges but also the curvature, which we neglected in our model so far. Figure 7.5e shows the resulting spatially and spectrally resolved field intensities inside the microtube wall along the tube axis. These calculations agree very well with both the experiment and the above-described model and demonstrate that our approximations are warrantable. Note that the computing time for the FDTD calculations is more than two magnitudes larger than for the model using the adiabatic separation.

As elaborated above, a parabolic lobe of a microtube leads to a parabolic quasi potential, which appears squared in the quasi-Schrödinger equation. Naturally, the question arises, why the *square* of a parabolic potential should lead to an equidistant spacing of the eigenenergies of the axial modes. The potential in the case of a parabolic lobe can generally be expressed as $V_{\text{eff}}(z) = k_{\text{circ}}(z) = az^2 + b$, with the curvature of the lobe a and the wave-vector offset $b = k_{\text{circ}}^{(m)}(0)$ of the m th azimuthal mode. For microtubes with a parabolic lobe, typical values of the curvature and the wave-vector offset are $a \approx 5 \cdot 10^{15} \text{m}^{-3}$ and $b \approx 6 \cdot 10^6 \text{m}^{-1}$. Thus, neglecting the forth order term in $V_{\text{eff}}^2(z) = k_{\text{circ}}^2(z) = a^2 z^4 + 2abz^2 + b^2$ does not change $V_{\text{eff}}^2(z)$ with an accuracy better than 99.9% for $|z| < 6 \mu\text{m}$. The quasi-Schrodinger equation then reads

$$-\frac{1}{n^2} \frac{\partial^2}{\partial z^2} \Psi(z) + 2abz^2 \Psi(z) = (k^2 - b^2) \Psi(z). \quad (7.5)$$

The analytical solution yields

$$k^2 - b^2 = \left(h + \frac{1}{2}\right) \sqrt{8ab/n}, \quad (7.6)$$

with the axial mode number $h = 0, 1, 2, \dots$. Since the right-hand side of the equation is small compared to b^2 , a Taylor series yields in first approximation

$$k \approx b + \left(h + \frac{1}{2}\right) \sqrt{2a/b/n} \quad (7.7)$$

with an accuracy of about 99.9% for the first 20 modes. This consideration indeed demonstrates that a squared parabolic potential leads to approximately equidistant axial modes for microtubes with parabolic lobes. Figure 7.5d depicts the first 20 analytically obtained solutions of an infinite parabolic quasi potential. It is seen that at least the low-lying axial modes correspond to our measurements and to the numerically obtained solutions for the finite potential induced by the lobe.

Additionally to the above-described experiments on microtubes with parabolically shaped lobes, we also prepared and investigated microtubes with triangular and rectangular lobes. Consequently, differently shaped lobes should lead to different potentials in the quasi-Schrödinger equation 7.4 and thus to different eigenmodes and -energies. Figure 7.6a, b show PL spectra corresponding to the triangular and rectangular lobe, respectively. Again, one observes groups of axial modes, but, unlike to the parabolic lobe (see Fig. 7.5a), the axial mode spacings decrease with higher energies for the triangular lobe and increase for the rectangular one. This behavior is even more explicit in Fig. 7.6a–c, which depict the measured axial mode dispersions (red circles) for parabolic, triangular, and rectangular lobes, respectively. The modes' spacings are in direct accordance to the electronic counterparts of differently shaped potentials discussed in textbooks on quantum mechanics. The insets in Fig. 7.6a, b illustrate the different level spacings for a triangular and a rectangular potential. We also calculated the axial mode dispersions using the adiabatic

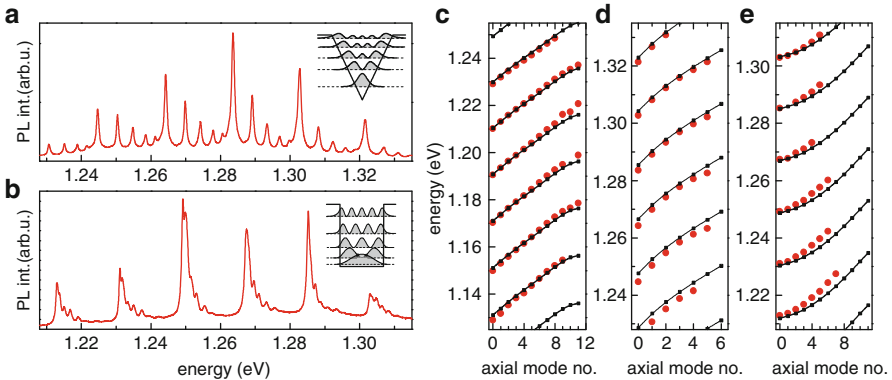


Fig. 7.6 (a,b) PL spectra of microtube bottle resonators with a (a) triangular and (b) rectangular lobe. The insets sketch the level spacings in a (a) triangular and (b) rectangular potential. (c)–(e) Axial mode dispersions for a (c) parabolic, (d) triangular, and (e) rectangular lobe. Measured (calculated) values are depicted as red circles (black squares). Connecting lines serve as guides for the eye. (Following [21])

separation described above, taking into account the exact geometry of the tubes from SEM images. Results are depicted as black squares in Fig. 7.6c–e. One can see the overall agreement of experiment and theory. These three types of lobes stand exemplarily for a manifoldness of lobes and mode dispersions: It is possible to tailor the mode dispersion of the axial modes precisely just by a predefined modulation of the rolling edge.

We emphasize that our above-described method is a reliable and robust method to tailor the three-dimensional light confinement as also proved in experiments by other researchers which perfectly reproduce our results [27].

7.7 Conclusion and Outlook

We have reviewed selected experiments concerning the light confinement in InGaAs-based microtube resonators. At the first glance, one can regard such microtubes as two-dimensional ring resonators. Indeed, the ringlike shape of the cross-section of the microtube most strongly influences the optical properties. It allows for azimuthal wave guiding and constructive interference of light, leading to resonant modes, which can be characterized by an azimuthal mode number m . These ringlike modes are most prominent in optical spectra of microtube resonators. However, already in the first measurements proving these ring modes, signatures of a confinement of light also along the tube axis has been observed [13]. Later it has been shown that the two rolling edges of multiwalled microtubes have a strong impact on the confined optical modes. Structured rolling edges can lead to an axial confinement of light [19]. The inside edge is proven to induce an axial line of preferential [19] and directional [22] emission of resonant light. Motivated by the experiments on microtubes with uncontrolled structured rolling edges, it was shown that the three-dimensional confinement of light can in fact be precisely tailored by a controlled preparation of specially shaped lobes in the rolling edges [21]. These lobes turn the microtubes into the so-called bottle resonators. The mode energies and axial field distributions can be calculated by a straight and intuitive model using an adiabatic separation of the circulating and the axial light propagation. Both experimental and theoretical results are in good agreement with FDTD simulations that take into account the exact tube geometry.

The beauty of these microtube bottle resonators and their description within the adiabatic separation is that different field patterns and mode dispersions for a desired application can be precisely tailored. One might, e.g., couple two or more lobes in order to fabricate photonic molecules or crystals. Microtube bottle resonators can act as two port devices such as optical filters: using the evanescent fields in the classical axial turning points one can couple light in and out by two wave guides. The full control of the optical properties of these microcavities is of course invaluable when one wants to utilize them, e.g., in micro lasers or as optical elements in lab-on-chip devices.

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Chapter 8

Scanning Tunneling Spectroscopy of Semiconductor Quantum Dots and Nanocrystals

Giuseppe Maruccio and Roland Wiesendanger

Abstract Quantum dots (QDs) and nanocrystals (NCs) have attracted great attention for applications in nano- and opto-electronics, quantum computation, biosensing, and nanomedicine. Three-dimensional electronic confinement can be achieved based on lateral or vertical QDs in a two-dimensional electron gas, by strain-induced QDs, or by colloidal NCs.

In this chapter, we will focus on tunneling spectroscopy on semiconductor QDs and NCs. First, in Sect. 8.2, we will provide a brief introduction on the electronic structure and single-particle wavefunctions of QDs and NCs. Section 8.3 will be dedicated to the fundamentals of electron transport through QDs and NCs: tunneling spectroscopy, Coulomb blockade, shell-tunneling, and shell-filling spectroscopy. In Sects. 8.4 and 8.5, we will report on the status of research in scanning tunneling microscopy and spectroscopy applied to semiconductor QDs and NCs. Key results and recent research directions on wavefunction mapping of individual electronic confinement states will be highlighted. Finally, we will draw conclusions in Sect. 8.6.

8.1 Introduction

Quantum dots (QDs) and nanocrystals (NCs) have attracted great attention in the last years as an exceptional class of materials in which three-dimensional electronic confinement leads to novel phenomena and enables new applications, from nano- and opto-electronics to quantum computation, biosensing, and nanomedicine [1–4]. They are commonly called artificial atoms as they share many similar features related to the electronic properties of atoms, an analogy further reinforced by the theoretical prediction of atomic-like symmetries for the electron wavefunctions. These peculiar electronic properties can be investigated with a number of different techniques such as optical spectroscopies, electrochemical techniques, capacitance measurements, magnetotunneling experiments and tunneling spectroscopies.

Moreover, QDs can be prepared by different approaches using a variety of materials.

A first, general approach consists in defining *lateral QDs* in a two-dimensional electron gas (2DEG) at the interface region of a semiconductor heterostructure. In this case, metal surface gate electrodes are appositely fabricated and employed to apply an electrostatic potential, which further confines the electrons to a small region (dot) in the interface plane. Since the electron phase is preserved over distances that are large compared with the size of the system, new phenomena based on quantum coherence appear. As a result, quantum interference devices can be fabricated, and applications in quantum computation (e.g. using spin states as qubits [5]) are envisioned (for a recent review see [6]). Alternatively, still starting from a 2DEG, *vertical QDs* can be defined by etching techniques in a related approach [7].

A second, largely-studied class concerns *strain-induced QDs* [2, 8], widely employed, for example, in optoelectronic applications. In this case, epitaxial techniques (such as molecular beam epitaxy (MBE)) are used to grow the QDs. The In-GaAs/GaAs material system is the most widely investigated due to the possibility of achieving emission at wavelengths of interest for telecommunication [9–14] (e.g. in QD lasers).

An inexpensive chemical route to produce semiconductor NCs is provided by *colloidal synthesis* [15]. Here, the electronic structure of NCs can be finely tailored by tuning size, shape, and composition [15]. For instance, size-dependent spectroscopies evidence higher energy as the QD size decreases, as expected because of quantum confinement. However, the confined states and their energies are also influenced by the environment, as we will discuss later.

Beyond these classes, QDs have also been formed inside semiconducting nanowires, carbon nanotubes or heterostructures [16–20], or were represented by single molecules trapped between electrodes [21]. Moreover metal [22], superconducting [23, 24], or ferromagnetic nanoparticles [25] were also investigated.

In this chapter, we will focus on tunneling spectroscopy on semiconductor QDs and NCs [26]. Firstly, in Sect. 8.2, we will provide a brief introduction on electronic structure and single-particle wavefunctions of QDs and NCs. Sect. 8.3 will be dedicated to the fundamentals of electron transport through QDs and NCs: tunneling spectroscopy, Coulomb blockade, shell-tunneling and shell-filling spectroscopy. In Sects. 8.4 and 8.5, we will report (without claiming to be exhaustive) on the status of research worldwide in scanning tunneling microscopy and spectroscopy for semiconductor QDs and NCs. Key results, recent research directions, and results obtained at Hamburg University on wavefunction mapping will be highlighted. Finally, we will draw conclusions in Sect. 8.6.

8.2 Electronic Structure and Single-Particle Wavefunctions

The basic characteristic defining a QD is quantum confinement in all three spatial dimensions. After initial work on mesoscopic semiconductor structures with hundreds of electrons confined, technological progress has led to a breakthrough:

the few electron regimes, where analogies with atoms are reinforced by the evidence of a shell structure, making such artificial atoms an ideal model system for quantum mechanics and many-body theories [3]. In this respect, a key technique to investigate the electronic properties of QDs is conductance spectroscopy, which provides spectra defined by the interplay between Coulomb energies and the discrete spectrum of confined states. However, before discussing such results, it is worth briefly addressing the main parameters affecting the energy spectrum and the wavefunctions of confined carriers, as this will help in successively understanding experimental observations.

QD geometry is the first, determinant factor. However, as pointed out by Grundmann et al. [27], and Stier et al. [28] the (linear) piezoelectric effect has also to be considered. Moreover, Bester and Zunger [29] showed as, when modeling strain-induced QDs and NCs, that atomistic symmetry, atomic relaxation and piezoelectric effects have to be taken into account to appropriately calculate the electronic states and reproduce experimental observations such as non-degenerate p- and d-states and optical polarization anisotropy [see [29] and reference therein]. In fact, by simply assuming a naive shape symmetry for the confinement potential (i.e. cylindrical symmetry $C_{\infty v}$, or a C_{4v} symmetry in the case of squared-based pyramid QDs), continuous models based on the effective mass approximation or $\mathbf{k} \cdot \mathbf{p}$ methods are not able to reproduce these features, which can be explained only by postulating an irregular shape for the QDs or externally adding a piezoelectric potential to the Hamiltonian. On the other hand, splitting of p- and d-states and polarization anisotropy naturally emerge if the true (and lower) C_{2v} atomistic symmetry of the QD is considered, and the total potential is better described by adding contributions from (a) the short-range interfacial potential due to interfacial atomic symmetry lowering, (b) the displacement field resulting from atomic relaxation, and (c) the long-range piezoelectric field that originates in response to the displacement field [29]. Recently, a detailed work by Bester and Zunger provided deep insight into the effect of these terms on the energy spectrum and the single-particle wavefunctions by progressively adding them individually through four different levels of theory [29] to isolate and quantify their distinct contributions. Specifically, they considered QDs with different shapes and sizes (disk, truncated cone, lens, and pyramid). At level 1, as in classical effective mass or $\mathbf{k} \cdot \mathbf{p}$ approaches, a continuum model was used assuming shape symmetry, while strain was neglected or treated by continuum elasticity, and the piezoelectric field was not included. At level 2, they considered the atomistic nature of the structure (and the resulting interfacial potential), with unrelaxed atomic positions and piezoelectricity still neglected. In this frame, $[110]$ and $[1\bar{1}0]$ directions become already inequivalent, and the symmetry group to be used is C_{2v} , for which no degeneracy is automatically expected. The interface term affects the atomistic pseudopotential, which is now different along $[110]$ and $[1\bar{1}0]$ directions in proximity of the interfaces (short range) as illustrated in Fig. 8.1a for a square-based pyramid in which the effect is the strongest. As a measure of the actual confinement anisotropy and to address the effect on the wavefunction orientation, the energetic splitting $\Delta E = \varepsilon_{[100]} - \varepsilon_{[1\bar{1}0]}$ between the single-particle electron states along the $[110]$ and $[1\bar{1}0]$ directions was evaluated. As opposed to

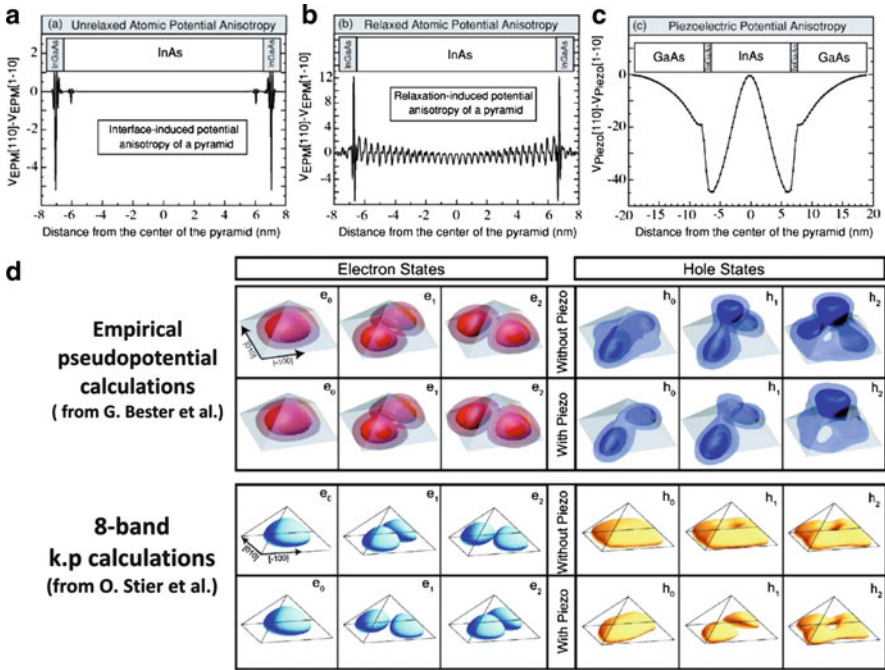


Fig. 8.1 (a–c) Evolution of the pseudopotential difference along the $[110]$ and $[1\bar{1}0]$ directions for a square-based pyramid (11.3 nm base and 5.6 nm height) at increasing levels of accuracy: (a) atomistic pseudopotential with unrelaxed atomic positions, (b) relaxed case, (c) Difference between the piezoelectric potential along the $[110]$ and $[1\bar{1}0]$ directions in the relaxed case, (d) Comparison of the first three single-particle electron and hole squared WFs as calculated for a pyramidal InAs QD by means of empirical pseudopotential (*top*) and 8-band $k \cdot p$ calculations (*bottom*). [Reprinted with permission from (a–d, *top*) G. Bester et al., Phys. Rev. B 71, 045318 (2005); (d, *bottom*) O. Stier et al., Phys. Rev. B, 59, 5688–5701 (1999). Copyright 2005 by the American Physical Society]

level 1, the splitting is not zero now, and the first electron p-state is predicted to have wavefunction aligned along the $[1\bar{1}0]$ direction. Proceeding further, at level 3, atoms are allowed to relax the stress due to lattice mismatch and, as a result, the difference in the atomistic pseudopotential is no longer confined at the interface, but propagates inside the nanostructure (Fig. 8.1b). Within this frame, the general trend reflects the interface contribution, but its magnitude is larger as the anisotropy spreads over a region where the confined states have the largest amplitude. Bester and also Zunger calculated the single-particle squared WFs, which are reported in Fig. 8.1d for the case of a pyramidal QD. Finally, at level 4, they also considered the piezoelectric potential which arises from strain along the $[111]$ direction and whose magnitude depends on the piezoelectric constant e_{14} . For this term also, there is a difference along $[110]$ and $[1\bar{1}0]$ directions, as illustrated in Fig. 8.1c, which now favors an orientation of the electron states along $[110]$. The consequence is, thus, a reduction of the energy splitting induced by the first two terms (interface and

stress relaxation effects), but the wavefunction associated with the lower energy electron p-state remains oriented along $[1\bar{1}0]$ for a pyramidal QD (Fig. 8.1d). As far as a comparison with other reports is concerned, without piezoelectricity, these results agree with previous calculations based on the empirical pseudopotential method [30, 31] in wavefunction orientation and p-level splitting within 10%, while with piezoelectricity, the wavefunction orientation is different with respect to $\mathbf{k} \cdot \mathbf{p}$ results, which miss the atomistic splitting. A first p-state oriented along $[1\bar{1}0]$ is in agreement with the experimental results [32–34]. More recently, the importance of considering quadratic (second order) piezoelectric effects [35, 36] was theoretically demonstrated. They were found to be opposed to the first order term and could lead to a mutual cancellation depending on QD geometry and composition profile [37]. Later we will see how electron-electron interactions and correlation effects have to be considered to correctly describe QDs with more (interacting) electrons [38, 39], as experimentally observed in light scattering [40], capacitance spectroscopy and high source-drain voltage spectroscopies [41].

8.3 Electron Transport Through Quantum Dots and Nanocrystals

8.3.1 Tunneling Spectroscopy

A powerful technique to probe the electronic properties of QDs and NCs is tunneling spectroscopy, in which the dot is coupled to and exchanges electrons via tunnel barriers with reservoirs (Fig. 8.2). The Bardeen formalism can be employed [42] to the first order to describe tunneling between two leads (in our case, the tip and the sample) by solving the time-dependent Schrödinger equation using a perturbation approach. The first step consists of calculating the single-particle wavefunctions for the tip ψ_μ^t and the sample ψ_ν^s , considered as separated and independent entities and having eigenvalues E_μ^t and E_ν^s , respectively. Then, for an elastic tunneling, the current can be calculated on the basis of Fermi's “golden rule” and expressed by means of the tunneling matrix element $M_{\mu\nu}$, which connects the unperturbed tip states ψ_μ^t to sample states ψ_ν^s :

$$I = \frac{2\pi e}{\hbar} \sum_{\mu,\nu} \{f(E_\mu^t)[1 - f(E_\nu^s + eV)] - f(E_\nu^s + eV)[1 - f(E_\mu^t)]\} \\ \times |M_{\mu\nu}|^2 \delta(E_\nu^s - E_\mu^t)$$

where the delta function guarantees energy conservation (elastic tunneling), while $f(E)$ and V are the Fermi function and the applied sample-voltage, respectively. In other words, this equation states that the tunneling current is proportional to the square of the matrix element connecting the various initial/final states times the

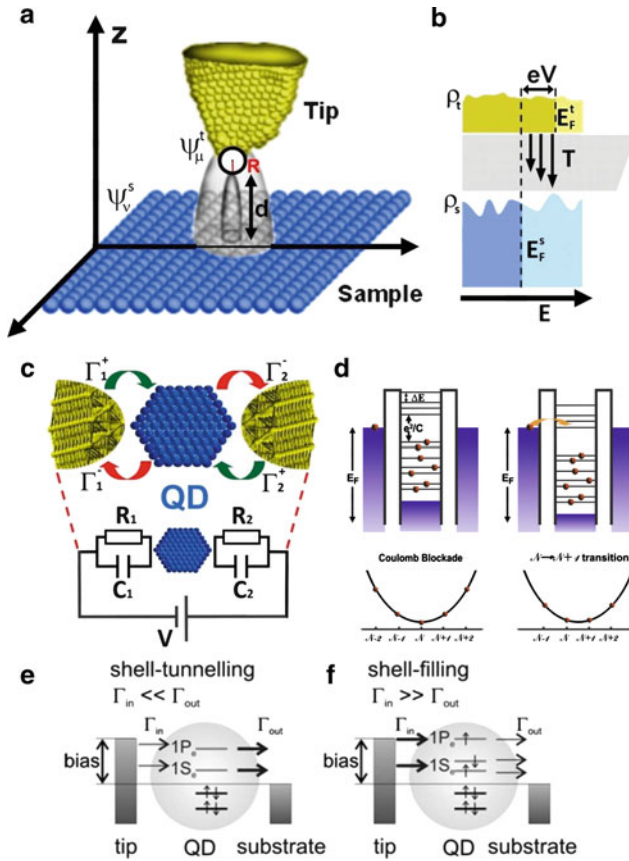


Fig. 8.2 (a–b) Scanning tunneling spectroscopy and corresponding energy diagram. For positive sample bias voltage, unoccupied (electronic) states of the samples are probed. (c) double tunnel barrier junction and corresponding electrical scheme. (d) Coulomb blockade and dependence of electrostatic energy on the quantized charge on the island. [Reprinted with permission from Y. Alhassid, *Review of Modern Physics* 72, 895 (2000). Copyright 2000 by the American Physical Society] (e–f) Sketches of shell tunneling and shell filling regimes. [P. Liljeroth et al., *Phys. Chem. Chem. Phys.* 8, 3845–3850 (2006), Reproduced by permission of the PCCP Owner Societies]

probability of finding an occupied state on one side and an empty state on the other. However, the term corresponding to reverse tunneling needs to be considered only at high temperatures. Within the limits of low temperature and small voltage, it is possible to write:

$$I = \frac{2\pi}{\hbar} e^2 V \sum_{\mu, \nu} |M_{\mu\nu}|^2 \delta(E_\nu^s - E_F) \delta(E_\mu^t - E_F)$$

Here, the main difficulty is the evaluation of $M_{\mu\nu}$, which is the term hiding the dependencies on the tunnel barrier height and width, and the orbital character of the

tunneling electrons. According to Bardeen:

$$M_{\mu\nu} = \frac{\hbar^2}{2m} \int dS \cdot \left(\psi_{\mu}^{t*} \nabla \psi_{\nu}^s - \psi_{\nu}^s \nabla \psi_{\mu}^{t*} \right)$$

which is the integral of the current operator over any surface lying entirely within the vacuum (barrier) region separating the two leads [43].

This expression was further simplified by Tersoff and Hamman [43]. As a first, rough approximation, a point probe (arbitrarily small) can be considered and the result is a matrix element proportional to the modulus squared of the sample wavefunctions ψ_{ν}^s at the tip position. A more precise description is, however, obtained by assuming s-like states for a tip with a spherical shape (centered at $\mathbf{r}_0$ and having radius R). By expanding the tip and sample wavefunctions, and evaluating the matrix elements one finds:

$$I \propto V \rho_t(E_F) e^{2kR} \sum_{\nu} \left| \psi_{\nu}^s(\mathbf{r}_0) \right|^2 \delta(E_{\nu}^s - E_F) = V e^{2kR} \rho_t(E_F) \rho_s(\mathbf{r}_0, E_F)$$

By increasing the bias V , the region of energetically overlapping occupied and unoccupied states has to be considered (Fig. 8.2b), and thus an integration over energy is required:

$$\begin{aligned} I &\propto \int_0^{eV} \rho_t(E - eV) \rho_s(\mathbf{r}_0, E) dE \\ &\propto \int_0^{eV} \rho_t(E - eV) \rho_s(x, y, E) T(E, eV, z = d + R) dE \end{aligned}$$

where $\rho_{t,s}$ are the densities of states (DOS) of the tip and the sample. In the last step, the transmission coefficient T was introduced to relate the sample DOS at the tip position $\mathbf{r}_0$ to the sample local density of states (LDOS) at $z = 0$, i.e. on the sample surface. The last one is the most interesting quantity and can be evaluated from the differential tunneling conductivity:

$$\begin{aligned} \frac{dI}{dV}(x, y, V) &\propto e \rho_t(0) \rho_s(x, y, E = eV) T(E = eV, eV, z) \\ &+ \int_0^{eV} \rho_t(E - eV) \rho_s(x, y, E) \frac{dT(E, eV, z)}{dV} dE \\ &+ \int_0^{eV} \frac{d\rho_t(E - eV)}{dV} \rho_s(x, y, E) T(E, eV, z) dE \end{aligned}$$

Among these terms, the first one dominates at low bias while the others account for less than 10% [44]. Thus, the energy-resolved LDOS can be spatially mapped by acquiring bias-dependent and spatially-resolved dI/dV images using a lock-in technique. In the case of a system described by discrete states $\psi_{\nu}^s(E_{\nu}, x, y)$, the LDOS is given by $\sum_{\delta E} |\psi_{\nu}^s(E_{\nu}^s, x, y)|^2$. As a consequence, if the energy resolution δE is

less than the energy level spacing, the LDOS reduces to a single term and spatially-resolved dI/dV maps display the detailed spatial structure of $|\psi_v^s(E_v^s, x, y)|^2$ at the corresponding energy eV .

The transmission coefficient T can be evaluated by the WKB method or measured using $I(V, z) \propto T(E = eV, V, z)$. In both cases, the well-known exponential form is obtained:

$$T(E = eV, eV, z) \propto \exp\left(-A\sqrt{\hat{\phi} - e|V|/2z}\right)$$

with $A = \sqrt{8m/\hbar}$ and $\hat{\phi}$ depending on the tip [44]. To compensate for possible changes in the tip position z on the sample surface (x, y) , the normalized conductance $(dI/dV)/(I/V)$ is sometimes employed (although the division can introduce additional noise) as the ratio of conductance to current is nearly independent of tip-sample separation [44, 45].

8.3.2 Coulomb Blockade

Another important phenomenon observed in electron transfer through QDs and NCs is Coulomb blockade. While in the case of large islands the charge can be considered as a continuous variable, when dealing with small systems, it is necessary to take into account charge quantization effects ($Q = Ne$, with e = electron charge), which dominate transport in QDs. Before discussing the orthodox theory, it is worth noting that a basic understanding can be achieved by simple electrostatic considerations. In fact, if we consider a conductive metal island characterized by a capacitance C , the total classical electrostatic energy associated with N electrons in this dot (and thus stored in the junction) is:

$$E(N) = \frac{(Ne)^2}{2C} - NeV_{\text{ext}} = \frac{(Q - Q_{\text{ext}})^2}{2C} + \text{const}$$

where V_{ext} is the electrostatic potential and $Q_{\text{ext}} = CV_{\text{ext}}$, the externally induced charge. If the island is small, this energy is significant and can be higher than the thermal energy $k_B T$. In this size range, the charge can no longer be considered as a continuous variable, but needs to be expressed in units of electronic charge (Fig. 8.2d). Thus, the charge stored in the island changes discontinuously and at low bias, the energy change associated with the tunneling of a single electron can be energetically unfavorable and no current will flow through the junction. This is the Coulomb blockade (CB) region, and the number of electrons in the dot for a given V_{ext} can be determined by minimizing $E(N)$ and is the integer nearest Q_{ext}/e . The voltage at which the charge on the dot can increase by one electron corresponds to situations in which $E(N + 1) = E(N)$. This condition occurs periodically (Coulomb oscillations) for $Q_{\text{ext}} = (N + 1/2)e$ and results in peaks in the

dI/dV spectrum (maximal conductance) with a spacing between adjacent maxima equal to e/C . The lower C is, the bigger ΔE and the corresponding temperature to observe CB effects. Increasing the temperature above $k_B T \approx E_c = e^2/2C$ (charging energy), the “Coulomb staircase” is gradually smeared out by thermal fluctuations.

In the presence of a discrete level spectrum, the previous equation becomes:

$$E(N) = \frac{(Ne)^2}{2C} - NeV_{\text{ext}} + \sum_{i=1}^N \varepsilon_i(B)$$

where the sum in the last term is over the occupied single-particle states $\varepsilon_i(B)$, which are the solutions of the single-particle Schrödinger equation and the only terms depending on the magnetic field [7]. The addition energy of the dot is thus:

$$E_{\text{ADD}}(N) = E_c + \Delta E(N),$$

where $E_c = e^2/2C$ is the electrostatic charging energy to add an electron to the dot, while $\Delta E(N)$ is the difference in the single-particle energies for N and $N - 1$ electrons on the dot [7]. In other words, the separations among the different peaks are determined by the single-electron charging energies (addition spectrum) and the spacing between the discrete levels (excitation spectrum).

Finally, it is worth mentioning that, in devices, a third gate electrode, capacitively coupled to the conductive island/dot, can be introduced in addition to the two junctions system described earlier. As a consequence, the condition to have conduction can be also reached by acting on the gate voltage (V_G). In a two-dimensional plot of the dI/dV as a function of V and V_G , Coulomb diamonds are obtained where the number of electrons in the island is fixed. This is the basis to fabricate single electron transistors, which – thanks to their rapid conductance variation – are ideal devices for high-precision electrometry, as they are very sensitive to changes in gate voltage when the bias voltage is close to the Coulomb blockade value. For further details on Coulomb blockade, a number of reviews are available [46–52].

8.3.3 Shell-Tunneling and Shell-Filling Spectroscopy

The typical experimental configuration used in tunneling spectroscopy on QDs and NCs (Fig. 8.2c) consists of a double barrier tunnel junction (DBTJ). Here, tunnel barriers provide the decoupling necessary to investigate the inherent electronic properties of the system under analysis and to obtain direct images of the dot’s WFs disentangled from the electronic structure of the substrate. In the weak-coupling limit, the tunneling current is, in fact, determined by the resonant tunneling through the energy levels of the dot when they align with the Fermi level of the tip or

substrate. However, this process depends on the overall dynamics and the specific tunneling rates into and out of the dot, which determine the tunneling regime¹ [53].

In the frame of the orthodox theory [46, 49–52], it is possible to calculate the I-V characteristics from the tunneling rates. Specifically, the current can be expressed as:

$$\begin{aligned} I &= \sum_{N=-\infty}^{+\infty} e[\Gamma_1^+(N, V) - \Gamma_1^-(N, V)]\rho(N, V) \\ &= \sum_{N=-\infty}^{+\infty} e[\Gamma_2^-(N, V) - \Gamma_2^+(N, V)]\rho(N, V) \end{aligned}$$

where Γ_i^+ and Γ_i^- are the rates of electron tunneling into and out of the QD for the i th junction, respectively, while $\rho(N, V)$ is the probability of finding N electrons in the dot. In turn, Γ_i^+ and Γ_i^- can be evaluated by Fermi's "golden rule". In the case of Γ_i^+ , this means the integration over energy of the square of the tunneling matrix element coupling the initial and final state at energy E times the number of occupied initial states and unoccupied final states [49]:

$$\begin{aligned} \Gamma_1^+ &= \frac{2\pi}{\hbar} \int_{-\infty}^{+\infty} |M_{1,QD}|^2 \rho_1(E - E_1) f(E - E_1) \rho_{QD}(E - E_{QD}) \\ &\quad \times (1 - f(E - E_{QD})) dE \end{aligned}$$

where $f(E)$ is the Fermi distribution function, $\rho_1(E)$ and $\rho_{QD}(E)$ are the density of states of the first electrode and the dot, respectively, and E_1 and E_{QD} are their Fermi energies. If the DOS and the tunneling matrix elements are assumed to be energy independent ($\rho_1(E) = \rho_{1,0}$, $\rho_{QD}(E) = \rho_{QD,0}$, $|M_{1,QD}|^2 = |M_{1,QD,0}|^2$), all the four tunneling rates can be written in a compact way [49]:

$$\Gamma_i^\pm(N, V) = \frac{1}{e^2 R_i} \left(\frac{-\Delta E_i^\pm}{1 - e^{\Delta E_i^\pm / k_B T}} \right)$$

where R_i is the tunneling resistance of the i th junction defined by the constants introduced above and $\Delta E^\pm$ is the change in energy when an electron tunnels across the barrier and can be evaluated from electrostatic considerations ($e > 0$) [50]:

$$\begin{aligned} \Delta E_1^\pm &= \Delta U^\pm \pm \frac{eC_2}{C_\Sigma} V, \\ \Delta E_2^\pm &= \Delta U^\pm \mp \frac{eC_1}{C_\Sigma} V, \end{aligned}$$

¹ The tunneling rate from the NC to the substrate is usually much larger than the typical tunneling rate from the tip to the NC, thus to switch from a regime to the other different stabilization currents I_{stab} (corresponding to different tip-dot distances) can be used. Smaller distances (i.e. higher currents keeping a fixed voltage) correspond to gradually increasing tunneling rates into the NC which at some stage starts to be charged.

where Q is the charge on the dot, C_i the capacitance of the i th junction, $C_\Sigma = C_1 + C_2$ the total capacitance, while $\Delta U^\pm$ is the change in electrostatic energy when an electron tunnels through one barrier:

$$\Delta U^\pm = \frac{(Q \pm e)^2}{2C_\Sigma} - \frac{Q^2}{2C_\Sigma},$$

The second term in $\Delta E^\pm$ is, instead, the voltage drop across the i th junction.

Finally, the probability $\rho(N, V)$ of finding N electrons can be obtained by the master equation for $\rho(N, V)$ evaluated in the steady state where $\frac{\partial \rho(N, V, t)}{\partial t} = 0$, i.e. the net probability of making a transition between two adjacent states (for instance N and $N + 1$) is zero:

$$[\Gamma_1^+(N, V) + \Gamma_2^+(N, V)]\rho(N, V) - [\Gamma_1^-(N + 1, V) + \Gamma_2^-(N + 1, V)]\rho(N + 1, V) \equiv 0$$

This equation has to be solved with the normalization condition: $\sum_{N=-\infty}^{+\infty} \rho(N, V) = 1$ and the result is:

$$\rho(N, V) = \frac{\left(\prod_{i=-\infty}^{N-1} [\Gamma_1^+(i, V) + \Gamma_2^+(i, V)] \right) \left(\prod_{i=N+1}^{+\infty} [\Gamma_1^-(i, V) + \Gamma_2^-(i, V)] \right)}{\sum_{j=-\infty}^{+\infty} \left(\prod_{i=-\infty}^{j-1} [\Gamma_1^+(i, V) + \Gamma_2^+(i, V)] \right) \left(\prod_{i=j+1}^{+\infty} [\Gamma_1^-(i, V) + \Gamma_2^-(i, V)] \right)}$$

Thus, in summary, the tunneling rates are related to the corresponding resistances R_i , while the relative capacitances of the two barriers determine the charging energy and the potential distribution in the DBTJ, which has to be known in order to extract quantitative information. On the other hand, at a fixed bias, the probability $\rho(N, V)$ and thus, the number of additional electrons on the dot depends on the ratio between the tunneling rates into and out the QD . If we neglect reverse tunneling now, we can consider only two tunneling rates, Γ_{in} and Γ_{out} . Two limiting scenarios (with many possible intermediate cases) exist [54]:

- The shell tunneling regime ($\Gamma_{\text{in}} \ll \Gamma_{\text{out}}$), where the probability of having an electron in the LUMO (hole in the HOMO) is zero and electrons (holes) tunnel one by one, without interacting. In this case, single-particle states with their degeneracy are probed.
- The shell filling regime ($\Gamma_{\text{in}} \gg \Gamma_{\text{out}}$), where carriers accumulate in the dot and Coulomb interaction among them influences tunneling, lifting orbital degeneracy and resulting in charging multiplets [55–57]. In this case, the spacing between the peaks within the multiplets can change significantly with the number of electrons in the dot and, in general, decreases with increasing total angular momentum from s- to p- and d-type states, depending on the mutual overlapping

of the orbitals and the decrease of Coulomb interaction when higher orbitals with broader wavefunctions are occupied [58].

A modification of the orthodox model was also reported to account for the discrete level spectrum of QDs [59,60].

8.4 MBE-Grown Quantum Dots

8.4.1 *Scanning Tunneling Microscopy and Cross-Sectional STM*

Scanning tunneling microscopy and spectroscopy are among the most employed methods to investigate the electronic properties of QDs and NCs. They are particularly important because, in comparison for example, to optical spectroscopic techniques, they allow the direct probing of the electronic states instead of measuring transitions among them. Moreover, STM makes it possible to gain important structural information needed to improve theoretical models describing QD formation (size, shape, and density) and growth phenomena in general (e.g. surface reconstruction, island nucleation and growth, elemental segregation) [8].

A key issue concerns the determination (and prediction) of the equilibrium shape of the QDs, which would allow the improvement of sample uniformity and device performances (electronic properties depend strongly on the geometry). Morphological characterization is possible by atomic force or scanning tunneling microscopy on uncapped QDs. In this respect, Hasegawa et al. [61] observed InAs QDs with a shape elongated in the $[1\bar{1}0]$ direction and preferred (113), (114) faceted planes (as estimated by the inclination angle, 25.2° and 19.5° , respectively, from the (001) substrate). These facets are more stable, having smaller surface energy. They also evidenced steps on the wetting layer as preferential nucleation sites and an increase of the island size with the deposited InAs ML. In a different in situ STM study, Marquez et al. [62] were able to obtain atomically-resolved images of InAs QDs and identify dominating facets with high Miller indices, thanks to the examination of the atomic features within the facets (Fig. 8.3). Specifically, reflection high energy electron diffraction (RHEED), used for initial characterization, evidenced chevron-like spots when the electron beam was directed along the $[1\bar{1}0]$, attributed to facets having a 20° – 25° inclination angle with the substrate, while no specific facet orientation was observed along $[110]$. This observation was explained by structural data from STM because a clear pyramidal shape was observed with four pronounced facets. Electrons directed along $[1\bar{1}0]$ are diffracted from the $\{137\}$ facets, while a beam traveling along $[110]$ probes the rounded QD profile along $[1\bar{1}0]$. However, these high-index facets are thermodynamically stable only up to a certain island volume [62]. Moreover, if applications are targeted, QDs should normally be embedded in a matrix material and QD shape and composition can change during capping/overgrowth, while the different confining barriers can also influence electronic properties.

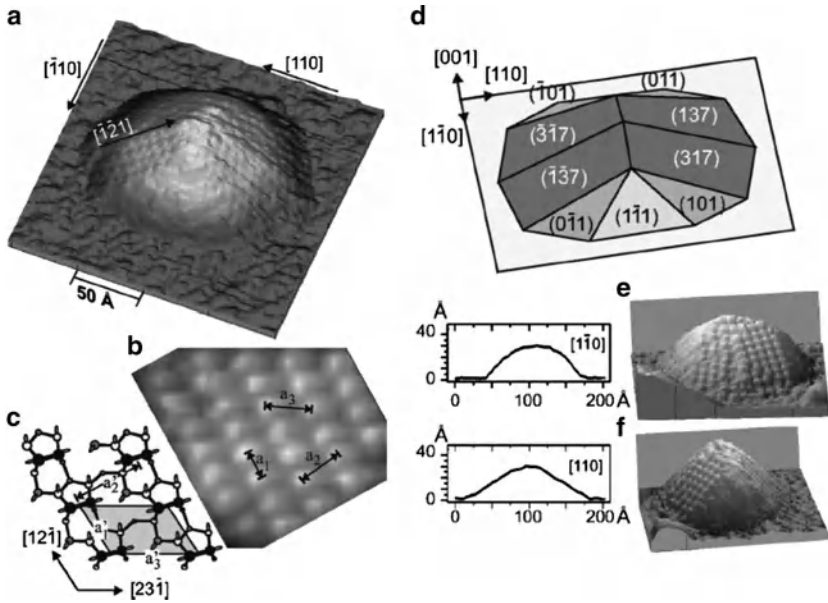


Fig. 8.3 (a–b) Atomic resolution STM image of an uncapped InAs QD and one of its {137} facets, whose structure is shown in (c), where As atoms are gray, and In atoms are white circles. (d) Schematics of the three dimensional structure of the quantum dot (QD). (e–f) Anisotropy of QD shape and different line profiles along [110] and $[1\bar{1}0]$ directions. [Reprinted with permission from J. Marquez et al., *Appl. Phys. Lett.* 78, 2309–2311 (2001). Copyright 2001, American Institute of Physics]

For this reason, capped/buried QDs were also intensively investigated by STM [63] and cross-sectional STM [64–68], where the latter technique (X-STM) works on cleavage edges and requires atomically smooth cleavage planes. The main factors determining image contrast for X-STM on QDs are (a) strain relaxation which induces accessible topographic protrusions and (b) electronic effect. As a consequence, due to the smaller band gap and larger atomic size, In-rich regions appear brighter than Ga-rich counterparts [64]. This elemental sensitivity makes X-STM very useful, and it allowed Lita et al. [65] to study InAs segregation and Eisele et al. [66] to investigate size changes in stacked QDs. A detailed study was carried out by Liu et al. [64] who evidenced an In-rich core with an inverted-cone shape in trapezoidal QDs. Bruls et al. [67] performed similar studies to investigate the growth dynamics of stacked QDs which, for small spacing layers, are vertically aligned as the formation of a dot in the strain field of the previous one is energetically favored. A deformation of the dots through the stack was observed due to a lower GaAs growth rate above the dot.

Beyond structural information, electronic properties are also accessible by scanning tunneling spectroscopy. In particular, Grandidier et al. [68] were the first to observe standing wave patterns associated with confined states in stacked QDs by acquiring current images that correspond to maps of the integrated LDOS up to

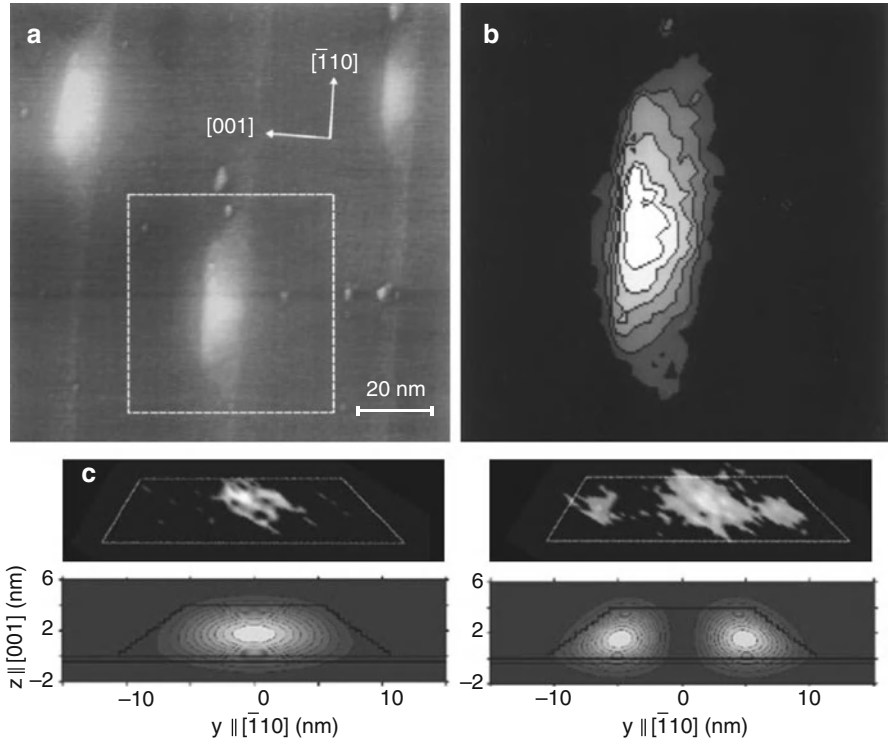


Fig. 8.4 (a) Cross-sectional STM image showing stacked QDs on a (110) cleavage surface. The brighter triangular regions in the QDs correspond to In-rich cores. (b) Contour plot of the marked QD in (a). (c) current images at +0.69 and +0.82 V showing, respectively, the ground state alone (*left*) and superimposed to the first excited states (*right*). The corresponding calculated maps are also reported for comparison. [Reprinted with permission from (a–b) N. Liu et al., Phys. Rev. Lett. 84, 334–337 (2000); (c) B. Grandier et al., Phys. Rev. Lett. 85, 1068–1071 (2000). Copyright 2000 by the American Physical Society]

the acquisition bias. As a consequence, the symmetry is that of the ground state for the image at +0.69 V, while at +0.82 V it is a superposition of the ground state and the first excited state in the dot (Fig. 8.4). More recently, dI/dV maps were acquired to image the confined states separately. The QDs were embedded in a p-type buffer layer in order to probe valence band states made empty by tip-induced band bending [69]. QD wavefunctions were also mapped indirectly in the reciprocal space by magnetotunneling spectroscopy [70–72]. Finally, it is worth mentioning the interesting work of Jacobs et al. [73] who measured electroluminescence with high spatial resolution by using an STM tip to locally inject carriers in a GaAs p-i-n sample with InGaAs QDs.

8.4.2 Wavefunction Mapping of MBE-Grown InAs Quantum Dots

The wave functions (WFs) of electrons and holes confined in the QDs are the most basic features ultimately determining all QD properties. Notably, in 2003, the possibility of mapping the dot's WF in the one-electron regime by means of spatially resolved tunneling spectroscopy images was demonstrated in Hamburg by Maltezopoulos et al. [32]. As discussed above, the differential tunneling conductivity $dI/dV(V, x, y)$ is to a good approximation proportional to the local density of independent-electron states (LDOS), and, in a system with discrete states $\psi_v^s(E_v, x, y)$, the LDOS $\left(\sum_{\delta E} |\psi_v^s(E_v, x, y)|^2\right)$ reduces to a single term $|\psi_v^s(E_v, x, y)|^2$, provided a sufficient energy resolution δE is achieved (i.e. less than the energy level spacing). Thus, voltage-resolved dI/dV images provide maps of the squared WF at the corresponding energy eV , and this technique was successfully applied to obtain spectacular images of WFs of isolated nano-objects such as QDs [32, 33, 68], carbon nanotubes [17, 74, 75], and even single molecules [76, 77] and atomic chains [78].

In Hamburg, strain-induced InAs QDs grown by MBE were investigated [32, 33]. Specifically, two series of samples (S and L in the following) were studied in more detail. They consisted of the following layers: (a) an n-doped GaAs buffer layer ($N_D \approx 2 \times 10^{18} \text{ cm}^{-3}$) deposited at a temperature of about 600°C on n-doped GaAs(001) substrates; (b) an undoped tunneling barrier ($N_A < 10^{15} \text{ cm}^{-3}$) to work in the weak coupling limit and to investigate the inherent electronic properties of the QDs; and (c) the uncapped QDs. In series $S(L)$, the buffer layer was 400 nm (200 nm) thick, the tunnel barrier 15 nm (5 nm) thick, and the QDs grown at 495°C (500°C) by depositing 2.0 ML (2.1 ML) of InAs at a growth rate of 0.05 ML/s. In situ QD formation was monitored by RHEED, which evidenced a transition from a streaky to a spotty pattern (indicating the onset of three-dimensional islanding) and chevron-like spots associated with QD facets [62]. The base pressure of the MBE and STM chambers was below 10^{-10} mbar, and the samples were transferred among them without being exposed to air, by means of a mobile ultra-high vacuum transfer system at $p < 10^{-9}$ mbar to avoid contamination.

The sample structure and experimental setup are sketched in Fig. 8.5, along with the band profile along the direction of tunneling as estimated by means of a 1D-Poisson solver neglecting 3D confinement [32]. Experiments were carried out using a low-temperature STM operating at $T = 6 \text{ K}$ and having a maximum energy resolution of $\delta E = 2 \text{ meV}$ [79]. Both W and PtIr tips were employed and acquisition of STM images was performed in constant-current mode with a typical sample bias in the range of 2–4 V and tunneling currents of 20–40 pA. The $dI/dV(V, x, y)$ signal was measured using a lock-in technique by superimposing a modulation voltage V_{mod} with frequency around 1 kHz and an amplitude in the range of 5–20 mV. Finally, WF mapping was carried out by stabilizing the tip-surface distance at each point (x, y) at voltage V_{stab} and current I_{stab} , switching off the feedback and recording a dI/dV curve from V_{start} to V_{end} ($V_{\text{start}} \leq V_{\text{stab}}$) [32]. As a result, WF mapping produces a three-dimensional array of dI/dV data, which allows (a) obtaining

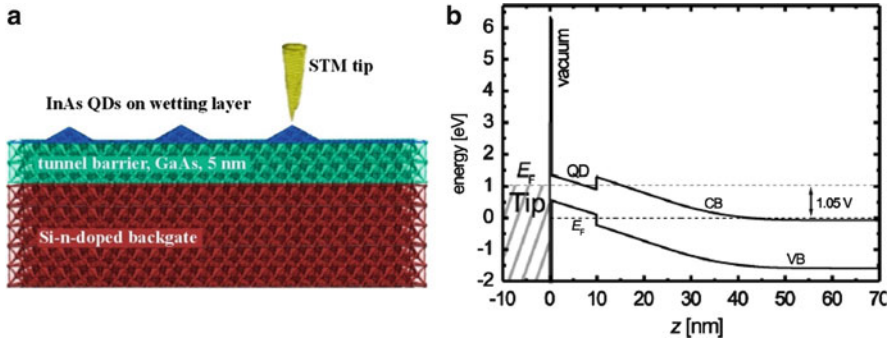


Fig. 8.5 (a) Sample structure and experimental setup. (b) band profile along the tunneling direction as calculated with a 1D-Poisson solver. [Reprinted with permission from T. Maltezopoulos et al., Phys. Rev. Lett. 91, 196804 (2003). Copyright 2003 by the American Physical Society]

spatially resolved dI/dV images at different values of V_{sample} and (b) extracting the dI/dV spectra at specific positions corresponding to specific topographic features.

A QD density of $2.5\text{--}5.0 \times 10^{10} \text{ cm}^{-2}$ was estimated from large-scale constant-current STM images (Fig. 8.6). Here, not only are QDs visible as bright spots but also steps in the wetting layer (WL) can be observed which at atomic resolution exhibit a superstructure compatible with a disordered (2×4) reconstruction (Fig. 8.6b). Most of the QDs exhibit similar sizes as shown in further detail in the small area STM image of Fig. 8.6e. As is well known, structural features and QD density depend on the growth conditions. Among the investigated samples, we observed smaller dots ($21 \pm 2 \text{ nm}$ along $[1\bar{1}0]$ and $16 \pm 2 \text{ nm}$ along $[110]$) with higher density and a larger shape asymmetry in series *S* than in series *L* where most of the dots had a pyramidal shape with well-defined facets and a fairly sharp summit, typical lateral extension of 30 nm along both $[110]$ and $[1\bar{1}0]$ directions, and an average height of $5\text{--}6 \text{ nm}$. However, they exhibited a pronounced shape anisotropy as shown in the 3D view of Fig. 8.6g. This anisotropy can be further evaluated by comparing the height profiles: triangular along $[110]$ and rounded along the $[1\bar{1}0]$, as in the report of Marquez et al. [62]. The inclination angle between the facets and the substrate is approximately 19° , in line with (114) planes.

To examine the QD electronic structure, scanning tunneling spectroscopy was performed. In particular, unoccupied states of the sample were investigated at positive sample-voltage. Samples of series *L* were probed at low stabilization currents, corresponding to a situation where electrons tunnel through an empty QD (shell-tunneling spectroscopy). In Fig. 8.7, typical I - V and dI/dV - V curves acquired above a QD and the WL are reported. While no spectral features are observed in WL spectra, steps in the tunneling current and corresponding peaks in the differential tunneling conductivity are revealed by curves on individual QDs. As far as the peak width is concerned, from a fundamental point of view, it is related to the lifetime of the electrons in the confined states, having an upper boundary in the tunneling rate through the undoped GaAs tunnel barrier, which is larger than the rate

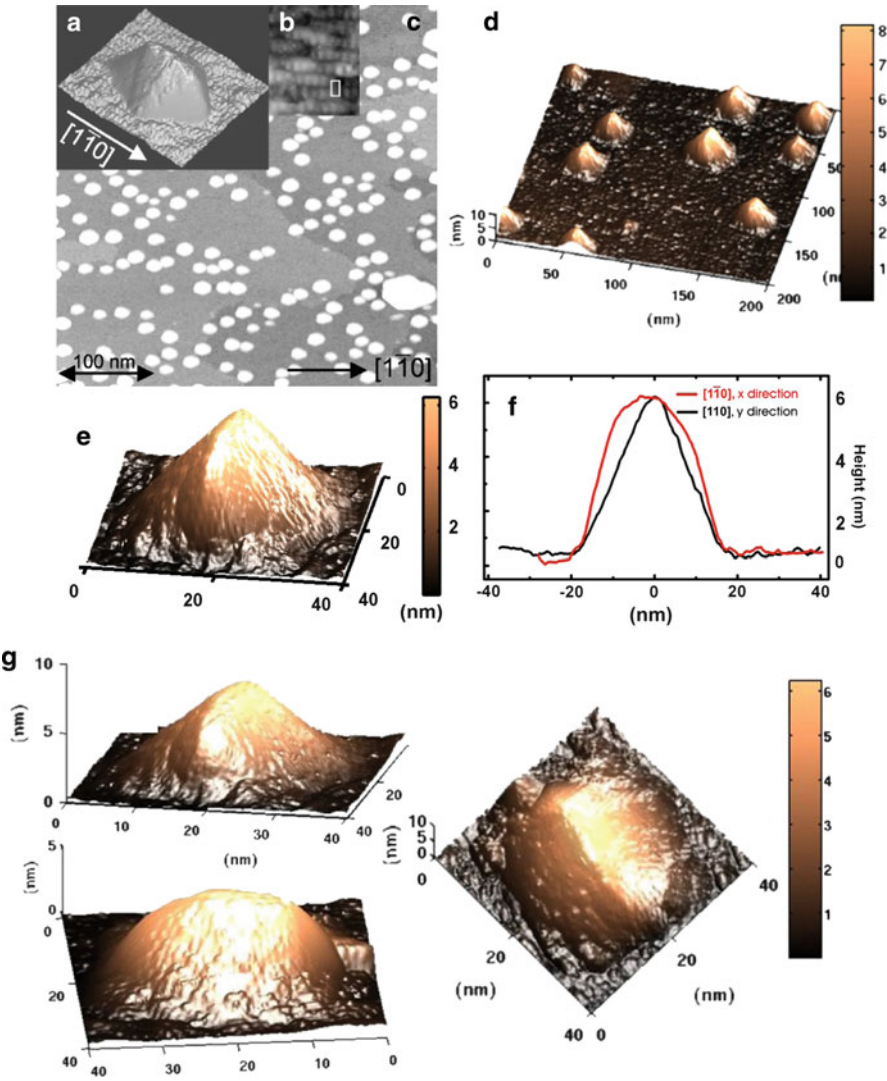


Fig. 8.6 (a–e) STM image of uncapped QD samples grown on a n-doped GaAs(001) substrate. Steps in the wetting layer, its disordered (2×4) reconstruction and anisotropies in QD shape are clearly visible. (f) The different height profiles of a typical QD along the $[110]$ and $[110]$ directions are also shown. (g) Different views of the same QD. [Reprinted with permission from (a–c) T. Maltezopoulos et al., Phys. Rev. Lett. 91, 196804 (2003). Copyright 2003 by the American Physical Society; (d–e) G. Maruccio et al., Nano Lett. 7, 2701–2706 (2007). Copyright 2007 by the American Chemical Society]

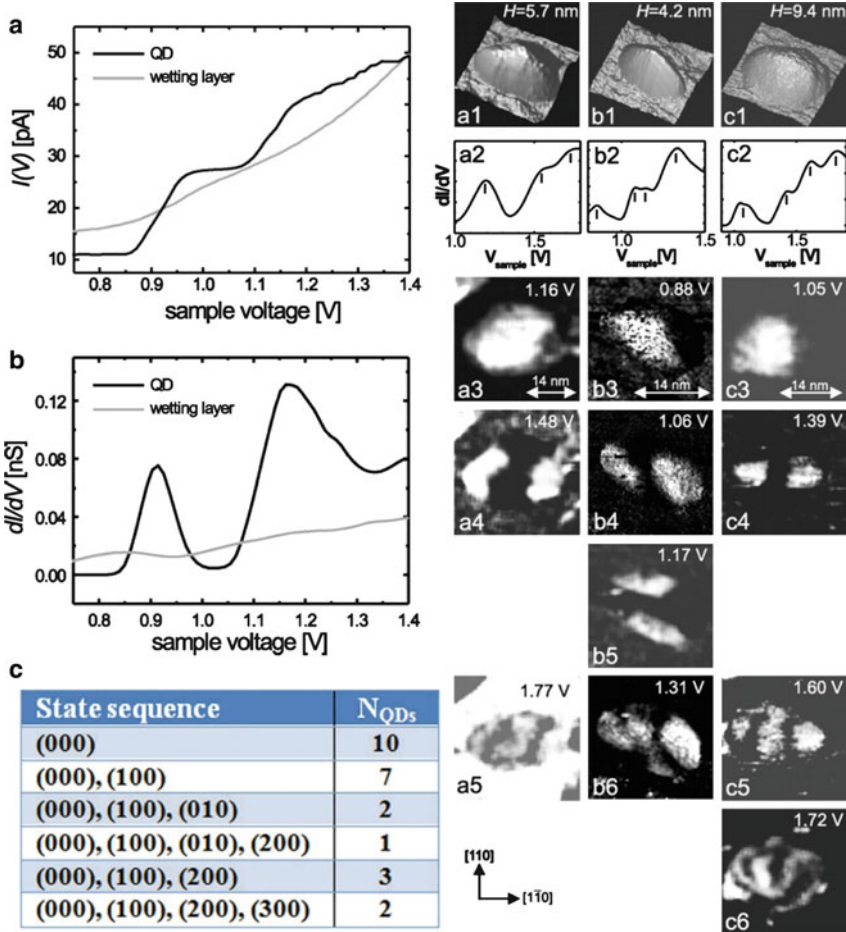


Fig. 8.7 (left, a–b) Current and conductance spectra from a typical QD (black curve) and the wetting layer (gray curve). (right) Wavefunction mapping of independent-particle electronic states in three different QDs. Maps correspond to peak positions denoted by vertical lines in a2, b2 and c2. In this case a low stabilization current was used (50–70 pA, with $V_{\text{stab}} = 1.6$ – 2.4 V) to work in the shell tunneling regime. (left, c) Table summarizing the energetic state sequence for an ensemble of 25 different QDs. [Reprinted with permission from T. Maltezopoulos et al., Phys. Rev. Lett. 91, 196804 (2003). Copyright 2003 by the American Physical Society]

from the tip, and can be estimated as in [80]. The result is an intrinsic lifetime broadening of about 20 meV, which corresponds to approximately 110 mV once the voltage/energy conversion factor² (≈ 5.5) is taken into account. Moving from the

² This factor depends on the potential distribution in the tunnel junction, which is determined by the relative capacitances of the two barriers in a DBTJ, as discussed in Sect. 8.3. It can be estimated from a lever-arm-rule by means of 1D-Poisson calculations or by roughly considering the ratio between the tunneling barrier thickness and the total thickness.

QD center to its sides, the intensity of the low-energy peak decreases while the others increase in weight. A small blue shift of the whole spectrum to higher energies was also observed, probably due to the increased band bending at a smaller distance between the tip, and the degenerately doped GaAs backgate in the case of curves from the QD edge [32]. However, since the resulting peak shifts are small, dI/dV images still largely represent the peak intensity as a function of position [32].

To map single-particle squared WFs, spatially resolved dI/dV images were recorded on tens of QDs from different samples using low stabilization currents (shell tunneling regime). In Fig. 8.7, data from three QDs with more than two confined states are presented. Topographies (first line of figure) again show an elongation of the QDs along the $[1\bar{1}0]$ direction. The spectra in line 2 are averaged on the whole QD area (and for this reason have larger width than single point spectroscopies). As expected, the first state is s-like and exhibits a circular symmetry, while the second one has a p-like shape with a node in the center. Higher energy states with increasing total angular momentum were also observed. These WFs have to be compared with the calculated states discussed in Sect. 8.2. The results of a statistical analysis of the observed WF symmetries and sequences are summarized in Fig. 8.7c. The agreement is good, but the observed state sequences demonstrate the presence of an electronic anisotropy. This can be associated with the shape asymmetry, which induces a stronger confinement along $[110]$ and is thus expected to lift the degeneracy of the p-like states. In fact, the low energy p-state is oriented along the $[1\bar{1}0]$ direction and (200) and (300) states, with nodes in the $[1\bar{1}0]$ direction sometimes appearing in the absence of a (010) state. A qualitative explanation of this somewhat surprising result can be obtained by taking into account the experimentally observed large aspect ratio (around 1.5) in theoretical calculations [32], although shape anisotropy cannot elucidate all the details and, as addressed in a series of theoretical publications (see Sect. 8.2), atomistic symmetry, atomic relaxation and piezoelectric effects have to be considered to achieve a more accurate description. Moreover, different profiles along $[1\bar{1}0]$ and $[110]$ can also contribute to defining a preferential confinement direction.

8.4.3 Coulomb Interactions and Correlation Effects

QDs, however, can be strongly-interacting objects. In particular, in the shell filling regime, the presence of more electrons in a dot [72, 81] adds in the Hamiltonian a mutual Coulomb interaction, which affects the energy spectrum and the WFs of confined carriers, leading to novel ground and excited states that change with the number N of electrons and are distorted by correlation effects. As a consequence, a better understanding of few-particle interactions in strongly correlated systems would be crucial for applications of QDs in single-electron devices, spintronics, and quantum information encoding. Evidence of large correlation effects was first reported in light scattering [40] as well as in high source-drain voltage spectroscopies [41]. On the other hand, most of the reports on WF mapping (both

in real [32, 68, 82, 83] and reciprocal space [70, 84–86] interpreted images in terms of independent-electron orbitals although, as pointed out by Rontani et al. [38, 39], wavefunction mapping is expected to be sensitive to correlation effects. Recently, this was experimentally demonstrated by Maruccio et al. [33].

The samples are similar to those described before (series *L*) but were now investigated at high tunneling currents where electron–electron interaction and correlation effects play a crucial role, as validated by simulation performed by Rontani et al. using the many-body tunneling theory combined with full configuration interaction (FCI) calculations [see 87]³. To proceed further, let us remember that in WF mapping, $dI/dV(V, x, y)$ is proportional to the tunneling probability. Yet, in the case of strong correlation effects, this is not given any more by a sum of the squared single-particle orbitals within the energy resolution as a many-body state is now probed and tunneling leads to a transition among the QD ground states with N and $N + 1$ electrons (where each electron number has its own set of eigenstates). However, extending Bardeen’s formalism and using the many-body tunneling theory, it can be demonstrated that now dI/dV is proportional to the interacting local density of states:

$$dI/dV(V, x, y) \propto -\frac{1}{\hbar\pi} \text{Im } G(x, y; x, y; eV) = |\varphi_{QD}(E_v, x, y)|^2$$

where G is the interacting retarded Green’s function (or single-electron propagator) resolved in both energy and space [38, 39, 88] and the imaginary part of the spectral density $-G/\hbar\pi$ may be regarded as the modulus squared of a quasi particle WF $\varphi_{QD}(E_v, x, y)$, which generalizes the single-particle WF $\psi_v^s(E_v, x, y)$ in the case of strongly correlated systems and can considerably deviate from its independent-particle counterpart [38, 39].

Experimentally, STS spectra (Fig. 8.8) exhibited four clear peaks associated with resonances in the QD spectral density and marked A at 840 mV, B at 1,040 mV, C at 1,140 mV, and D at 1,370 mV. Their full widths at half maximum (FWHM) are about 30, 25, 40, and 75 mV, respectively, and merit some discussion. First of all, it is worth noting that the voltage/energy conversion factor has to be taken into account when converting V_{peak} into the energy of the corresponding quantized state. In particular, due to the smaller tunnel barrier thickness in series *L*, the FWHM appears smaller now than in the previous case (see below for further discussion). The symmetry of the corresponding squared WFs was determined by spatially-resolved mapping performed using high stabilization currents (shell filling regime). Results are shown in Fig. 8.8 and reveal the following approximate symmetries from low to high energy: one s-like (A) and two (or possibly three) p-like (B, C, and D). While states A and B exhibit a standard shape, surprisingly state C again shows a p-like symmetry in the $[1\bar{1}0]$ direction, as before, instead of $[110]$ as expected for the second p-like orbital [32]. As a consequence, it is not possible to explain the WF sequence in terms of one-electron orbitals either, in which case two orthogonal

³ For full details on our FCI method, its performances, and ranges of applicability, see [87].

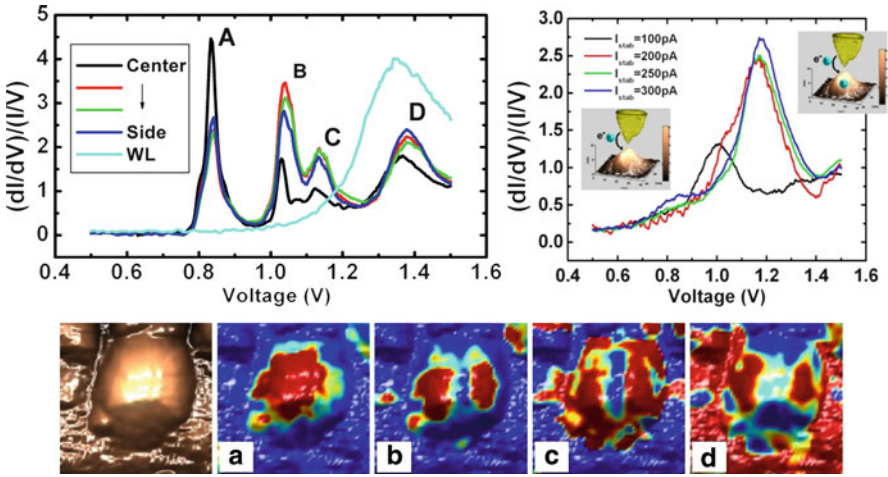


Fig. 8.8 *Top.* STS spectra from a single QD at (*left*) different positions moving from the center to the side and (*right*) at various stabilization currents to change the QD occupancy (in this case, spectra were acquired at the QD side, where states B and C are dominant). Bottom panels. STS spatial maps ($30 \times 30 \text{ nm}^2$) of a single representative QD, taken at 840, 1,040, 1,140, and 1,370 mV, for resonances A, B, C, and D, respectively (2nd–5th panel), in a partial shell filling regime ($I_{\text{stab}} = 100 \text{ pA}$, $V_{\text{stab}} = 1.5 \text{ V}$). The color code represents the STS signal with respect to the topographic STM image on the left hand side (1st panel), increasing from blue to red. [Reprinted with permission from G. Maruccio et al., *Nano Lett.* 7, 2701–2706 (2007). Copyright 2007 by the American Chemical Society]

p-like states should be observed. Additionally, charging of the same p-like orbital with a second electron having opposite spin can be excluded as well, as in this case also one would expect to observe the charging of the s-like orbital, resulting in a second state with circular symmetry. For similar reasons, C is not a phonon replica. In other words, dI/dV maps considerably deviate from the non-interacting WFs.

To explain these observations, correlation effects have to be considered as in the many-body picture developed by Rontani et al. [38,39]. First of all, since states B, C, and D are oriented along the usual preferential direction ($[1\bar{1}0]$), a C_{2v} symmetry has to be used for the effective potential, as described in Sect. 8.2. FCI calculations[18] were thus performed using a fully interacting Hamiltonian for different electron numbers and taking into account anisotropy, electron correlation, and the effect of dielectric mismatch (expected to be strong and to enforce correlation effects). Then the quasi particle WF maps [38, 39] were obtained from the computed correlated states for N and $N - 1$ electrons. As a first result, an asymmetric QD was found to lead to a calculated STS map for the ground state $\rightarrow$ ground state tunneling transition $N = 1 \rightarrow N = 2$ characterized by two peaks along the major axis, while its non-interacting counterpart is simply an elongated gaussian. Figure 8.9 shows (from left to right) the experimental STS energy spectrum (first column) and the typical predicted maps calculated separately for the charging processes corresponding to the injection of the first (second column) and second (third column)

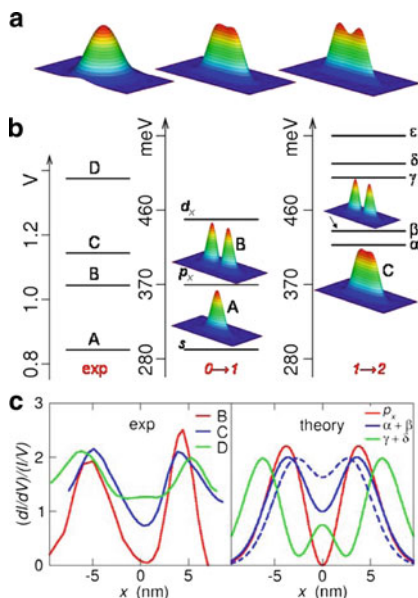


Fig. 8.9 (a) Calculated STS maps for tunneling transition $N = 1 \rightarrow N = 2$, as a function of QD anisotropy with major/minor axes ratios 1, 2.5, and 5, respectively. (b) Experimental STS energy spectrum (*left column*) and calculated states for the tunneling processes $N = 0 \rightarrow N = 1$ (*center column*) and $N = 1 \rightarrow N = 2$ (*right column*). The predicted images of experimentally relevant states are also shown. (c) Profiles of STS maps (*left*) and predicted probability densities (*right*) along a QD volume slice. The predicted *solid blue* (*green*) curve corresponds to the overlap of α and β (γ and δ) states, mixed with a 1:1 ratio, which cannot be resolved at the experimental energy resolution. The dashed blue line is the 1:1 overlap of s and p_x non-interacting orbitals. [Reprinted with permission from G. Maruccio et al., *Nano Lett.* 7, 2701–2706 (2007). Copyright 2007 by the American Chemical Society]

electron into the QD. A comparison among theory and experiments suggests the ascribing of states A and B to the two lowest-energy states predicted for the tunneling process $N = 0 \rightarrow N = 1$. Then, in response to the increased current at larger voltages/energies, the tunneling regime appears to switch from $N = 0 \rightarrow N = 1$ to $N = 1 \rightarrow N = 2$, and states C and D can be associated with $(\alpha + \beta)$ and $(\gamma + \delta)$, respectively, which cannot be distinguished in the tunneling process due to an insufficient energy resolution. A comparison of experimental and theoretical line profiles reinforces this conclusion. As far as the energy scale is concerned, theoretical results are consistent with experimental ones once the voltage/energy conversion factor is considered. If the stabilization current I_{stab} is changed to modify the tip-dot distance and thus the corresponding tunneling rate and regime, further support for this interpretation is obtained. In fact, Fig. 8.8 reports dI/dV spectra collected at increasing I_{stab} on the QD sides where states B and C dominate. Notably, the first (second) peak corresponding to the first (second) p-like orbital gradually disappears (appears), suggesting that at low (high) values of the stabilization current, the energy spectrum of

an uncharged (charged) QD is probed, while for intermediate values, both peaks are observed (as in the WF maps). These observations demonstrate the sensitivity of STS to electron correlation and should be considered for the interpretation of other physical properties of QDs and for their application in devices. Unfortunately, it was not possible to achieve WF mapping with a still higher I_{stab} .

8.5 Colloidal Nanocrystals

8.5.1 *Electronic Properties, Atomic-Like States, and Charging Multiplets*

Colloidal NCs [15] are particularly interesting as their preparation by colloidal techniques is simpler, cheaper and more flexible than physical nanofabrication or epitaxy approaches. Beyond optical techniques (such as photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopies), the effect of the quantum confinement has also been largely investigated by STS [55–57,83]. In the following, without pretending to be exhaustive, we summarize results from a short selection of relevant studies.

Despite previous spectroscopic investigations demonstrating a discrete, atomic-like spectrum [89–92], the character of the individual states was first identified by Banin et al. [55] in 1999 on the basis of experimental observation of two- and six-fold charging multiplets in STS spectra associated with s-like and p-like states, respectively. Specifically, they investigated InAs nanoparticles with radii ranging between 10 and 40 Å and immobilized on a gold substrate using hexanedithiol molecules. In Fig. 8.10a–b, an I–V curve and the corresponding numerical dI/dV spectrum are presented. In the last plot, the above mentioned multiplets are clearly visible and the relevant energies (E_C and the $1P_e$ – $1S_e$ level spacing) can be estimated: the first as the intramultiplet separation, and the last by subtracting E_C from the separation between the two groups. As expected, the energy gap is observed to increase when decreasing the NC size (Fig. 8.10c), in good quantitative agreement with PLE-measured excitonic band gaps corrected to take into account for the excitonic Coulomb interaction absent in the case of STS (Fig. 8.10d). Data analysis also allowed the identification of the levels involved in optical transition (see also [57]). In a different work [93], the same authors discussed in detail the tunneling peak width (see also [94]), which they found to be determined by the electron dwell time on the NC. Millo et al. [83] also investigated InAs/ZnSe core/shell nanocrystals and found an influence of the shell growth on the s–p spacing (Fig. 8.10e). The reason is that while the s-state confined in the core does not shift, the p-state that extends in the shell undergoes a red-shift with shell growth. In the same work, a first evidence of the WF symmetries was reported in current images acquired using the CITS (current image tunneling spectroscopy) technique (Fig. 8.11). This method was also employed by Grandidier et al. for QDs [68] and provides maps of the integrated

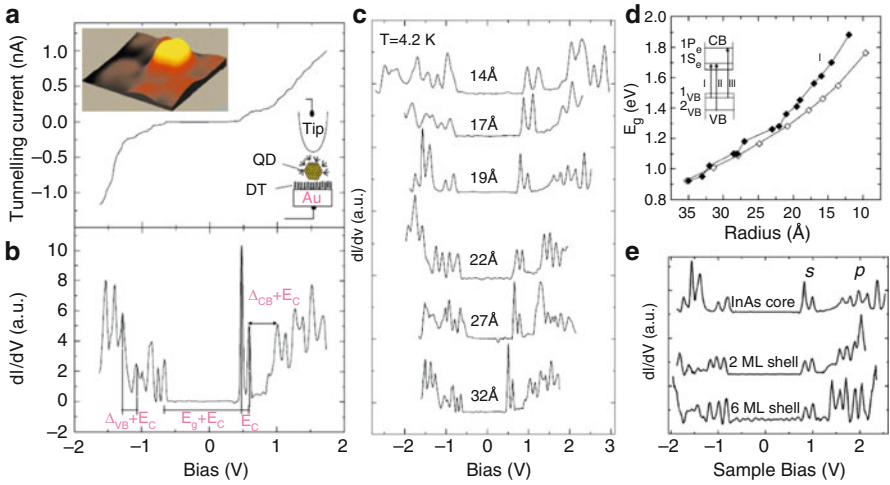


Fig. 8.10 (a–b) Current and conductance spectra acquired on an isolated InAs NC with radius 32 Å (see topographic image in the inset). (c) Size-dependent dI/dV spectra from nanocrystals (NCs) with different radii. (d) Dependence of the STS- and PLE-measured gap (transition I in the inset) on the NC radius. [a–d: Reprinted by permission from Macmillan Publishers Ltd: U. Banin et al., Nature 400, 542 (1999). Copyright 1999]; (e) Evolution of the dI/dV spectra with the shell thickness in core/shell InAs/ZnSe NCs. [Reprinted with permission from O. Millo et al., Phys. Rev. Lett. 86, 5751 (2001). Copyright 2001 by the American Physical Society]

LDOS up to the acquisition bias. Consistent with the previous discussion, they observed a s-like WF with spherical shape localized mainly at the NC core and p-like WFs also extending into the shell region. As the STM geometry and a preferential tunneling through the in-plane p-components (p_x and p_y) lifted the degeneracy between p_x , p_y -states and the p_z -state, the first observed p-state exhibited a toroidal shape and was associated with the combination ($p_x^2 + p_y^2$), while at higher energy the p_z -state was manifested. These assignments were supported by calculations of the envelope functions. Finally, in the same group, hybrid metal-semiconductor nanostructures (Au–CdSe nanodumbbells) were investigated as well [95] with the purpose of gaining insight into the relevant issue of contacts in nanoelectronic devices [96]. In such nanostructures, a significant broadening and a quenching of PL with respect to normal CdSe nanorods were reported [97] as a consequence of the Au growth on their apexes. Such indication for a strong coupling between the Au dots and the CdSe nanorod was further supported by STS studies. Specifically, Steiner et al. [95] acquired spatially resolved spectra along the nanodumbbell, moving on its axis from the metallic dot at the apex to the nanorod center. The spectra were found to be influenced by the coupling through the heterojunctions and non-equidistant Coulomb peaks were observed on the Au dot, while subgap structures manifested on the CdSe part, especially in proximity to the interface. Both features were attributed to localized interface states.

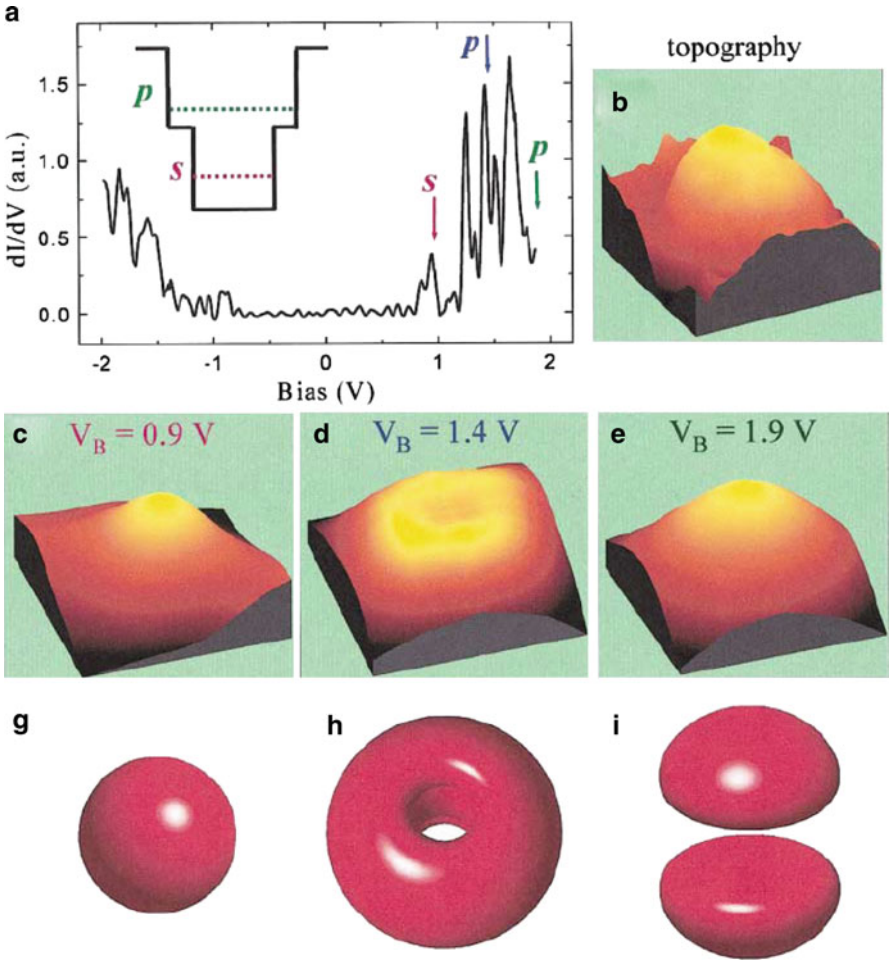


Fig. 8.11 (a–b) dI/dV spectrum and topography acquired on an InAs/ZnSe core/shell NCs with a 6 ML shell. (c–e) Current image tunneling spectroscopy (CITS) maps displaying the integrated LDOS up to the specified acquisition bias, indicated by an arrow in the dI/dV spectrum in (a). (g–i) Corresponding isoprobability surfaces (s^2 , $p_x^2 + p_y^2$ and p_z^2 respectively) calculated for a spherical QD using the radial potential as sketched in the inset of (a). [Reprinted with permission from O. Millo et al., Phys. Rev. Lett. 86, 5751 (2001). Copyright 2001 by the American Physical Society]

Other relevant experiments were carried out by Liljeroth et al. [56], who investigated the coupling among semiconductor NCs in superlattices (also called QD-solids). Previous investigations concentrated on superlattices of Ag NCs, where Coulomb blockade peaks were observed to disappear [98]. In the case of semiconductor NCs, the scenario is made different by the presence of a discrete spectrum of widely-spaced confined states. In this case, STS spectra are significantly different for isolated NCs or NCs inside the superlattice. In particular, resonances

at negative bias (hole states) were found to undergo minor changes while electronic states exhibited a significant broadening, attributed to band-selective coupling among nearest neighbors (as no differences among arrays with long-range and local order were observed). In some cases, Liljeroth et al. also observed an apparent delocalization in the two-dimensional array. More recently, the same group compared the behavior of NCs in monolayers and bilayers [99].

In summary, tunneling spectroscopy has proven to be a very useful technique to investigate the electronic properties of NCs. Moreover, strong experimental evidence exists that the confined states of NCs and their energies are strongly influenced by the local environment, such as (a) the surrounding medium which may cause a leakage of the wavefunctions in neighboring layers of buried QDs [12] as well as core-shell NCs [100]; (b) the organic capping ligands bound to the surface of colloidal NCs; (c) the quantum mechanical coupling with neighboring NCs in QD solids [56, 101–103]. Moreover, NCs can be doped and very small clusters (with dimensions of a few nanometers) have been reported to exhibit a molecular-like behaviour [104].

8.5.2 *Electronic Wavefunctions in Immobilized Semiconductor Nanocrystals*

In all previously mentioned studies, s- and p-type states were identified by their appearance as two- and six-fold charging multiplets [55–57] in STS spectra, as a consequence of the atom-like Aufbau principle of sequential energy level occupation [55], with some information on WF symmetries provided by the CITS technique in the form of current images; these, however, display only the integrated LDOS up to the acquisition bias. When compared to larger MBE-grown QDs, there are additional difficulties in STS studies on NCs due to their smaller size. Additionally, the sample preparation from solution requires a stable immobilization of the NCs upon deposition on a suitable substrate. Maps of the individual WFs of NCs were only recently obtained by Maruccio et al. [105] who resolved s- and p-like WFs in individual NCs. They also investigated the influence of the coupling of the NCs with the surrounding molecules and the gold substrate on the WF energies and symmetry.

Specifically, InP nanoparticles with sizes of 4–6 nm were investigated. After exchanging the original capping molecules with shorter ones (hexanethiols), NCs were immobilized on Au(111) by means of hexanedithiols as shown in Fig. 8.12 where STM images of the dithiol layer and an immobilized NC are presented. Most of the molecules lie flat and organize in stripes oriented along the three main crystallographic directions of the Au(111) substrate with bright features corresponding to sulfur-containing endgroups [106–108] with a separation being in good agreement with the molecular length. However, some molecules in a “standing up phase” are also visible (brighter stripe in Fig. 8.12b). After dipping this thiolated Au substrate in a toluene solution containing the NCs, they can be observed as additional features having lateral dimensions and heights comparable with the NC size (4–6 nm) as determined by transmission electron microscopy (TEM).

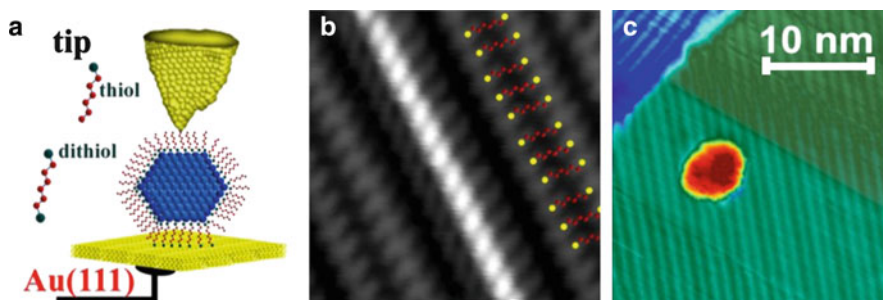


Fig. 8.12 (a) Experimental setup with InP nanocrystals immobilized on a Au(111) surface via hexanedithiols. (b–c) STM images of the hexanedithiol layer and an immobilized NC. The measured length of the adsorbed molecules and the size of the NC are in good agreement with the expected values. [G. Maruccio et al., *Small*, 5, 808–812 (2009). Copyright Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission]

Concerning the electronic properties of the immobilized NCs studied by STS, a non-conducting gap and various confined states were observed with the Fermi level closer to the electron states in the conduction band (Fig. 8.13a). Moreover, the annealing conditions were found to influence the measured band gap: in the case of as-deposited NCs, an agreement with values around 1.7–1.8 eV reported in optical studies for InP NCs of the same size was found (black curve), while after a single annealing step, the gap reduced to 1.43 V and the s-state lost in weight (red curve) as if the WF were extended into the neighboring layers (capping molecules and Au substrate). This situation changes if an additional annealing step is carried out, in which case almost equally spaced peaks are exhibited in the dI/dV spectrum (green curve). As the NCs appear more strongly immobilized on the surface after annealing, these observations can be ascribed to the appearance of subgap states induced at the metal-semiconductor junction [95,96] due to an enhanced binding of the NCs to the underlying substrate through the dithiol molecules, resulting in a change of the coupling with the environment [105]. Different charging multiplets were also observed in the shell filling regime (Fig. 8.13) and assigned to the successive charging of s-, p-, and d-states which all exhibit specific charging energies decreasing with increasing total angular momentum due to a decrease of Coulomb interaction when higher orbitals with broader wavefunctions are occupied (in agreement with theoretical predictions [58]).

Electronic WF symmetries were also successfully mapped in the case of NCs after a single annealing step and using a lower stabilization current to work under shell tunneling conditions, and to obtain stable immobilization⁴ and tunneling conditions during the long time necessary for data acquisition (approximately 12 h). As a measure of the energy-resolved LDOS, bias-dependent and spatially-resolved dI/dV images on individual NCs were acquired using a lock-in technique. Results

⁴ A WF mapping experiment typically takes more than 10 h and it is essential to avoid any accidental contact between the STM tip and the sample during that time period.

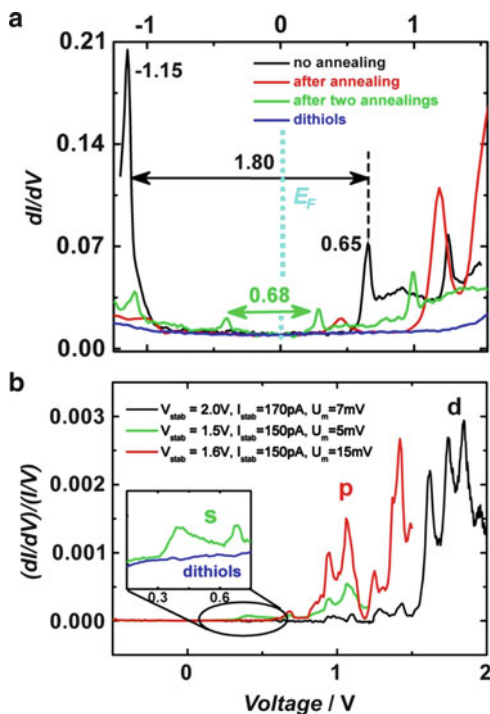


Fig. 8.13 STS spectra of immobilized InP NCs. (a) Shell tunneling regime and dependence of the gap on the annealing conditions, (b) Shell filling regime: individual spectra measured on a single NC moving from the NC center (green curve) to its sides (red and black curves). An almost featureless STS curve on the dithiol layer (blue curve in the inset) is also shown for comparison. [G. Maruccio et al., *Small*, 5, 808–812 (2009). Copyright Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission]

from three different NCs with similar sizes are shown in Fig. 8.14. Topographies (scale bar = 5 nm) are reported along with superimposed STS maps, with the color scale increasing from blue to red as in the visible spectrum. For tunneling conditions corresponding to the energy gap of the NCs (Fig. 8.14b), the LDOS inside the NCs is negligible and the position occupied by the NC appears blue. On the other hand, when resonant with the first two peaks in the dI/dV spectra, s- and p-symmetries are observed (Fig. 8.14). Apart from some differences in the extension of the s-state, such results are similar for different NCs. When compared to results from Millo et al. [83], no hybridization of p_x and p_y states was observed, but rather two clear p-like lobes with a pronounced node in between (Fig. 8.14), indicating a further lifting of p-state degeneracy. Among the different possible explanations, an elongated shape of the NCs [29, 33] was ruled out according to TEM and STM images, while a piezoelectric field [29] could be excluded as well, as no strain is expected for such NCs (in contrast to MBE-grown QDs). Thus, a coupling with the environment (dithiols and gold substrate) [56] remains the most plausible reason for the selection

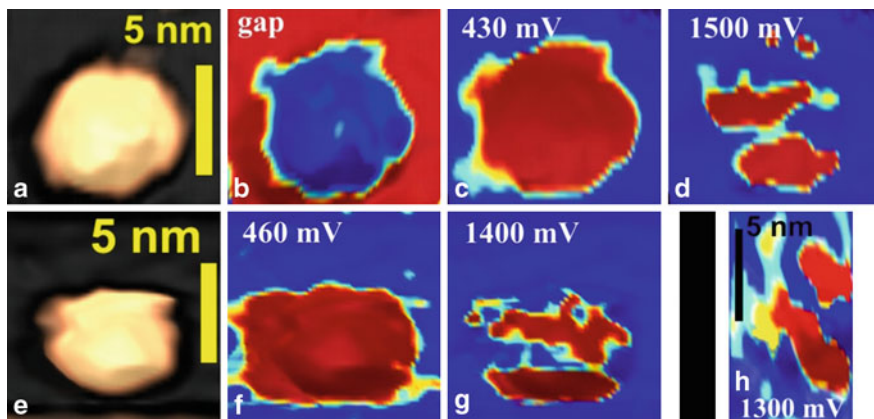


Fig. 8.14 Wavefunction mapping of immobilized InP NCs. (a) Topography; (b–d) Simultaneously acquired STS maps of a single representative NC, taken in the band gap, at 430 and 1,500 mV respectively (2nd–4th panel), corresponding to peaks in the $(dI/dV)/(I/V)$ spectrum. The color code represents the STS signal with respect to the topographic STM image on the left hand side (1st panel), increasing from *blue* to *red*. The scale bar is 5 nm. (e–g) Topography and STS maps showing the s- and p- states of another NC. (h) a p-state from a third NC with a different orientation. [G. Maruccio et al., *Small*, 5, 808–812 (2009). Copyright Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission]

of the p-state oriented along the Au(111) main crystallographic directions (also followed by the dithiol stripes). The same argument would also explain the differences observed in the extension of the s-states, the smaller weight and increased FWHM of the s-peak, and the dependence of the gap on the annealing steps. No clear images of higher energy states (such as d-states or the p_z -state) were reproducibly obtained due to the need of higher bias voltages, resulting in unstable tunneling conditions.

8.6 Conclusions

In conclusion, QDs and NCs represent an important field of current fundamental and applied research, with continuously evolving applications. Here, experimental results on tunneling spectroscopy on semiconductor QDs and NCs have been discussed, showing the potential of this technique and its evolution, e.g., spatially resolved WF mapping to perform detailed investigations of the electronic states of QDs and more complex systems, such as hybrid and shape-controlled NCs [105].

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Chapter 9

Scanning Tunneling Spectroscopy on III–V Materials: Effects of Dimensionality, Magnetic Field, and Magnetic Impurities

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Abstract We review low-temperature scanning tunneling spectroscopy (STS) investigations of the local electron density of states (LDOS) of different electron and hole systems in III–V semiconductors. By cleavage of InAs or InSb, a clean (110) surface can be prepared, with no intrinsic surface states in a range of ± 1 eV around the band edges, which is the relevant energy window for STS. This allows the study of the electronic properties of the simple parabolic, s-like conduction band, thus giving access to effects induced by interaction. Systems in different dimensions and in an applied magnetic field have been studied in real space on the atomic scale in order to disentangle the interesting but complex interaction of electrons with disorder. We focus on a comparison between the three-dimensional (3D) electron system and the two-dimensional (2D) electron system with and without magnetic fields. While without a magnetic field, the electronic wave functions are much more complex in 2D than in 3D, an appealing similarity has been found in high magnetic fields. In 2D, the imaged states can be clearly identified with the states responsible for integer quantum Hall transitions. The origin of the 3D states appearing in the extreme quantum limit is still not clear. Furthermore, by doping the semiconductor with magnetic acceptors like Mn, the properties of the bound hole and its interaction with the tip-induced potential can be studied on the local scale. The LDOS of the hole has a strongly anisotropic shape, which is further disturbed by interaction with the (110) surface.

9.1 Introduction

Since the discovery of the scanning tunneling microscope (STM) [1], a huge number of different solid state samples have been investigated to understand the properties of the systems down to the atomic scale [2]. Basically, a metallic tip is brought close to a sample surface, i.e. the distance between tip and sample is about 5 Å. At this distance, a stable tunneling current between the tip and the sample is established, coupling the electronic states of the tip to the electronic states of the sample. Scanning the tip across the surface thus allows the study of the local electronic properties

at the sample surface, i.e. provides insight into the local distribution of the electronic states. By measuring the differential conductivity as a function of the sample voltage in the so called scanning tunneling spectroscopy (STS), one can separate the electronic states at different energies [3]. Provided that the tip injects electrons of a preferential spin orientation, which can be achieved by coating the tip with a thin layer of magnetic material, one can even get additional information on the spin orientation of the corresponding electronic states [4, 5].

The (110) surface of the III–V semiconductors InAs and InSb is especially well suited for an STS study of electron or hole systems of different dimensions and of buried dopants because of three reasons: (1) As it is the natural cleavage plane, a largely defect free and flat surface that typically shows no steps over several hundreds of nanometers can easily be prepared in ultra high vacuum; (2) STS is usually sensitive to the local density of states (LDOS) *at* the surface. However, since the (110) surface has no intrinsic surface states in a range of ± 1 eV around the band edges, which is the relevant window for STS, the electronic states of the bulk three-dimensional electron system (3DES) or of dopants lying up to ten layers below the surface are still detectable; (3) Confined electron systems of lower dimensionality can be obtained by depositing adsorbates (2DES), below step edges (1DES), or just below the local gate built by the tip of the STM (0DES). Thus, the local electronic properties of electrons in all dimensions as well as of bulk dopants can be studied on the atomic scale.

Here, we review the efforts to understand the versatile electronic properties of low gap III–V semiconductors concentrating on 3D and 2D systems with and without magnetic field, which gives an unprecedented direct insight into the interaction of electrons with potential disorder (Sect. 9.3), and the investigation of magnetic impurities in III–V's (Sect. 9.4), which are the base of the promising sample system of ferromagnetic semiconductors. We start with a section on the interpretation of STM and STS data (Sect. 9.2), with a focus on the effect of the tip-induced band bending.

9.2 Interpreting STM and STS Data

In scanning tunneling microscopy (STM), a metallic tip is positioned close to a sample surface and the tip is moved parallel to the surface as shown in Fig. 9.1a. One detects the tunneling current I as a function of applied voltage V and lateral position of the tip with respect to the surface (x, y) [3]. For elastic tunneling, which is the major tunneling channel in usual STM/STS experiments [6], and z distances where tip density of states (DOS) and sample DOS are not mutually influenced, a matrix approach developed by Bardeen is appropriate to describe I [7]. As the resulting expression is still complicated, Tersoff and Hamann introduced the additional assumption that the tip exhibits a DOS consisting of s-like states [8, 9]. This

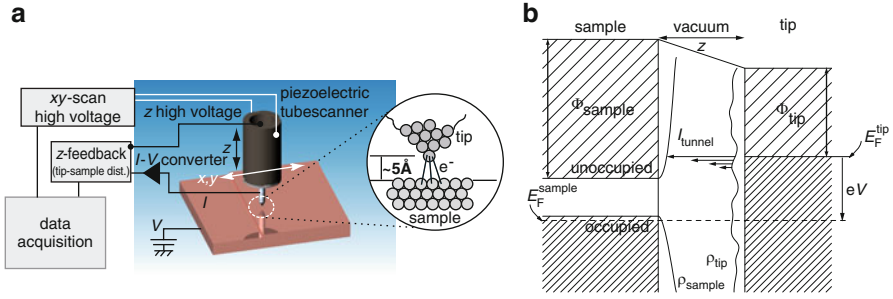


Fig. 9.1 (a) Schematic drawing of the working principle of a scanning tunneling microscope (STM) with the bias voltage V applied to the sample resulting in a tunneling current I . Tip movement in the x, y plane and displacement z relative to the sample surface normal are indicated by arrows. (b) Energy diagram of the tunneling gap between a p-doped semiconductor and a metallic tip. Φ_{tip} and Φ_{sample} are the work functions of tip and sample. Here, the sample voltage is positive, resulting in a tunneling current from occupied states of the tip with density ρ_{tip} into unoccupied states of the sample with density ρ_{sample} . The tip-induced band bending is ignored

led to the further simplified expression:

$$I(V, x, y, z) \propto \int_0^{eV} \rho_{\text{tip}}(E - eV) \cdot \rho_{\text{sample}}(E, x, y) \cdot T(E, V, z(x, y)) dE \quad (9.1)$$

Here $\rho_{\text{tip}}(E)$ is the tip DOS, $\rho_{\text{sample}}(E, x, y)$ is the sample's local density of states (LDOS), and $T(E, V, z(x, y))$ is a transmission coefficient basically describing the spatial overlap of states from sample and tip. The integral covers the region of energetically overlapping occupied and unoccupied states, as illustrated in Fig. 9.1b for the example of a p-doped semiconductor and a metallic tip.

To extract the sample LDOS, $T(E, eV, z)$ has to be known. One can measure it using $I(V, z) \propto T(E = eV, V, z)$, which is valid as long as $V \ll \hat{\Phi}$, where $\hat{\Phi}$ is the effective barrier height at $V = 0$ mV (usually 2–4 V). Measuring $I(V, z)$ confirms the normally assumed exponential dependency on the distance z

$$T(E = eV, V, z) \propto \exp(-A \cdot \sqrt{(\hat{\Phi} - e|V|/2) \cdot z}) \quad (9.2)$$

with $A = \sqrt{8m_e}/\hbar$ (m_e : free electron mass, $\hbar$: Planck's constant). Obviously, $\hat{\Phi}$ depends on the tip. It is mostly found to be smaller than the work functions of the tip Φ_{tip} and the sample Φ_{sample} [10–13]. The reason for the small values of $\hat{\Phi}$ is not completely clear, but it is likely that image charge effects play an important role. In this work, $\hat{\Phi}$ is regarded as a measurable quantity. $I(V, z)$ curves are recorded for each set of measurements to determine the actual $\hat{\Phi}$. Mostly, $\hat{\Phi} \simeq 1.4$ eV is found [14].

Direct access to the LDOS is given by the differential conductivity dI/dV , i.e.

$$\begin{aligned} \frac{dI}{dV}(V, x, y, z) &\propto \rho_{\text{tip}}(0) \cdot \rho_{\text{sample}}(eV, x, y) \cdot T(E = eV, V, z) \\ &+ \int_0^{eV} \rho_{\text{tip}}(E - eV) \cdot \rho_{\text{sample}}(E, x, y) \cdot \frac{dT(E, V, z)}{dV} dE \\ &+ \int_0^{eV} \frac{d\rho_{\text{tip}}(E - eV)}{dV} \cdot \rho_{\text{sample}}(E, x, y) \cdot T(E, V, z) dE. \end{aligned} \quad (9.3)$$

The second and third term remain small at low V and can thus be neglected. A quantitative estimate shows that they contribute less than 10% to dI/dV as long as $V \leq 200$ mV [14]. Thus, the lateral variation of $dI/dV(x, y)$ at a particular V would directly reflect the sample LDOS at the corresponding energy E , if z is kept constant. For practical reasons, images are usually obtained not with constant tip distance but by stabilizing the tip at each (x, y) point, so $z(x, y)$ fluctuates. By recording $z(x, y)$ parallel to $dI/dV(x, y)$ and also measuring the $I(z)$ -dependence of the corresponding tip at V , one can compensate for this error according to [15]

$$\text{LDOS}(E = eV, x, y) = \rho_{\text{sample}}(eV, x, y) \propto \frac{dI/dV(V, x, y)}{I(V, z(x, y))}. \quad (9.4)$$

Thus, the lateral dependence of the LDOS can be directly measured. Often, it is not necessary to use (9.4); it is sufficient to assume $\text{LDOS}(eV, x, y) \propto dI/dV(V, x, y)$. This has the advantage that one does not introduce additional noise to the original dI/dV data by the division. As a rule of thumb, one can keep in mind that corrugations in $dI/dV(x, y)$ of less than 10% have to be normalized according to (9.4), while larger corrugations are not sensitive to a changing $z(x, y)$.

To compare different LDOS images, it is useful to define the strength of the corrugation C_{meas} :

$$C_{\text{meas}} = \frac{\text{LDOS}_{\text{mean}} - \text{LDOS}_{\text{min}}}{\text{LDOS}_{\text{mean}}}, \quad (9.5)$$

where min and mean refer to the smallest and average values of the LDOS in an image area, respectively.

Remarkably, the measured quantity $\text{LDOS}(E = eV, x, y)$ is directly related to the electronic wave functions at the sample surface, i.e. one measures the squared wave functions at the selected energy:

$$\text{LDOS}(E = eV, x, y) \propto \sum |\Psi(E, x, y)_{\text{surface}}|^2. \quad (9.6)$$

Experimentally, one has to consider the finite temperature and the fact that dI/dV is usually measured by lock-in-technique, utilizing a modulation of the bias voltage with the amplitude V_{mod} (rms-value). Both limit the energy resolution, which can be approximated by a Gaussian broadening with a full width at half maximum (FWHM) $\Delta E = \sqrt{(3.3 \cdot kT)^2 + (2.5 \cdot V_{\text{mod}})^2}$ [16, 17].

9.2.1 Assumptions

Because the restriction to s-like tip states is questionable, the assumptions used in the above derivation have to be discussed. Chen has shown that tip states of higher orbital momentum would lead to a replacement of ρ_{sample} in (9.1) and (9.3) by its spatial derivative [18–20]. In particular, for p_x -, p_y - or p_z -parts of the tip state, the first derivative along x , y or z should be used. For d -states, one needs the corresponding second derivatives, and so on. As tunneling into higher orbital tip states requires a strong orientation of the states towards the surface, i.e. along z , one usually detects a derivative of ρ_{sample} along z . Anyway, at large enough tip-surface distances, the z -dependence of the LDOS is largely described by $e^{-\alpha z}$ (a typical value is $\alpha = 1.4 \text{ \AA}^{-1}$). In case of higher orbital tip states, this mainly leads to an additional numerical constant in (9.1) and (9.3), if α does not depend on (x, y) . The latter is indeed evidenced on the larger length scales by measuring the spatial dependence of $I(z)$. In contrast, atomic scale images are influenced by the derivative effect. Here, the apparent corrugation is largely a consequence of a laterally changing α .

A word is in order with respect to the interpretation of $dI/dV(V)$ -curves. Besides the LDOS(E), they are influenced by two effects. First, $T(E, V, z)$ depends on V . From (9.2), one sees that $T(E, V, z)$ gets larger for increasing $|V|$. Thus, one should keep in mind that an increase of dI/dV with increasing $|V|$ is larger than the corresponding increase of the LDOS(E). Between $|V| = 0 \text{ mV}$ and $|V| = 100 \text{ mV}$, this effect can be estimated to be below 25%. Furthermore, strong features in the DOS of the tip, $\rho_{\text{tip}}(E)$, can change the appearance of dI/dV -curves. These features can usually be identified, and the corresponding tips are not used for measurements.

Images and curves presented in this work are either normalized to adequately represent the LDOS(E, x, y) or it has been checked that this normalization is not relevant for the conclusions taken from the data.

9.2.2 Tip-Induced Band Bending

So far we have not taken into account the fact that the tip can influence the sample LDOS by its electrical field. Fields caused by potential differences $\Delta\Phi$ between sample and tip are only screened within the screening length λ_s of the sample. As semiconductors exhibit λ_s up to several 10 nm, an extended band bending in the area below the tip is the result [22, 23]. If the tip work function is lower than the sample work function, which is usually true for W-tips with GaAs(110), InAs(110) or InSb(110) samples, the band bending is downwards for $V_{\text{bias}} = 0$.

For n-doped material, this results in the formation of a tip-induced quantum dot (QD) in the area below the tip as sketched in Fig. 9.2 [14, 24]. The QD has quantized states that are strongly confined along z (left panel) and less strongly confined along (x, y) (inset of right panel). The corresponding state energies lead to peaks in dI/dV -curves as shown in the main part of the right panel. Vice versa, the

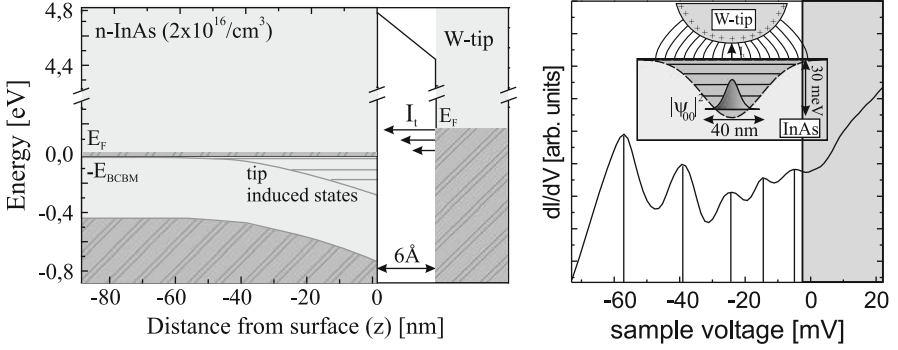


Fig. 9.2 *Left panel:* Sketch of the confined states of the tip-induced quantum dot for an n-doped sample ($2 \cdot 10^{16} \text{ cm}^{-3}$) at $V_{\text{bias}} \approx 0.1 \text{ V}$. The surface conduction band minimum E_{SCBM} is shifted relative to the bulk conduction band minimum E_{BCBM} due to tip-induced band bending $\Phi_{\text{BB}} = E_{\text{BCBM}} - E_{\text{SCBM}}$; the tip-surface distance ($6 \text{ \AA}$) is arbitrary, but does not strongly influence the shape of the band bending; *lengths of the arrows* indicate different transmission coefficients for the tunneling current. *Right panel:* Resulting spatially averaged dI/dV -spectrum ($I_{\text{stab}} = 300 \text{ pA}$, $V_{\text{stab}} = 100 \text{ mV}$, $V_{\text{mod}} = 1.2 \text{ mV}$). *Vertical lines* mark peak positions, and the *grey area* corresponds to the bulk conduction band of InAs. *Inset:* Sketch of the corresponding quantum dot potential (*dark grey area*); *horizontal lines* mark the quantized states and $|\Psi_{00}|^2$ represents the shape of the lowest energy state [21]

measured peak voltages can be used to determine the shape of the QD. Considering the fact that the potential difference between sample and tip depends on $\Delta\Phi$ and V_{bias} , Hartree calculations can be used for different trial potentials to reproduce the measured state energies [24]. In general, it turns out that a Gaussian potential along (x, y) adequately reproduces the data. Thus, two parameters to reproduce the spectra remain: $\Delta\Phi$, which mainly determines the energy of the lowest QD state and σ , the width of the Gaussian, which determines the energy distance between adjacent QD states. Both parameters depend on the actual tip. It is believed that changes in the local atomic arrangement at the tip apex modify the tip work function, and that changes in the tip radius modify the extension of the band bending. For some tips and moderate bias voltages, there might even be no tip-induced QD, and the interpretation of STS data taken on the 3DES and 2DES can be done straightforwardly, without taking into account tip-induced QD states.

As the tip-induced potential also depends on the applied bias voltage, the band bending changes from downward band bending over flat band condition V_{FB} to upward band bending by applying a sufficiently large positive V_{bias} as sketched in Fig. 9.3 for a p-doped sample. For large bias voltages, this effectively shifts the LDOS of the sample $\rho_{\text{sample}}(E)$ with regard to the tip by the amount $\Phi_{\text{BB}}(V_{\text{bias}})$. The canonical equation for the tunneling current in the Tersoff–Hamann model (9.1) therefore has to be extended to

$$I(V, x, y, z) \propto \int_0^{eV} \rho_{\text{tip}}(E - eV) \cdot \rho_{\text{sample}}(E + \Phi_{\text{BB}}(V), x, y) \cdot T(E, V, z(x, y)) dE. \quad (9.7)$$

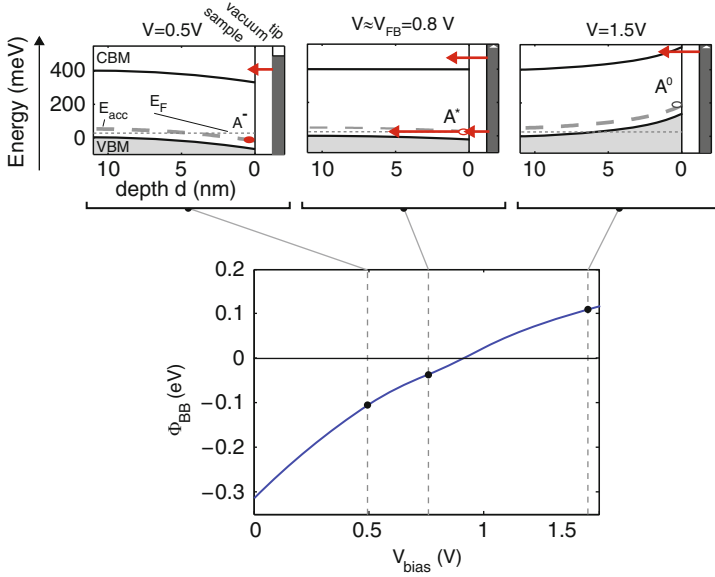


Fig. 9.3 Tip-induced band bending in a p-doped material. *Top panels:* Sketches of the depth-dependent conduction band minimum (CBM) and valence band maximum (VBM) resulting from a 1D poisson model for p-doped InAs ($5 \cdot 10^{18} \text{ cm}^{-3}$) at different V_{bias} using the work function of the W tip ($\Phi_{tip} = 4.5 \text{ eV}$), the band gap ($E_{gap} = 0.41 \text{ eV}$) and electron affinity ($E_A = 4.9 \text{ eV}$) of InAs(110), and a tip-surface distance of $6 \text{ \AA}$. The acceptor level E_{acc} and the Fermi level E_F are indicated. Going from lower to higher bias (*left to right*), a surface acceptor is switched from charged A^- to neutral A^0 configuration creating an additional tunneling path. *Bottom panel:* Calculated Φ_{BB} as a function of V_{bias}

As a consequence, while increasing the bias voltage during an STS experiment, additional tunneling paths can be created by shifting parts of the LDOS of the sample across the Fermi energy. This is illustrated in the top panels of Fig. 9.3 for the case of a surface acceptor. The bands become flat at about $V_{FB} = (E_A + E_{gap} - E_{acc}/2 - \Phi_{tip})/e = 0.8 \text{ V}$, for a tip work function $\Phi_{tip} = 4.5 \text{ eV}$, electron affinity $E_A = 4.9 \text{ eV}$, and band gap $E_{gap} = 0.41 \text{ eV}$ of InAs, and an acceptor energy for Mn in InAs $E_A = 28 \text{ meV}$. At this voltage, the acceptor level is pushed above E_F , opening an additional tunneling path from the tip through the acceptor level into the bulk of the semiconductor. As will be shown in Sect. 9.4, the corresponding LDOS of the hole bound to the acceptor can be imaged under these conditions. In order to obtain an estimate of the extent and amount of $\Phi_{BB}(V)$, a Poisson solver specialized for a one-dimensional model of an STM tunnel junction on a semiconductor sample can be used [25]. The calculated Φ_{BB} as a function of the applied bias voltage assuming an acceptor concentration of $5 \cdot 10^{18} \text{ cm}^{-3}$ and a tip-surface distance of $6 \text{ \AA}$ is shown in the bottom panel of Fig. 9.3.

9.2.3 Experimental Procedures

Depending on the desired material, different commercial n- and p-doped rods as cut from semiconductor wafers are available, in which the dopant density and the degeneracy of the electron system is checked by van-der-Pauw measurements. After in-situ cleavage at a base pressure below 1×10^{-8} Pa, the sample is transferred into the STM and moved into the cryostat. The procedure results in a clean (110)-surface with an STM-detectable adsorbate density of about $10^{-7} \text{ \AA}^{-2}$, an even lower surface vacancy density, and a step density well below $1 \mu\text{m}^{-1}$. The adsorbate density does not increase within weeks. The as-cleaved samples can be used to study the QD and the 3DES as well as for the investigation of the magnetic acceptors buried below the (110) surface. In order to induce a 2DES with a well defined amount of filling and disorder, different adsorbates are deposited from an e-beam evaporator (Fe, Co, Nb) [26–30] or from a dispenser (Cs) [31]. The coverage can be determined by imaging the surface and counting the atoms and is given with respect to the unit cell of the substrate.

For STS measurements, an ex-situ etched W-tip can be further prepared in-situ by field emission and by applying voltage pulses between the tip and a W(110)-sample. Constant-current images are then taken with the voltage V_{bias} applied to the sample. The differential conductivity dI/dV is recorded by lock-in technique ($f = 1.5 \text{ kHz}$, $V_{\text{mod}} = 0.4 - 20 \text{ mV}_{\text{rms}}$). The $dI/dV(V)$ curves are measured at fixed tip position with respect to the surface stabilized at a current I_{stab} and a voltage V_{stab} . Maps of the LDOS result from (x, y) -arrays of adequately normalized $dI/dV(V)$ values.

For the work presented in this chapter, two UHV low-temperature scanning tunneling microscopes have been used, which are described in detail elsewhere [16, 32]. The first one works down to 6 K and in magnetic fields up to 6 T perpendicular to the sample surface with a spectral resolution in STS of $\Delta E \simeq 1.5 \text{ mV}$ [32]. The second has a base temperature of 300 mK and a maximum field of 12 T perpendicular to the sample surface, with a resolution limit of $\Delta E \simeq 0.1 \text{ mV}$ [16].

9.3 Electrons in Different Dimensions

9.3.1 Overview

The understanding of interacting electron systems is a major challenge in solid state physics. Often the interacting systems are not spatially uniform and a local technique like STS can yield indispensable insight into the behaviour of the system [33–42]. It is well known that a rather systematic study of interaction effects can be performed on degenerately doped III–V semiconductors [43, 44]. Here, one deals with only one band exhibiting a nearly parabolic dispersion and the influence of the interaction parameters like dimensionality, potential disorder, electron density, and magnetic field can be varied systematically. Ionized dopants provide the potential

disorder, i.e. deviations from the periodicity of the crystal potential. A low electron density, tuned, e.g., by a gate, increases the significance of electron–electron interactions. Finally, the magnetic field can be used to quench the kinetic energy (Bloch wave energy). This can create systems that are largely determined by the interaction of the electrons with potential disorder and/or mutual electron–electron interactions [45]. Many different electron phases have been identified, leading to a variety of physical effects, e.g., metal–insulator transitions [46], quantum Hall transitions [47], composite fermion phases [48], Wigner crystals [49] or Luttinger liquids [50]. Even quantum Hall ferromagnets [51] and quantum Hall superconductors [52] have been found. Such electron phases are intensively studied by macroscopic means such as transport, magnetization and optical spectroscopy [53]. The microscopic properties, on the other hand, have been probed less extensively. Local properties are rather important, as detailed predictions exist in theory (see e.g. [54–56]). It is, therefore, favorable to apply scanning probe methods that allow the study of microscopic properties on a nm scale [57–59]. A recent review on scanning probe approaches is given in [60]. Here, we summarize part of a systematic study of such III–V semiconductors (InAs/InSb) with varying magnetic fields and dimensionalities [15, 21, 24, 28, 30, 31, 62–66].

There are several advantages of the low-gap materials InAs or InSb for these kind of studies: having a low effective mass and a high g -factor, which results in large Landau and spin splittings in a magnetic field. Moreover, the cleavage plane (110) does not exhibit any surface states within the band gap and within the area of the parabolic conduction and valence band due to the relaxation of the surface atoms shown in Fig. 9.4a. The As-atom moves outwards to realize a configuration close to $s+p^3$, while the In atom moves inwards, leading almost to an (sp_2^3+p) configuration. The resulting surface states that are unoccupied for the In dangling bond and occupied for the As dangling bond are marked as crosses within the band structure of Fig. 9.4b, which was calculated by density functional theory using the local density approximation [67]. The surface states can also be identified by comparing the density of states within the muffin tin parts of the calculation corresponding to atoms at the surface and within the bulk of the material. The latter is demonstrated in Fig. 9.4c, d. This feature of surface states far away from the band edges, which is not present on GaAs-surfaces, is the key allowing systematic modification of the dimensionality of the electron system.

9.3.2 Three-Dimensional Electron System (3DES)

We start by describing STS measurements of the three-dimensional electron system belonging to the InAs conduction band and reaching up to the surface of InAs(110). The corresponding band structure is simple, i.e. the conduction band is nearly parabolic and isotropic, and the symmetry of the atomic wave functions is s -like [67]. The single-particle wave functions $\Psi(\underline{x}, t)$ can be described as Bloch waves:

$$\Psi(\underline{x}, t) = u_s(\underline{x}) \cdot e^{i(\underline{k} \cdot \underline{x} - \omega \cdot t)} \quad (9.8)$$

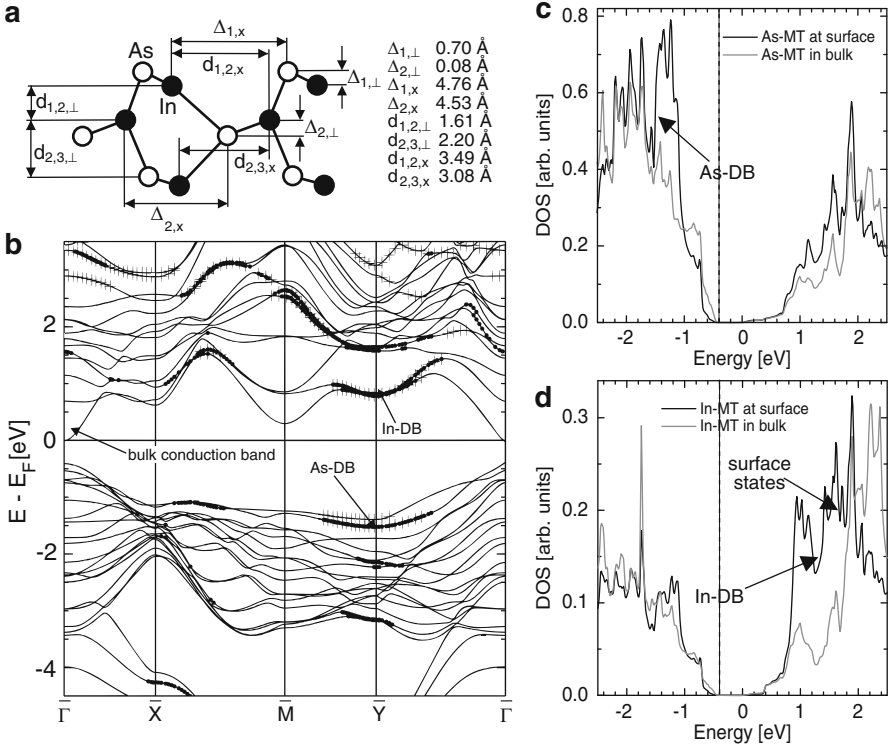


Fig. 9.4 Calculated relaxation, band structure and density of states of the InAs(110) surface [67]. **(a)** Relaxation of the InAs(110) surface with indicated atomic distances (black dots: In, white dots: As). The calculated values of the relaxed distances are listed on the right. **(b)** InAs(110) band structure. Large symbols mark states that lie more than 80% in the upper two layers, crosses (+) mark states with more than 15% probability in the vacuum. The nearly parabolic bulk conduction band at $\bar{\Gamma}$ is marked. States corresponding to the dangling bonds of the In and As atoms are at least 0.75 eV away from E_F . **(c, d)** Local density of states spatially integrated over muffin tin (MT) regions of As **(c)** and In **(d)**. Black lines correspond to MTs directly at the surface and grey lines to atoms in the middle of the slab. The zero energy level is positioned at the conduction band minimum. The vertical dashed line indicates the valence band maximum. Regions corresponding to three different surface states are marked

with energy

$$E = \frac{(\hbar \cdot \underline{k})^2}{2 \cdot m^*} \quad (9.9)$$

Here, $u_s(\underline{x})$ is the atomically periodic part of the Bloch wave, $\underline{k}$ is the wave vector and m^* is the effective mass of the InAs conduction band ($m^* \simeq 0.023 \cdot m_e$). The atomically periodic part $u_s(\underline{x})^2$ can be directly seen in dI/dV -images as shown in Fig. 9.5a, c. The function $u_s(\underline{x})^2$ can be calculated by density functional theory within the local density approximation. The result is shown in Fig. 9.5b, d. The calculated patterns show good agreement with the measured ones [67]. The wave

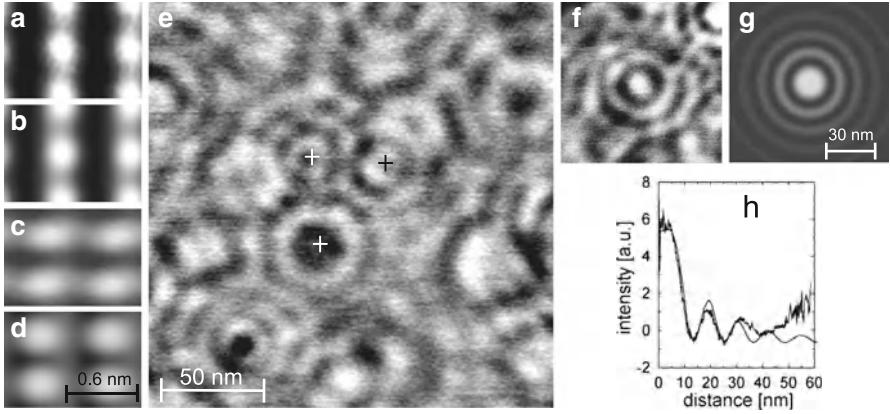


Fig. 9.5 (a) dI/dV -image of InAs(110) measured at $V = 100$ mV. (b) Calculated dI/dV -image corresponding to (a). (c) dI/dV -image of InAs(110) measured at $V = 900$ mV. (d) Calculated dI/dV -image corresponding to (b). (e) dI/dV -image measured at $V = 50$ mV; crosses mark positions of dopants. (f) dI/dV image measured at $V = 50$ mV showing the wave pattern around a defect located 14.3 nm below the surface. (g) Calculated wave pattern corresponding to (f). (h) Height profiles of the measured and the calculated pattern; the x axis starts from the center of the circular pattern (circular line section [15]); the measured pattern has been normalized in order to establish constant tip-surface distance; scale bar in (d) belongs to (a)–(d), scale bar in (g) belongs to (f), (g) [15, 67]

functions in Fig. 9.5a, b belong to the InAs conduction band and the wave functions in Fig. 9.5c, d belong to a surface band at a higher energy [67]. While the image at low voltage is dominated by the As atoms being lifted with respect to the In atoms, the image at high voltage highlights the position of the In dangling bonds, as can be deduced from the calculations.

The long range part $e^{i\vec{k}\cdot\vec{x}}$ can only be seen if the phase of the plane wave is fixed [68], i.e. if the superposition of $e^{i\vec{k}\cdot\vec{x}}$ and $e^{-i\vec{k}\cdot\vec{x}}$ states leads to a corrugation in real space. This can be realized by introducing defects into the atomically periodic structure of the crystal. One can argue that the incoming wave is scattered at the defect, and the interference between the incoming and the reflected wave results in a standing electron wave. In semiconductors, the natural defects are dopants. Figure 9.5e shows a dI/dV image of InAs(110), which exhibits circular standing waves around the sulphur dopants. The circular structure is a direct consequence of the isotropic band structure of the InAs conduction band [69]. The image displays the sum of all scattered waves at the corresponding energy. These are all wave functions with the same $|\vec{k}|$, but with $\vec{k}$ pointing in different directions. As one measures only the surface part of the scattered wave functions, different diameters of the circular structures result, which depend on the depth of the scatterer below the surface. A detailed analysis reveals that the measured patterns result from scatterers down to 25 nm below the surface [15]. The individual circular structures can be reproduced by a simple scattering theory, as shown in Fig. 9.5f–h.

9.3.3 Comparison of 2DES and 3DES

2D electron systems (2DESs) can either be prepared by the growth of InAs on GaAs(111) [70] or by depositing minute amounts of adsorbates on InAs(110) [71] or InSb(110) [31, 72]. In the former case, the interface to the GaAs realizes the 2D confinement, while in the latter cases, a surface accumulation layer on n-type material and an inversion layer on p-type material is formed. Both systems provide relatively large electron densities ($\simeq 10^{12} \text{ cm}^{-2}$) and a moderate disorder strength (5–20 meV) [30]. However, the influence of the interaction of the electrons with the disorder, which is known to lead to weak localization, can be measured on the local scale. Figure 9.6a shows a dI/dV image of a 2DES [30]. A complex wave pattern with a preferential wave length is visible. The Fourier transform of the real space data is shown in the right inset. It is a representation of the wave vectors contributing to the dI/dV image. The apparent ring structure demonstrates that the majority of the contributing wave vectors have the same length.

Figure 9.6c shows a histogram of all dI/dV values obtained in Fig. 9.6a. The histogram is broad, i.e. one finds many different $\text{LDOS}(x, y, E)$ values within the investigated sample area. $\text{LDOS}(x, y, E)$ fluctuates spatially between large values and small values, i.e. the $\text{LDOS}(x, y, E)$ corrugation as defined earlier is large. A single particle calculation using the measured disorder potential (left inset in Fig. 9.6a) can reproduce the features of the dI/dV -image [30].

As the pristine 3D InAs(110) system has also been measured [15], a direct comparison between 2D and 3D behaviour of electrons is possible. A dI/dV image of the 3D system and its histogram are shown in Fig. 9.6b,d. Note that both systems, 2D and 3D, are measured at the same temperature, with the same disorder strength, and at the same energy with respect to the band edge. Thus, the comparison is rather direct. Obviously, the 3D pattern is much more regular, consisting of self-interference rings around individual scatterers. Moreover, the corrugation of the 3D system is lower by an order of magnitude. This is a direct visualization of the fact that 2D systems tend to localize weakly, while 3D systems do not do so [73]. In 3D, the pattern results only from scattering processes at individual dopants, while in 2D, more complex scattering paths, including several dopants, lead to the observed complex pattern. The fact that the scattering paths are closed in 2D [73] leads to the complete phase fixing of all electronic states, resulting in the observed strong corrugation, and it induces the well-known weak localization in 2D.

As an important thermodynamic parameter, a magnetic field of up to 6 T has been applied to systems from 0D to 3D. Landau and spin quantization has been observed for 2D and 0D systems [31, 61]. More interestingly, the transformation of the wave functions in a magnetic field was observed in real space. While 0D and 1D systems did not show pronounced changes due to the relatively large confinement, a distinct change has been observed for 2D [31, 64] and 3D [65]. In both cases, serpentine structures exhibiting strong corrugation appear in Fig. 9.6e,f. The full width at half maximum (FWHM) of the serpentine widths is exactly the magnetic length $l_B = \sqrt{\hbar/(eB)}$. Moreover, in 2D, the patterns are periodic in energy, having a periodicity of the Landau energy. Thus, the theoretically predicted drift states [54],

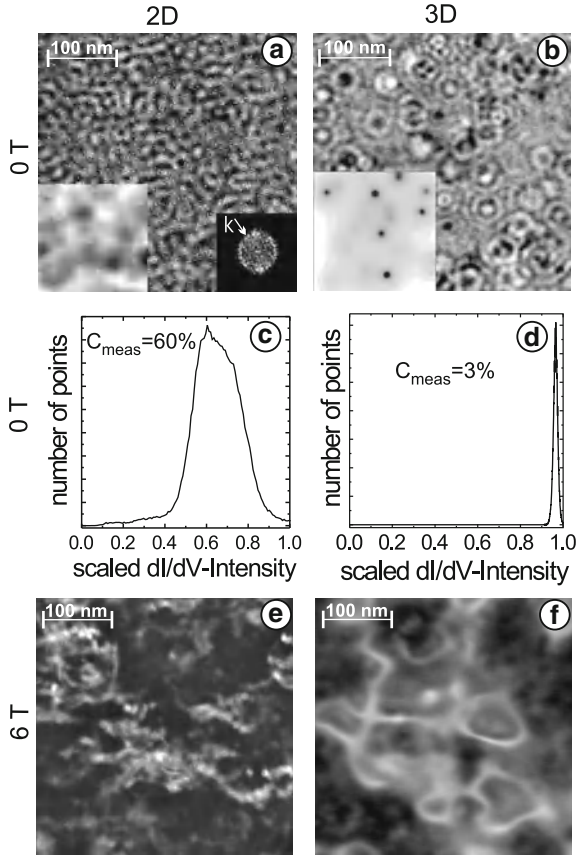


Fig. 9.6 (a) dI/dV map of a 2DES at $B = 0$ T; *insets*: *left*: Measured disorder potential of the 2DES; *right*: Fourier transform of the LDOS with dominating $\vec{k}$ -vector length indicated [30]. (b) dI/dV map of a 3DES measured at the same kinetic energy as the 2DES at $B = 0$ T; *inset*: measured disorder potential of the 3DES at the surface [15]. (c, d) Histograms of the data of (a) and (b) with indicated values of relative LDOS(x, y, E) corrugation (C_{meas} [21]). (e) dI/dV map of the 2DES at $B = 6$ T [64]. (f) dI/dV map of the 3DES at $B = 6$ T [65]; $T = 6$ K

which arise due to the interaction of Lorentz forces and electrostatic disorder, are experimentally confirmed. The states run along equipotential lines of the disorder potential as predicted [74] and shown in Fig. 9.7. The fact that drift states also appear in 3D was surprising and could be linked to the appearance of a Coulomb gap at the Fermi level [75]. Probably, a partial localization of the electrons parallel to the magnetic field arises in the extreme quantum limit, prior to magnetic freeze out. This could lead to local 2D properties of the electrons [76]. Thus, electrons running along equipotential lines of the disorder also exist in 3D at sufficiently high magnetic fields.

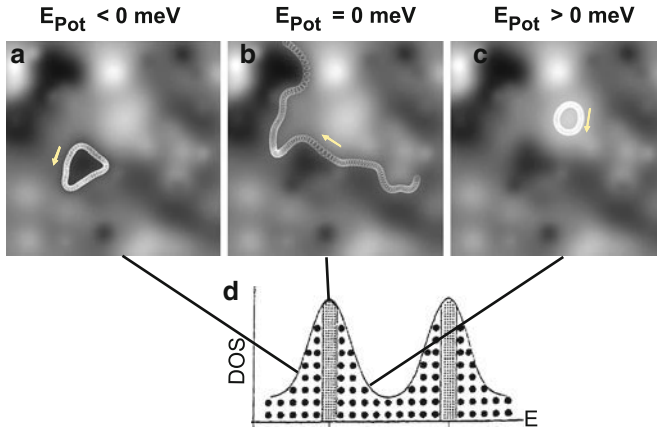


Fig. 9.7 (a, b, c) Calculated electrostatic disorder potential resulting from the dopant density of the sample shown in Fig. 9.6e is displayed in grey scale, and a possible classical electron path within this disorder potential is drawn as a white line; $B = 6$ T. The cycloid paths run along equipotential lines at low (a), middle (b) and high (c) electrostatic potential E_{Pot} . (d) Sketch of the density of states (DOS) of a pair of Landau levels with lines marking the positions of the drift states in (a), (b) and (c)

9.3.4 2DES in a Magnetic Field

In this section, we give a more detailed description of the drift states in 2D systems. Figure 9.7d shows the density of states of a 2DES in a magnetic field. It consists of several Landau levels with index n marking the allowed kinetic energies, which are broadened due to the electrostatic potential disorder dominated by charged impurities. If the cyclotron radius $r_c = \sqrt{(2n + 1)\hbar/(eB)}$ is significantly smaller than the length scale of lateral potential fluctuations, and if the strength of the fluctuations is smaller than the Landau level separation, so-called drift states evolve [54]. They are qualitatively explained by the classical motion of a 2D electron within perpendicular magnetic and electric fields, which leads to cycloid paths within the electrostatic disorder potential running along equipotential lines of the disorder potential as displayed in Fig. 9.7a–c. Thus, at low (high) potential energy E_{Pot} , the electrons circle around the valleys (hills) of the disorder representing localized states, while only at a central potential energy, a percolating cycloid path including saddle points of the potential traverses the whole sample. This extended state is conductive and represents the critical state of the insulator–metal–insulator transition appearing between two quantum Hall plateaus [77, 78]. Quantum mechanically similar states result, as has first been shown by Ando [74], i.e. the probability distribution of the states $|\Psi(\mathbf{x})|^2$ is elongated along the equipotential lines with a FWHM of about the cyclotron radius. The quantum phase transition has been theoretically analyzed in detail, showing a critical exponent of about $7/3$ [77] instead of the classically expected value of $4/3$, which is partly attributed to lateral quantum tunneling processes at the saddle points of the potential [77]. The critical exponent is

in agreement with temperature dependent and frequency dependent transport results [79, 80]. Also, the statistical properties of the critical wave function, which is predicted to exhibit a universal multifractal spectrum, have been analyzed theoretically [77, 78]. Thus, scanning tunneling spectroscopy of the states of this quantum phase transition is valuable for the elucidation of the theoretical predictions in real space.

The 2DES in this section was prepared in ultrahigh vacuum (UHV) by depositing 0.01 monolayer Cs on cleaved n-InSb(110) [31, 81]. STS was performed with a carefully selected W-tip exhibiting a minimum of tip-induced band bending by trial and error [31]. Figure 9.8a shows a color scale plot of the local differential conductivity $dI/dV(V_s)$ of the 2DES measured along a straight line across the sample (left) at $B = 0$ T (sample voltage: V_s). The corresponding spatially averaged dI/dV curve is shown on the right. They represent the energy dependence of the LDOS (energy-position plot) and of the macroscopically averaged LDOS, i.e. the DOS [21]. The energy-position plot shows two apparent boundaries coinciding with two step-like features in the averaged dI/dV curve at $V_s = -115$ meV and -47 meV. They are signatures of the first (E_1) and second (E_2) subband edges and are in excellent agreement with the subband energies resulting from a self-consistent calculation (Fig. 9.8b) [31, 82]. Notice that the irregularity of the onset line of E_1 in Fig. 9.8a is a signature of the potential disorder [64]. Figure 9.8c shows a set of dI/dV curves measured at the same position at different B -fields. At $B = 6$ T, the dI/dV curve already exhibits distinct LLs with a pronounced twofold spin splitting. Repeating the measurement using more B -field steps reveals the continuous evolution of the spin-split LLs, i.e. the LL fan diagram (Fig. 9.8d). The green (red) dashed lines in Fig. 9.8d mark the four (two) spin-down LLs of the first (second) subband. The accompanying spin-up LLs are visible at correspondingly higher energies as marked by blue spin arrows for the lowest LL. LLs of different subbands cross without anti-crossing, indicating orthogonality and, thus, negligible interaction between E_1 and E_2 subbands.

The separation of spin- and Landau-levels increases with B -field reaching $\Delta E_{\uparrow\downarrow} = 24$ meV and $\Delta E_{LL} = 72$ meV, respectively, at $B = 12$ T (Fig. 9.8c). The LLs are separated by regions of $dI/dV \approx 0$, evidencing complete quantization of kinetic energy. Indeed, spin-resolved integer quantum Hall plateaus up to filling factor six were recently observed by magnetotransport on an adsorbate-induced 2DES on InSb(110) [72].

From the peak distances, we deduce the effective mass m^* and the absolute value of the g-factor $|g|$ via $\Delta E_{LL} = \hbar e B / m^*$ ($\hbar$: Planck's constant, e : electron charge) and $\Delta E_{\uparrow\downarrow} = |g| \mu_B B$ (μ_B : Bohr magneton), to be $m^*/m_e = 0.019 \pm 0.001$ and $|g| = 39 \pm 2$ for the lowest energy peaks at $B = 6$ T. This is close to the known values at the band edge $m^*/m_e = 0.014$ and $|g| = 51$ with slight deviations due to the non-parabolicity of the InSb conduction band [83] and the increased energetic distance to the spin-orbit split valence band [84]. The large $|g|$ -factor and low m^* inherent to InSb are the key to a direct measurement of spin-resolved LLs by STS, while a clear energetic resolution of the much smaller spin splitting in GaAs would require the very recently developed capacitance spectroscopy technique [85], which has not been shown to offer sufficient spatial resolution.

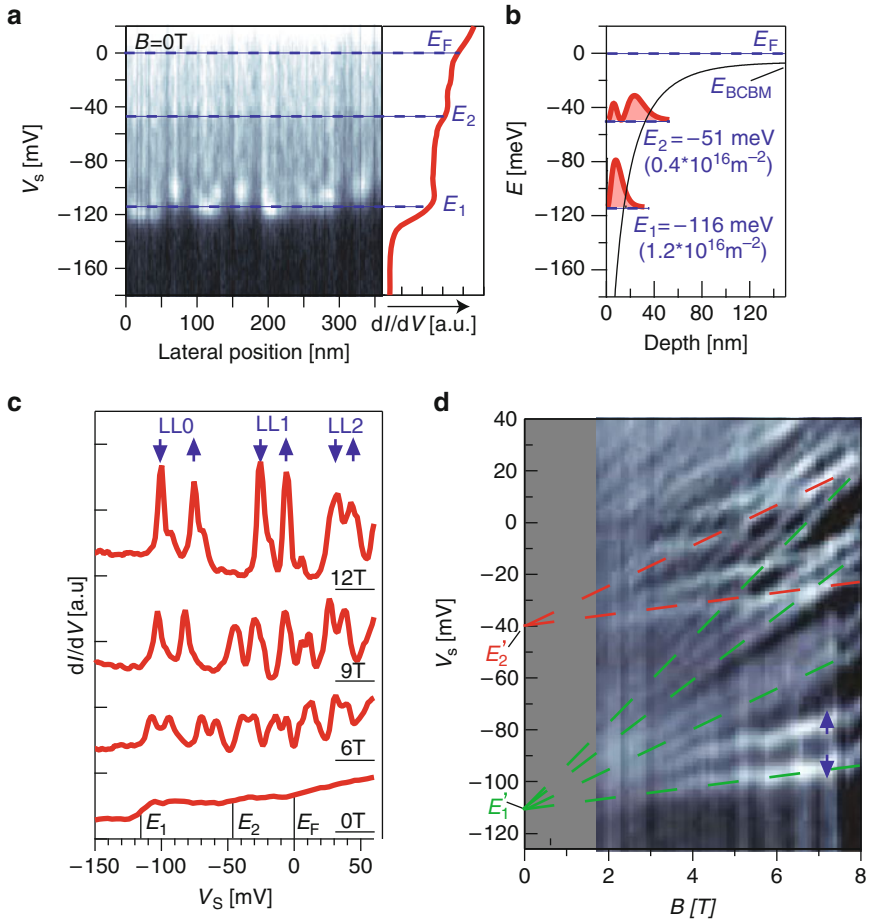


Fig. 9.8 STS of Cs induced 2DES in InSb ($I_{\text{stab}} = 0.13$ nA (a), (c), and 0.10 nA (d), $V_{\text{stab}} = 150$ mV, $V_{\text{mod}} = 2.0$ mV (a), 1.5 meV (d)) (a) Color scale plot of dI/dV (V_s) measured along a straight line at $B = 0$ T (left), and corresponding spatially averaged dI/dV curve (right). Blue lines: E_F and subband energies of the 2DES (E_1 , E_2). (b) Self-consistently calculated band bending (black solid line) and confined states (red areas). Resulting E_i and corresponding electron densities n_i are marked. (c) dI/dV curves at different B -fields recorded at the same lateral position ($V_{\text{mod}} = 2.0$ mV (0 T), 1.3 mV (6 T), 1.0 mV (9 T) and 0.9 mV (12 T)). E_1 , E_2 and E_F (bottom), ($dI/dV = 0$)-lines (right), as well as LLs with index and spin directions (top) are marked. (d) Experimentally determined Landau fan diagram (see the text). Green (red) dashed lines: Spin-down LLs of the first (second) subband. Arrows: different spin levels of the lowest LL. Reprinted figure with permission from [31]. Copyright (2009) by the American Physical Society

Now we address the real-space behavior of the LDOS across a quantum-Hall transition. Figure 9.9a–g present the dI/dV images recorded at different V_s in the lowest spin-down LL at $B = 12$ T. The corresponding, spatially averaged dI/dV curve is shown in Fig. 9.9h. The continuous change of the LDOS with energy can be found in [31]. In the low-energy tail of the LL at $V_s = -116.3$ mV (Fig. 9.9a), we

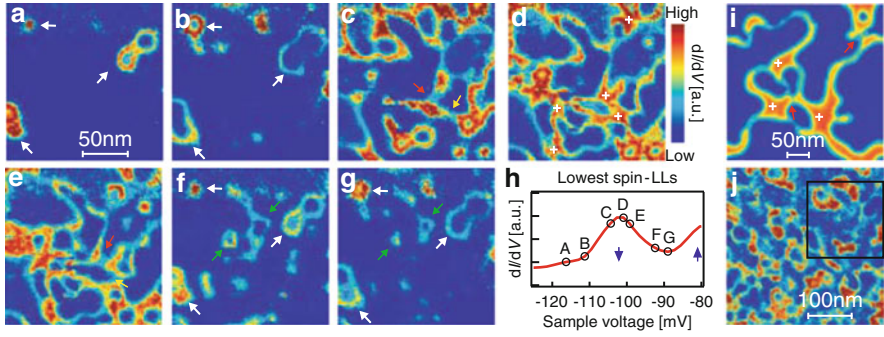


Fig. 9.9 Reprinted figure with permission from [31]. Real-space LDOS at the lowest LL. (a)–(g) Measured dI/dV images at $B = 12$ T and $V_s = -116.3$ mV (a), -111.2 mV (b), -104.4 mV (c), -100.9 mV (d), -99.2 mV (e), -92.4 mV (f) and -89.0 mV (g) ($I_{\text{stab}} = 0.10$ nA, $V_{\text{stab}} = 150$ mV, $V_{\text{mod}} = 1.0$ mV). The same dI/dV -color scale is used for each image. Green (white) arrows mark drift states encircling potential maxima (minima) [(a), (b), (f), and (g)]. Red and yellow arrows mark tunneling connections existing at the identical position in (c) and (e). Crosses mark extended LDOS areas at the saddle points (d). (h) Spatially averaged dI/dV curve with circles marking V_s corresponding to (a)–(g). (i) Calculated LDOS at the center of the spin-down LL0 ($B = 12$ T). Red arrows mark tunneling connections at the saddle points. Crosses mark extended areas. (j) dI/dV image measured at $B = 6$ T and $V_s = -99$ mV close to the center of spin-up LL0. The image includes the area of (a)–(g) within the marked rectangle. Reprinted figure with permission from [31]. Copyright (2009) by the American Physical Society

observe spatially isolated closed-loop patterns. The averaged FWHM of the closed loop in the top right of Fig. 9.9a is 6.9 nm close to the cyclotron radius $r_c = 7.4$ nm. Thus, we attribute the isolated patterns to localized spin-down drift states aligning along the equipotential lines around a potential minimum. Accordingly, at slightly higher energy (Fig. 9.9b), the area encircled by the drift states increases, indicating that the drift states probe a longer equipotential line at higher energy within the potential valley. In contrast, the ring patterns at the high-energy LL tail marked by green arrows in Fig. 9.9f,g encircle an area decreasing in size with increasing voltage. They are attributed to localized drift states around potential maxima. Notice that the structures in Fig. 9.9a,b appear almost identical to those in Fig. 9.9f,g as marked by the white arrows. The latter structures are the low-energy spin-up states localized around potential minima, which energetically overlap with the high-energy spin-down states localized around potential maxima. When the voltage is close to the LL center (Fig. 9.9c,e), adjacent drift states become partly connected (probably at saddle points of the potential) and a dense network is observed directly at the LL center (Fig. 9.9d). This corresponds exactly to the expected behavior of an extended drift state at the integer quantum-Hall transition, which carries the current through the whole sample. Figure 9.9j shows another extended state recorded on a larger area at different B .

Interestingly, the extended drift states observed around the LL center indicate quantum tunneling of drift states at the saddle points. Within the classical percolation model, the adjacent drift states are connected only at a singular energy at each

saddle, eventually leading to the wrong localization exponent $\nu = 4/3$. Quantum tunneling between classically localized drift states [74, 77] can explain the different value of $7/3$ found numerically [77] and experimentally [79, 80]. The tunneling connections are indeed visible in our data. As an example, the red and yellow arrows in Fig. 9.9c, e mark the same connection point at $V_s = -104.4$ mV and -99.2 mV. LDOS is faintly visible at both positions in both images and surprisingly rotates by about 90° between the images. The reason is simply that the tunneling interconnection mediates between valley states at energies below the LL center and between hill states at energies above the LL center, while hills and valleys are connected via nearly orthogonal lines. The weak links are also reproduced by Hartree-Fock calculations taking the disorder into account [31] as marked by the red arrows in Fig. 9.9i, which represents the LDOS including an extended state. Notice that the intrinsic energy resolution of the experiment is 0.1 meV [16], while peaks in the LL fan diagram exhibit a FWHM of 2.5 meV, probably due to life-time effects. Both values are smaller than the energy region exhibiting intensity at the saddle point. Although the restricted energy resolution can partly account for intensity at the saddles within a larger energy range, it cannot explain the change of orientation. Another intriguing aspect is the observation of LDOS areas larger than r_c around the saddles, again visible in both, calculation (crosses in Fig. 9.9i) and experiment (crosses in Fig. 9.9d). This is probably due to the flat potential at the saddles leading to slow drift speed and, thus, extended LDOS intensity. Note that the observed spatial and energetic spreading of the charge density at the saddle is consistent with previous quantum mechanical calculations [43, 77].

9.4 Magnetic Acceptors

9.4.1 Overview

In the last decade after the invention of the scanning tunneling microscope, there has been an extensive study of surface defects and dopants of semiconductors using STS [86–88]. Recently, there is renewed interest in the shape of the hole bound to magnetic acceptors such as Mn in III–Vs due to its relevance for an atomic scale understanding of the coupling mechanism in diluted magnetic semiconductors [89–91]. Interestingly, STM revealed a strongly anisotropic shape of the topographic signature of the magnetic acceptor in III–Vs as shown in Fig. 9.10a for the example of Mn in InAs. If this shape was directly reflecting the charge density of the hole bound to the acceptor, the relative orientation of an acceptor pair would have an effect on the overlap of their holes and accordingly, an influence on their magnetic coupling. However, as described in Sect. 9.2.2, the interpretation of the STS results is complicated by the strong tip-induced band bending, which leads to a shift of the surface band structure with changing bias voltage. As a consequence, there is an

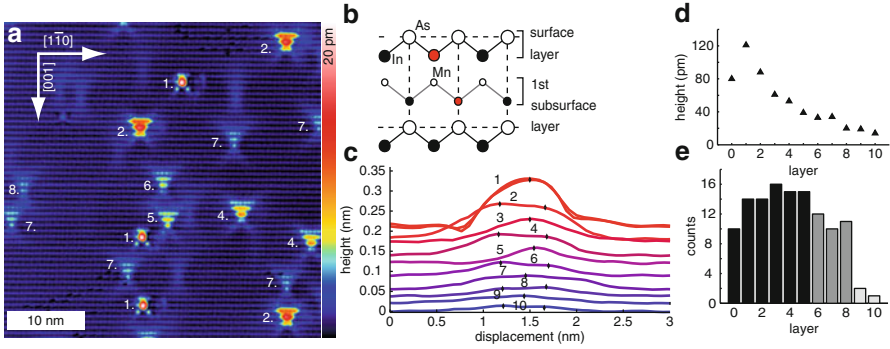


Fig. 9.10 Determination of the embedding depth of Mn acceptors. **(a)** Constant current image taken on the (110)-surface of Mn-doped InAs ($5 \cdot 10^{18} \text{ cm}^{-3}$, $V_{\text{stab}} = 1.1 \text{ V}$, $I_{\text{stab}} = 0.5 \text{ nA}$). Depending on their embedding depths, the Mn acceptors appear as triangular or bow-tie protrusions. The depth below the surface is determined as explained in the text and indicated by the numbers (0: in the surface). **(b)** Sketch of the surface unit cell (black dots: In, white dots: As) and of the first subsurface layer. Substitutional Mn in the surface and subsurface layer are indicated in red. **(c)** Line profiles taken in $[1\bar{1}0]$ direction over the Mn in different depths. The alternating symmetry due to the position of the Mn relative to the surface As atoms is visible: odd layer Mn has one maximum, even layer Mn has two maxima. **(d)** Apparent height of Mn in increasing embedding depths. **(e)** Histogram over the number of Mn assigned to the different layers

ongoing debate whether the anisotropic shape is related to the acceptor state itself, or to tunneling processes at the valence band edge.

Similar anisotropic shapes in topography and spectroscopy have been found for different magnetic and nonmagnetic acceptors such as Zn [88, 92–94, 102], Cd [93], Be [94], Mn [95–100], C [101, 102] and Si [103] in GaAs, Mn in InAs [104–106], and Cd in GaP [107]. The shape ranges from a bow-tie to a triangle, most probably depending on the interplay of three parameters: (1) the binding energy of the acceptor, (2) the embedding depth below the surface, and (3) the amount of tip-induced band bending, which determines the available tunneling paths responsible for the appearance of the acceptor in STM images. For the deep acceptor Mn in GaAs (binding energy $E_A = 113 \text{ meV}$), the interpretation of the asymmetric shape is still under debate [108]. For the shallower Mn in InAs ($E_A = 28 \text{ meV}$), it is now quite settled that it is directly related to the shape of the hole bound to the acceptor. In this section, we will review our effort towards an understanding of the local electronic structure of the bound hole and its coupling to the host states of the semiconductor.

9.4.2 Determining the Depth Below the (110) Surface

Figure 9.10a shows a typical STM topograph of an Mn-doped InAs sample taken at a bias voltage close to flat band condition V_{FB} , where the acceptor level lines up with the bulk sample Fermi level (see Fig. 9.3, top panels). Due to the outward relaxation

of the surface As sublattice and the missing In dangling bond surface states (see Fig. 9.4a, d), only the periodic rectangular As sublattice sketched in Fig. 9.10b is imaged at bias voltages up to about 1.5 eV. This is also proven by taking bias dependent topographs, where a shift from the As to the In sublattice imaging is observed not until 1.4 eV. Due to the statistical distribution of Mn on substitutional In positions in the crystal, the acceptors are found in different embedding depths below the surface layer and are imaged as protrusions of different shape and apparent height superimposed on the As surface lattice. The depth of the corresponding Mn atoms below the surface can be deduced accurately by taking into account the following facts: (1) in line profiles taken along $[1\bar{1}0]$, the maximum of the protrusion is expected on top (in between) the As rows for Mn in odd (even) layers below the surface (surface layer counted as 0, see Fig. 9.10b, c); (2) due to the exponential decay in the charge density of the hole, the topographic height is decreasing as a function of distance from the Mn, as shown in Fig. 9.10d. Note that the Mn in the surface layer (0) appears lower than expected from the exponential decay, indicating a different bond formation. The depth determined accordingly is given in Fig. 9.10a by numbers showing Mn atoms down to at least eight layers below the surface. A histogram of the relative frequency of the Mn in Fig. 9.10e reveals the expected equipartition in the different layers.

9.4.3 Acceptor Charge Switching by Tip-Induced Band Bending

A typical STS spectrum taken on the bare InAs(110) surface in Fig. 9.11a has zero differential conductivity in the bulk band gap region and then rises in the bulk conduction ($V_{\text{bias}} > 0.4$ V) and valence ($V_{\text{bias}} < 0$ V) bands. The Mn acceptor level is expected to lie 28 meV above the valence band edge. In contrast, in an STS spectrum on top of a Mn (Fig. 9.11a), the pronounced peak signature of the acceptor level appears far inside the bulk conduction band. The reason for this shift is the downward bending of the surface electronic states by the tip-induced potential (Sect. 9.2.2), which requires a large positive bias voltage $V_{\text{bias}} \approx 0.7$ V for flat band conditions in order to allow for direct tunneling through the acceptor level. At the same voltage where direct tunneling through the acceptor occurs, its ground state is also pushed above E_F . The acceptor thus changes from negatively charged for $V_{\text{bias}} < 0.7$ V to neutral for $V_{\text{bias}} > 0.7$ V. The negative acceptor is surrounded by a screened Coulomb potential, which leads to an upward band bending of the conduction band, and consequently to a reduction in the conduction band tunneling, in the vicinity of the acceptor. The acceptor switching occurs in a situation where the current is already dominated by tunneling into the conduction band. Therefore, the switching process leads to a step in the tunneling current, and a peak in dI/dV [109].

Depending on the size of the tip-induced potential, the charge switching still happens when a Mn atom is up to several nanometers off from the center of the tip-induced potential (Fig. 9.2, right panel). As the acceptor's Coulomb potential extends over several nanometers, the spot on the surface from which the tunneling

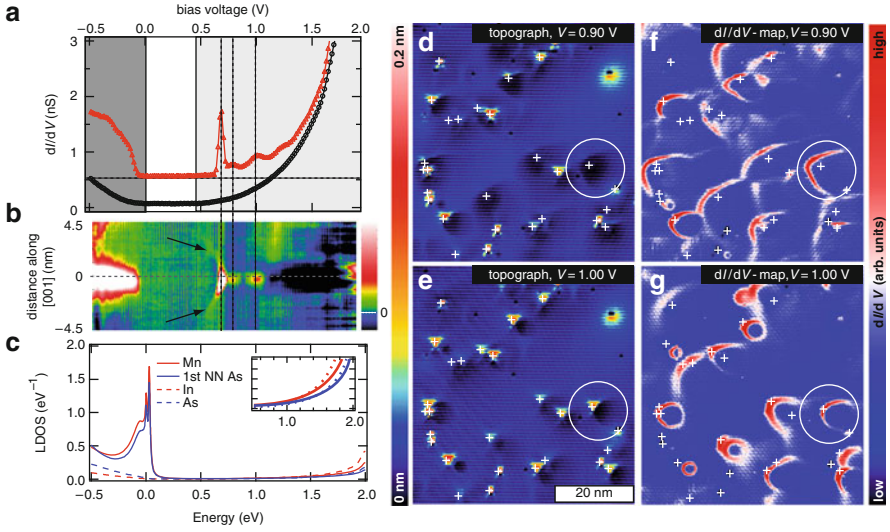


Fig. 9.11 (a) dI/dV curves from the bare InAs (black) and on a second subsurface layer Mn (red) ($n \approx 5 \cdot 10^{18} \text{ cm}^{-3}$). Curves are offset for clarity with dotted zero lines. The bulk valence (dark grey area) and conduction (light grey area) bands are indicated. (b) Voltage dependent section of the relative differential conductivity after subtraction of the InAs curve [105]. Acceptor-related peaks are marked by dashed vertical lines; the shifting of the main peak to lower voltages as a function of the distance to the acceptor is marked by arrows ($V_{\text{stab}} = 2 \text{ V}$, $I_{\text{stab}} = 2 \text{ nA}$, $V_{\text{mod}} = 20 \text{ mV}$). (c) Calculated LDOS at the Mn site, the first-nearest-neighbor sites, and at the bare In and As sites. The inset shows the reduction of the conduction band DOS. (d, e) STM-topographs taken at bias voltages V slightly below and above the peak in dI/dV curves obtained with a tip that has a different work function than in (a, b), $I = 0.5 \text{ nA}$. (f, g) dI/dV maps of same area with V as in (d), (e) ($V_{\text{mod}} = 10 \text{ mV}$). The center of the evolving ring is slightly shifted away from the acceptor's positions, which are marked with white crosses. This can, for example, be seen in the evolution of the ring from one acceptor, marked by white circles in (d)–(g) [109]

electrons are collected in this case will still show a peak, accordingly at a lower voltage and with less intensity. This is the reason for the shifting of the STS peak position to lower voltages as a function of the distance from the acceptor position shown in Fig. 9.11b by black arrows, and for the ring shaped dI/dV intensity found in the dI/dV maps in Fig. 9.11f, g [109]. Exactly at the bias voltage where direct tunneling through the Mn acceptor state is possible, i.e. where the dI/dV ring crosses the position of the Mn marked by white crosses, the triangular feature appears in the STM topographs (see white circles in Fig. 9.11d, e). This is a proof for the assumption that the asymmetric features observed in the topograph (Fig. 9.10a) are indeed direct images of the charge density of the holes bound to Mn acceptors of different depths below InAs(110).

The impact of charge switching on conduction band tunneling in STS experiments has also been observed for conductive grains and Co clusters on the surface of InAs [110, 111] and for Si donors in GaAs [112, 113]. It has been shown that such experiments can be used to deduce the shape and strength of the tip-induced band

bending [109], the strength of the dopant's Coulomb potential [112], and to study the effect of the surface on the dopant's binding energy [113].

9.4.4 Properties of the Hole Bound to the Mn Acceptor

Tight-binding model calculations for Mn in bulk InAs or GaAs show that the LDOS close to the Mn acceptor is spin-orbit split into three states close to the valence band maximum as shown in Fig. 9.11c [105, 114]. For InAs, the $J = 1$ ground state lies 28 meV above the valence band edge. The higher-energy spin states are located at ~ 25 meV and ~ 75 meV below the $J = 1$ ground state [105]. The additional weaker peaks that occur about 125 mV and 375 mV above the acceptor ground state peak in Fig. 9.11a,b could be related to tunneling through the excited spin-orbit split states (see dashed vertical lines). Their order is reversed and their splittings to the ground state are increased by a factor of 5 due to tip-induced band bending. From Fig. 9.11b, their lateral extensions are deduced to be only slightly smaller than the size of the $J = 1$ ground state. This is indeed confirmed by the bulk tight-binding model calculations (not shown).

Besides the peaks, an increase in valence band LDOS by up to 400% and a 10% reduction in conduction band LDOS is observed in Fig. 9.11a,b. The conduction band suppression and valence band enhancement have about the same extension of 2 nm as the acceptor state, and depend only slightly on energy. The same trend is found in the tight-binding model calculations shown in Fig. 9.11c, and is due to a strong effect of the p - d exchange interaction on the valence band and a weaker one on the conduction band [105].

Most importantly, Fig. 9.12 shows a systematic study of the shape of the bound-hole charge density as a function of the depth below the [110] surface, in comparison to the acceptor-level LDOS in different distances to the Mn as calculated from the bulk tight-binding model. In the experimental data, a crab-like shape is observed for Mn in the surface and first subsurface layer, which is very reminiscent of the shape

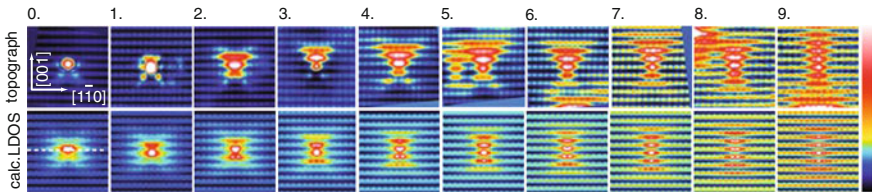


Fig. 9.12 *Top row:* Topographs of the charge density of Mn holes at different layer depths. The layer number below the surface where the Mn is located is marked (surface layer counted as 0). *Bottom row:* LDOS at the acceptor energy at different distances from the Mn as calculated from the bulk tight-binding model. The LDOS is plotted on a logarithmic scale with a spatial broadening factor of $2 \text{ \AA}$. The dashed horizontal line indicates the (001) plane. (Tunneling parameters: $V_{\text{stab}} = 1 \text{ V}$, $I_{\text{stab}} = 0.5 \text{ nA}$) [105]

observed for surface Mn in GaAs [89]. The hole in the second to fourth subsurface layer has a triangular shape, and then gradually changes to a bow-tie shape when the distance is increased to nine layers from the surface. Interestingly, the charge density of the hole down to the seventh layer shows a strong asymmetry with regard to the (001) mirror plane. As visible in Fig. 9.10b, the (001) plane is indeed no mirror plane of the lattice. In the tight-binding model data, the extension and the general shape of the acceptor state are largely reproduced. However, obvious discrepancies are found, in particular, with respect to the (001) mirror plane asymmetry. For example, the states in the 2nd layer appear more intense above the (001) plane within the STM data but less intense within the tight-binding model data. The agreement between the tight-binding model and STM improves with increasing depth and the shapes are nearly identical in the eighth and ninth layers. This depth corresponds to about half the lateral extension of the acceptor wave function. Obviously, the relaxed InAs(110) surface sketched in Fig. 9.4a, which is not included in the calculations, has a significant influence on the spatial distribution of the Mn hole state down to about seven layers.

A similar change from bow-tie to triangular shape for Mn close to the surface has recently been observed for GaAs, both experimentally [98, 99] and theoretically [99, 115], and this seems to be a general trend for acceptors in III–Vs. There are two effects competing with each other, which can explain the reduced symmetry: (1) the impact of the strain field of the surface relaxation on the hole charge-density and (2) the hybridization with surface states. Recent calculations have indeed shown that effect (1) could explain the asymmetry found in Mn in InAs [106] and by taking into account effect (2), the asymmetry of Mn in GaAs can be excellently reproduced [99].

9.5 Conclusions and Outlook

We have reviewed a detailed investigation of the real space properties of conduction-band electron systems in narrow band-gap III–V semiconductors in two and three dimensions using scanning tunneling spectroscopy. Our experiment performed away from E_F is the first direct observation of wave functions across purely non-interacting integer quantum-Hall phase transitions, which are a hallmark in the theoretical description [43, 77, 78, 116]. In principle, the study can be extended to measurements probing 2DES states at E_F in p-type samples, which are currently underway. Thereby, one can probe the role of electron–electron interactions, which are known to provide a wealth of further quantum phases and their transitions [78, 117].

Furthermore, we have reviewed the recent research on the properties of magnetic acceptors in III–V semiconductors revealing that the hole bound to Mn has a strongly anisotropic shape, which is further disturbed by the presence of the surface. It is well known that the asymmetry of the bound hole will have a strong impact on the magnetic exchange interaction between pairs of acceptors of different orientation [89]. Consequently, the magnetic properties, such as Curie temperature and

magnetic anisotropy of materials in the high doping regime, will be altered at the surface and probably also at interfaces of heterostructures where similar effects are expected. It remains for the future to prove this expectation by magnetic sensitive techniques, such as spin-resolved scanning tunneling spectroscopy [118].

An appealing approach for future experiments would be to combine the two systems of 2DES and magnetic acceptors in order to study the effect of hole depletion or inversion on the magnetic interaction between acceptors. Promising first results by magneto-transport measurements have been recently obtained and indicate a spin-glass ordering of magnetic adatoms and their effect on electron scattering in the inversion layer [72, 119].

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Chapter 10

Magnetization of Interacting Electrons in Low-Dimensional Systems

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Abstract In this article, we review selected experiments on the magnetization of low-dimensional electron systems in GaAs-based semiconductor heterostructures. The magnetization monitors the ground state energy of the electron system and is thus of fundamental interest. We discuss the experimental advances in highly sensitive magnetometry that made these experiments possible. Following a short introduction to magnetic quantum oscillations, i.e., the de Haas–van Alphen effect in two-dimensional electron systems, we review key experimental results with particular emphasis on the effects of electron–electron interaction in the regime of the integer and fractional quantum Hall effects. Magnetization experiments on quantum wires and quantum dots created by a top-down approach from two-dimensional systems highlight the effects of external confining potentials and the electron–electron interaction on the ground state energy.

10.1 Introduction

The de Haas–van Alphen (dHvA) effect discovered in 1930 [1] has proved to be an excellent method to determine the Fermi surface of three-dimensional metals. Investigations on *low-dimensional* electron systems (LDES) in *semiconductors*, however, are rare due to the very weak signal strength associated with the orbital moments of the dilute electron systems. The magnetization M is a thermodynamic quantity and hence is a powerful tool to determine the electronic properties of LDES. This reaches far beyond the determination of Fermi surface cross sections and effective masses, since M is for $T \rightarrow 0$ given by the negative derivative of the internal energy U with respect to the magnetic field B , i.e., $M = -\partial U / \partial B|_{T=0}$. A magnetization measurement at sufficiently low temperature thus directly monitors the evolution of the system’s ground state energy. Detailed information about the energy spectrum of the system in thermodynamic equilibrium can be gained. This includes in particular the effects of quantum confinement and the electron–electron interaction.

In this review, we outline the development of the field of magnetometry on LDES: In Sect. 10.2, we describe the advances in highly sensitive magnetometry that enabled these investigations in the first place. Particular emphasis will be placed on the evolution of micromechanical techniques to measure the torque $\boldsymbol{\tau} = \boldsymbol{M} \times \boldsymbol{B}$ acting on a magnetic moment $\boldsymbol{M}$ in an external magnetic field $\boldsymbol{B}$. In Sect. 10.3, we will briefly introduce the basic theory of the equilibrium magnetization of two-dimensional electron systems (2DESs) in quantizing magnetic fields. Selected experiments on 2DESs in high-mobility AlGaAs/GaAs heterostructures will be discussed in Sect. 10.4 with emphasis on the effects of the electron–electron interaction on M . The effect of additional lateral confinement in one and two dimensions, i.e., quantum wires and quantum dots, will be discussed in Sect. 10.5.

10.2 Highly Sensitive Magnetometry

Since the orbital magnetic moment of the quasi-free electrons in a typical semiconductor heterostructure is of the order of a few 10^{-13} J/T per mm^2 its detection is an experimental challenge. The first observation of the dHvA effect in a 2DES was thus reported on stacked layers of an AlGaAs/GaAs heterostructure using a commercial SQUID (Superconducting Quantum Interference Device) magnetometer with a sensitivity of 10^{-10} J/T [2]. The authors detected the magnetic response of an effective 2DES area of 240 cm^2 by stacking several pieces cleaved out of a wafer containing 173 quantum wells grown on top of each other and using an averaging time of up to 30 min per data point. The experiment yielded dHvA oscillations that were about a factor 30 smaller than anticipated. This was attributed to variations in the carrier densities in the individual quantum wells.

Because commercial magnetometers up to date are not sensitive enough to resolve the dHvA effect in single-layer 2DES, a number of groups have developed dedicated magnetometers for this task. In the following, we will review the different approaches and their particular advantages and drawbacks. Due to space limitations, we cannot give an extensive review of all experimental setups that have been reported. Instead, we will strive to give an overview and point out possible ways of further advancement.

10.2.1 *Figures-of-Merit*

Different figures-of-merit have been chosen in the literature [3, 4] to compare the sensitivity of magnetometers. The choice depends on the physics in the focus of the discussion. We do not use a single number here, but plot the change in magnetic moment (in J/T) that can be detected at a given field in a measurement bandwidth of 1 Hz in Fig. 10.1a. We choose this particular representation to (1) keep it independent of the material system under investigation and (2) take into account the different – and in some cases complementary – dependence of the sensitivity on the external field. In comparing the magnetometer performance for large-area 2DESs,

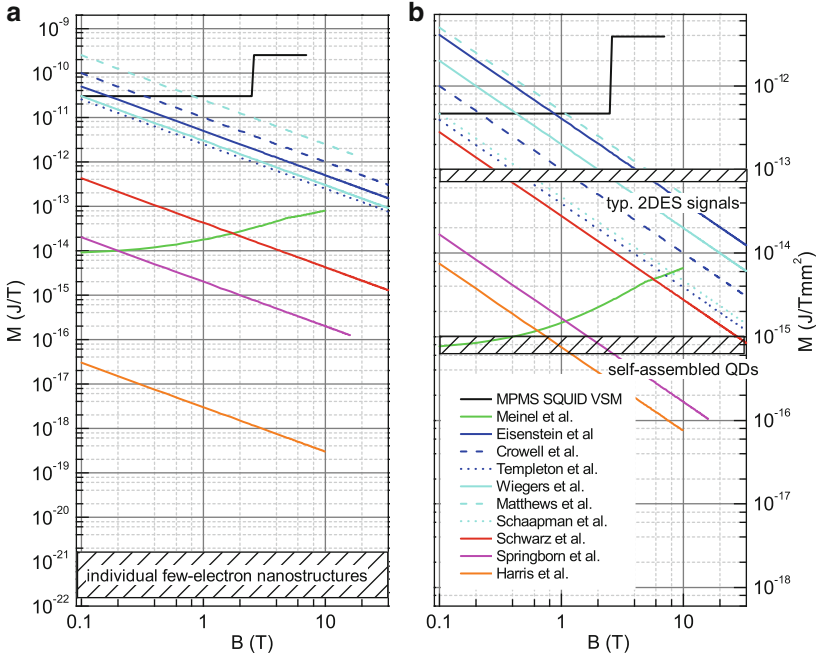


Fig. 10.1 Magnetic moment sensitivity of different magnetometers reported in the literature. Torsion balances are depicted in shades of *blue* and cantilevers in shades of *red*. The *green* line denotes the SQUID of Meinel et al. [5]. In *black*, the specified sensitivity of the commercial Quantum Design MPMS SQUID VSM is shown. (a) Magnetic moment (J/T) that can be detected in a measurement bandwidth of 1 Hz as a function of magnetic field. Note that SQUIDs and torque-based magnetometers are complementary with respect to the field dependence of their sensitivity: the torque $\tau = \mathbf{M} \times \mathbf{B}$ increases with B , while the SQUID performance deteriorates. (b) As in (a), but scaled with the maximum available sample area. Typical signal strengths associated with different electronic systems are depicted as *hatched areas*. The references will be given in the text

it is also useful to scale the sensitivity with the maximum available sample area, since the dHvA effect is proportional to the number of electrons in the system. Figure 10.1b shows the values from (a) scaled with the available sample area in mm². Having in mind the magnetization of electronic nanostructures, however, the absolute moment sensitivity is in the focus of interest, allowing for ever smaller and more homogeneous arrays of nanostructures and – as a visionary goal – the magnetic characterization of individual few-electron structures such as, e.g., a single quantum dot. Indicated as hatched areas are typical (expected) magnetic signal strengths of 2DESs and electronic nanostructures. We point out explicitly here that the representation in Fig. 10.1 is not sufficient as a characterization of the individual instrument's advantages and drawbacks. We therefore briefly discuss the merits of the different systems in the following.

10.2.2 SQUID Magnetometer

A dedicated SQUID susceptometer was developed in the Hamburg group [5]. The system is integrated into a ^3He cryostat with a base temperature of 300 mK that can be operated in standard superconducting solenoids in fields up to $B = 10\text{ T}$ [6]. A sketch of the setup is shown in Fig. 10.2. SQUIDs are quantum limited sensors of changes in magnetic flux. This is, however, only true in background magnetic fields far below the critical field of the Josephson junctions, which is in the mT range. The challenge is thus the design of a SQUID system that works in high magnetic fields. The design concepts of high-field susceptometers have been discussed in detail in [8]. The Hamburg group uses thin-film DC-SQUIDs with an integrated multiturn input coil and NbN–MgO–NbN Josephson junctions [9]. A first order gradiometer wound of NbTi wire on a ceramic sample holder is connected to the SQUID input coil using a superconducting bonding technique. The gradiometer is placed symmetrically with respect to the field center and has a baseline and loop diameter of 10 mm each, adapted to the homogeneity of the magnet. In addition to the input coil and the gradiometer, the flux transformer circuit contains a feedback

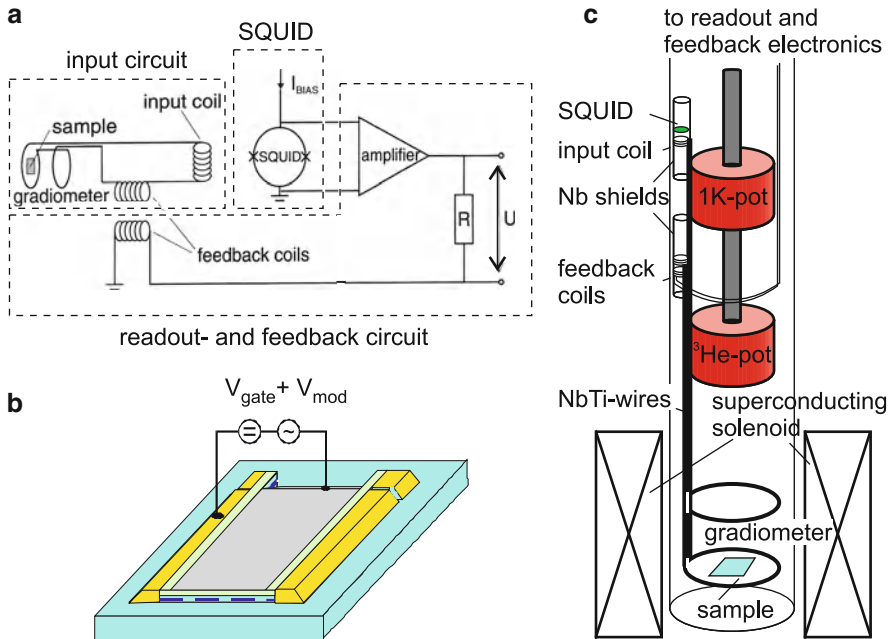


Fig. 10.2 (a) Sketch of the SQUID readout and feedback circuit. Adapted from [6]. (b) Sample design. The mesa area (gray) is $4 \times 4\text{ mm}^2$. The 2DES is provided with alloyed ohmic contacts and a metal top gate allowing for simultaneous magnetotransport and magnetocapacitance measurements. After [7] (c) Sketch of the magnetometer setup integrated into a commercial ^3He cryostat

coil in series as shown in Fig. 10.2a. The readout and feedback loop uses the SQUID output to balance the signal current in the input circuit (null detector). Figure 10.2c shows a sketch of the SQUID magnetometer integrated into a commercial ^3He cryostat. The sample is mounted on a ceramic sample holder attached to the cold finger of the ^3He system. The SQUID and feedback coil are located outside the superconducting magnet. They are thermally anchored to the 1K pot held at $T = 1.5\text{ K}$ regardless of the sample temperature and shielded from the magnets stray field by superconducting Nb tubes (Nb shields). The SQUID and the superconducting input circuit can be warmed above their critical temperature using wire-wound heaters whenever flux penetration occurs and deteriorates the performance.

The system is calibrated using a lithographically defined current loop at the sample position. Magnetization studies on 2DES are performed using a dynamical method, i.e., by modulating the charge density n_s via the top gate voltage with a small rms amplitude $\Delta V_{\text{mod}} = 4\text{ mV}$ and standard lock-in detection. This approach has already been proposed by Shoenberg [10]. The sensitivity of the magnetometer is shown as a green line in Fig. 10.1a. For comparison, the specification of the commercially available MPMS SQUID VSM [manufacturer: Quantum Design, USA (2009)] is drawn as a black line. We point out here that this is the sensitivity guaranteed by the manufacturer, and not the typical best value as given for the magnetometers designed by research groups. Figure 10.1b depicts the same values scaled with the available sample area in mm^2 . The SQUID sensitivity deteriorates with increasing magnetic field. The SQUID output voltage V_{SQ} normalized by the rms amplitude of the voltage modulation is proportional to $\partial M / \partial n_s$. From a measurement of $\partial M / \partial n_s$ vs n_s , the oscillatory part of the magnetization can be calculated. The dynamic readout enhances the resolution. This technique is very sensitive, but limited to LDES, where the electron density can be modulated, for example, by gating.

10.2.3 Concepts of Torque Magnetometry

Torque magnetometers have been very successful in magnetization measurements on LDES, because the magnetic anisotropy of most LDESs allows for a straightforward interpretation of the signals. For example, in a 2DES the direction of the orbital magnetization can, for most practical cases, be assumed to be fixed in the direction perpendicular to the 2DES area, c.f. [11, 12]. The magnetic moment sensitivity *increases* linearly with increasing magnetic field, making torque magnetometers ideal for high-field applications. The devices have in common that the torque acting on the sample's magnetic moment is converted into a deflection of a flexible element on which the sample is mounted.

10.2.4 Torsion-Balance Magnetometers

The discovery of the dHvA effect in metals [1] as well as the first successful measurement of the quantum oscillations of the magnetization $\mathbf{M}$ in a single-layer 2DES was carried out using torsion-balance magnetometers. The principle of operation is sketched in Fig. 10.3a. A sample with or without a sample holder is suspended on a thin wire and placed in a magnetic field pointing in a direction perpendicular to the wire but tilted away from the direction of $\mathbf{M}$ by an angle α . For all quasi-static torque measurements discussed below, this angle is assumed to be $\alpha \approx 15^\circ$. The resulting torque rotates the sample to align $\mathbf{M}$ and $\mathbf{B}$, thereby twisting the wire. Most torsion balance magnetometers reported in the literature use capacitive detection of the deflection. Here, the change in capacitance between an electrode mounted on the sample holder (rotor) and a fixed counterelectrode (stator) is measured (Fig. 10.3b).

Eisenstein et al. [14] employed a semicircular rotor electrode in the plane perpendicular to the wire. Two pie-shaped stator electrodes were placed parallel to the rotor electrode forming a differential capacitor that was read out using an AC voltage bridge. The response of the instrument is linear even for large deflections. The sensitivity of the instrument is depicted as a solid blue line in Fig. 10.1.

Templeton et al. [13] used a different capacitor geometry where two stator electrodes were placed parallel to the rotor electrode. This design allowed one to apply a DC bias for calibration purposes and to operate a feedback loop keeping the rotor position fixed (dotted blue line in Fig. 10.1).

Wiegers et al. [15] reported a torsion balance that was optimized with respect to minimal mechanical coupling to external vibrations, for example originating from high-field Bitter magnets. This was achieved by a highly symmetric design, where a cylindrical rotor with evenly spaced capacitance electrodes was suspended on the torsion wire and a corresponding stator counterpart. Faulhaber et al. [16] constructed

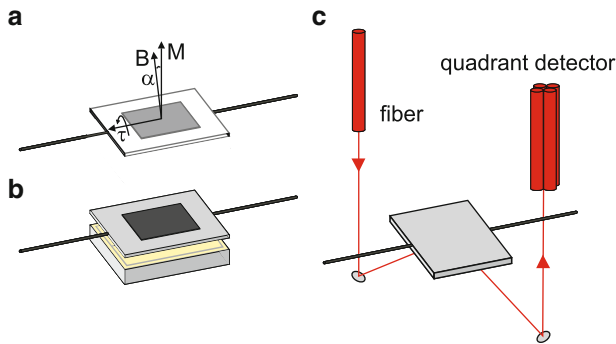


Fig. 10.3 Torsion-balance magnetometers. (a) Principle of operation. (b) Readout via a differential capacitor setup as in the Templeton design [13]. (c) Optical-lever readout as realized in the Maan group [3]

a magnetometer following Wiegers' design but with additional wiring for a gate electrode and transport contacts. The magnetometer performance was comparable (solid cyan line in Fig. 10.1).

Matthews et al. [17] employed a magnetometer with a rotor including the 2DES and a reference sample, both tilted in opposite directions with respect to $\mathbf{B}$ to cancel out the magnetic background of the substrate (dashed cyan line in Fig. 10.1). They realized an in situ adjustment of the stator capacitance electrodes using a piezoelectric stick-slip drive.

Schaapman et al. [3] were the first to use an optical detection scheme for their torsion-balance, sketched in Fig. 10.3c. The light from a fiber-coupled 790 nm laser was collimated by a ball lens and focused on the substrate side of the 2DES. The direction of the reflected light was detected using a quadrant detector consisting of four optical fibers that were monitored by four identical photodiodes (dotted cyan line in Fig. 10.1).

A resonant readout scheme for a torsional oscillator has been reported by Crowell et al. [18] (dashed blue line in Fig. 10.1). Here, the torsional oscillator with thin-film electrodes was micromachined from a Si wafer. The oscillator chip was glued to a substrate containing two thin-film electrodes and a guard ring. Crowell et al. used a standard phase-locked loop to drive the torsional oscillator at its resonance frequency by applying a bias voltage. The shift in resonance frequency due to the additional restoring torque $\Delta\tau = \mathbf{m} \times \mathbf{B}$ was detected using a frequency counter.

10.2.5 Cantilever Magnetometers

An alternative approach to torque magnetometry is the use of micromechanical cantilever magnetometers (MCMs). Here, the sample is attached or incorporated at the free end of a flexible, singly clamped beam. A torque or force acting on the sample is converted into a deflection of the cantilever that can be detected, e.g., capacitively or by interferometric techniques (see Fig. 10.4).

Cantilevers designed using the precision engineers toolbox are used routinely for dHvA measurements on bulk systems. Recent experiments considered "layered" organic metals [20] and unconventional superconductors [21]. The cantilever beams are typically made of thin CuBe plates with a thickness of 20–50 μm and lateral dimensions in the mm range leading to a sensitivity on the order of $10^{-9} - 10^{-12}$ J/T in high magnetic fields (cf. Fig. 10.5a). A reduction of the size of the MCMs leads to a higher sensitivity in absolute units, since the spring constants of the MCMs decrease with the third power of the beam thickness t , the square of the beam length l , and scale linearly with the width w . Downscaling, however, is not necessarily an advantage for measurements on bulk systems, since the sample volume that the sensor can accommodate scales down likewise. For measurements on LDES, however, the magnetic signal is proportional to the number of electrons that scales with the square of the LDES's lateral dimensions, while a linear reduction of sensor

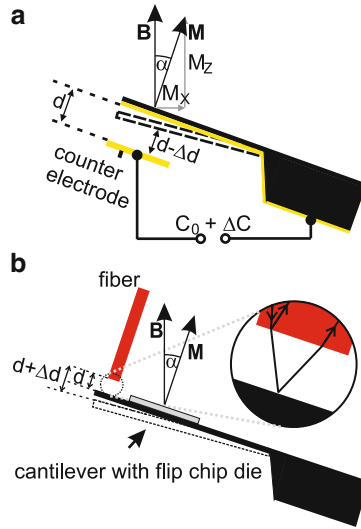


Fig. 10.4 Schematic sideviews. (a) Capacitive deflection readout. The MCM normal $\mathbf{n}$ is tilted by an angle α with respect to $\mathbf{B}$. A torque $\boldsymbol{\tau} = \mathbf{M} \times \mathbf{B}$ is acting on an anisotropic magnetic moment $\mathbf{M} \parallel \mathbf{n}$. The backside of the sensor is metallized, forming a plate capacitor with a fixed counterelectrode on the substrate. (b) Interferometric readout. The MCM deflection is detected using the interference of light reflected back from the Au-coated cantilever surface and the cleaved edge of the fiber, respectively. After [19]

and LDES in all three dimensions leads to a stronger decrease of the spring constant, thereby enhancing the sensitivity.

The CuBe-spring cantilevers can be seen as the starting points of MCMs based on the toolbox of the MEMS (microelectromechanical systems) designers. MEMS-based MCMs are now used for measurements on LDES (Fig. 10.5b–e). Such sensors have been pioneered by M.J. Naughton [24]. The first capacitive MCM used to measure the dHvA effect in a 2DES was developed in the present authors groups by Schwarz et al. [25]. The sensor, shown in Fig. 10.5b is micromachined from a GaAs-based heterostructure that incorporates the 2DES. For the preparation, special etch-stop layers grown by MBE (molecular beam epitaxy) and selective wet etching techniques are used to define the beam thickness with atomic precision. The monolithic design allows for very low spring constants due to the minimized mass of the sample. A capacitive readout as sketched in Fig. 10.4a is employed using an Andeen-Hagerling capacitance bridge. The sensors are calibrated by passing a current through a lithographically defined thin-film coil around the 2DES mesa. Figure 10.5c shows a design that provides a sensitivity which is improved by about one order of magnitude [26] (red line in Fig. 10.1) if compared to CuBe-spring cantilevers. This results from the downscaling (upper left in Fig. 10.5c). The cantilever has a spring constant of 0.06 N/m. The right sensor in Fig. 10.5c contains a separate sample, which is thinned down to $\approx 10 \mu\text{m}$ and attached on top of the beam. This

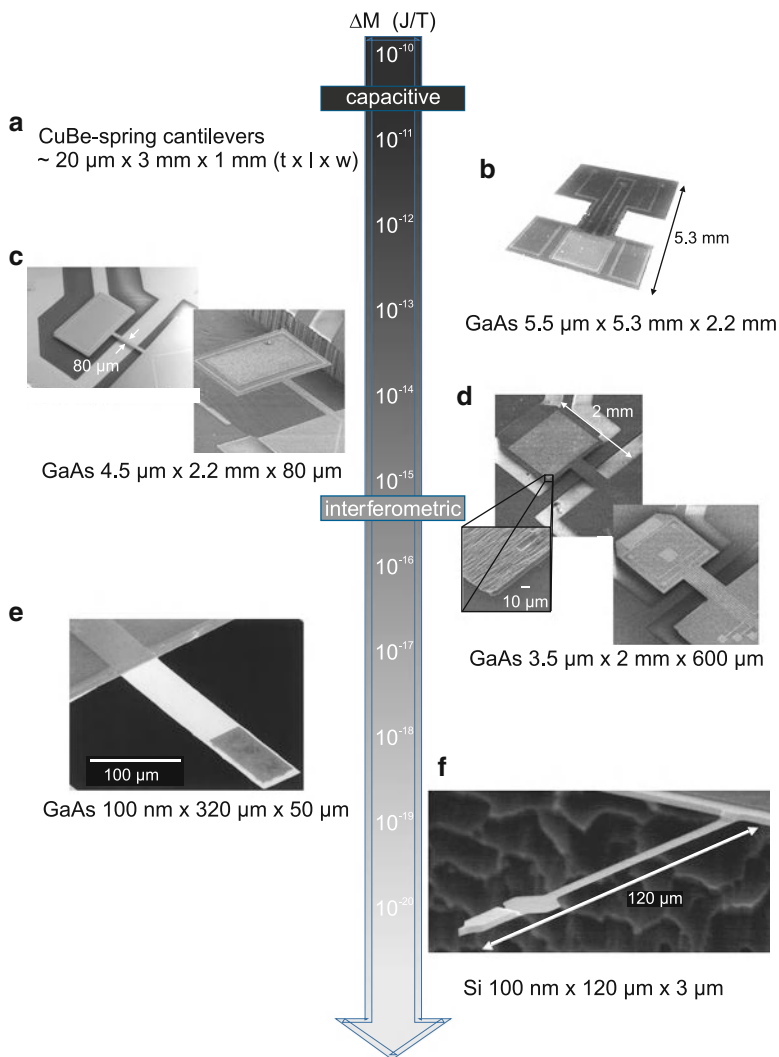


Fig. 10.5 Micromechanical cantilevers. (a) Typical dimensions of CuBe-spring cantilevers used for dHvA measurements on bulk systems. (b) Capacitive MCM successfully employed to measure the dHvA effect in a semiconductor 2DES. (c) Improved design with integrated (left) or separately applied samples (right). (d) Cantilevers for interferometric readout. A micromachined sample chip containing the electron system is mounted in flip-chip configuration on the sensor. Contacts to the thin-film leads on the sensor (lower right image) are made using a conductive-epoxy bonding technique. (e) GaAs microcantilever optimized for resonant interferometric detection. (f) Si cantilever for magnetic resonance force microscopy applications. This sensor design has not yet been used for cantilever magnetometry but demonstrates the potential of the technique in terms of sensitivity. Figures (e) and (f) reprinted with permission from [22] and [23]. Copyright 1999 (2007) by the American Physical Society

fiber-to-cantilever distance. A photograph of the sample head with the piezo tube and the coarse approach mechanism on the basis of a slip-stick drive is shown on the left. The magnetic moment sensitivity is given as magenta line in Fig. 10.1 and corresponds to a displacement resolution of the interferometer of about 8 pm. A second feedback loop operating in a frequency band well above the cutoff frequency of the null-detector loop is used to actively damp the fundamental mode of vibration of the cantilever, a process termed “feedback cooling” in the optomechanics community [23]. Here, a derivative controller is employed to provide a velocity-proportional signal that is fed into the single-turn coil on the cantilever, thereby “cooling” the fundamental vibrational mode.

A resonant readout for MCMs was developed by Harris et al. [29]. The authors employed an interferometer setup similar to that described above. The cantilevers were glued to a piezoelectric actuator and excited at their resonance frequency. The laser intensity incident on the photodiode was thus modulated with the sensor eigenfrequency. The signal was fed back into the piezoelectric actuator using a phase-locked loop, and the frequency was measured using an oven-stabilized frequency counter. In this setup, the frequency shift $\Delta\nu$ due to the additional restoring torque $\Delta\tau = \mathbf{M} \times \mathbf{B}$ is measured. An advantage of the technique is that measurements under arbitrary (average) tilt angles including zero are possible and the sensitivity can be increased. However, the lever motion can induce eddy currents in the sample that can complicate the measurement of the equilibrium magnetization. Harris et al. developed miniaturized MCMs with integrated electron systems shown in Fig. 10.5e [22], where the beam thickness was only 100 nm. The sensors were prepared from GaAs heterostructures containing AlAs sacrificial layers [30]. The sensitivity is shown as an orange line in Fig. 10.1.

In Fig. 10.5f, taken from [23], a state-of-the-art Si cantilever developed for magnetic resonance force microscopy is shown. The sensor has a thickness of 100 nm and a width of only 3 μm . The paddle near the tip is used as a mirror for the interferometric readout. The sensor has a spring constant of 86 $\mu\text{N/m}$. This design has not been used for cantilever magnetometry so far. However, a calculation assuming the current sensitivity for quasi-static interferometric readout of about 8 pm suggests a sensitivity of $\approx 2 \times 10^{-20}$ J/T at $B = 10$ T. Employing a resonant readout, a magnetic moment sensitivity in the 10^{-21} J/T range (corresponding to about seven effective Bohr magnetons in GaAs) seems feasible, thus indicating that magnetization measurements on individual electronic semiconductor nanostructures might soon come within experimental reach.

10.3 Theory of Magnetic Quantum Oscillations

In the following, the theoretical foundations of magnetic quantum oscillations in 2DESs are briefly introduced.

10.3.1 Thermodynamics Definition of Magnetization

The Helmholtz free energy F of a thermodynamic system is given by $F = U - TS$, with the internal energy U , the entropy S , and the temperature T . The differential of the free energy is given by $dF = -SdT - MdB + \chi dN$, where N and χ denote the particle number and the chemical potential, respectively. This yields for the magnetization M of the system

$$M = - \left(\frac{\partial F}{\partial B} \right) \bigg|_{T,N}, \quad \text{and} \quad M = - \left(\frac{\partial U}{\partial B} \right) \bigg|_{T=0,N}. \quad (10.1)$$

The derivative is taken at constant temperature and constant particle number, and the right hand side is valid for $T = 0$. It follows that for $T \rightarrow 0$, the magnetization directly monitors the evolution of the *ground state energy* U of the system with the magnetic field. Thus an experimental determination of M as a function of an externally applied magnetic field B yields direct access to the ground state energy of the system in thermodynamic equilibrium.

For a 2D fermion system with fixed particle number $N = n_s A_s$, with area A_s and sheet density n_s , the chemical potential can be determined numerically from

$$n_s = \int f(E, \chi, T) D(E) dE, \quad (10.2)$$

where $D(E)$ is the density of states (DOS) of the system per unit area and $f(E, \chi, T)$ is the Fermi–Dirac distribution function.

Equation 10.1 allows the numerical calculation of the magnetization of LDESs for a fixed particle number $N = n_s A_s$ and temperature T by evaluating

$$F = \chi N - k_B T A_s \int D(E) \ln \left[1 + \exp \left(\frac{\chi - E}{k_B T} \right) \right] dE. \quad (10.3)$$

10.3.2 DHvA Effect in 2DESs

The zero-field DOS of a spin-degenerate 2DES is given by $D_0(E) = m^*/\pi\hbar^2$ per occupied subband, where $m^* = 0.065m_e$ is the effective mass in GaAs, determined below. The Hamiltonian for noninteracting electrons in a uniform magnetic field $\mathbf{B} = B\mathbf{e}_z$ along the z axis in the Landau gauge $\mathbf{A} = xB_z\mathbf{e}_y$ can be transformed into the equation of a harmonic oscillator using a separation ansatz:

$$\left[-\frac{\hbar^2}{2m^*} \frac{\partial^2}{\partial x^2} + \frac{m^* \omega_c^2}{2} (x - x_0)^2 \right] \phi_x(x) = E_{xy} \phi_x(x). \quad (10.4)$$

Here, $\omega_c = \frac{eB}{m^*}$ is the cyclotron frequency, $x_0 = -\hbar k_y / m^* \omega_c = -k_y l_B^2$ is the guiding center coordinate of a cyclotron orbit, and $l_B = (\hbar / eB)^{1/2}$ is the magnetic length. k_y is the y component of the wavevector $\mathbf{k}$. The corresponding eigenenergies, i.e., the energies of the Landau levels (LLs), are given by

$$E_j = \left(j + \frac{1}{2}\right) \hbar \omega_c, \quad j = 0, 1, 2, \dots \quad (10.5)$$

The energy eigenvalues are degenerate with respect to $\mathbf{k}$. Since in a rectangular sample with edge lengths L_x, L_y and area $A_s = L_x \cdot L_y$ the distance between two guiding centers in the Landau gauge is $\Delta x_0 = \Delta k_y l_B^2 = (2\pi / L_y)(\hbar / eB)$, the number of states with the same energy is $N = L_x / \Delta x_0 = A_s eB / h$. The degeneracy of a LL per unit area is hence $N_L = (eB / h) g_s$, where $g_s = 2$ for a spin degenerate system. For a given carrier density n_s , the filling factor is defined as $\nu = n_s / (eB / h)$.

For the ideal 2DES, χ jumps discontinuously between two adjacent LLs at even ν . The jump in χ crosses an energy gap $\Delta E = \hbar \omega_c = 2\mu_B^* B$ in the single-particle spectrum, with the effective Bohr magneton $\mu_B^* = e\hbar / 2m^*$. The Maxwell relation $(\partial M / \partial \chi)|_B = (\partial N / \partial B)|_\chi$ can be simplified for the case of a 2DES with δ - or box-shaped LLs, where N depends linearly on B according to $N_L = g_s eB / h$ [31]:

$$\frac{\Delta M}{N} = \frac{\Delta \chi}{B}. \quad (10.6)$$

This relation predicts a peak-to-peak dHvA amplitude per electron of an ideal 2DES at zero temperature of $\Delta M / N = \Delta \chi / B = 2\mu_B^*$. In order to achieve a more realistic description of a 2DES, one has to account for the effects of finite temperature and residual disorder in the sample. The disorder leads to a broadening of the ideally δ -peak shaped LLs, and the single-particle energy gaps will be reduced by the level broadening. In the following, we refer to the energy difference extracted from the relation $\Delta E = \Delta M B / N$ as thermodynamic energy gap.

In Fig. 10.7a, the DOS and the chemical potential χ are shown for Gaussian broadened LLs with half-width $\Gamma = 0.3 \text{ meV/T}^{1/2} \times (B[\text{T}])^{1/2}$ at different temperatures. The finite temperature reduces the oscillation amplitude and smears out the sawtooth waveform expected for the ideal system (cf. Fig. 10.7). The corresponding free energy F and magnetization M per electron are depicted in Fig. 10.7c,d, respectively. The period $\Delta(1/B)$ of the oscillations is related to the carrier density n_s according to $\Delta(1/B) = g_s e / h n_s$.

10.4 Experimental Results on 2DESs

In Fig. 10.8a, the experimental magnetization of a 2DES residing in an AlGaAs/GaAs heterostructure is shown for different temperatures. DHvA oscillations at even and odd filling factors ν are resolved. They correspond to the chemical potential jumping between adjacent LLs and between sublevels with opposite spin

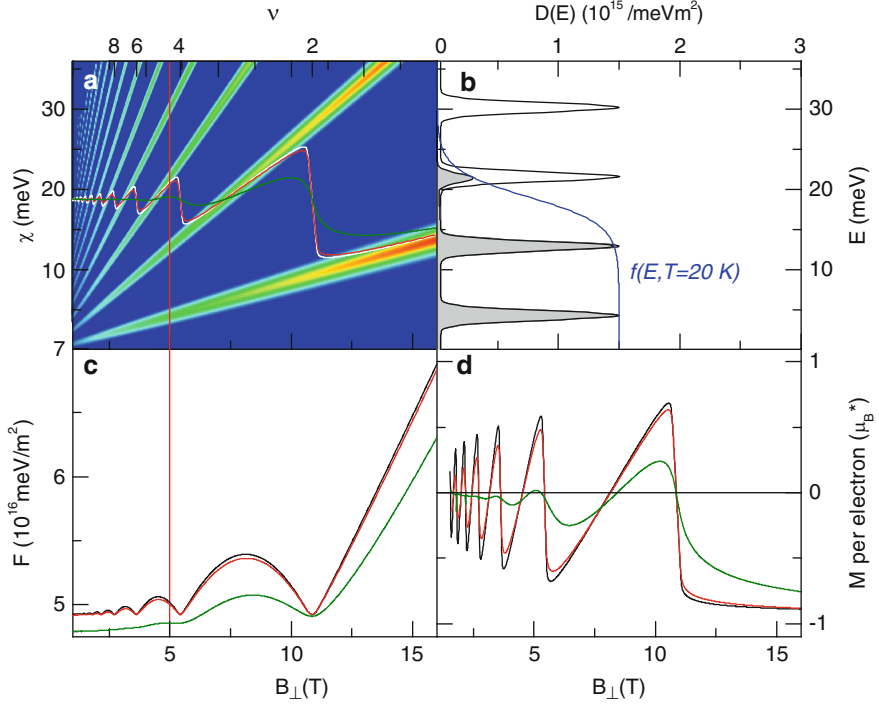


Fig. 10.7 (a) DOS (color plot) assuming Gaussian broadened LLs with $B^{1/2}$ dependence of the broadening parameter Γ . The chemical potential χ is shown for $T = 0.3$ K (white), $T = 2$ K (red), and $T = 20$ K (green). (b) Cut through the DOS at $B = 5$ T. The Fermi distribution function (blue) gives the level occupation. (c) Free energy calculated from the model DOS and χ using (10.3). (d) Magnetization M of the 2DES calculated from (c)

within the same LL, respectively. From the oscillation amplitudes at the lowest temperature, the thermodynamic energy gaps can be recalculated using $\Delta E = \Delta MB/N$. From the temperature dependence of the dHvA amplitude at even ν , shown in Fig. 10.8b, the effective mass is determined to be $m^* = (0.065 \pm 0.001)m_e$ [19]. Solid lines denote the amplitudes from the model calculation outlined in Fig. 10.7d. By comparison with model calculations, a detailed picture of the DOS in a magnetic field can be gained. For a review see [19] and [4].

10.4.1 DOS and Energy Gaps at Even Integer ν

Following Wiegers et al. [31], the finite slope of the dHvA oscillations allows the evaluation of the DOS D_g in the gap *between* Landau levels: in a small interval ΔB around an even integer filling factor ν , the magnetization exhibits a negative linear

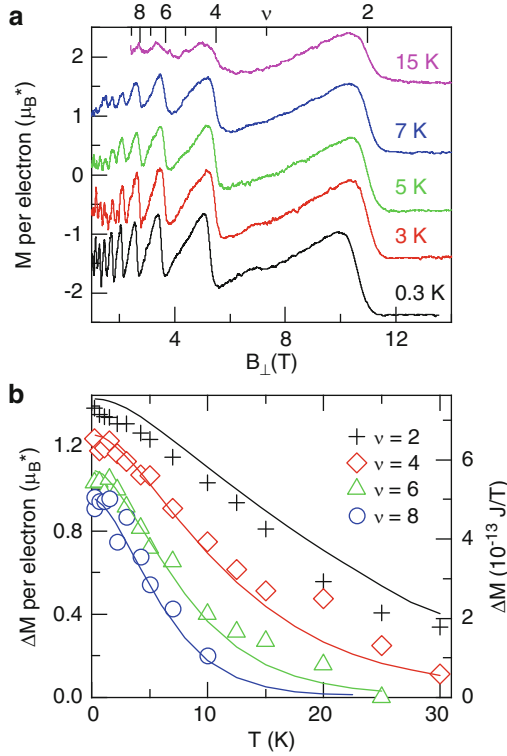


Fig. 10.8 (a) Experimental magnetization for a 2DES in an AlGaAs/GaAs heterostructure. Curves are offset for clarity. Pronounced sawtooth-like oscillations are observed at even ν , where the chemical potential jumps between adjacent LLs. Oscillations at odd ν correspond to spin gaps within LLs. (b) Temperature dependence of the peak-to-peak dHvA amplitude ΔM . Symbols: Experimental values. Lines: result of the model calculations. Excellent agreement is achieved for $\nu \geq 2$. From the T -dependence of the dHvA amplitude, the effective mass is determined to be $m^* = (0.065 \pm 0.001)m_e$. After [32]

slope from the local maximum to the local minimum. We use the ratio $\Delta B/B_\nu = n_g/n_s$, i.e., the relative number of states n_g/n_s between adjacent Landau levels in a 2DES, to estimate the average DOS $D_g = \Delta E/n_g$ between the levels. The value of D_g normalized to D_0 is shown in Fig. 10.9a for samples from three different wafers of AlGaAs/GaAs heterostructures grown by MBE. D_g increases linearly with $1/B_\perp$. Strikingly, there is a sample of highest purity where the dHvA oscillations are indeed discontinuous [34], as predicted in a theoretical model by Peierls 70 years ago [35]. Figure 10.9b shows the corresponding DOS fraction for a sample where n_s could be varied via a gate electrode. Within the experimental error, different ν at the same B exhibit the same D_g , indicating that the DOS in the energy gap D_g is *not* a function of ν , as was speculated earlier [26]. Instead, the DOS between levels might resemble the density of impurity induced states [A.V. Chaplik, Private communication]

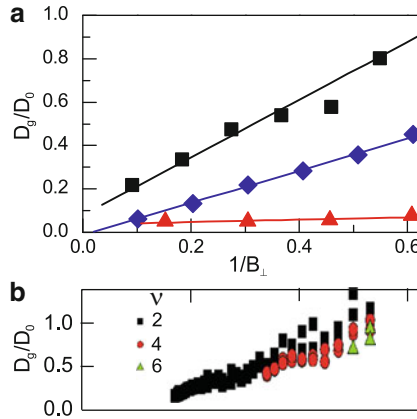


Fig. 10.9 (a) Normalized DOS *between* LLs, D_g/D_0 , for three different AlGaAs/GaAs heterostructures. Here n_s was constant for a given heterostructure and ν was varied by varying B . A linear dependence on $1/B_{\perp}$ is observed. (b) D_g/D_0 for a gated heterostructure. Here, n_s was varied in the measurement. The same linear dependence on $1/B_{\perp}$ is observed. However, for a given B , the DOS between levels is the same within the experimental resolution for different ν . (b) After [33]

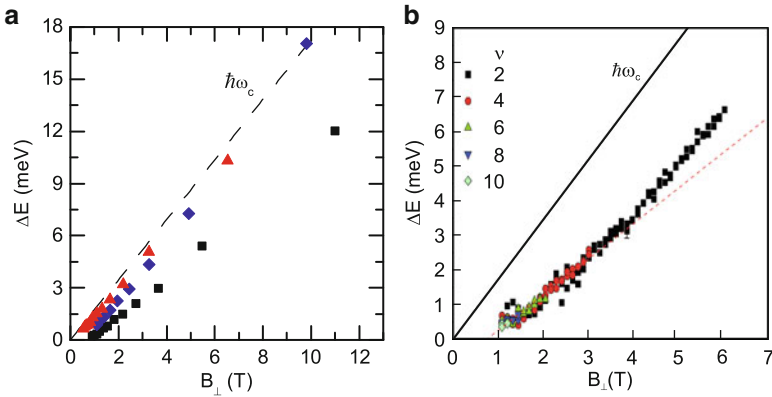


Fig. 10.10 (a) Thermodynamic energy gaps at even ν for three different samples with high (black squares), intermediate (blue diamonds), and very low (red triangles) amounts of disorder. (b) Energy gaps for different even ν in a sample where n_s was varied. The gap size does not depend on ν . (b) After [33]

In Fig. 10.10, the thermodynamic energy gaps at even ν are plotted for (a) the same three samples of different purity and (b) one sample with tunable density. The gap values are systematically smaller than $\hbar\omega_c$ expected for the ideal system (dashed line). We determined the effective mass with high accuracy from the T dependence of ΔM and found that m^* did not vary from sample to sample. Therefore, we attribute the reduced gap values to the different amounts of disorder in the samples, leading to a different LL broadening. As can be seen in Fig. 10.10b, the

gap size does not depend on the filling factor, but on the magnetic field value. These results suggest that in the carrier density regime of Fig. 10.10b, the energy gaps at *even* integer ν are not dominated by electron–electron interaction. Instead, disorder broadening of the LLs governs the observed energy gaps.

10.4.2 Energy Gaps at Odd Integer ν

The thermodynamic energy gaps for spin filling factors $\nu = 1$ –13 are shown in Fig. 10.11a for the highest purity sample, i.e., the sample exhibiting discontinuous dHvA oscillations at high B . The energy gaps increase strongly with increasing B and decreasing ν . The dashed line corresponds to the bare Zeeman energy with $|g| = 0.44$ in GaAs. The effective g -factor g^* evaluated from the measured gap at $\nu = 1$ is $g^* = 7.7$. That is the energy gap is enhanced by a factor of ~ 17 . This is attributed to exchange enhancement of the spin energy gap. In Fig. 10.11b, the experimentally observed gap in the quantum limit at $\nu = 1$ is plotted as a function of B . The gap value shows a *linear* dependence on B . This linear dependence has also been deduced from other experiments on exchange interaction-enhanced energy gaps [37–39] and is in strong contrast to the square-root dependence predicted by a straightforward Hartree–Fock theory. The measured thermodynamic energy gaps between spin and also valley [11, 40] sublevels in 2DESs are strongly dominated by the electron–electron interaction.

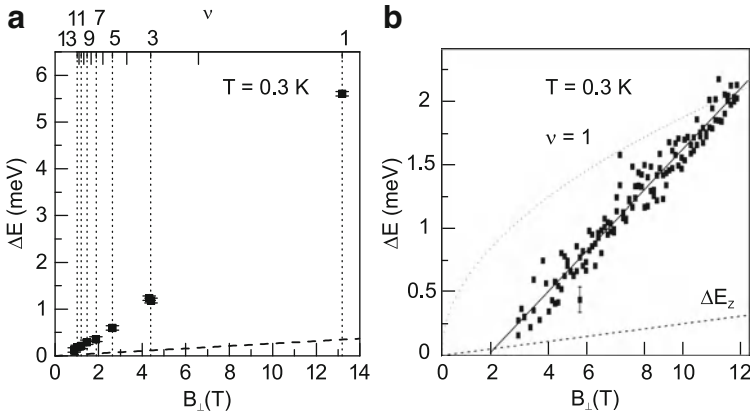


Fig. 10.11 (a) Thermodynamic energy gaps at odd ν ranging from 1 to 13. The gap value increases strongly with increasing B and decreasing ν . The gap value at $\nu = 1$ corresponds to an effective g -factor $g^* = 7.7$. As a dashed line, we show the bare Zeeman energy assuming a GaAs band-structure g -factor of -0.44 . The strong enhancement of the spin gap is attributed to exchange interaction. Adapted from [19] (b) Energy gap at $\nu = 1$ for different densities. A linear dependence of the exchange-enhanced gap value on the magnetic field is observed. This is in strong contrast to the $\sqrt{B}$ -dependence predicted by a straightforward Hartree–Fock theory [36]. Adapted from [27]

10.4.3 Fractional QHE Gaps

The fractional quantum Hall effect is due to correlation effects between electrons in the same Landau level and is thus a pure many-body phenomenon. In Fig. 10.12a, $\partial M/\partial n_s$ measured with the dynamic SQUID technique introduced in Sect. 10.2.2 is plotted versus the carrier density. Pronounced signatures are present at $\nu = 2/3$ and $\nu = 1/3$. Integration of the data yields the magnetization M shown in Fig. 10.12b for $B = 7$ T. A sawtooth-like signal is observed at $\nu = 2/3$ and $\nu = 1/3$, providing direct evidence for a gap in the ground state energy spectrum. The signal amplitude corresponds to $0.32\mu_B^*$ per electron at $\nu = 1/3$, corresponding to a gap value of 1.9 meV at $B = 7$ T. This is in good agreement with the theoretical predictions of Geller and Vignale [42] who obtained $\Delta M \approx 0.56\mu_B^*$ at 7 T and zero temperature neglecting disorder. From excitation spectroscopy, a slightly smaller value 1.2 meV has been found for $\nu = 1/3$ at 10 T [43].

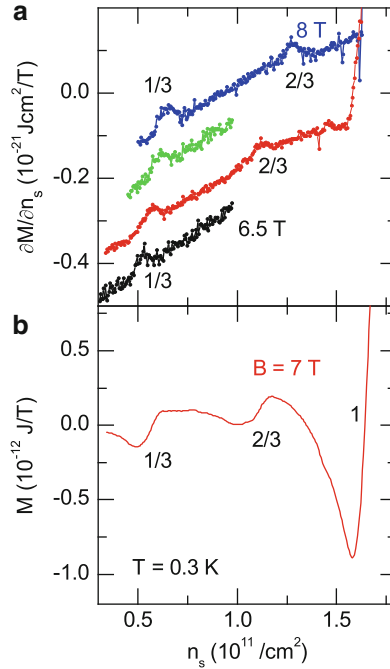


Fig. 10.12 (a) SQUID measurement of $\partial M/\partial n_s$ revealing magnetic signals at fractional filling factors $\nu = \frac{1}{3}$ and $\nu = \frac{2}{3}$. Curves are offset for clarity. (b) Sawtooth-like magnetization of $\nu = \frac{1}{3}$ and $\nu = \frac{2}{3}$ vs n_s for $B = 7$ T, obtained from the red curve in (a) by integration with respect to n_s and subtraction of the magnetic background. This corresponds to an abrupt change in the magnetic field dependent ground state energy at the FQH states. The amplitude corresponds to $\Delta M = 0.32\mu_B^*$ ($0.12\mu_B^*$) per electron for $\nu = \frac{1}{3}$ ($\nu = \frac{2}{3}$), yielding an energy gap $\Delta E = 1.9$ meV (0.7 meV). After [7, 41]

10.5 Magnetization of Nanostructures

Magnetization measurements on 2DESs have proven to be a powerful tool for the investigation of the DOS in quantizing magnetic fields and in particular of the effects of electron–electron interaction on the systems ground state. The direct measurement of M is a noninvasive technique that does not require electrical contacts to the electron system. Magnetization measurements are thus ideally suited for the investigation of laterally confined electron systems such as quantum wires and quantum dots. Even more so than in the case of 2DESs, the inherently low magnetic signal strength of electronic nanostructures has restricted the number of experiments. In the following, we discuss selected magnetization experiments on arrays of quantum wires and quantum dots.

10.5.1 Magnetization of AlGaAs/GaAs Quantum Wires

To investigate the effect that an additional lateral confinement potential has on M , periodic arrays with different electronic wire widths w_e ranging from 160 nm to 380 nm (depending on the width in nm, the wire samples are named $w160$, $w320$, and $w380$ in the following) have been prepared by laser interference lithography and reactive-ion etching starting from an AlGaAs/GaAs heterostructure. The data are compared with a reference 2DES from the same wafer and with model calculations on the basis of a parabolic confinement potential for the quantum wire. Figure 10.13a shows scanning-electron micrographs of a wire array. All data were taken after brief illumination with a red light emitting diode, resulting in a saturation carrier density of $n_s = 5.25 \times 10^{11}/\text{cm}^2$ in the 2DES, and many occupied one-dimensional subbands in the wires. The electronic width $w_e = w_g - 2w_{de}$ was calculated from the geometric width w_g assuming a depletion length of $w_{de} = 120$ nm extracted from further experiments on nanostructured LDESs.

Experimental magnetization data taken at $T = 0.3$ K are shown in Fig. 10.13b. The magnetization of the 2DES exhibits the expected sawtooth-like oscillations discussed above. Two observations with respect to samples $w160$ to $w380$ are striking: first, the coarse shape of the magnetic oscillations in the wire arrays is still sawtooth like and thus similar to the behavior of the 2DES. They are periodic in $1/B$. Second, the absolute signal strengths are of the same order.

It is instructive to compare our data to a 1DES model calculation assuming noninteracting electrons: In the quantum wires, the external potential arises from the interplay of the negative surface charges at the sidewalls and the positively charged ionized donors in the doping layer. Following Riege et al. [44], we derive the external confinement potential for our quantum wires and find it to be well described by a parabolic approximation (e.g., $\hbar\omega_0 = 5.4$ meV for $w160$). In the parabolic approximation, the energy spectrum in a perpendicular magnetic field can be derived analytically: The external harmonic potential $V(\mathbf{r}) = \frac{1}{2}m^*\omega_0^2x^2$ leads to a Hamiltonian that can, as in the 2DES case, be transformed by a separation

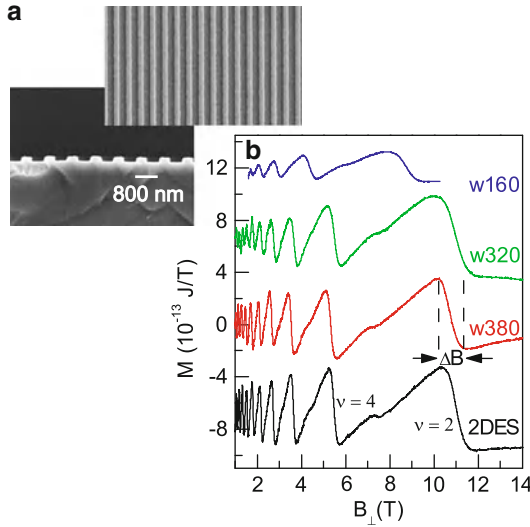


Fig. 10.13 (a) Scanning-electron micrographs of a quantum-wire array. The area covered by the wire array was 1.53 mm^2 . Taking the period $\simeq 1 \mu\text{m}$ the total length of the wires under investigation sum up to 2 m. Assuming a lateral depletion length of 120 nm the electronically active area is estimated to be 20% of the total array area. (b) Experimental magnetization for samples w160–w380 and the reference 2DES, all prepared from the same wafer. The curves are offset for clarity. After [32]

ansatz into the equation of the harmonic oscillator, albeit with oscillator frequency $\omega = \sqrt{\omega_c^2 + \omega_0^2}$, guiding center coordinate $x_0^* = -\hbar k_y \omega_c / m^* \omega^2$ and the effective magnetic mass $m_y^*(B) = m^* \frac{\omega^2}{\omega_0^2}$. The energy eigenvalues are

$$E_j(k_y) = \hbar \omega \left(j + \frac{1}{2} \right) + \frac{\hbar^2 k_y^2}{2m_y^*(B)}, \quad j = 0, 1, 2, \dots, \quad (10.7)$$

and the density of states takes the form

$$D_{1D}(E) = \sum_{j=0}^{\infty} \frac{\sqrt{2m_y^*(B)}}{h} \frac{\Theta(E - E_j)}{\sqrt{E - E_j}}, \quad (10.8)$$

where $\Theta(x)$ is the Heaviside function and j is the subband index. The magnetization is calculated as outlined in Sect. 10.3, where the corresponding 2D quantities have to be replaced by their one-dimensional counterparts.

Figure 10.14a shows the 1D DOS as a color plot for $\hbar \omega_0 = 5.4 \text{ meV}$. Here (10.8) has been convoluted with a Gaussian with $\Gamma = 0.26 \text{ meV/T}^{1/2} \times (B[\text{T}])^{1/2}$ extracted from the reference 2DES to account for level broadening. The red line in

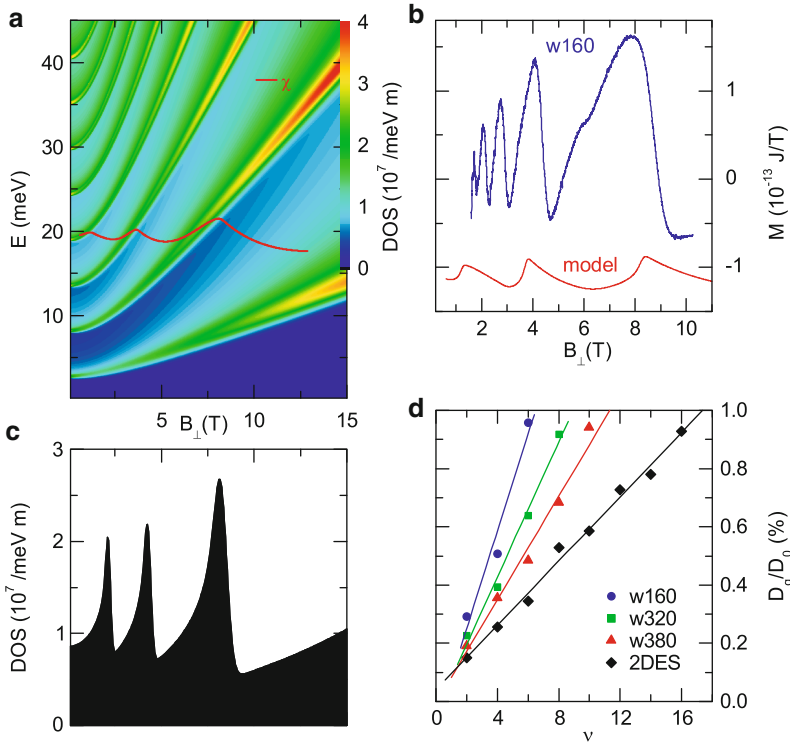


Fig. 10.14 (a) Density of states and chemical potential χ for a 1DES with parabolic lateral confinement. The ideal 1D DOS is convoluted with a Gaussian to qualitatively account for the effects of disorder. The parameters for the calculation were $T = 0.3$ K, $\hbar\omega_0 = 5.4$ meV, and line density $\lambda_1 = 1.4 \times 10^8 \text{ m}^{-1}$. (b) Comparison of the calculated magnetization (red line) with the experimental curve for sample w160 (blue line). (c) cut through the density of states along a line of constant energy highlighting the absence of spectral gaps in the 1D case. (d) Average DOS between the 1D subband edges D_g of the quantum wire samples w160–w380 and the reference 2DES normalized to the zero-field DOS D_0 . A roughly linear increase with filling factor is found for all samples. D_g increases monotonically with increasing lateral confinement potential, i.e., decreasing wire width. After [32]

(a) denotes the chemical potential χ for a 1D carrier density $\lambda_1 = 1.4 \times 10^8 \text{ m}^{-1}$ at $T = 0.3$ K. In (b), the corresponding calculated magnetization M is depicted in red and compared with the experimental result for sample w160 (blue).

The most important outcome of the theory is that the lateral confinement introduces a continuous DOS function without gaps in the spectrum. This is highlighted in Fig. 10.14c where a cut through the calculated DOS along a line of constant energy is shown. Even in the case of $T = 0$ and $\Gamma = 0$ (no disorder broadening), the gapless DOS function provokes oscillations of χ and M that do not exhibit the sharp sawtooth-like shape, but show spike-like maxima and rounded minima. The oscillation amplitude is significantly reduced if compared to the 2DES

case. These conclusions are consistent with calculations in [45–47]. The sharp maxima occur in M whenever a subband with index j is completely depopulated with increasing magnetic field. However, a discontinuity is no longer expected due to the electron states between subsequent subband edges. We observe a completely different shape and, surprisingly, a much larger amplitude ΔM in the experiment. For samples $w320$ and $w160$, the oscillations at even ν in high magnetic field consist of a rounded local maximum and a minimum that is still relatively sharp. The measured oscillation amplitudes are roughly a factor of 4 larger than the theoretical values.

To quantitatively investigate the effect of the lateral patterning, we performed the evaluation of D_g introduced in Sect. 10.4 for all four samples in Fig. 10.13b. The values are summarized in Fig. 10.14d. For each sample, D_g/D_0 increases roughly linearly with ν consistent with the results on 2DESs. Comparing the results for the different wire samples at a given ν , we see that D_g increases strongly with decreasing wire width.

The magnetic oscillations in the quantum wire arrays show three characteristic features: (1) the relative density of states D_g/D_0 is larger than in the 2DES and increases as a function of decreasing wire width, (2) the traces $M(B)$ are sawtooth-like with amplitudes, which (3) are larger than the calculated ones. We believe that the first observation is a clear signature of the 1D characteristics in the magnetization. For the narrowest wires, we find at low ν an increase in D_g/D_0 of more than a factor of two with respect to the widest wires and to the 2DES. This number reflects the larger DOS between subsequent 1DES subband edges due to lateral confinement. We rule out that disorder introduced by the deep-mesa etching is responsible for this by comparison with the results of Raman spectroscopy [48] where no broadening of the resonance lines of single-particle excitations was observed. The increase D_g/D_0 can hence be understood in the framework of noninteracting electrons. This is not possible for the observations (2) and (3). We suggest that they are a consequence of the electron–electron interaction. Using a mean field approximation incorporating many-body effects, Fogler et al. [45] calculated a magnetic behavior qualitatively like that observed in our experiment, i.e., rounded maxima and sharp cusp like minima. The microscopic reason is that in wires with many occupied subbands, the Coulomb interaction partly screens the external potential such that the effective Hartree potential is not parabolic but flattens off in the wire center [49]. As a result, electron states very similar to Landau levels are formed in the bulk part of the wire.

The T dependence of ΔM for a given field position is nearly identical for all wire arrays and for the 2DES as shown exemplarily in Fig. 10.15 for the sample $w320$ and the 2DES. Both datasets exhibit the same temperature dependence, indicating that the underlying energy gap is $\hbar\omega_c$ in both cases. This result is in striking contrast to our results on quantum dots of comparable lateral size [50], as will be discussed below.

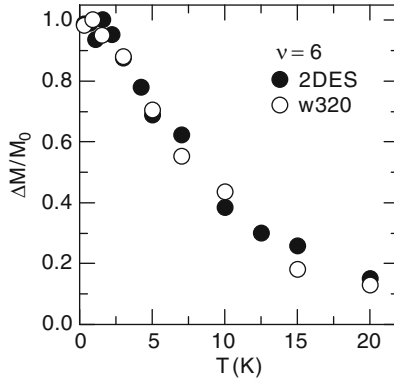


Fig. 10.15 Temperature dependence of the normalized oscillation amplitudes $\Delta M/M_0$ of the quantum wires w320 (solid symbols) and the 2DES at $\nu = 6$ (open symbols). After [32]

10.5.2 Magnetization of AlGaAs/GaAs Quantum Dots

An array of 10^6 quantum dots was integrated into an MCM by patterning a 2DES using laser interference lithography and deep-mesa etching. A scanning-electron micrograph of the etched dots is shown in Fig. 10.16a. They have a circular shape with an average geometric diameter of $2r_g = 550$ nm. Assuming an edge-depletion length of $w_d = 120$ nm, the estimated number of electrons confined to each dot is $N \approx 230$.

The experimental magnetization is shown in Fig. 10.16b. At $T = 0.3$ K, pronounced oscillations with a sawtooth-like shape are observed at $B = 1.20$ T, 6.7 T, and 13.4 T. The peak-to-peak amplitude ΔM normalized to the electron number is a few μ_B^* . Surprisingly, this value is comparable to the dHvA amplitudes per electron of a large area 2DES prepared from the same wafer shown in Fig. 10.16c. The oscillation observed at $B = 1.2$ T depends only weakly on temperature and remains almost unchanged up to $T = 30$ K. The oscillations in the high-field regime, at 6.7 T and 13.4 T, exhibit a strong temperature dependence and have almost vanished at 8 K, which is in strong contrast to the dHvA effect of the 2DES. The temperature dependence of the normalized oscillation amplitude for the quantum dot at 6.7 T and for the 2DES at 4.9 T is compared in Fig. 10.17.

For the detailed interpretation of the data, it is again instructive to model the magnetization of a quantum dot in a single-particle approach. In the following, the energy spectrum of electrons in a magnetic field is discussed for the case of a parabolic and of a hard-wall confining potential $V(r)$, both with a circular symmetry. In the first case, the problem can be solved analytically [52, 53]. In the second case, the energy spectrum has to be computed numerically [54]. We choose cylindrical coordinates (r, ϕ) and the symmetric gauge $\mathbf{A} = Br\mathbf{e}_\phi/2$. In the resulting Schrödinger equation, the variables r and ϕ can be separated by setting $\psi(r, \phi) = 1/\sqrt{2\pi} \exp(il\phi)R(r)$ with the angular momentum quantum number

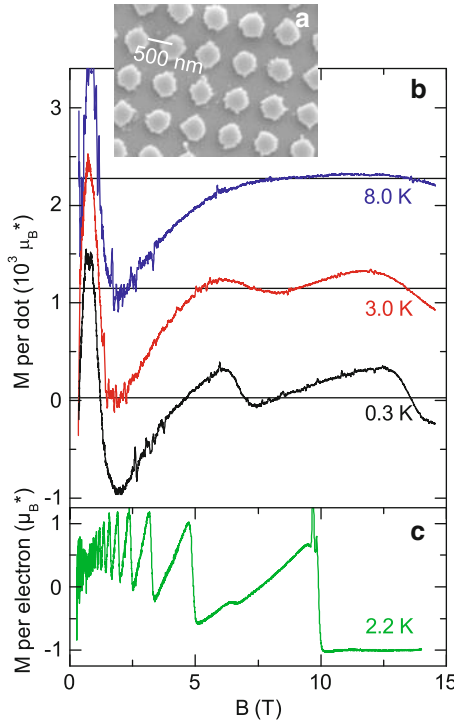


Fig. 10.16 (a) Scanning-electron micrograph of the dot array with a dot diameter of 550 nm. The total area covered by the dot array was $A_t = 1.28 \text{ mm}^2$. Assuming a depletion length of 120 nm, the total effective dot area is estimated to be about 12% of A_t (b) Experimental magnetization of the dot array. Curves recorded at different temperatures are offset for clarity. (c) Experimental magnetization of a large area 2DES from the same wafer after illumination. After [51]

$l = 0, \pm 1, \pm 2, \dots$. For a parabolic potential $V(r) = (1/2)m^*\omega_0^2 r^2 = m^*\omega_0^2 l_B^2 x$, the resulting eigenenergies are the Fock–Darwin levels given by

$$E_{j,l} = \left(j + \frac{|l|}{2} + \frac{1}{2} \right) \hbar\omega + \frac{l}{2} \hbar\omega_c, \quad j = 0, 1, 2, \dots \quad (10.9)$$

with $\omega = \sqrt{\omega_c^2 + 4\omega_0^2}$ [52, 53]. The energy levels calculated from (10.9) for a confinement of $\hbar\omega_0 = 2.3 \text{ meV}$ are plotted in Fig. 10.18a. At zero magnetic field, they form the 2D harmonic oscillator spectrum with levels separated by $\hbar\omega_0$. In the limit of high magnetic fields, the Fock–Darwin levels with a negative angular momentum quantum number l approach the j -th Landau level with energy $(j + 1/2)\hbar\omega_c$ indicated by dotted lines.

For an empty dot, the confinement potential is to a good approximation parabolic [44]. Calculations by Kumar et al. [55] have shown that the self-consistent potential,

Fig. 10.17 Temperature dependence of the normalized oscillation amplitudes of the quantum dots at 6.7 T (*solid symbols*) and of the large-area 2DES at 4.9 T (*open symbols*). The magnetic signal of the quantum dots shows a drastically stronger T dependence. After [51]

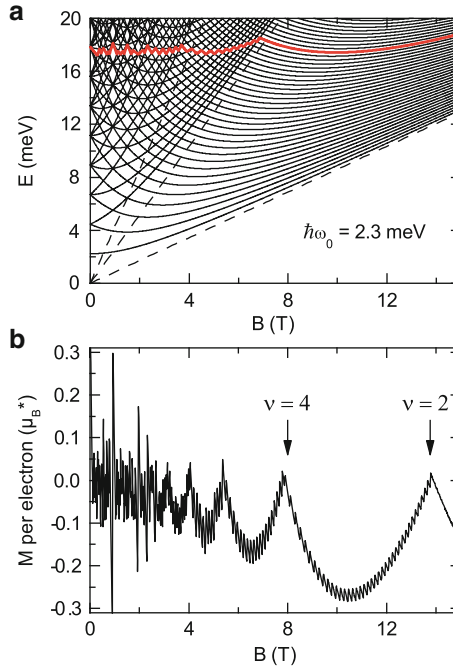
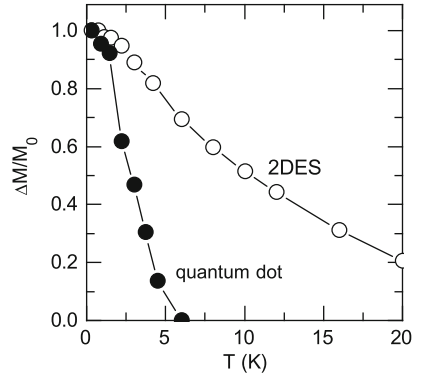


Fig. 10.18 (a) Fock–Darwin energy levels for a parabolically confined dot with $\hbar\omega_0 = 2.3$ meV. The *dashed lines* indicate the Landau level energies with $j = 0, 1, 2$. The *red line* highlights the highest occupied level for 60 spin degenerate electrons in the ground state. (b) Calculated magnetization for a parabolic dot containing $N = 230$ electrons and $\hbar\omega_0 = 3.05$ meV as estimated for the experimental situation. *Arrows* indicate the magnetic field positions corresponding to the $\nu = 4$ and $\nu = 2$ transitions. After [51]

which in a dot with many electrons marks the effective single-particle potential, flattens at the center of the dot, and becomes steeper at the edges if compared to the parabolic case. This suggests that the self-consistent potential for a dot with many electrons can be expected to be hard-wall like rather than parabolic.

A hard-wall confining potential is defined by $V(r) = 0$ for $r \leq R_0$ and $V(r) = \infty$ for $r > R_0$ where R_0 is the radius of the confined electron system. In this case, using the same ansatz as in the parabolic case leads to solutions in the form of the confluent hypergeometric function ${}_1F_1$. The eigenenergies are determined by the boundary condition $\psi(r = R_0, \phi) = 0$, which is equivalent to

$${}_1F_1(-\alpha, \gamma, x_0) = 0, \quad (10.10)$$

with $x_0 = R_0^2/2l_B^2$. When α_{jl} are the values for which condition (10.10) is fulfilled, the energy levels can be expressed as

$$E_{jl} = \left(\alpha_{jl} + \frac{l + |l|}{2} + \frac{1}{2} \right) \hbar \omega_c, \quad (10.11)$$

with $j = 0, 1, \dots$. For a large magnetic field or a large radius, α_{jl} approaches j and the case of an infinite 2DES is recovered. In general, the values α_{jl} have to be determined numerically. In particular, for large absolute values of the angular momentum quantum number $|l|$, the value of α_{jl} will deviate from the integer value given by j . Comparing two adjacent values of l , one finds $\alpha_{j|l|} < \alpha_{j|l|+1}$, which for the energy levels leads to the relation

$$E_{j|l|} < E_{j|l|+1}. \quad (10.12)$$

In Fig. 10.19a, the density of the levels in the (B, E) -plane is depicted as color plot for a confinement with $R_0 = 80$ nm. At low magnetic field, the spectrum is quite complicated. With increasing magnetic field, quasi-degenerate Landau levels emerge. This happens much faster than for a parabolic confinement (c.f. Fig. 10.18a). In this regime, the trace of the highest occupied level energy strongly resembles the oscillations of the chemical potential obtained in Fig. 10.7a for an extended 2DES.

For the calculations assuming a parabolic confinement, the experimental dot potential is approximated by $\hbar \omega_0 = 3.05$ meV, which is in reasonable agreement with values found in far-infrared spectroscopy on similar quantum dots [56]. The numerically calculated magnetization for $N = 230$ in a parabolic quantum dot is shown in Fig. 10.18b. Two types of oscillations can be distinguished: A slowly varying dHvA-type oscillation and a fast inverse sawtooth-like oscillation, superimposed onto the first one. The fast oscillation is due to single-electron transitions from a state with radial quantum number $j + 1$ to a state with j and can be regarded as Aharonov–Bohm (AB) type [57].

The magnetization exhibits an upward cusp whenever all levels approaching a Landau level with index $j + 1$ are depopulated with increasing field. The agreement between experiment and calculation regarding the magnetic-field position, shape, and amplitude of the oscillation is not satisfactory. In particular, the experimentally observed oscillation amplitude is larger by almost one order of magnitude.

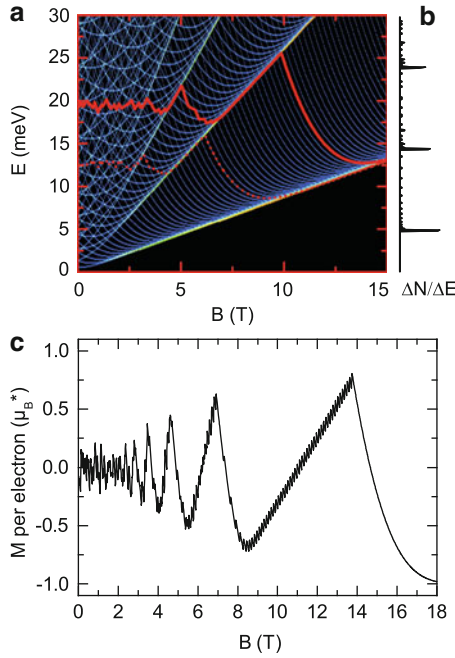


Fig. 10.19 (a) Color plot of the level density $\Delta N/\Delta E$ of an electronic dot confined by a hard wall with radius $R_0 = 80$ nm. With increasing magnetic field, quasi-degenerate Landau levels are formed. Note that the *white lines* are not artificially added to the graph but arise from the high density of states accumulating at energies $E_j = (j + 1/2)\hbar\omega_c$. The *solid (dotted) red line* marks the highest occupied level for $N = 96$ ($N = 60$). (b) Level density profile at $B = 5.5$ T. (c) Magnetization calculated for a dot containing $N = 230$ electrons confined to a cylindrical hard wall with radius $R_0 = 105$ nm. Note that the absolute amplitude per electron is increased by almost one order of magnitude if compared to the parabolic confinement (Fig. 10.18). After [51]

The magnetization calculated for 230 electrons in a hard-wall potential is shown in Fig. 10.19b for $R_0 = 105$ nm. One finds oscillations which have an amplitude that approaches $2\mu_B^*$ per electron with increasing field. The positions of the upward cusps are found at $\nu = 2$ at 13.8 T and $\nu = 4$ at 7 T. These positions are remarkably close to the positions of the experimentally observed oscillations. This outcome suggests that a hard-wall potential can be used to qualitatively simulate the self-consistent potential of a quantum dot with many electrons.

At lower magnetic fields, the calculations and the experiment deviate significantly. In the latter, only one very strong oscillation at $B = 1.2$ T occurs. In the calculation, the dHvA-like signatures appear down to $B = 3$ T. While the faster oscillations visible in the calculation might be smeared out by ensemble averaging in the experiment, there is currently no theory predicting the strong oscillation at $B = 1.2$ T. One may speculate, however, that this feature has a semiclassical origin connected to single-particle level crossings of classical electron trajectories [50].

The AB-type oscillations in the model calculations are not observed in the experiment. However, for an inhomogeneous array of n dots, the total amplitude is predicted to increase as $\sqrt{n}$ instead of n , which for the present case of 10^6 dots, yields a relative reduction by a factor of 1,000 if compared to an perfectly homogeneous array [57].

Finally, we discuss the temperature dependence of the oscillation amplitude in the quantum-dot magnetization. The temperature dependent dHvA data on the large-area 2DES at even filling factor demonstrate that here the damping of the oscillation scales with the cyclotron energy as predicted by the Lifshitz–Kosevich theory [58], that is, the excitation energy of the system is given by $\hbar\omega_c$. For the quantum dots a significantly stronger temperature dependence is observed in Fig. 10.17. We interpret this as a consequence of a small excitation energy.

The importance of the electron–electron interaction to the general properties of quantum dots, has already been pointed out by Maksym and Chakraborty [59]. A couple of theoretical predictions exist on the magnetization of dots containing a small number of interacting electrons [60–62]. On such few-electron quantum dots magnetization measurements have not yet been such successful that it has been possible to experimentally verify the predictions. For quantum dots with a large number of electrons, only few predictions exist regarding their equilibrium properties when interactions are taken into account [45, 63]. Experiments in this regime are feasible as outlined here and help to gain insight into the systems properties. Detailed magnetization data might help to improve the theoretical models. Experimental evidence for the interaction effects in the ground state properties of quantum dots with up to 50 electrons comes from single-electron capacitance spectroscopy [64].

10.6 Conclusions

The developments in the field of highly sensitive magnetometry that form the basis of magnetic investigations on LDES have been reviewed. For future experiments on individual electronic nanostructures, progress in the field of nanoelectromechanical systems (NEMs) is in particular promising. Attonewton force sensitivity has already been demonstrated [65]. However, the applicability of novel sensors and detection schemes for magnetometry in high fields has yet to be proven.

The dHvA effect in 2DES has been shown to be powerful for studying the density of states in quantizing magnetic fields. The results have important consequences for the understanding of the quantum Hall effect. The magnetization provides direct access to thermodynamic energy gaps, i.e., gaps in the ground state energy spectrum, including interaction-induced renormalization. At even integer ν , the results can largely be explained by a single-particle picture including lifetime broadening due to disorder. The spin splitting at odd integer ν is found to be strongly dominated by exchange interaction. Enhancement factors of ~ 17 are found in the quantum limit if compared to the single-particle Zeeman splitting. The experimentally observed linear field dependence of the gap is in contrast to straightforward Hartree–Fock

theory. A more detailed review is given in [19] and [4]. The observation of dHvA-type oscillations at fractional quantum Hall states is a direct proof for a many-body correlated ground state and yields access to the thermodynamic energy gap.

Magnetization experiments on quantum wires and quantum dots reveal a strong influence of the lateral confinement on the ground state energy spectrum. In case of the wires, electron–electron interaction dominates the magnetization in high magnetic fields in the sense that self-consistent screening of the external potential restores the sawtooth-like dHvA effect. In the quantum dots, the magnetic signal differs strongly from the 2D behavior even in the highest accessible fields of $B = 16$ T. The very strong temperature dependence of the magnetic oscillations in high fields together with their high amplitude of $\sim 2\mu_B^*$ per electron can only be interpreted as a consequence of a strongly interaction-renormalized gaps in the ground state energy spectrum.

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Chapter 11

Spin Polarized Transport and Spin Relaxation in Quantum Wires

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Abstract We give an introduction to spin dynamics in quantum wires. After a review of spin-orbit coupling (SOC) mechanisms in semiconductors, the spin diffusion equation with SOC is introduced. We discuss the particular conditions in which solutions of the spin diffusion equation with vanishing spin relaxation rates exist, where the spin density forms persistent spin helices. We give an overview of spin relaxation mechanisms, with particular emphasis on the motional narrowing mechanism in disordered conductors, the D'yakonov–Perel' spin relaxation. The solution of the spin diffusion equation in quantum wires shows that the spin relaxation becomes diminished when reducing the wire width below the spin precession length L_{SO} . This corresponds to an effective alignment of the spin-orbit field in quantum wires and the formation of persistent spin helices whose form as well as amplitude is a measure of the particular SOC's, the linear Rashba and the linear Dresselhaus coupling. Cubic Dresselhaus coupling is found to yield in diffusive wires an undiminished contribution to the spin relaxation rate, however. We discuss recent experimental results which confirm the reduction of the spin relaxation rate. We next review theoretical proposals for creating spin-polarized currents in a T-shape structure with Rashba-SOC. For relatively small SOC, high spin polarization can be obtained. However, the corresponding conductance has been found to be small. Due to the self-duality of the scattering matrix for a system with spin-orbit interaction, no spin polarization of the current can be obtained for single-channel transport in two-terminal devices. Therefore, one has to consider at least a conductor with three terminals. We review results showing that the amplitude of the spin polarization becomes large if the SOC is sufficiently strong. We argue that the predicted effect should be experimentally accessible in InAs. For a possible experimental realization of InAs spin filters, see [1].

11.1 Introduction

Spin-dependent electronic transport is attracting considerable attention because of possible applications to spintronics. Many of the proposals for two-dimensional (2D) spintronic devices are based on the presence of spin-orbit coupling (SOC)

in the 2D electron system (2DES) semiconductor heterostructure. In III–V semiconductors, the inversion asymmetry of the zinc-blende structure results in the Dresselhaus-spin-orbit-coupling. The effective electric field, originating from the asymmetry of the potential confining the 2DES, results in the Rashba-SOC. Since the strength of the Rashba-SOC can be controlled via external gates, 2DESs have become most promising for spintronic applications. In order to realize such devices, one needs to induce spin polarized electrons in the 2DES. One can generate spin polarized electrons by injecting a current with ferromagnetic metallic leads into the 2DES. However, it has been found that in practice the efficiency of such spin injection is poor because of the conductivity mismatch. Therefore, the direct generation of spin polarized electrons via SOC is favorable. Here, we review recent theoretical progress on spin polarization and spin relaxation in quantum wires with spin-orbit interaction. We give an introduction to spin dynamics and review the SOC mechanisms in semiconductors. The spin diffusion equation with SOC is reviewed. In particular, the existence of persistent spin helix solutions with vanishing spin relaxation rates is shown. We give an overview of all spin relaxation mechanisms, with particular emphasis on the motional narrowing mechanism in disordered conductors, the D'yakonov–Perel' spin relaxation (DPS). We then present solutions of the spin diffusion equation in quantum wires, and show that there is an effective alignment of the spin-orbit field in wires whose width is smaller than the spin precession length L_{SO} , resulting in the reduction of the spin relaxation rate. This can be measured optically or by a change in the sign of the quantum corrections to the conductivity. This effect is very favorable for spintronic applications, since the itinerant electron spin keeps precessing on the length scale L_{SO} , while the spin relaxation is suppressed for wires with width smaller than L_{SO} , which can exceed several μm . We review recent experimental results which confirm the decrease of the spin relaxation rate in wires whose width is smaller than L_{SO} . We then review results on creating spin polarized currents in a T-shape structure with Rashba-SOC. For relatively small SOC, high spin polarization can be obtained. However, the corresponding conductance has been found to be small. Due to the self-duality of scattering matrix for the system with spin-orbit interaction, no spin polarization of the current can be obtained for single-channel transport in two-terminal devices. Therefore, one has to consider at least a conductor with three terminals. Also, one can make use of the fact that the D'yakonov–Perel' spin relaxation is suppressed already in wires with many channels. We review results showing that the amplitude of the spin polarization becomes large if the SOC is sufficiently strong. We argue that the predicted effect should be experimentally accessible in InAs.

11.2 Spin-Dynamics in Semiconductor Quantum Wires

11.2.1 Spin-Orbit Interaction in Semiconductors

In semiconductors with broken inversion symmetry like the III–V-semiconductors GaAs, InAs, or InSb, this bulk inversion asymmetry (BIA) results in SOC, the

Dresselhaus-spin-orbit-coupling (DSOC), which is anisotropic in the electron momentum $\mathbf{k}$ as given by [2],

$$H_D = \gamma_D [\sigma_x k_x (k_y^2 - k_z^2) + \sigma_y k_y (k_z^2 - k_x^2) + \sigma_z k_z (k_x^2 - k_y^2)], \quad (11.1)$$

where γ_D is the Dresselhaus-spin-orbit coefficient. We set $\hbar = 1$ here, and in the following. The confinement of electrons in quantum wells of width a on the order of the Fermi wave length λ_F yields then a spin-orbit interaction which depends strongly on growth direction. Taking the expectation value of (11.1) in the direction normal to the plane grown in [001] direction, one finds with $\langle k_z \rangle = \langle k_z^3 \rangle = 0$, [2]

$$H_{D[001]} = \alpha_1 (-\sigma_x k_x + \sigma_y k_y) + \gamma_D (\sigma_x k_x k_y^2 - \sigma_y k_y k_x^2), \quad (11.2)$$

with the linear Dresselhaus parameter $\alpha_1 = \gamma_D \langle k_z^2 \rangle$. For narrow quantum wells, where $\langle k_z^2 \rangle \sim 1/a^2 \geq k_F^2$, the linear terms exceeds the cubic ones. A special situation arises for quantum wells grown in the [110] direction, where the spin-orbit field is pointing normal to the quantum well, as shown in Fig. 11.1, so that an electron whose spin is initially polarized along the normal of the plane, remains polarized as it moves in the quantum well.

In quantum wells with asymmetric electrical confinement, the inversion symmetry perpendicular to the quantum well is broken. This structural inversion asymmetry (SIA) can be deliberately modified by changing the confinement potential with a gate voltage. The resulting SOC, the SIA coupling, or Rashba-SOC (RSOC) [3] is given by

$$H_R = \alpha_2 (\sigma_x k_y - \sigma_y k_x), \quad (11.3)$$

where α_2 depends on the asymmetry of the confinement potential $V(z)$ in the direction z , the growth direction of the quantum well, and can thus be deliberately changed by the application of a gate potential. At first glance, the expectation value of the electrical field $\mathcal{E}_c = -\partial_z V(z)$ seems to vanish in the symmetric ground state of the quantum well. The coupling to the valence band [4, 5], the discontinuities in the effective mass [6], and corrections due to the coupling to odd excited states [7] yield, however, a sizable coupling parameter which depends, albeit in a non-trivial way, on the asymmetry of the confinement potential [5, 8]. This dependence allows one, in principle, to control the electron spin with a gate potential, which can therefore be used as the basis of a spin transistor [9].

All SOC's can be combined in the form of an effective Zeeman term

$$H_{SO} = -\gamma_g \mathbf{s} \mathbf{B}_{SO}(\mathbf{k}), \quad (11.4)$$

where the spin vector is $\mathbf{s} = \boldsymbol{\sigma}/2$ and γ_g is the gyromagnetic ratio. However, the spin-orbit field $\mathbf{B}_{SO}(\mathbf{k})$ is antisymmetric, $\mathbf{B}_{SO}(-\mathbf{k}) = -\mathbf{B}_{SO}(\mathbf{k})$ under the time reversal operation, so that it does not break time reversal symmetry as the spin changes sign as well under the time reversal operation, $\mathbf{s} \rightarrow -\mathbf{s}$. The electron spin

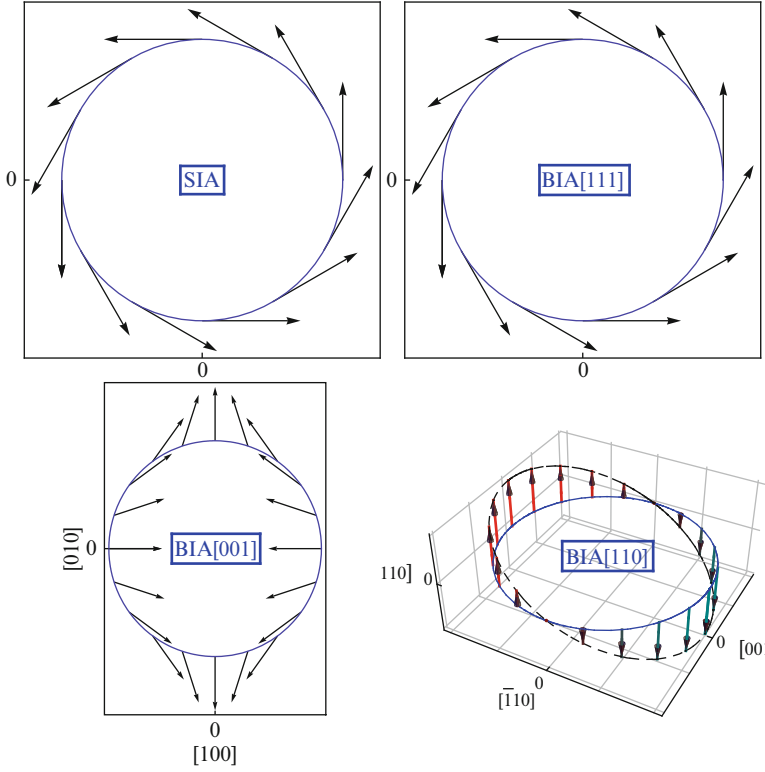


Fig. 11.1 The spin-orbit vector fields for linear bulk inversion asymmetry (BIA) SOC for quantum wells grown in [001], [110], and [111] direction, and for linear structure inversion asymmetry (Rashba) coupling, respectively

operator $\hat{\mathbf{s}}$ is for fixed electron momentum $\mathbf{k}$ governed by the Bloch equations with the spin-orbit field,

$$\frac{\partial \hat{\mathbf{s}}}{\partial t} = \gamma_g \hat{\mathbf{s}} \times (\mathbf{B} + \mathbf{B}_{\text{SO}}(\mathbf{k})) - \frac{1}{\hat{\tau}_s} \hat{\mathbf{s}}. \quad (11.5)$$

We set $\gamma_g = 1$ in the following. The spin relaxation tensor $\hat{\tau}_s$ is in the presence of spin-orbit interaction not necessarily diagonal. If it is diagonal, $\tau_{sxx} = \tau_{syy} = \tau_2$, is the spin dephasing time, and $\tau_{szz} = \tau_1$ the spin relaxation time.

In narrow quantum wells where the cubic DSOC is weak compared to the linear SOC, the spin-orbit field is thus given by

$$\mathbf{B}_{\text{SO}}(\mathbf{k}) = -2 \begin{pmatrix} -\alpha_1 k_x + \alpha_2 k_y \\ \alpha_1 k_y - \alpha_2 k_x \\ 0 \end{pmatrix}. \quad (11.6)$$

Thus, both its amplitude $|\mathbf{B}_{\text{SO}}(\mathbf{k})| = 2\sqrt{(\alpha_1^2 + \alpha_2^2)k^2 - 4\alpha_1\alpha_2k_xk_y}$ and direction change as the direction of the momentum $\mathbf{k}$ is changed. Accordingly, the energy dispersion is anisotropic as given by

$$E_{\pm} = \frac{1}{2m^*}k^2 \pm \alpha k \sqrt{1 - 4\frac{\alpha_1\alpha_2}{\alpha^2} \cos \theta \sin \theta}, \quad (11.7)$$

where $k = |\mathbf{k}|$, $\alpha = \sqrt{\alpha_1^2 + \alpha_2^2}$, and $k_x = k \cos \theta$.

When an electron is initially injected with energy E along the $[100]$ direction, its wave function becomes a superposition of plain waves with the positive momenta $k_{\pm} = \mp \alpha m^* + m^*(\alpha^2 + 2E/m^*)^{1/2}$. The momentum difference $k_- - k_+ = 2m^*\alpha$ causes a rotation of the electron eigenstate in the spin subspace. When at $x = 0$, the electron spin was polarized up spin, with the Eigenvector $\psi(x = 0) = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$, then in a distance x , it rotated the spin as described by the Eigenvector

$$\psi(x) = \frac{1}{2} \left(\frac{1}{\frac{\alpha_1 + i\alpha_2}{\alpha}} \right) e^{ik_+x} + \frac{1}{2} \left(\frac{1}{-\frac{\alpha_1 + i\alpha_2}{\alpha}} \right) e^{ik_-x}. \quad (11.8)$$

In Fig. 11.2, we plot the corresponding spin density for pure RSOC, $\alpha_1 = 0$. The spin points again in the initial direction, when the phase difference between the plain waves is 2π , which gives the condition $2\pi = (k_- - k_+)L_{\text{SO}}$. Thus, when the electron is moving in $[100]$ direction, for linear SOC's,

$$L_{\text{SO}} = \pi/m^*\alpha. \quad (11.9)$$

We note that the period of spin precession changes with the direction of the electron momentum since the spin-orbit field, (11.6), is anisotropic.

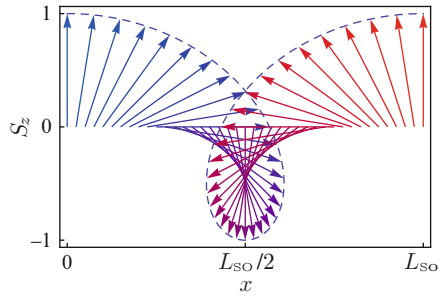


Fig. 11.2 Precession of a spin injected at $x = 0$, polarized in z direction, as it moves by one spin precession length $L_{\text{SO}} = \pi/m^*\alpha$ through the wire with linear Rashba SOC α_2

11.2.2 Spin Diffusion

Translational invariance is broken by the presence of disorder because of impurities and lattice imperfections in the conductor. As the electrons scatter from the disorder potential elastically, their momentum changes in a stochastic way, resulting in diffusive motion. That results in a change of the local electron density $\rho(\mathbf{r}, t) = \sum_{\sigma=\pm} |\psi_{\sigma}(\mathbf{r}, t)|^2$, where $\sigma = \pm$ denotes the orientation of the electron spin, and $\psi_{\sigma}(\mathbf{r}, t)$ is the position and time dependent electron wave function amplitude. On length scales exceeding the elastic mean free path l_e , it is governed by the diffusion equation $\partial_t \rho = D_e \nabla^2 \rho$, where the diffusion constant D_e is related to the elastic scattering time τ by $D_e = v_F^2 \tau / d_D$, where v_F is the Fermi velocity, and d_D the Diffusion dimension of the electron system. On average the variance of the distance, the electron moves after time t is $\langle (\mathbf{r} - \mathbf{r}_0)^2 \rangle = 2d_D D_e t$, yielding the diffusion length at time t , $L_D(t) = \sqrt{D_e t}$. We can write the density as $\rho = \langle \psi^\dagger(\mathbf{r}, t) \psi(\mathbf{r}, t) \rangle$, where $\psi^\dagger = (\psi_+^\dagger, \psi_-^\dagger)$ is the two-component vector of the up (+), and down (−) spin fermionic creation operators, and ψ the 2-component vector of annihilation operators, respectively, $\langle \dots \rangle$ denotes the expectation value. Accordingly, a diffusion equation governs also the spin density $\mathbf{s}(\mathbf{r}, t)$, which is defined by

$$\mathbf{s}(\mathbf{r}, t) = \frac{1}{2} \langle \psi^\dagger(\mathbf{r}, t) \boldsymbol{\sigma} \psi(\mathbf{r}, t) \rangle, \quad (11.10)$$

where $\boldsymbol{\sigma}$ is the vector of Pauli matrices, $\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$, $\sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$, and $\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$, the z component of the spin density being naturally half the difference between the density of spin up and down electrons, $s_z = (\rho_+ - \rho_-)/2$, which is the local spin polarization of the electron system.

Scattering by imperfections changes the electron momentum, and thereby the direction of the spin-orbit field $\mathbf{B}_{SO}(\mathbf{k})$ as the electron moves through the sample. Thereby, the electron spin direction is randomized, the spin precession dephases, and the spin polarization relaxes. Also the spin precession term is modified from the ballistic Bloch-like equation, (11.5), as the momentum $\mathbf{k}$ changes randomly, The spin diffusion equation, which can be derived semiclassically [10, 11], by diagrammatic expansion [12], or by using simple and intuitive random walk arguments as detailed in [13], is given by

$$\frac{\partial \mathbf{s}}{\partial t} = -\mathbf{B} \times \mathbf{s} + D_e \nabla^2 \mathbf{s} + 2\tau \langle (\nabla \mathbf{v}_F) \mathbf{B}_{SO}(\mathbf{p}) \rangle \times \mathbf{s} - \frac{1}{\tau_s} \mathbf{s}, \quad (11.11)$$

where $\langle \dots \rangle$ denotes the average over the Fermi surface. Spin polarized electrons injected into the sample spread diffusively, and their spin polarization, while spreading diffusively as well, decays in amplitude exponentially in time. Since, between scattering events, the spins precess around the spin-orbit fields, one expects also an oscillation of the polarization amplitude in space. One can find the spatial

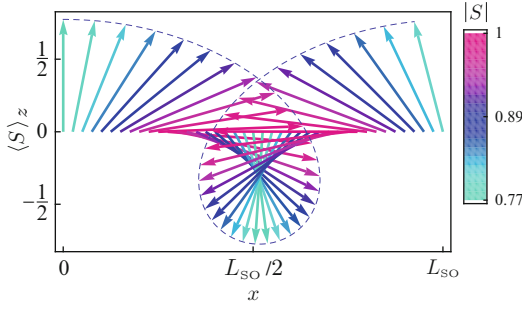


Fig. 11.3 The spin density for linear Rashba coupling which is a solution of the spin diffusion equation with the relaxation rate $1/\tau_s = 7/16\tau_{s0}$. Note that, compared to the ballistic spin density, Fig. 11.2, the period is slightly enhanced by a factor $4/\sqrt{15}$. Also, the amplitude of the spin density changes with the position x , in contrast to the ballistic case. The color is changing in proportion to the spin density amplitude

distribution of the spin density which is the solution of (11.11) with the smallest decay rate Γ_s . As an example, the solution of (11.11) is in 2D for linear Rashba coupling, [11]

$$\mathbf{s}(\mathbf{x}, t) = (\hat{e}_q \cos \mathbf{q}\mathbf{x} + A\hat{e}_z \sin \mathbf{q}\mathbf{x}) e^{-t/\tau_s}, \quad (11.12)$$

with $1/\tau_s = 7/16\tau_{s0}$ where $1/\tau_{s0} = 2\tau k_F^2 \alpha_2^2$ and where the momentum $\mathbf{q}$ is fixed by $D_e q^2 = 15/16\tau_{s0}$, $A = 3/\sqrt{15}$, and $\hat{e}_q = \mathbf{q}/q$. In Fig. 11.3, we plot the linearly independent solution obtained by interchanging cos and sin in (11.12), with the spin pointing initially in z direction, and $\hat{e}_q = \hat{e}_x$. The period of precession is enhanced by the factor $4/\sqrt{15}$ in the diffusive wire, and the amplitude of the spin density is modulated, between one 1 and $A = 3/\sqrt{15}$.

In analogy to the density diffusion current, one can define for the spin components s_i with $i = x, y, z$, spin diffusion currents as

$$\mathbf{j}_{s_i} = -\tau \langle \mathbf{v}_F (\mathbf{B}_{SO}(\mathbf{k}) \times \mathbf{s})_i \rangle - D_e \nabla s_i. \quad (11.13)$$

Thereby, we get the spin continuity equation

$$\frac{\partial s_i}{\partial t} = -D_e \nabla \mathbf{j}_{s_i} + \tau \langle \nabla \mathbf{v}_F (\mathbf{B}_{SO}(\mathbf{k}) \times \mathbf{s})_i \rangle - \frac{1}{\tau_{sij}} \mathbf{s}_j. \quad (11.14)$$

There are two additional terms due to the SOC. The last one is the spin relaxation tensor which will be considered in detail in the next section. The other term arises from the spin precession. This has important physical consequences, resulting in the suppression of the spin relaxation rate in quantum wires and quantum dots as soon as their lateral extension is smaller than the spin precession length L_{SO} , as we will see below.

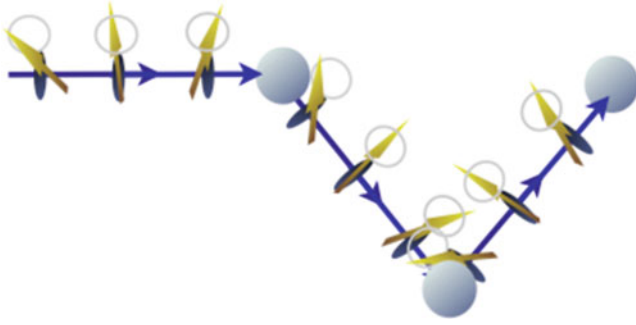


Fig. 11.4 Elastic scattering from impurities changes the direction of the spin-orbit field around which the electron spin is precessing

11.2.3 Spin Relaxation Mechanisms

The intrinsic SOC itself causes the spin of the electrons to precess coherently as the electrons move through a conductor, defining the spin precession length L_{SO} , (11.9). Since impurities in the conductor randomize the electron momentum, the impurity scattering is transferred into a randomization of the electron spin by the spin-orbit interaction, which thereby results in spin dephasing and spin relaxation. This *D'yakonov–Perel' spin relaxation* (DPS) can be understood qualitatively in the following way: The spin-orbit field $\mathbf{B}_{\text{SO}}(\mathbf{k})$ changes its direction randomly after each elastic scattering event from an impurity, after a time of about the elastic scattering time τ as sketched in Fig. 11.4.

Thus, the spin has the time τ to perform a precession around the present direction of the spin-orbit field and can change its direction by an angle $\mathbf{B}_{\text{SO}}\tau$. After a time t and $N_t = t/\tau$ scattering events, the spin will therefore change its angle by $|\mathbf{B}_{\text{SO}}|\tau\sqrt{N_t} = |\mathbf{B}_{\text{SO}}|\sqrt{\tau t}$. The spin relaxation time τ_s is the time by which the spin direction has changed by an angle of order one. Thus, $1/\tau_s \sim \tau\langle\mathbf{B}_{\text{SO}}(\mathbf{k})^2\rangle$, where the angular brackets denote integration over all angles. Remarkably, this spin relaxation rate becomes smaller, the more scattering events take place, because the smaller the elastic scattering time τ is, the less time the spin has to change its direction by precession. Such a behavior is also well known as *motional narrowing* of magnetic resonance lines [14]. The spin relaxation length, L_s , is given by $L_s = \sqrt{D_e\tau_s}$. A more rigorous derivation for the kinetic equation of the spin density matrix yields also nondiagonal elements of the spin relaxation tensor [15],

$$\frac{1}{\tau_{sij}} = \tau \left(\langle \mathbf{B}_{\text{SO}}(\mathbf{k})^2 \rangle \delta_{ij} - \langle B_{\text{SO}}(\mathbf{k})_i B_{\text{SO}}(\mathbf{k})_j \rangle \right). \quad (11.15)$$

These nondiagonal terms correspond to interference terms and can result in a reduction of the spin relaxation. As an example, we consider a narrow quantum well grown in [001] where the linear SOC's are dominating. The energy dispersion is

anisotropic, as given by (11.7), and the spin-orbit field $\mathbf{B}_{\text{SO}}(\mathbf{k})$ changes its direction and its amplitude with the direction of the momentum $\mathbf{k}$: Diagonalizing the spin relaxation tensor, one finds three eigenvalues $\frac{1}{\tau_s}(\alpha_1 \pm \alpha_2)^2/\alpha^2$ and $\frac{2}{\tau_s}$ where $\alpha^2 = \alpha_1^2 + \alpha_2^2$, and $\frac{1}{\tau_s} = 2k^2\tau\alpha^2$. One of these eigenvalues vanishes when $\alpha_1 = \alpha_2 = \alpha_0$. In that case, the spin-orbit field does not change its direction with the momentum:

$$\mathbf{B}_{\text{SO}}(\mathbf{k})|_{\alpha_1=\alpha_2=\alpha_0} = 2\alpha_0(k_x - k_y) \begin{pmatrix} 1 \\ 1 \\ 0 \end{pmatrix}, \quad (11.16)$$

The spin density $\mathbf{S} = S_0(1, 1, 0)$ does not decay in time, since its vector is parallel to $\mathbf{B}_{\text{SO}}(\mathbf{k})$, (11.16) [16]. It turns out, that there are two more modes which do not decay in time for $\alpha_1 = \alpha_2$ [12]. These modes are inhomogeneous in space, and correspond to precessing spin densities, called *persistent spin helix* [17, 18] with the period L_{SO} . We can get these *persistent spin helix modes* analytically by solving the spin diffusion equation (11.11) with the spin relaxation tensor given by (11.15) [12, 13].

The momentum scattering can also be due to *electron–phonon or electron–electron scatterings* [19–22], yielding the total scattering rate as defined by $1/\tau = 1/\tau_0 + 1/\tau_{\text{ee}} + 1/\tau_{\text{ep}}$, where $1/\tau_0$ is the elastic scattering rate. In degenerate semiconductors and metals, the electron–electron scattering rate is the Fermi liquid inelastic electron scattering rate $1/\tau_{\text{ee}} \sim T^2/\epsilon_F$. The electron–phonon scattering time $1/\tau_{\text{ep}} \sim T^5$ decays faster with temperature. Thus, at low temperatures the DP spin relaxation is dominated by τ_0 .

Since the SOC mixes spin Eigenstates a nonmagnetic impurity potential V can thus change the electron spin, which results in another source of spin relaxation [23, 24], the *Elliott–Yafet spin relaxation*. For degenerate III–V semiconductors, one finds [25, 26]

$$\frac{1}{\tau_s} \sim \frac{\Delta_{\text{SO}}^2}{(E_G + \Delta_{\text{SO}})^2} \frac{E_k^2}{E_G^2} \frac{1}{\tau}, \quad (11.17)$$

where E_G is the gap between the valence and the conduction band of the semiconductor, and Δ_{SO} is the spin-orbit splitting of the valence band. Thus, the EYS can be distinguished, being proportional to $1/\tau$, and thereby to the resistivity, in contrast to the DP spin scattering rate, (11.15), which is proportional to the conductivity. Since the EYS decays in proportion to the inverse of the band gap, it is negligible in large band gap semiconductors like Si and GaAs. In nondegenerate semiconductors, $1/\tau_s \sim \tau T^3/E_G$ attains a stronger temperature dependence.

As SOC arises whenever there is a gradient in an electrostatic potential, the impurity potential itself gives rise to a spin-orbit interaction $V_{\text{SO}}(\mathbf{r})$. The corresponding spin relaxation rate is proportional to the concentration of impurities n_i and increases with the atomic number Z of the impurity element as Z^2 , being stronger for heavier element impurities.

The exchange interaction J between electrons and holes in p-doped semiconductors results in *Bir–Aronov–Pikus spin relaxation* (BAP) [27]. Its strength is

proportional to the density of holes p and depends on their itineracy. Localized holes act like magnetic impurities. The spin of the conduction electrons is transferred by the exchange interaction to itinerant holes, where the spin-orbit splitting of the valence bands causes spin relaxation of the hole spin.

Magnetic impurities with spin $\mathbf{S}$ interact with the conduction electron spins by the exchange interaction J , resulting in fluctuating local magnetic Zeeman fields. The resulting spin relaxation rate is $\frac{1}{\tau_{\text{MS}}} = 2\pi n_{\text{M}} \nu J^2 S(S+1)$, where n_{M} is the density of magnetic impurities, ν the density of states at the Fermi energy, and S is the spin quantum number. Antiferromagnetic exchange interaction J results in the *Kondo effect*, which enhances the spin scattering from magnetic impurities maximally at the Kondo temperature $T_{\text{K}} \sim E_{\text{F}} \exp(-1/\nu J)$ [28]. At large concentration of magnetic impurities, the RKKY-exchange interaction between the magnetic impurities quenches the spin quantum dynamics, so that $S(S+1)$ is reduced to the classical value S^2 . In Mn-p-doped GaAs, the exchange interaction between the Mn dopants and holes can compensate the hole spins and suppress the BAP spin relaxation [29]. The *hyperfine interaction* between nuclear spins $\mathbf{I}$ and the conduction electron spin $\hat{s}$ results in a local Zeeman field, and its spatial and temporal fluctuations result in spin relaxation proportional to its variance.

As magnetic field changes, the electron momentum due to the Lorentz force, the spin-orbit field changes, which results in motional narrowing and thereby a reduction of DPS [30], $1/\tau_{\text{s}} \sim \tau/(1 + \omega_{\text{c}}^2 \tau^2)$. This can be used to identify the spin relaxation mechanism, since the EYS does have only a weak magnetic field dependence due to the Pauli paramagnetism.

Dimensional Reduction of Spin Relaxation. Confinement of conduction electrons reduces the dimension of their motion. In *quantum dots*, the energy conservation restricts relaxation processes between their discrete energy levels. Thus, absorption or emission of phonons is necessary, yielding spin relaxation rates proportional to the electron–phonon scattering rate [31]. In GaAs quantum dots, spin relaxation is dominated by hyperfine interaction [32–34]. A similar conclusion can be drawn in low density n-type GaAs, where electron localization in the *impurity band* results in spin relaxation dominated by hyperfine interaction [35, 36]. Although wires have a continuous energy spectrum, spin relaxation can still be diminished as we review in the next section.

11.2.4 Spin Dynamics in Quantum Wires

In one dimensional wires with one conducting channel, impurities can only reverse the momentum $p \rightarrow -p$, resulting merely in a change of sign of the spin-orbit field, and the D'yakonov–Perel' spin relaxation vanishes [37]. In quasi-one-dimensional wires with more than one channel occupied, $W > \lambda_{\text{F}}$, the spin relaxation depends on the ratio between W and two more length scales, the spin precession length L_{SO} , (11.9), and the elastic mean free path l_{e} . For wide wires, the spin relaxation rate is expected to converge to a finite value, while for $W \rightarrow \lambda_{\text{F}}$ it vanishes. On which

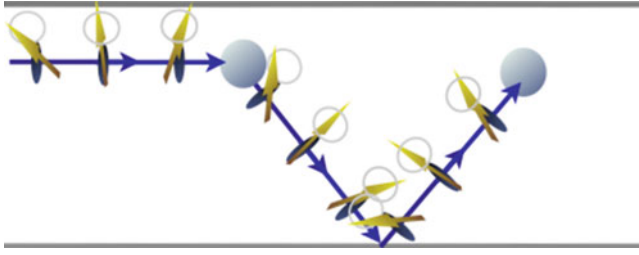


Fig. 11.5 The direction of the spin-orbit field changes due to scatterings from impurities and from the boundary of the wire

length scale, this crossover occurs is of great practical importance for spintronic applications (Fig. 11.5).

Suppression of spin relaxation in ballistic wires has been obtained numerically in [10, 38–42]. For diffusive wires, one can analytically derive the spin relaxation rate and find that it is diminished as soon as W is smaller than the spin precession length L_{SO} to [43],

$$\frac{1}{\tau_s}(W) = \frac{1}{12} \left(\frac{W}{L_{SO}} \right)^2 \delta_{SO}^2 \frac{1}{\tau_s} + D_e (m^* \epsilon_F \gamma_D)^2, \quad (11.18)$$

where $1/\tau_s = 2p_F^2(\alpha_2^2 + (\alpha_1 - m^* \gamma_D \epsilon_F/2)^2)\tau$. We introduced the dimensionless factor, $\delta_{SO} = (Q_R^2 - Q_D^2)/Q_{SO}^2$ with $Q_{SO}^2 = Q_D^2 + Q_R^2$ where Q_D depends on DSOC, $Q_D = m^*(2\alpha_1 - m^* \epsilon_F \gamma)$, while Q_R depends on RSOC: $Q_R = 2m^* \alpha_2$. Thus, for negligible cubic DSOC, the spin relaxation length increases when decreasing the wire width W as,

$$L_s(W) = \sqrt{D_e \tau_s(W)} \sim \frac{L_{SO}^2}{W}, \quad (11.19)$$

Equation (11.18) is obtained by solving the spin diffusion equation, imposing the condition that the spin current vanishes normal to the boundary, $\mathbf{j}_{s_i} \cdot \mathbf{n}|_{\text{Boundary}} = 0$. These boundary conditions effectively align the spin-orbit fields in the direction they would have in a one-dimensional wire. For wires grown along the [010] direction, one finds,

$$\mathbf{B}_{SO}(\mathbf{k}) = -2k_y \begin{pmatrix} \alpha_2 \\ \alpha_1 \\ 0 \end{pmatrix}, \quad (11.20)$$

not changing its direction when the electrons are scattered. Indeed the spin diffusion equation has the persistent solution [12] $\mathbf{S} = S_0(\alpha_2, \alpha_1, 0)$. This is remarkable, since this alignment already occurs in wires with many channels, where the diffusion is two-dimensional, and the transverse momentum k_x can be finite. It turns out that there are also two persistent spin helix solutions [12, 43] which oscillate periodically with the period $L_{SO} = \pi/m^* \alpha$. In quantum wires of width $W < L_{SO}$, these solutions are persistent for arbitrary α_1, α_2 and are given by

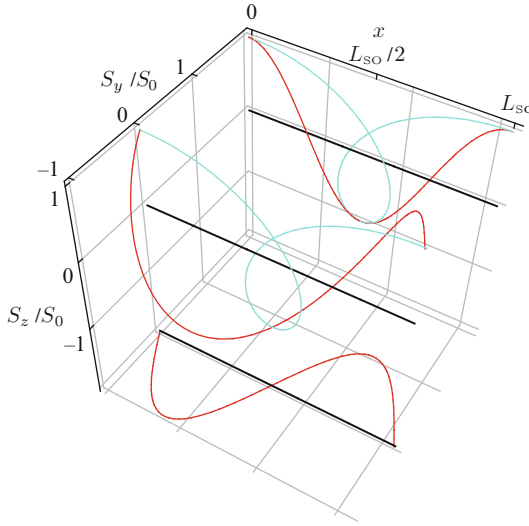


Fig. 11.6 Persistent spin helix in a diffusive quantum wire with spin precession length L_{SO} larger than the wire width W for pure linear Rashba (blue curve) and pure linear Dresselhaus coupling (red curve)

$$\mathbf{S} = S_0 \begin{pmatrix} \frac{\alpha_1}{\alpha} \\ -\frac{\alpha_2}{\alpha} \\ 0 \end{pmatrix} \sin\left(\frac{2\pi}{L_{SO}}y\right) + S_0 \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \cos\left(\frac{2\pi}{L_{SO}}y\right), \quad (11.21)$$

and the linearly independent solution, interchanging cos and sin in (11.21). Thus, the spin precesses as the electrons diffuse along the quantum wires with the period L_{SO} , forming a *persistent spin helix*, whose x component is proportional to the linear Dresselhaus-coupling α_1 while its y component is proportional to the Rashba coupling constant α_2 , see Fig. 11.6. Solving the spin-diffusion equation for larger W , one finds that the spin relaxation rate oscillates on the scale L_{SO} in analogy to Fabry–Pérot resonances [43]. For pure linear Rashba coupling, in the approximation of a homogenous spin density in transverse direction, the relaxation rate is given by

$$\frac{1}{\tau_s}(W) = D_e \frac{1}{2} Q_{SO}^2 \left(1 - \frac{\sin(Q_{SO}W)}{Q_{SO}W}\right), \quad (11.22)$$

where $Q_{SO} = 2\pi/L_{SO}$. Taking into account the transverse modulation of the spin density, one finds for $W > L_{SO}$ edge modes with a lower relaxation rate than the bulk modes [11, 12], whose relaxation rate $1/\tau_s = 0.31/\tau_{s0}$ is smaller than the one of bulk modes $1/\tau_s = 7/16\tau_{s0}$.

Quantum Corrections. Quantum interference of electrons in low-dimensional, disordered conductors results in corrections to the electrical conductivity $\Delta\sigma$, as the quantum return probability to a given point $\mathbf{x}_0$ after a time t differs from the classical

return probability. This weak localization effect is very sensitive to dephasing and symmetry breaking [44] and increases the lower the dimension of the conductor is. An electron can be scattered back and move on closed orbits, clockwise or anticlockwise with equal probability. Thus, the probability amplitudes of both add coherently, if the orbit length is smaller than the dephasing length L_φ . In a magnetic field, the electrons acquire a magnetic flux phase which changes sign with the circulation direction on the closed path, so that the quantum corrections are diminished. In the presence of SOC, the sign of the quantum correction changes to weak antilocalization [45]. SOC suppresses interference of time reversed paths in spin triplet configurations, while interference in singlet configuration remains unaffected. Since singlet interference reduces the electron's return probability it enhances the conductivity, the weak antilocalization effect. Weak magnetic fields suppress these singlet contributions, reducing the conductivity and resulting in negative magnetoresistance. When the dephasing length L_φ is smaller than the wire width W , the quantum corrections to the conductivity increase logarithmically with L_φ which increases itself as the temperature is lowered, as $L_\varphi \sim T^{-1/2}$ at low temperatures, where the electron–electron scattering is dominating.

In quasi-one-dimensional quantum wires which are coherent in transverse direction, $W < L_\varphi$ the weak localization correction is further enhanced, and increases linearly with the dephasing length L_φ . Thus, for $WQ_{SO} \ll 1$, the weak localization correction is [43], as shown in Fig. 11.7.

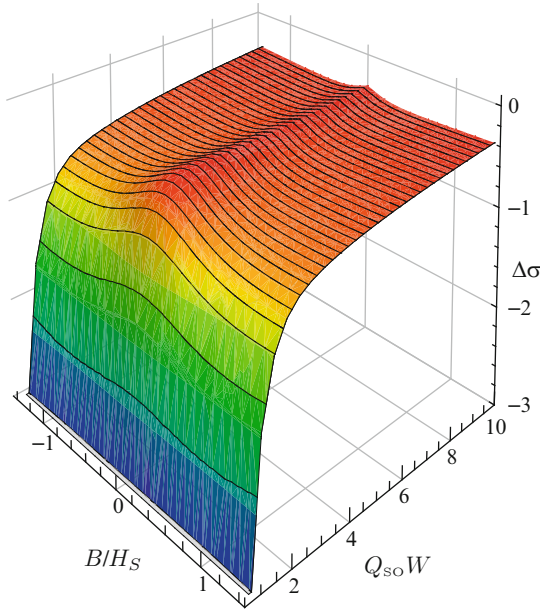


Fig. 11.7 The quantum conductivity correction in units of $2e^2/h$ as function of magnetic field B (scaled with bulk relaxation field H_s), and the wire width W (scaled with $L_{SO}/2\pi$), for pure Rashba coupling, $g_{SO} = 1$

$$\Delta\sigma = \frac{\sqrt{H_W}}{\sqrt{H_\varphi + \frac{1}{4}B^*(W) + \frac{2}{3}H_{Ms}}} - \frac{\sqrt{H_W}}{\sqrt{H_\varphi + \frac{1}{4}B^*(W) + H_s(W) + \frac{2}{3}H_{Ms}}} - 2\frac{\sqrt{H_W}}{\sqrt{H_\varphi + \frac{1}{4}B^*(W) + \frac{1}{2}H_s(W) + \frac{4}{3}H_{Ms}}}, \quad (11.23)$$

in units of e^2/h . All parameters are rescaled to dimensions of magnetic fields: $H_\varphi = 1/(4eL_\varphi^2)$, $H_W = \hbar/(4eW^2)$ the spin relaxation field due to spin orbit relaxation $H_s(W) = \hbar/(4eD_e\tau_s(W))$ [46], and its 2D limit H_s . The spin relaxation field due to magnetic impurities is $H_{Ms} = \hbar/(4eD_e\tau_{Ms})$, where $1/\tau_{Ms}$ is the magnetic scattering rate from magnetic impurities. The first term does not depend on the DP spin relaxation rate. This term originates from the interference of time reversed paths, which contributes to the quantum conductance in the singlet state, $|S = 0; m = 0\rangle = (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)/\sqrt{2}$. The minus sign is the origin of the change in sign in the weak localization correction. The other three terms are due to interference in triplet states, $|S = 1; m = 0\rangle = (|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)/\sqrt{2}$, $|S = 1; m = 1\rangle$, $|S = 1; m = -1\rangle$ which do not conserve the spin symmetry. Thus, at strong SOC spin relaxation, these terms are suppressed, and the sign of the quantum correction switches to weak antilocalization. We defined the effective magnetic field,

$$B^*(W) = \left(1 - 1/\left(1 + \frac{W^2}{3l_B^2}\right)\right) B. \quad (11.24)$$

The spin relaxation field $H_s(W)$ is for $W < L_{SO}$,

$$H_s(W) = \frac{1}{12} \left(\frac{W}{L_{SO}}\right)^2 \delta_{SO}^2 H_s, \quad (11.25)$$

suppressed in proportion to $(W/L_{SO})^2$. In analogy to the effective magnetic field, (11.24), the SOC acts in quantum wires like an effective magnetic vector potential [V.L. Fal'ko, private communication (2003)]. One can expect that in ballistic wires, $l_e > W$, the spin relaxation rate is suppressed in analogy to the flux cancellation effect, which yields the weaker rate, $1/\tau_s = (W/Cl_e)(D_eW^2/12L_{SO}^4)$, where $C = 10.8$ [47–49].

11.2.4.1 Comparison with Experiments

Optical Measurements. With optical time-resolved Faraday rotation (TRFR) spectroscopy [50], spin dynamics in an array of n-doped InGaAs wires was probed [51, 52]. Spin aligned charge carriers were created by absorption of circularly polarized light, and the time evolution of the spin polarization was measured with a linearly polarized pulse [51], fitting well with an exponential decay $\sim \exp(-\Delta t/\tau_s)$.

The thus measured lifetime τ_s at fixed temperature $T = 5$ K of the spin polarization was found to be enhanced when the wire width W is reduced [51]: For $W > 15 \mu\text{m}$, it is $\tau_s = (12 \pm 1) \text{ps}$, it increases for channels grown along the [100] direction to $\tau_s = 30 \text{ps}$ and in [110] direction to $\tau_s = 20 \text{ps}$. Wires aligned along [100] and [010] show equivalent spin relaxation times, which are longer than those of wires patterned along [110] and $[\bar{1}10]$. The dimensional reduction was seen for wire widths smaller than $10 \mu\text{m}$, which is much wider than both the Fermi wave length and the elastic mean free path l_e . This agrees well with the predicted reduction of the DP scattering rate, (11.18). From the measured 2D spin diffusion length $L_s(2D) = (0.9 - 1.1) \mu\text{m}$ and its relation to the spin precession length (11.9), $L_{SO} = 2\pi L_s(2D)$, we expect the crossover to occur on a scale of $L_{SO} = (5.7 - 6.9) \mu\text{m}$ as observed [51]. From $L_{SO} = \pi/m^*\alpha$, we get with $m^* = .064m_e$ a SOC $\alpha = (5 - 6) \text{meV}\text{\AA}$. According to $L_s = \sqrt{D_e\tau_s}$, the spin relaxation length increases by $\sqrt{30/12} = 1.6$ in the [100], and by $\sqrt{20/12} = 1.3$ in the [110] direction. However, the spin relaxation time has been found to attain a maximum at about $W = 1 \mu\text{m} \approx L_s(2D)$, decaying appreciably for smaller widths. While a saturation of τ_s could be expected according to (11.18) for diffusive wires, due to cubic Dresselhaus-coupling, a decrease is unexpected. Schwab et al., [11], noted that with wire boundary conditions which do not conserve the spin of the conduction electrons one can obtain such a reduction. This could occur in wires with smooth confinement. The magnetic field dependence follows the expected form, confirming that DPS is the dominant spin relaxation mechanism in these wires.

Transport Measurements. A dimensional crossover from weak antilocalization to weak localization and a reduction of spin relaxation has recently been observed experimentally in n-doped InGaAs quantum wires [53] [F.E. Meijer, private communication (2005)], in GaAs wires [54], as well as in AlGaIn/GaN wires [55]. The crossover indeed occurred in all experiments on the length scale of the spin precession length L_{SO} . Wirthmann et al., [53], did measure the magnetoconductivity of inversion-doped InAs quantum wells with a density of $n = 9.7 \times 10^{11}/\text{cm}^2$, and an effective mass of $m^* = .04m_e$. In wide wires, the magnetoconductivity showed a pronounced weak antilocalization peak fitting well with the 2D theory [46, 56] with a spin-orbit-coupling parameter of $\alpha = 9.3 \text{meV}\text{\AA}$. They observed a diminishment of the antilocalization peak which occurred for wire widths $W < 0.6 \mu\text{m}$, at $T = 2$ K, indicating a dimensional reduction of the DP spin relaxation rate. Very recently, Kunihashi et al. [57] observed the crossover from weak antilocalization to weak localization in gate controlled InGaAs quantum wires. The asymmetric potential normal to the quantum well could be enhanced by the application of a negative gate voltage, yielding an increase of the SIA coupling parameter α , with decreasing carrier density, as was obtained by fitting the magnetoconductivity of the quantum wells to 2D weak localization corrections [56]. Thereby, the spin relaxation length $L_s = L_{SO}/2\pi$ was found to decrease from 0.5 to $0.15 \mu\text{m}$, which with $L_{SO} = \pi/m^*\alpha$ corresponds to an increase of α from $(20 \pm 1) \text{meV}\text{\AA}$ at electron concentrations of $n = 1.4 \times 10^{12}/\text{cm}^2$ to $\alpha = (60 \pm 1) \text{meV}\text{\AA}$ at electron concentrations of $n = 0.3 \times 10^{12}/\text{cm}^2$. The magnetoconductivity of a sample with

95 quantum wires in parallel showed a clear crossover from weak antilocalization to localization. Fitting the data to (11.23), a corresponding decrease of the spin relaxation rate was obtained, which was observable already at large widths of the order of the spin precession length L_{SO} in agreement with the theory (11.18). However, a saturation was obtained theoretically in diffusive wires, due to cubic BIA coupling was not observed. This might be due to the limitation of (11.18), to diffusive wire widths, $l_e < W$, while in ballistic wires a suppression also of the spin relaxation due to cubic BIA coupling can be expected, since it vanishes identically in 1D wires. The dimensional crossover has also been observed in the heterostructures of the wide gap semiconductor GaN [55]. A saturation of the spin relaxation rate could not be observed, suggesting that the cubic BIA coupling is negligible in these structures. We note that in none of the transport experiments an enhancement of the spin relaxation rate was observed as in the optical experiments of narrow InGaAs quantum wires [51].

11.3 Spin Polarized Currents in Quantum Wires

So far, we have discussed the spin relaxation in nanowires in the presence of SOC. Now let us turn our attention to the generation of a spin-polarized current by SOC. We first show that the spin-polarized current *cannot* be obtained for the single-channel transport in two-terminal geometry, due to the scattering matrix property of the system with SOC. We then consider the nonuniform SOC and the three-terminal geometry in order to avoid this no-go theorem. Both of them can be used for the spin filtering without magnetic field.

11.3.1 Self-Duality and Spin Polarization

In this section, we show that the spin-polarized current cannot be obtained for the single-channel transport in two-terminal geometry. It is known that the system with SOC belongs to the so-called symplectic universality class and the property of the scattering matrix is described by self-duality [58]. The self-duality is defined as follows. If a particular 2×2 spin-dependent element of the scattering matrix is represented by

$$S_{ij} = \begin{matrix} & \begin{matrix} |\uparrow\rangle & |\downarrow\rangle \end{matrix} \\ \begin{matrix} \langle\uparrow| \\ \langle\downarrow| \end{matrix} & \begin{pmatrix} A & B \\ C & D \end{pmatrix} \end{matrix}, \quad (11.26)$$

then its reverse elements can be given by

$$S_{ji} = \begin{pmatrix} |\uparrow\rangle & |\downarrow\rangle \\ \langle\uparrow| & \langle\downarrow| \end{pmatrix} \begin{pmatrix} D & -B \\ -C & A \end{pmatrix}. \quad (11.27)$$

Under this condition, the scattering matrix for the single-channel transport in two-terminal geometry is given by

$$S = \begin{pmatrix} r_{\uparrow\uparrow} & r_{\uparrow\downarrow} & t'_{\uparrow\uparrow} & t'_{\uparrow\downarrow} \\ r_{\downarrow\uparrow} & r_{\downarrow\downarrow} & t'_{\downarrow\uparrow} & t'_{\downarrow\downarrow} \\ t_{\uparrow\uparrow} & t_{\uparrow\downarrow} & r'_{\uparrow\uparrow} & r'_{\uparrow\downarrow} \\ t_{\downarrow\uparrow} & t_{\downarrow\downarrow} & r'_{\downarrow\uparrow} & r'_{\downarrow\downarrow} \end{pmatrix} = \begin{pmatrix} e & 0 & d & -b \\ 0 & e & -c & a \\ a & b & f & 0 \\ c & d & 0 & f \end{pmatrix}, \quad (11.28)$$

where $r_{\sigma\sigma'}$ and $t_{\sigma\sigma'}$ denote the spin-dependent reflection and transmission coefficient, respectively. Due to the unitarity of the scattering matrix, $S^\dagger S = I$, the matrix elements have the following relations,

$$|a|^2 + |b|^2 - |c|^2 - |d|^2 = 0, \quad (11.29)$$

$$ac^* + bd^* = 0. \quad (11.30)$$

By using these relations, one can evaluate the spin polarizations,

$$P_x = \frac{\text{Tr}(t^\dagger \sigma_x t)}{\text{Tr}(t^\dagger t)} = \frac{ac^* + bd^*}{|a|^2 + |b|^2 + |c|^2 + |d|^2} + \text{c.c} = 0, \quad (11.31)$$

$$P_y = \frac{\text{Tr}(t^\dagger \sigma_y t)}{\text{Tr}(t^\dagger t)} = \frac{i(ac^* + bd^*)}{|a|^2 + |b|^2 + |c|^2 + |d|^2} - \text{c.c} = 0, \quad (11.32)$$

$$P_z = \frac{\text{Tr}(t^\dagger \sigma_z t)}{\text{Tr}(t^\dagger t)} = \frac{|a|^2 + |b|^2 - |c|^2 - |d|^2}{|a|^2 + |b|^2 + |c|^2 + |d|^2} = 0. \quad (11.33)$$

11.3.2 Spin Filtering Effect by Nonuniform Rashba SOC

One can expect that the spin separation of the conduction electrons is realized when the effective magnetic field has a spatial gradient like in the Stern–Gerlach experiment [59]. In this section, we suggest a spin-filtering device using the nonuniform RSOC fabricated in a two-dimensional electron gas (2DEG) [60]. In contrast to the Stern–Gerlach experiments, there is no Lorentz force due to the applied magnetic field. One can easily obtain the spin-dependent force acting on the particle with an electric charge.

The important feature of RSOC is that one can control the strength of RSOC by tuning the gate voltage. It makes it possible to consider a position dependent

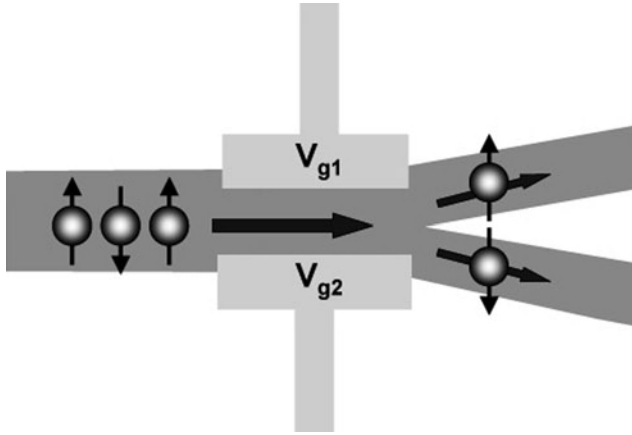


Fig. 11.8 Top view of Stern–Gerlach spin filter. V_{g1} and V_{g2} are gate voltages to produce a spatial gradient of RSOC. Stern–Gerlach type spin separation occurs when unpolarized electrons go through the nonuniform RSOC region between the two gate electrodes

strength of RSOC using a nonuniform gate voltage. In [60], we have proposed the spin-filtering device as shown in Fig. 11.8. We consider the nanowire structure consisting of the electron waveguide and two gates, fabricated at the edge of the nanowire. We apply different voltages on each gates, and the gradient of the strength of RSOC is achieved perpendicular (the y direction) to the propagation direction (the x direction) of the conduction electrons. The Hamiltonian of the proposed system is described by

$$H_{\text{SG}} = \frac{1}{\hbar} \left\{ \alpha(y) p_x \sigma_y - \frac{(\alpha(y) p_y + p_y \alpha(y)) \sigma_x}{2} \right\}, \quad (11.34)$$

where $\alpha(y)$ is the strength of RSOC. From the Heisenberg equation of motion, $\dot{p}_y = [p_y, H]/i\hbar$, one finds that the conduction electrons are accelerated in the y direction and that the direction is opposite for up and down spins. We take the spin quantization axis in the y direction.

In order to examine this suggestion, we performed numerical simulations by employing the equation of motion method to calculate the time evolution of the wave packet. Figure 11.9 shows the initial wave packet with average momentum in the x direction. The charge density $\sum_{\sigma} (|\langle \uparrow | \psi_{\sigma} \rangle|^2 + |\langle \downarrow | \psi_{\sigma} \rangle|^2)$ of the initial wave packet is plotted, where the initial wave function with spin σ is

$$\psi_{\sigma}(t=0) = A \sin\left(\frac{\pi y}{L_y + 1}\right) \exp\left(ik_x x - \frac{\delta k_x^2 x^2}{4}\right) \chi_{\sigma}, \quad (11.35)$$

with

$$\chi_{\uparrow} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ i \end{pmatrix}, \quad \chi_{\downarrow} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -i \end{pmatrix}, \quad (11.36)$$

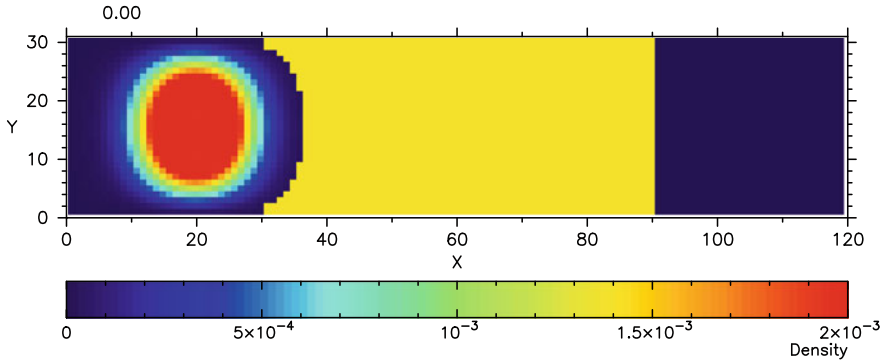


Fig. 11.9 Initial wave packet ($t = 0$) propagating to the right. *Yellow region* indicates the area where the nonuniform RSOC is present

where L_y is the width of the nanowire, $k_x = 0.5$, and $\delta k_x = 0.2$. We consider that the nonuniform RSOC exists at the middle of the electrode as shown in Fig. 11.8. We set the change of RSOC between the two gates $\Delta\alpha = 0.02\pi$. It corresponds to the experimentally obtained value, $0.4 - 0.8 \times 10^{-11}$ eVm.

The wave packet after time evolution is shown in Fig. 11.10. The charge density splits into the upper and the lower parts with opposite spin polarization. The maximum value of $\sum_{\sigma} (|\langle \uparrow | \psi_{\sigma} \rangle|^2 + |\langle \downarrow | \psi_{\sigma} \rangle|^2)$ and that of $|\sum_{\sigma} (|\langle \uparrow | \psi_{\sigma} \rangle|^2 - |\langle \downarrow | \psi_{\sigma} \rangle|^2)|$ are almost the same, which means that nearly 100% spin filtering has been achieved.

11.3.3 Generation of the Spin-Polarized Current in a T-Shape Conductor

In Sect. 11.3.1, we have shown that spin polarization cannot be obtained by single-channel transport in a two-terminal geometry. This no-go theorem does not hold for a multi-terminal geometry. In this section, we show that the spin-polarized current can be obtained in three-terminal geometry in the presence of *uniform* SOC. By evaluating the time derivative of the velocity operator, one can show that the Rashba and Dresselhaus SOC (RSOC and DSOC) work as an effective spin-dependent magnetic field perpendicular to the 2DEG [61],

$$\tilde{\mathbf{B}} = \left(0, 0, \frac{2m^* (\alpha^2 - \beta^2)}{e\hbar^3} \sigma_z \right). \quad (11.37)$$

In contrast to the Stern–Gerlach type spin-filter based on the nonuniform RSOC (Sect. 11.3.2), this field induces the spatial separation of the out-of-plane spin

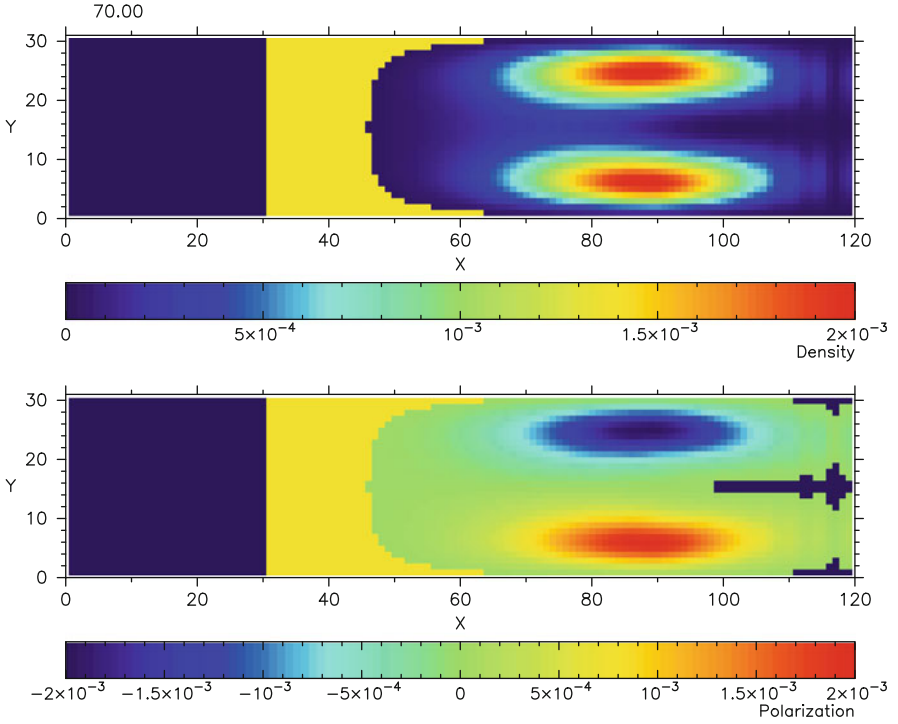


Fig. 11.10 Wave packet after time evolution ($t = 70 \cdot \hbar V_0^{-1}$) with V_0 defined in Eq. 11.40. The strength of RSOC is modulated in the y direction. *Upper*: the charge density $\sum_{\sigma} (|\langle \uparrow | \psi_{\sigma} \rangle|^2 + |\langle \downarrow | \psi_{\sigma} \rangle|^2)$, and *lower*: the corresponding polarization, $\sum_{\sigma} (|\langle \uparrow | \psi_{\sigma} \rangle|^2 - |\langle \downarrow | \psi_{\sigma} \rangle|^2)$

components. In the following, we focus on the transport in the presence of RSOC. For the results in the presence of both RSOC and DSOC, see [61].

We consider the T-shape conductor shown in Fig. 11.11 in the presence of RSOC. The sample region with RSOC is connected to three electron reservoirs by ideal leads. The electrons are injected into the sample from the reservoir 1 and go to reservoirs 2 or 3. The chemical potential at the reservoir 2 is equal to that at the reservoir 3. At small voltages, the currents I_{21} and I_{31} from the reservoir 1 to reservoirs 2 and 3, respectively, are proportional to the conductance G^{21} and G^{31} .

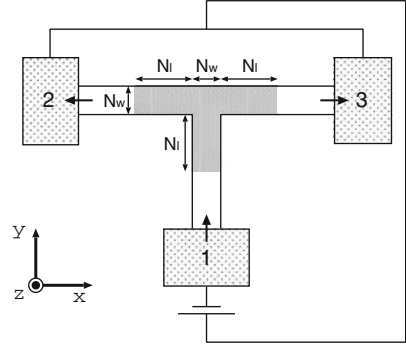
In the discrete lattice model, the effective Hamiltonian can be written as

$$H = \sum_{i,\sigma} W_i c_{i\sigma}^{\dagger} c_{i\sigma} - \sum_{\langle ij \rangle \sigma \sigma'} V_{i\sigma, j\sigma'} c_{i\sigma}^{\dagger} c_{j\sigma'}, \quad (11.38)$$

with

$$V_{i, i+\hat{x}} = V_0 \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix}, \quad (11.39)$$

Fig. 11.11 Schematic view of the T-shaped conductor. Current injected from reservoir 1 can go to reservoirs 2 or 3. *Shaded*: regions with nonvanishing SOC; parameters: $N_w = 10a$ and $N_l = 20a$ with a lattice spacing of tight-binding model



and

$$V_{i,i+\hat{y}} = V_0 \begin{pmatrix} \cos \theta & i \sin \theta \\ i \sin \theta & \cos \theta \end{pmatrix}, \quad (11.40)$$

where W_i denotes the random potential on the site i distributed uniformly in $[-W/2, W/2]$, and $V_{i,i+\hat{x}}(V_{i,i+\hat{y}})$ the hopping matrix elements in $x(y)$ directions, restricted to nearest neighbors. The hopping energy $V_0 = \hbar^2/2m^*a^2$, where m^* is the effective electron mass and a the tight-binding lattice spacing, is taken as the unit of the energy. The parameter θ represents the strength of RSOC, which is related to α by $\alpha = 2\theta V_0 a$ for $\theta \ll 1$.

The conductance and spin polarization from reservoirs J to I are

$$G^{IJ} = G_0 \text{Tr} t_{IJ}^\dagger t_{IJ}, \quad (11.41)$$

and

$$P_k^{IJ} = \frac{\text{Tr} t_{IJ}^\dagger \sigma_k t_{IJ}}{\text{Tr} t_{IJ}^\dagger t_{IJ}} \quad (k = x, y, z), \quad (11.42)$$

with the quantized conductance $G_0 \equiv e^2/h$. Here t_{IJ} denotes the transmission matrix from reservoirs J to I , which can be calculated by the recursive Green function method [62]. Below, we will focus on the transport between reservoirs 1 and 2 and omit the superscript of G^{21} and P_k^{21} . One can easily show that $G^{31} = G^{21}$, $P_x^{31} = P_x^{21}$, $P_y^{31} = -P_y^{21}$ and $P_z^{31} = -P_z^{21}$ via current conservation and the symmetry of the system [61]. In the following, we focus on the magnitude of spin polarization $|P| = (P_x^2 + P_y^2 + P_z^2)^{1/2}$ instead of P_z since that depends on N_l due to the spin precession.

First, we investigate the transport in the absence of impurities ($W = 0$). Figure 11.12 shows the energy dependence of conductance and spin polarization in the presence of RSOC. It is clearly shown that the spin polarization can be obtained by RSOC. Especially, a nearly perfect spin polarization is achieved in the single-channel energy regime ($-3.92V_0 < E < -3.68V_0$) while the polarization decreases

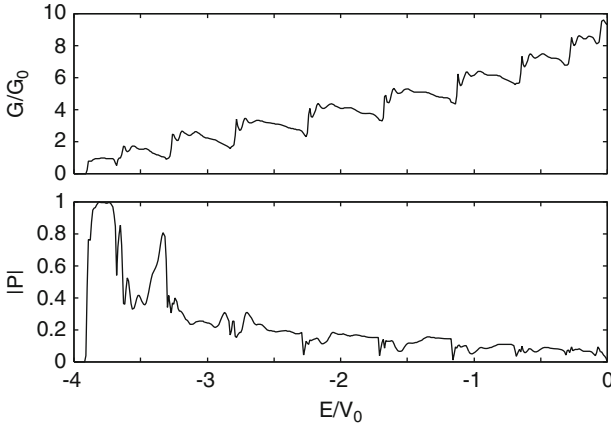


Fig. 11.12 Conductance G and spin polarization $|P|$ as a function of energy in the presence of RSOC ($\theta = 0.06\pi$). 100% spin polarization is obtained for the single-channel energy regime ($-3.92V_0 < E < -3.68V_0$)

rapidly as the number of channels increases. The condition for this 100% spin polarization is given by [63]

$$2l_{\bar{B}}(\theta) < N_w, \quad (11.43)$$

with

$$2l_{\bar{B}}(\theta) = \frac{2m^*v}{e\bar{B}(\theta)} = \frac{2L_{so}^2}{N_w}. \quad (11.44)$$

Here $2l_{\bar{B}}(\theta)$ denotes the cyclotron diameter of the spin-dependent effective field induced by RSOC, which is given by $\bar{B}(\theta) = 4\hbar\theta^2/e\pi a^2$ in the tight-binding description [cf. (11.37)]. The spin precession length is given by $L_{so} = \pi a/2\theta$. The condition (11.43) is satisfied if the spin precession length becomes shorter than the wire width ($L_{so} < N_w/\sqrt{2}$).

Figure 11.13 shows the current and spin density for the electron injected with up-spin in the case of perfect spin polarization. One can clearly see the spin-dependent deflection at the junction and the spin precession in the wire. The coupling of these two effects results in the snake motion of electrons.

We now consider briefly the effect of disorder on the spin polarization (Fig. 11.14). An ensemble average is performed over 10^4 samples. The suppression of the polarization by disorder becomes more prominent as the SOC becomes stronger. The mean free path of a 2DEG in the tight-binding model is given by $L_m = 48aV_0^{3/2}\sqrt{E+4V_0}/W^2$ [62]. One can use this estimate to distinguish the ballistic regime from the diffusive one. For the present system, we obtain for $W \simeq 1.53V_0$, $L_m = 50a$ (indicated by an arrow in Fig. 11.14). As seen in the figure, the sample size must be smaller than the mean free path in order to obtain high spin polarization.

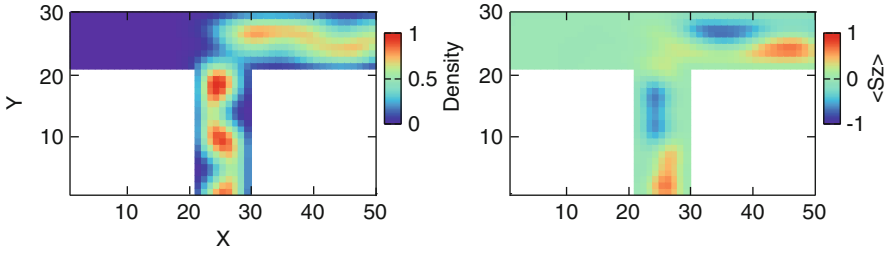


Fig. 11.13 Current and spin density for the up-spin injection in the presence of RSOC ($\theta = 0.06\pi$). The Fermi energy is set to be $E = -3.8V_0$. The conductive electrons are deflected at the junction due to the spin-dependent Lorentz force induced by RSOC (11.37)

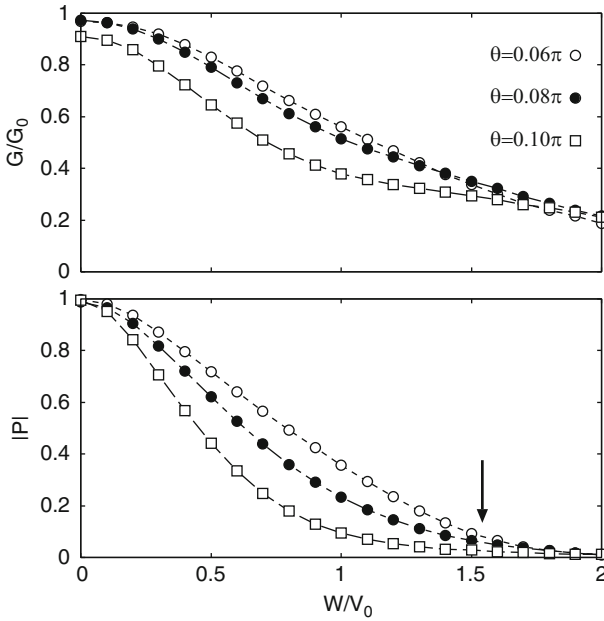


Fig. 11.14 Conductance G and spin polarization $|P|$ as functions of the strength of disorder W for several strengths of RSOC at $E = -3.8V_0$. Ensemble average has been taken over 10^4 samples. For stronger RSOC, the polarization becomes more sensitive to disorder. *Arrow*: crossover between ballistic and diffusive regimes

11.4 Critical Discussion and Future Perspective

The spin-dynamics and spin relaxation of itinerant electrons in disordered quantum wires with SOC is governed by the spin diffusion equation (11.11). The solution of the spin diffusion equation reveals the existence of persistent spin helix modes when the linear BIA and the SIA spin-orbit coupling are of equal magnitude. In quantum wires which are narrower than the spin precession length L_{SO} , there is

an effective alignment of the spin-orbit fields giving rise to long living spin density modes for arbitrary ratio of the linear BIA and the SIA spin-orbit coupling. The resulting reduction in the spin relaxation rate results in a change in the sign of the quantum corrections to the conductivity. Recent experimental results confirm the increase of the spin relaxation rate in wires whose width is smaller than L_{SO} , both the direct optical measurement of the spin relaxation rate as well as transport measurements. These show a dimensional crossover from weak antilocalization to weak localization as the wire width is reduced. Open problems remain, in particular in narrower, ballistic wires, where optical and transport measurements seem to find opposite behavior of the spin relaxation rate: enhancement and suppression, respectively. The reduction of spin relaxation in quantum wires opens new perspectives for spintronic applications, since the SOC and therefore the spin precession length remains unaffected, allowing a better control of the itinerant electron spin. Thus, creating spin polarized currents in a T-shape structure with Rashba-SOC may become experimentally possible. The observed directional dependence moreover can yield more detailed information about the SOC, enhancing the spin control for future spintronic devices further.

Acknowledgements

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Chapter 12

InAs Spin Filters Based on the Spin-Hall Effect

Jan Jacob, Toru Matsuyama, Guido Meier, and Ulrich Merkt

Abstract We give an overview of the generation of spin-polarized currents in all-semiconductor devices by utilizing the intrinsic spin-Hall effect. Two-staged cascades of Y-shaped three-terminal junctions of narrow quantum wires fabricated from InAs heterostructures with strong Rashba spin-orbit interaction allow all-electrical generation and detection of spin-polarized currents. We compare our low-temperature transport measurements to numerical simulations and find in both highly spin-polarized currents in the case of transport in the lowest one-dimensional subband.

12.1 Introduction

In the field of spintronics, the electron's spin is used as an additional information carrier besides its charge. This allows, for example, a quad-state logic, where '0' is represented by the lack of electrons, '1' by, e.g., spin-up electrons, '2' by spin-down electrons and '3' by a mixture of the two later states. Such a quad-state logic would largely enhance the capacity of a random access memory as well as the computation power of processors.

A mandatory prerequisite for spintronics is to create and to detect spin-polarized currents. While ferromagnets yield a high degree of spin polarization, semiconductors are ideal candidates for spin manipulation. Metal-based spintronic devices are already in use like hard disk read heads employing the giant magnetoresistance effect. However, spintronic devices based on semiconductors suffer from the lack of compatible spin-polarized sources. The injection of spin-polarized currents from ferromagnets into semiconductors is hardly achievable due to the conductivity mismatch and scattering at the interface. Optical injection and detection of spin polarizations in semiconductors is a proven concept but would need optical components in normally purely electric setups. It is preferable to develop all-semiconductor devices capable of generating and detecting spin-polarized currents all-electrically. This can be achieved by employing the intrinsic spin Hall effect caused by the spin-orbit interaction in three-terminal spin-filter devices with

strong spin–orbit interaction as demonstrated by several groups theoretically and by numeric simulations. We fabricated Y-shaped spin filters from InAs heterostructures. By cascading two filter stages, the all-electrically generated spin-polarized currents of the first stage are detected all-electrically by the second stage. Comparison of the results of our low-temperature transport measurements with numerical simulations indicates highly spin-polarized currents generated by the spin-filter cascades. The combined setup of generator and detector provides the possibility of the investigation of several important values and effects for semiconductor spintronics like the spin-precession length, spin-coherence length, or the zitterbewegung.

12.2 Spin–Orbit Coupling

Semiconductor spintronics relies on spin–orbit interaction, which arises from inversion asymmetry in the crystal structure or asymmetries of the confinement potential of low-dimensional electron or hole systems. For comparative reasons, we first describe the situation of a relativistic electron in vacuum. Then, different spin–orbit coupling mechanisms in semiconductors are explained.

12.2.1 Spin–Orbit Coupling in Vacuum

For a free electron in vacuum, the coupling of the spin and orbital degrees of freedom is a relativistic effect, which is described by the non-relativistic expansion of the Dirac equation in powers of the inverse speed of light. In second order, one obtains a spin-dependent term

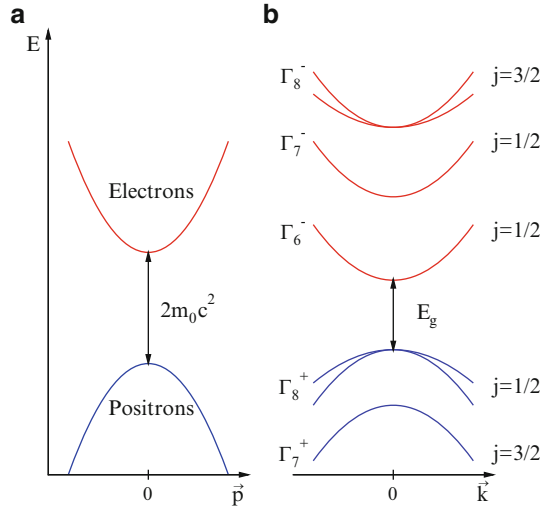
$$H_{\text{SO}} = \frac{1}{2m_0c^2} \mathbf{s} \left(\nabla V \times \frac{\mathbf{p}}{m_0} \right) \quad (12.1)$$

in the Hamiltonian, with the electron mass m_0 , the spin $\mathbf{s}$, the momentum $\mathbf{p}$ and an external potential V . The free Dirac equation for $V = 0$ yields two dispersion branches as shown in Fig. 12.1a, one for positive and one for negative energies

$$\epsilon(\mathbf{p}) = \pm \sqrt{m_0^2 c^4 + c^2 p^2}. \quad (12.2)$$

This corresponds to an energy gap of $2m_0c^2 \approx 1 \text{ MeV}$ between the dispersion branches. As described by Schrödinger, this leads to an oscillatory motion called zitterbewegung for a wavepacket that is a superposition of both solutions [1]. However, the effect has not yet been observed due to its small amplitude of $\approx 4 \cdot 10^{-3} \text{ \AA}$.

Fig. 12.1 Band structure of an electron in vacuum (*left*) and in a III–V semiconductor (*right*)



12.2.2 Spin–Orbit Coupling in III–V Semiconductors

The bandstructure of III–V semiconductors around the Γ -point with a parabolic s-type conduction band and p-type valence bands consisting of the light and the heavy hole band as well as the split-off band shows similarities to the result of the Dirac equation for a free electron as can be seen in Fig. 12.1b. As the energy gap is in the order of 1 eV or even less spin–orbit coupling effects are significantly more pronounced, which makes III–V semiconductors ideal candidates for spintronic devices [2]. For example, the amplitude of the zitterbewegung can be up to 100 nm [3]. Effective Hamiltonians that account for conduction-band electrons in different spatial dimensions can be obtained via the so-called $\mathbf{k} \cdot \mathbf{p}$ theory [4].

Dresselhaus Coupling in Bulk Material

For electrons in the s-type conduction band, the spin–orbit coupling of lowest order in the electron momentum has been derived by Dresselhaus [5]:

$$H_D^{\text{bulk}} = \frac{\gamma}{\hbar^3} [\sigma^x p_x (p_y^2 - p_z^2) + \sigma^y p_y (p_z^2 - p_x^2) + \sigma^z p_z (p_x^2 - p_y^2)] \quad (12.3)$$

with the Pauli matrix vector $\boldsymbol{\sigma}$ and the effective coupling parameter γ . As the zincblende lattice possesses no inversion center, the coupling parameter γ is different from zero for III–V semiconductors. This spin–orbit coupling due to bulk-inversion asymmetry is called Dresselhaus spin–orbit coupling.

Dresselhaus Coupling in Two-Dimensional Systems

In a quasi two-dimensional quantum well grown along the [001] direction, one introduces the expectation values $\langle p_z \rangle \approx 0$ and $\langle p_z^2 \rangle = \hbar^2 \langle k_z^2 \rangle$. Neglecting terms of order p_x^2, p_y^2 leads to a spin-orbit coupling that is linear in momentum [6]. It is described by the Hamiltonian

$$H_D = \frac{\beta}{\hbar} (p_y \sigma^y - p_x \sigma^x), \quad (12.4)$$

where $\beta = \gamma \langle k_z^2 \rangle$ is the Dresselhaus parameter for the lowest subband of the quantum well. The value for the Dresselhaus parameter in GaAs is well studied theoretically [7] and experimentally [8, 9] and is commonly given as $\gamma = 25 \text{ eV}\text{\AA}^3$. Depending on the width of the well this value results in a coupling strength β of up to 10^{-11} eV m . For InAs, similar coupling strengths are obtained.

Rashba Coupling in Two-Dimensional Systems

The second important contribution to the spin-orbit coupling in III-V semiconductor quantum wells arises from the confining potential of the well when that itself is lacking inversion symmetry. This is known as the Rashba spin-orbit interaction [10, 11] and is described by the term

$$H_R = \frac{\alpha}{\hbar} (p_x \sigma^y - p_y \sigma^x) \quad (12.5)$$

where α is the Rashba parameter, which is proportional to the gradient of the potential across the quantum well. This allows the tuning of the Rashba spin-orbit interaction by bending the potential landscape in the well with an external voltage. To achieve this, a gate voltage is applied to a pair of gate electrodes introducing an additional electric field across the quantum well [12, 13]. In heterostructures, another contribution to the Rashba spin-orbit coupling arises from asymmetric walls confining the well as the electron wave functions penetrate the finite walls differently [14, 15]. This contribution dominates the Rashba spin-orbit coupling. While the Rashba parameter α in GaAs is of the order of 10^{-12} eV m [16], it can be of the order of some 10^{-11} eV m in InAs [13, 17]. By comparing the strength of the Rashba and the Dresselhaus spin-orbit interaction, one can conclude that in InAs the resulting effective spin-orbit coupling is dominated by the Rashba contribution. Intuitively speaking the spin-orbit coupling terms result from a momentum dependent Zeemann field, which acts on the electron's spin as illustrated in Fig. 12.2. This leads to a dependence of the electron's spin state on its momentum. In the case of pure Rashba or pure Dresselhaus spin-orbit coupling, there are two parabolas shifted horizontally along the momentum axis instead of a vertical shift along the energy axis in the Zeemann case as shown in Fig. 12.3.

Fig. 12.2 Sketch of a quantum well with Rashba spin–orbit interaction. The asymmetry in the potential along the growth direction (z) is described by an electric field E_z . This field causes an effective magnetic field B_R in the rest system of the electron while it moves along its path in the x direction

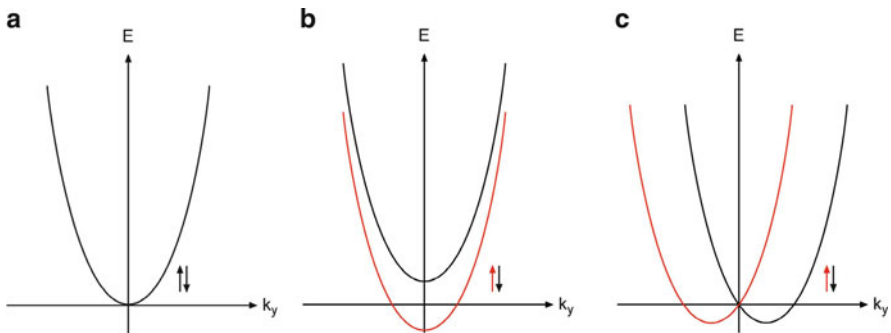
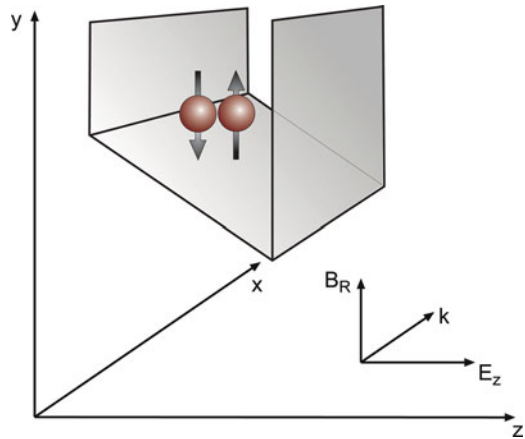


Fig. 12.3 (a) Spin degenerated dispersion relation. (b) Energy-shifted dispersions for the two spin subbands in a Zeemann field. (c) Momentum-shifted dispersion parabolas in the presence of spin–orbit interaction

12.3 Spin Hall Effect

The Hall effect describes the influence of the Lorentz force on a current flowing through a slab in a perpendicular magnetic field. The presence of the magnetic field causes a deflection of the electrons to one side of the sample. This can be measured as a voltage across the wire as can be seen in Fig. 12.4a. In ferromagnetic materials the Hall resistivity includes another contribution arising from the anomalous Hall effect, which depends on the magnetization of the material. The anomalous Hall effect is not due to the contribution of the magnetization to the total magnetic field but due to the magnetization of material itself. Here the electrons are again deflected to one side of the wire, but the direction depends on their spin. As, in a magnetized material, there are different numbers of spin-up and spin-down carriers, this results in a net Hall voltage. It is accompanied by a spin current transverse to the wire as can be seen in Fig. 12.4b [18]. In the absence of a magnetic field and without any magnetization of the material, there can still be a deflection of the electrons

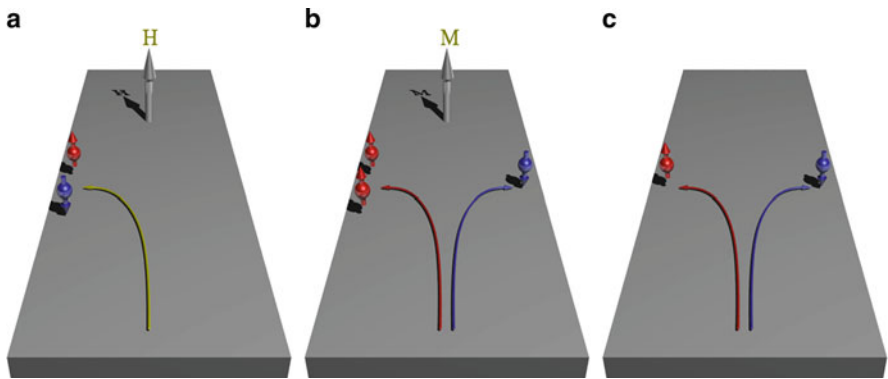


Fig. 12.4 (a) Sketch of the classic Hall effect, (b) the anomalous Hall effect, and (c) the spin Hall effect

transverse to the wire. This deflection is spin dependent and therefore called spin Hall effect. Like in unmagnetized materials without external magnetic fields, the numbers of spin-up and spin-down electrons moving to different sides of the wire are the same. There is no net Hall voltage present. Still a transverse spin current can be detected as can be seen in Fig. 12.4c. The first consideration of the concept that charge carriers with spin experience a spin-dependent drag perpendicular to the charge-dependent drag was given by Dyakonov and Perel [19] and attracted a lot of renewed interest in the late 1990s, especially by a publication of Hirsch [20]. In the following we will explain the extrinsic and the intrinsic spin-Hall effect as well as ways for its experimental detection.

12.3.1 *Extrinsic Spin Hall Effect*

In a paramagnetic metal or in a doped semiconductor, one considers charge transport of carriers with spin in the direction of an electric field in the absence of a magnetic field. The spin of the charge carriers and the same scattering mechanisms of the anomalous Hall effect in magnetized materials cause the scattered carriers with spin-up to move preferably into one direction, perpendicular to the electric field and carriers with spin-down to move in the opposite direction, which is known as skew scattering. This phenomenon is called the extrinsic spin Hall effect. Besides the skew scattering mechanism, also the side-jump mechanism is considered as a cause of the extrinsic spin Hall effect [21, 22]. An excellent overview of the mechanisms contributing to the extrinsic spin Hall effect is given by Dyakonov in [23].

12.3.2 *Intrinsic Spin Hall Effect*

In contrast to the extrinsic mechanism, the intrinsic spin Hall effect is entirely due to spin–orbit coupling without the need of scattering processes. The effect was predicted by Murakami et al. in 2003 [24]. Another prediction was made by Sinova et al. in 2004 [25]. While the first paper considers holes in a bulk semiconductor system, Sinova et al. investigate a two-dimensional electron gas with spin–orbit interaction of the Rashba type. Also the spin Hall transport of heavy holes in two-dimensional systems has been studied [3]. A survey on the intrinsic spin Hall effect is given by Schliemann in [26].

12.3.3 *Experimental Detection of the Spin Hall Effect*

Experimental investigations of the spin Hall effect have been carried out and additional experiments have been proposed. Most of them use the spin accumulation caused by the spin current to detect the spin transport. Recent theoretical studies on the accumulation of spins caused by the spin Hall effect include [27–32]. Kato et al. have studied spin Hall transport in n-doped bulk epilayers of GaAs and InGaAs, where they detected the spin Hall effect by Kerr microscopy in the presence of an external magnetic field in a Hanle-type setup [33]. Since the samples are clearly in the bulk regime, the intrinsic spin–orbit coupling is dominated by the Dresselhaus term, which has been studied as a possible intrinsic mechanism for the spin Hall effect [34]. The results from [34] have been applied to the experiments by Kato et al., but the agreement is not convincing. In addition, Kato et al. only find an extremely small dependence of the spin Hall transport on strain applied to the system. This is another indication disfavoring an intrinsic mechanism. Instead, reasonable agreement has been found between the results from Kato et al. and a theory of extrinsic spin Hall transport in GaAs based on impurity scattering [35]. Also for GaAs quantum wells, Kerr rotation experiments by Sih et al. strongly indicate an extrinsic mechanism [36]. Also the spin Hall transport of holes has been the subject of experiments. Wunderlich et al. investigated the spin Hall effect in a p-doped triangular well which is part of a p-n junction light-emitting diode [37]. Since the areal hole density in this experiment is significantly low so that only the lowest heavy-hole subband is occupied the authors conclude in a further publication that their results point to an intrinsic nature [38]. Hankiewicz et al. proposed an experiment for all-electrical detection of the intrinsic spin Hall effect in a ballistic H-shaped nanostructure by means of the inverse spin Hall effect. In their proposal, a voltage is induced perpendicular to a spin-current, which originates from the spin Hall effect [39]. The extrinsic spin Hall effect in aluminum has been investigated by all-electric means in lateral spin-valves by Valenzuela and Tinkham [40].

12.4 Spin Filters

Since the spin Hall effect spatially separates spin-up and spin-down electrons in a quasi one-dimensional wire, the idea to utilize this effect to generate spin-polarized currents in all-semiconductor devices has originated. All proposals share the same basic layout as they consist of three terminals. One terminal is used to inject an unpolarized current. At the other two terminals, oppositely spin-polarized currents leave the device. Besides the advantage of being all-semiconductor devices, these spin filters operate all-electrically as opposed to optical devices. Contrary to devices that use spin injection from ferromagnets which provide only a small amount of the spins at the output, in all-semiconductor spin-filters all of the spins from the input are available at the outputs. These three-terminal spin filters also provide both spin orientations at their outputs instead of blocking one spin orientation as it is case in quantum point contacts.

T-shaped Filters

In 2001, Kiselev and Kim proposed a T-shaped junction to separate electrons by their spin into two different outputs of the device utilizing spin-orbit interaction [41]. They assumed a symmetric T-shaped intersection of two quasi one-dimensional wires of 100 nm width with a pair of electrodes on top and beneath the junction. Using this front/backgate pair, the spin-orbit interaction of the Rashba type can be tuned at the junction. In their calculations, they neglect Dresselhaus spin-orbit interaction, electron-electron interactions and all types of relaxation processes. For incident electron energies in resonance with the quasi localized zero-dimensional states at the intersection, they obtain spin polarizations near 100% from numerical simulations. However, the transmission is less than 1/3 in that case. Later the authors expanded the device by a ballistic ring resonator of 200 nm diameter [42]. The latter device yields high transmission rates and high spin polarizations at low energies of the incident electrons. Higher energies corresponding to higher subbands lead to a decrease of the polarization.

Stern-Gerlach Filters

Another attempt to create an all-semiconductor device capable of generating spin-polarized currents was made by Ohe et al. in 2005 [43]. They proposed a device analogous to the Stern-Gerlach experiment where they replaced the gradient of the magnetic field from the experiment with silver atoms by a gradient of the spin-orbit coupling strength in a mesoscopic three-terminal device. In a Y-shaped device the spin-orbit interaction on the two sides of the input wire near the junction shall be tuned by a front/backgate pair on each side of the wire. By setting different voltages for each of the two electrode pairs, the spin-orbit coupling strength is tuned in different directions resulting in a gradient over the cross-section of the wire. Numerical

simulations of the propagation of electrons through this area with inhomogeneous spin-orbit interaction show a spatial separation of electrons with different in-plane spin components perpendicular to the wire. This leads to a separation of the input current into two oppositely spin-polarized currents in the output leads. They also concluded that the device can act as a detector of spin polarization when it is fed with an already polarized current.

More Detailed Geometries

Yamamoto et al. from the same group investigated spin-dependent transport through multi-terminal cavities with spin-orbit coupling for different geometries. Starting from a simple setup with a centered input lead on one side of the cavity and two symmetric output leads on the other side, they expanded the technique to create more complex devices [44, 45]. Their investigations revealed different mechanisms to be dominant for the filtering. For weak spin-orbit coupling, the separation is due to scattering by resonant states formed at the junction, i.e., by the extrinsic spin Hall effect. For strong spin-orbit coupling the intrinsic mechanism becomes dominant. The authors indicated two obstacles to be overcome in the fabrication of the filter device proposed by Kiselev and Kim: To achieve transport only in the lowest subband, which is mandatory to obtain a high spin polarization, the wires must be as narrow as 20 nm. And for this width and geometry, a Rashba parameter α of about 170×10^{-12} eVm would be necessary. This is much larger than the spin-orbit coupling strength in common materials like InAs where α is about 20×10^{-12} eVm. From a cooperative effort of the groups of Bernhard Kramer at the I. Institute of Theoretical Physics and Ulrich Merkt at the Institute of Applied Physics at the University of Hamburg a device, consisting of a Y-shaped intersection of three wires evolved, where each of the three wires is constricted by a quantum point contact to set the number of conducting subbands and to reach the one-dimensional quantum limit.

12.5 Device Layout

For an experimental realization of such an all-electrical, all-semiconductor three-terminal device for the generation of spin-polarized currents, we have modified the theoretically proposed devices. The main reasons for the modifications are limitations in the wire thickness and widths that can be prepared as well as possible ways to detect the spin polarization. The process of designing a device for experimental realization started with a concept for the detection of the spin polarization. In a second step, the size and geometry of the device have been addressed.

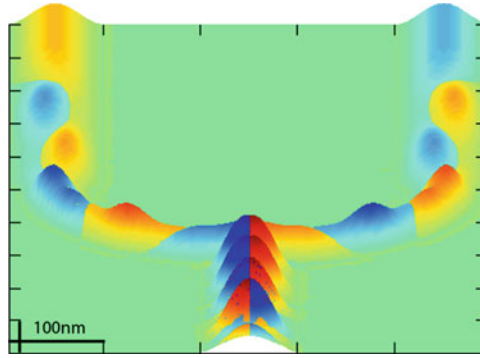


Fig. 12.5 Simulation of a single stage spin-filter by Cummings based on [46]. Spin-up and spin-down electrons are simultaneously injected at the bottom. The spin-resolved probability density shows an accumulation of spin-down electrons (colored blue) on the left and spin-up electrons (colored red) on the right of the input. At the T-shaped junction, the electrons are deflected into the two output wires with respect to their spatial separation in the input wire. This results in two oppositely spin-polarized currents in the output wires, where the spin precession and the zitterbewegung can be observed. The height indicates the probability, while the color indicates the spin orientation

Single Filter

Numerical simulations using a similar scattering matrix formalism as Yamamoto et al. have been performed by Cummings and coworkers [46, 47]. A result for a single spin filter is shown in Fig. 12.5. The device generates two oppositely spin-polarized currents, but there is no possibility to detect them all-electrically as they have the same magnitude. Optical detection is not feasible for spin-filter devices as the dimensions of the whole device are smaller than the size of a laser spot that would be used for the detection of a rotation of the polarization vector of the reflected light.

All-electrical Detection of the Spin Polarization

In cooperation with the Arizona State University, we have expanded the sample layout from a single spin filter to a two-staged cascade [48]. This cascade reveals the spin polarization generated by the first filter by different conductances of the outputs of the second filter stage as shown in Fig. 12.6. An unpolarized current composed of spin-up and spin-down electrons is injected into the input terminal at the bottom of the cascade. Due to the spin Hall effect, a spatial separation of spin-up and spin-down electrons is generated leading to an accumulation of spin-down electrons on the left and of spin-up electrons on the right side of the input wire. When reaching the first filter, the electrons are deflected into the two outputs with respect to their spatial position in the input wire. This creates two oppositely spin-polarized currents in the leads connecting the first and the second filter stage. In

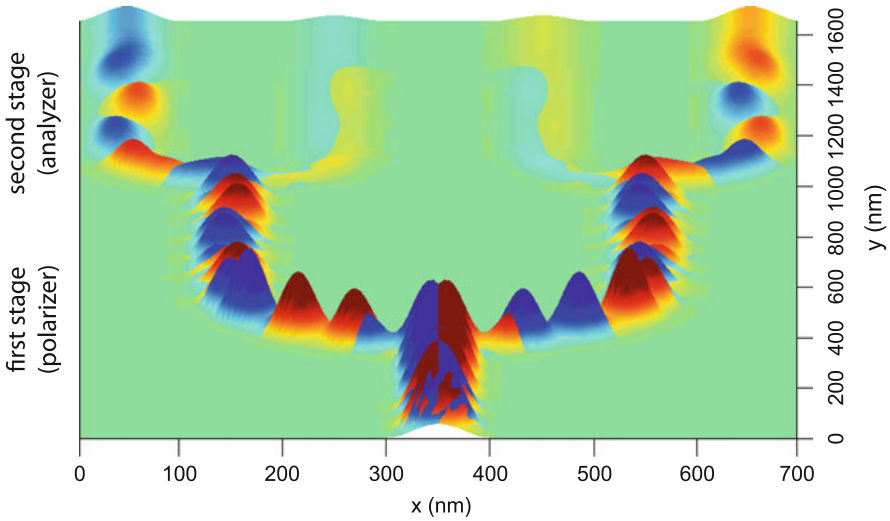


Fig. 12.6 Simulation of a two-stage cascade of spin filters. Shown is the spin-resolved probability density for spin-up (red) and spin-down (blue) electrons injected simultaneously at the input terminal

those two leads, the spin precession and the zitterbewegung can be observed. At the filters in the second stage of the cascade, there are more spin-down electrons arriving at the left filter and more spin-up electrons at the right filter. As spin-down electrons are shifted to the left and spin-up electrons to the right, they prefer different outputs of the filters. This leads to different conductances for the two outputs of each of the two second stage filters. The difference is proportional to the spin-polarization of the current arriving at the second filter stage. In that way, spin polarization can be converted to conductances and therefore be detected all-electrically.

Experimental Sample Layout

For experiments, it is advantageous to investigate a laterally symmetric device, which helps to distinguish between effects caused by imperfect preparation and those caused by the intrinsic spin Hall effect. Appending a filter to each of the two outputs of the first filter would yield redundant information. Therefore, a second filter stage is attached only to one of the two outputs. Y-shaped junctions are highly symmetric as can be seen in Fig. 12.7. The filter junctions are labeled X and Y. As it is a prerequisite that only the lowest subband is used for transport to get a well pronounced spin Hall effect, very narrow wires are needed. This cannot reliably be achieved by lithography. Therefore each of the five wires labeled 1-5 can be constricted individually by a quantum point contact formed by sidegates as indicated in Fig. 12.7 by orange triangles. In that way, the number of conducting channels in each arm can be set all-electrically.

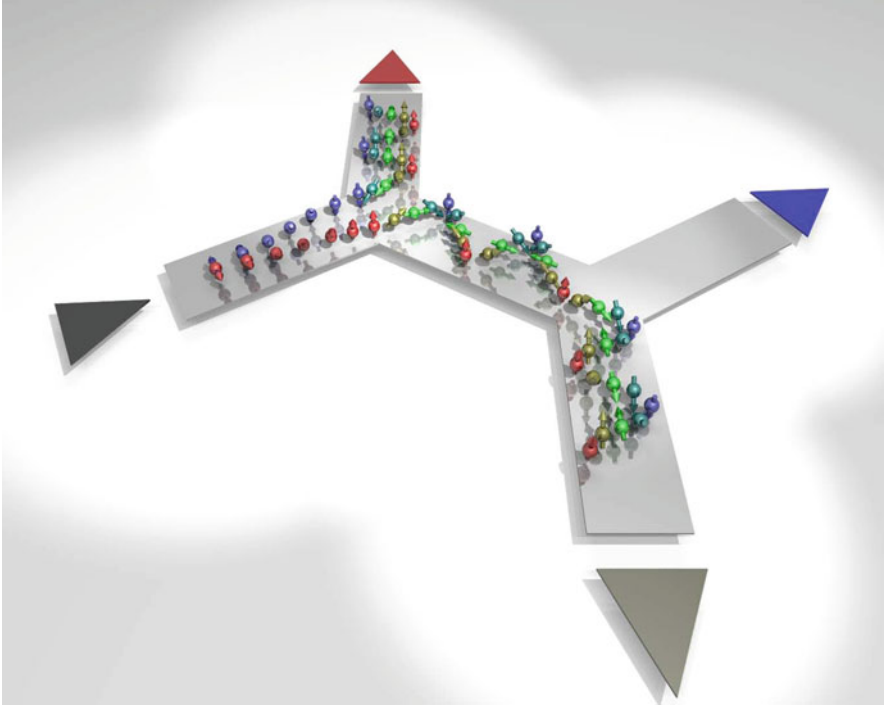


Fig. 12.8 Visualization of the spin-dependent transport through a two-stage spin-filter cascade

yield a conductance portion of 50% while the other output contributes 0% to the total conductance of the device as illustrated in Fig. 12.8.

Deviations from this perfect situation result in a smaller difference between the second filter's outputs and thus indicate a lower spin polarization arriving at the second filter. The filter efficiencies and the resulting spin polarizations in each lead are shown in Fig. 12.7 below the up and down arrows. Assuming n_0 electrons to enter the filter cascade at the input terminal A, there will be the same number $n_0/2$ of spin-up and spin-down electrons. For convenience, the numbers in Fig. 12.7 are normalized to $n_0/2$. After the first filter X, there will be $x n_0/2$ spin-up and $y n_0/2$ spin-down electrons in the center lead 3 and $(1 - x) n_0/2$ spin-up and $(1 - y) n_0/2$ spin-down electrons will exit the device at the output B. Due to the symmetric shape of the junction, the total number of electrons will be distributed equally among the two outputs resulting in $x + y = 1$. The efficiency of the first filter X is therefore given by $P_X = \frac{x-y}{x+y} = x - y$ and the polarizations of the currents in lead 3 and at output B are $P_3 = x - y$ and $P_B = -(x - y) = -P_3$. The current in lead 3 is split again with respect to the spin orientation of the electrons in the second filter Y, which in the ideal case is assumed to have the same polarization efficiency as the first filter: $P_Y = P_X$. If the distance between the two filters results in a spin precession angle of an integer multiple of 2π , i.e. the electrons are in the same spin state as at the first

junction, there are $x^2n_0/2$ spin-up and $y^2n_0/2$ spin-down electrons at output D. This corresponds to a polarization $P_D = \frac{x^2-y^2}{x^2+y^2}$. At output C $xy n_0/2$ spin-up and $yx n_0/2$ spin-down electrons arrive, which corresponds to $P_C = \frac{x(1-x)-y(1-y)}{x(1-x)+y(1-y)} = \frac{xy-yx}{xy+yx} = 0$. Not only the polarizations at outputs C and D are different but also the number of electrons. While $(x^2 + y^2)n_0/2$ electrons arrive at output D, there are $[x(1-x) + y(1-y)]n_0/2 = xy n_0/2$ electrons at output C. In experiments, this difference can be obtained from the conductances of the three outputs by taking the difference of the conductances from the outputs D and C normalized to the sum of all three outputs' conductances $(G_D/G) - (G_C/G)$ where $G = G_B + G_C + G_D$. It is possible to derive the filter's efficiencies and polarizations in all parts of the device by measuring the currents flowing through the three outputs of the cascade and measuring the voltage drops between the input and the three outputs in a four-point measurement.

12.6 Experiments

Several two-stage spin-filter cascades have been investigated in low-temperature transport measurements. Here we present results from a spin-filter cascade with a wire width of 150 nm and a filter distance of 1 μm , which has been fabricated by electron-beam lithography and reactive-ion etching from an InAs heterostructure wafer. The InAs quantum well is surrounded by InAlAs boundaries of 13.5 nm thickness above the channel and 2.5 nm below the channel. This means a significant asymmetry is introduced and a strong Rashba spin-orbit coupling of $20 \cdot 10^{-12}$ eVnm is achieved. Details about the layout of the quantum well in the heterostructure can be found in [49].

The experimental approach consists of three parts. First, the individual quantum point contacts are characterized to determine their threshold voltages. Second, the whole spin-filter cascade is characterized. The results in these two parts have been published in [50, 51]. Finally, the measured data is analyzed for conductance steps from the quantum point contacts and correlations between the number of occupied transport modes and the conductance portions are investigated.

12.6.1 Characterization of Single Quantum Point Contacts

To ensure that each of the five quantum point contacts is working correctly, the first step of the investigation of spin-filter cascade is to characterize the quantum point contacts individually. Besides obtaining the information that the quantum point contacts can constrict the corresponding arm of the spin-filter cascade and do not affect the rest of the cascade, the main goal of the characterization measurements is to obtain the individual threshold voltages that are needed for constriction. These

threshold voltages are used for the subsequent spin-filter measurement to ensure that there is the same number of occupied transport channels in each arm of the device and no asymmetry is induced by differences in the constriction of the arms. The characterization measurements are conducted in all four possible input configurations using all ports A, B, C, or D as input terminals of the cascade. In Fig. 12.9, the results for each of the five quantum point contacts are shown exemplarily in input configuration A. The input voltage is $100\text{ }\mu\text{V}$ and the frequency of the input signal is 531.3 Hz in all measurements.

It can be seen, that each quantum point contact can be constricted but different gate voltages are needed. For example, quantum point contact 1 is constricted at a gate voltage of about -0.8 V while quantum point contact 4 needs only about -0.5 V for total constriction. This emphasizes the importance of the single quantum point contact characterization as a prerequisite for the spin-filter measurements where all five quantum point contacts are constricted at the same time.

12.6.2 Characterization of Spin-Filter Cascades

For a spin-filter measurement, the previously determined threshold voltages of the five quantum point contacts are used to ensure homogeneous constriction of each arm of the cascade. The voltages applied to the quantum point contacts start at zero voltage and end at the same time at 100% of the individual threshold voltage of each quantum point contact. This application of different voltages to the different arms of the device results in the same number of occupied conductance channels for all arms at any time giving a symmetric potential landscape.

Results obtained in the four possible input configurations are shown in Fig. 12.10. The left panel presents the conductances of each of the three output terminals. In addition, the total conductance, i.e. the sum of the conductances of the three outputs, is shown in black. The conductance plots prove that all arms of the spin-filter cascade are constricted down to their individual threshold points. The conductance portions for the three outputs in the right panel are calculated by dividing the conductance of each output by the total conductance. The difference of the second filter's outputs is clearly visible. When no gate voltage is applied, the device behaves as a ballistic nanostructure resulting in a symmetric splitting of the input current at the first junction. This leads to a conductance portion of 50% for the output of the first filter. At the second junction, the current is again split equally among the two outputs of the second filter resulting in a conductance portion of 25% for these two outputs. If the transport through the cascade would be diffusive, different conductance portions for the outputs would be obtained. Assuming the same resistivity in each arm of the cascade Ohm's law predicts a conductance portion of 60% for the first filter's output and 20% each for the second filter's outputs. The conductance portions without gate voltages applied are therefore a good proof for the ballistic transport through the device. Over a wide range of gate voltages applied to the sidegates, this situation stays the same while the total conductance decreases.

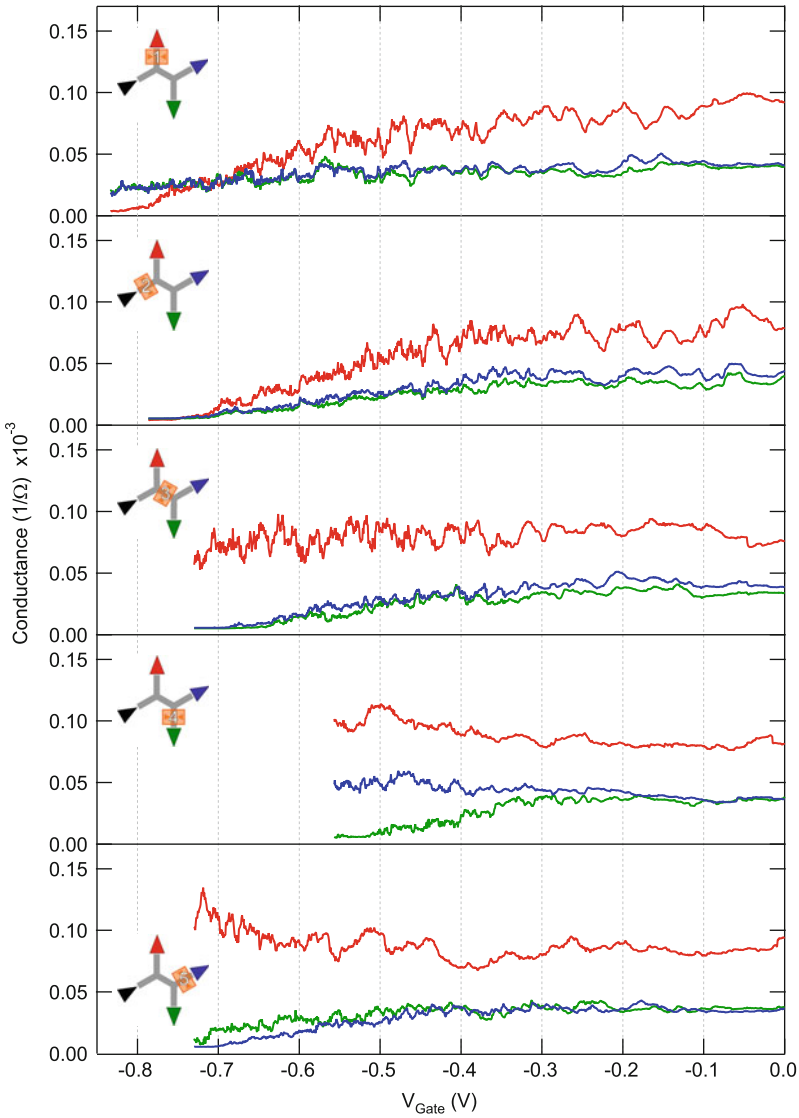


Fig. 12.9 Characterization of the five quantum point contacts in a two-stage spin-filter cascade with a filter distance of $1\ \mu\text{m}$ and a wire width of $150\ \text{nm}$. The icons indicate the position of the quantum point contact in the cascade as an orange rectangle. The *black arrow* indicates the input terminal and the colors of the *arrows* at the three output terminals correspond to the colors of the traces. For each quantum point contact, the conductance of each of the three output terminals is plotted versus the voltage applied to the sidegates

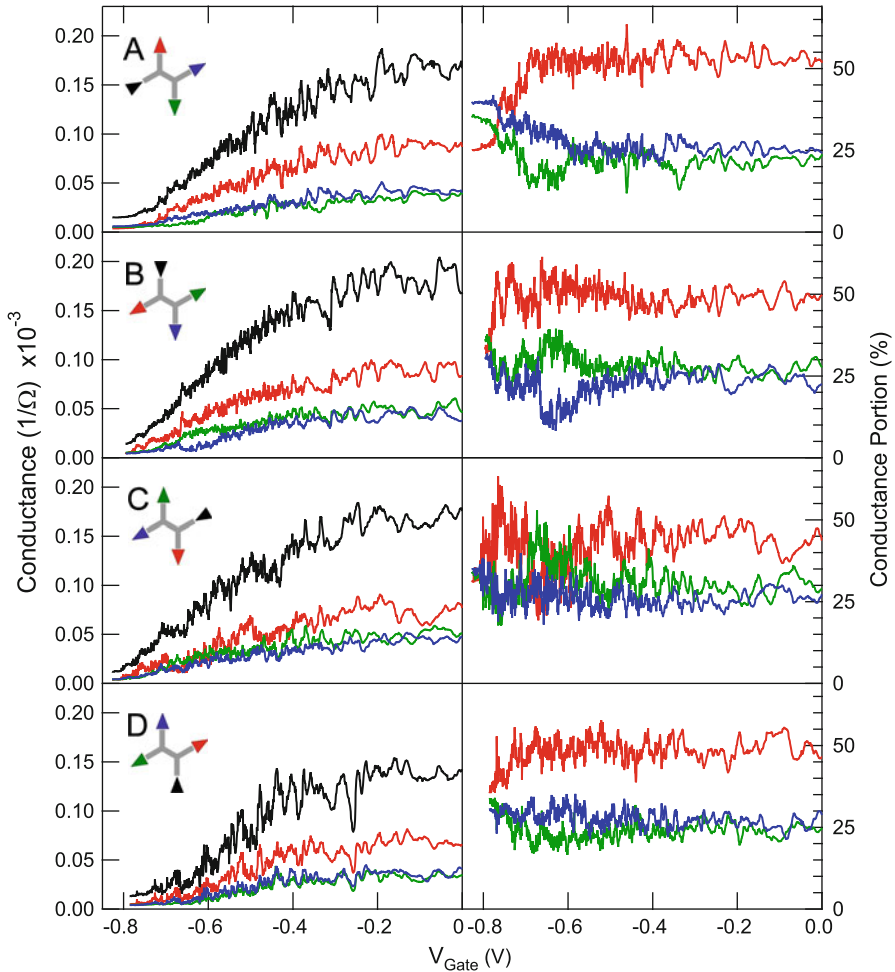


Fig. 12.10 Spinfilter measurements in the four possible input configurations. The icons indicate the input terminal used in the measurements. The colors of the output terminals correspond to the colors of the traces. In the left subpanels, the conductance of each output as well as the total conductance (colored black) of the device is plotted versus the gate voltage. The right subpanels show the conductance portions versus the gate voltage

In the regime of less than four conductance channels (about -0.45 V) first significant differences between the conductance portions of the second filter's outputs occur, which reach their maximum just before the gate voltages reach their thresholds and the wires of the spin-filter cascade are pinched off. Over the whole gate voltage range up to the thresholds the conductance portion of the first filter's output stays nearly constant at about 50% as expected. At low gate voltages, this is due to the symmetric splitting of the current at the first filter, at high gate voltages this is attributed to the filtering in the first junction due to the spin Hall effect. Then, there

are the same number of spin-up and spin-down electrons coming from the input terminal to the first filter resulting in two oppositely spin-polarized output currents with the same magnitude.

At the thresholds, the conductance portions all drop to about 33%. Due to the noise floor picked up by the measurement system, there is a finite residual value for the conductances. It is equal for all three outputs as the noise floor comes from the electrical environment and not from the sample itself. Also capacitive coupling in the measurement setup and the sample cause finite residuals that are equivalent for all three outputs. This leads to the same conductance contribution for all three outputs resulting in a conductance portion of 33% for all outputs.

12.6.3 Quantized Conductance

The conductance of a quantum point contact is a multiple of $2e^2/h$. In contrast to the well pronounced steps in the conductance of GaAs quantum point contacts the quantized conductance is not easily observed in InAs heterostructures [52]. In the results analyzed in the previous section, the steps in the conductance plots as well as corresponding features in the conductance portions are hidden under the noise. Therefore, the data are low-pass filtered to remove the noise cloaking the quantization features. To emphasise that the filtering of the data is reasonable and does not create or annihilate any features not already present, the original data are shown by symbols in light colors while the smoothed data are presented by dark solid lines in Fig. 12.11. Before smoothing the traces, the conductances are converted to units of $2e^2/h$ and the series resistivity is removed by setting an appropriate value for the constant c in the formula $G_{norm} = \left(\frac{1}{G} - \frac{c}{G_0}\right)^{-1}$. In this formula, G is the conductance at a certain gate voltage and G_0 the conductance without a gate voltage. The constant c is chosen such that the total conductance of the device in units of $2e^2/h$ without gate voltage corresponds to the number of conductance channels derived from the wire width, which is 9 for the investigated device. The gate voltage range is reduced to -0.4 V to -0.8 V , which corresponds to the last four transport modes. The smoothed data reveal steps in the conductance plots previously obscured by noise. The smoothing process is justified as the smoothed curves lie in a small band of the noisy original data as can be seen by the light colored symbols in the plots. In the spin-filter measurement, steps are not that clearly visible, but still some features can be recognized. The less pronounced steps in the spin-filter measurement could stem from the more complex potential landscape generated by constriction of all five quantum point contacts at the same time. Slightly imperfect coincidence of closing of a distinct conductance channel in the different arms of the device can lead to deviations in the total conductance making it harder to observe the distinct steps in the total conductance of the fully operated spin-filter cascade. Plateaus corresponding to multiples of $2e^2/h$ can be seen and are indicated by horizontal dashed lines. Oscillations in the conductance signal make it harder to define where a conductance channel is

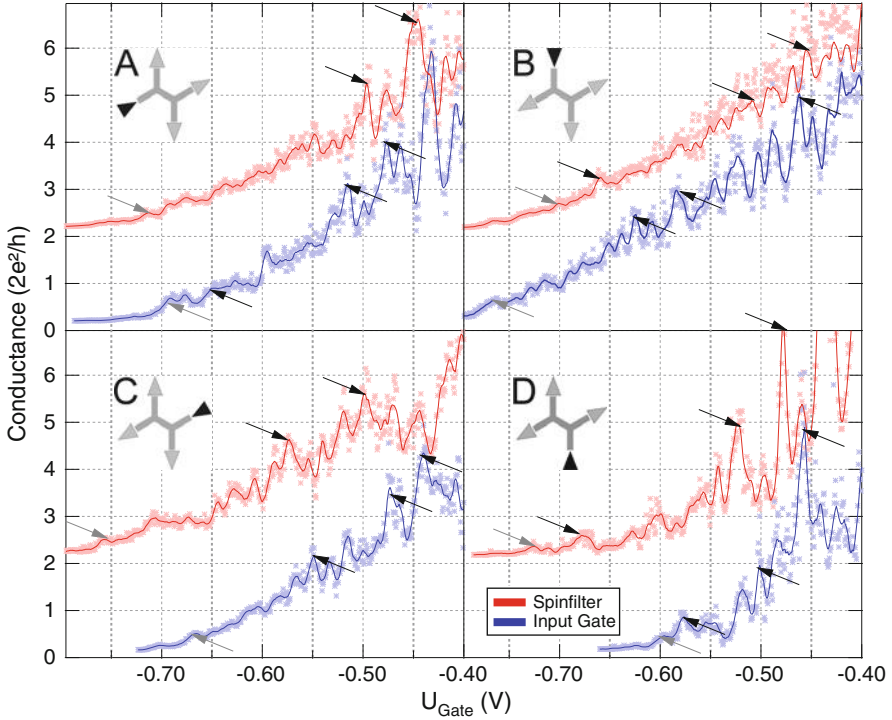


Fig. 12.11 Total conductance versus gate voltage for the four input configurations in the gate voltage range corresponding to the last four transport modes. The light colored markers in the background represent the original data. The dark colored solid lines in the foreground show filtered data that reveal conductance steps from the noisy signals. *Blue curves* are taken from the characterization measurements of the corresponding input gate, *red curves* are from the spin-filter measurement in that direction. horizontal dashed lines indicate the positions of conductance steps of $2e^2/h$. *Black arrows* indicate overshoot peaks at the beginning of each conductance step. The *grey arrows* indicate a steplike feature below one conductance channel

occupied. Especially for the higher modes, there is sometimes an overshoot right at the point where the occupation of that channel ends indicated by black arrows. The first four conductance steps are clearly visible in both the input gate and the spin-filter measurements. Also another feature below $2e^2/h$ is visible indicated by a gray arrow. Due to deviations from the ideal step distance of $2e^2/h$, it is impossible to decide whether this is the so called 0.7 feature observed by several groups [53, 54] or a 0.5 plateau indicating spin-polarized transport. When the total conductance of the spin-filter cascade is correlated with the conductance portions of the same measurement as shown in the next section, more information about that plateau is revealed. Long quantum channels as formed by the quantum point contacts in the spin-filter cascades tend to show less significantly pronounced conductance steps [55]. A tendency for resonance features as seen in the data is observed for elongated constrictions [56]. Due to the temperature of the measurements, which is well below

500 mK, additional resonances in the transmission are introduced to the plateaus [57–59]. They stem from reflections at the entrance and exit of the constriction [60, 61]. Also quantum interference associated with back scattering by impurities in the junctions has been discussed as a possible reason of the low-temperature noise [62].

12.6.4 Correlation Between Conductance Channels and Conductance Portions

The conductance portions show a significant correlation to the conductance steps as can be seen in Fig. 12.12. The best correlation can be found for direction B. The loss of a transport mode clearly changes the conductance portions. When the total conductance drops below a single transport channel, the previously present large difference between the conductance portions of the second filter's outputs nearly drops to zero. Opening more conductance channels reduces the difference of these two values. Remarkably, there are nodes visible in the conductance portions where a conductance step is observed, especially in input configuration A (see circles in Fig. 12.12). As none of the two second filter's outputs conductance portions drops to zero in the regime of the identified plateau below $2e^2/h$ this cannot be attributed to spin-polarized transport, rather it seems to be the 0.7 feature [63].

12.7 Summary

12.7.1 Conclusions

The generation of spin-polarized currents in all-semiconductor spin-filter cascades has been studied by all-electrical means. At the same time, the cascades are capable of detecting spin-polarized currents in an all-electrical way. Transport measurements on these spin-filter cascades based on InAs heterostructures have been performed at millikelvin temperatures. Based on proposed three-terminal devices utilizing the intrinsic spin Hall effect to split an unpolarized current into two oppositely spin-polarized currents, a Y-shaped geometry, whose dimensions are compatible with today's lithography capabilities, has been developed in close cooperation with A. W. Cummings from the Arizona State University, USA. The sample layout incorporates quantum point contacts to electrically narrow each lead of the device so that well defined numbers of occupied transport modes can be set. For all-electrical detection, a second Y-shaped filter stage is added to one of the outputs of the first filter. This stage is fed with the spin-polarized current from the first stage and therefore generates two output currents of different magnitude. This difference is taken as an electric measure of the spin-polarization generated by the first filter stage.

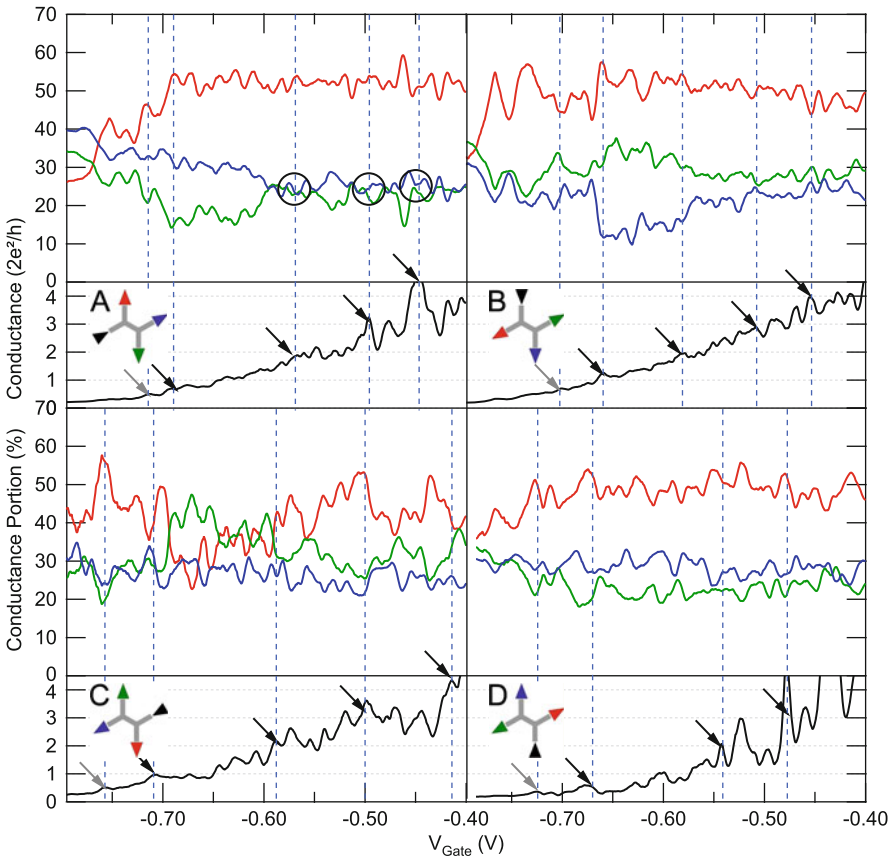


Fig. 12.12 Correlation between steps in the total conductance through the spin-filter cascade and distinct features in the conductance portions of the three outputs for the four possible input configurations. The icons indicate the input configuration. In the lower subpanel of each input configuration, the total conductance is shown as a *black* curve. The *black* arrows and vertical *blue* dashed lines indicate where a conductance channel is occupied. The upper subpanels show the conductance portions. The colors of the traces correspond to those of the output *arrows* in the icons. *Black* circles in input configuration A indicate nodes of the conductance portions of the second filter's outputs, which coincide with the opening of a new conductance channel

In addition, this second stage enhances the spin-polarization of the current flowing through this filter. To exclude asymmetries stemming from different numbers of occupied transport modes each quantum point contact is characterized on its own and its individual threshold is used in the spin-filter measurements. Additional confidence not to measure asymmetry effects is given by cyclic exchange of the input port of the cascade. Direct correlation between conductance differences and the spin polarization has to be treated with care as asymmetries induced by slight imperfections from the lithography will also contribute to asymmetric conductance portions. A proof of the functionality of the spin-filter cascades is given by a good correlation

of the occurrence of conductance steps and distinct features in the conductance portions of the three outputs and accordingly in the polarization. It is clearly shown that the reduction of occupied transport modes is related to an increasing difference in the conductance portions. This provides strong evidence for the spin Hall effect as the origin of the observed conductance asymmetries.

12.7.2 Outlook

Further investigation of the complex two-stage spin-filter cascades is needed for a more substantiated description, for example, of the interplay between intrinsic spin Hall effect, geometrical asymmetries, and disorder. To probe the spin-polarized current flowing between the two filter stages, the spin-precession length should be varied either by applying a magnetic field in the sample plane perpendicular to the center wire or by applying a voltage to a topgate-backgate pair to change the strength of the spin-orbit coupling. The change of the spin-precession length should result in an oppositely oscillatory change of the conductance portions of the second filter's outputs. As the electrons arriving at the second filter stage enter with either a positive or a negative spin-component in the z direction resulting in a different direction of the spin-current induced by the spin Hall effect, they will be deflected into different outputs. By using a magnetic field parallel or perpendicular to different axes of the spin-filter cascade, a detailed investigation of the magnetic properties of spin-filter cascades would be possible. For in-plane fields, the spin-precession length and the pattern of the zitterbewegung are changed in those arms that are perpendicular to the field [64]. This allows one to probe the presence of spin-polarized currents with the detector filter stage [65]. Also the dependence of the filter efficiency on the wire width and the filter distance should be studied. The spatial separation of spin-up and spin-down electrons generated by the spin Hall effect is a function of carrier density, effective mass, Rashba parameter, and wire width. Therefore varying the wire width should result in a maximum polarization at a certain width. By increasing the filter distance over the mean free path, the spin-filter effect should decrease significantly as the transport through the connecting wire of the two filter stages is no longer ballistic. Finally, changing the carrier density by means of topgates or backgates instead of sidegate quantum point contacts would drastically ease to simulate the device. Also the lateral confinement in the quantum wires would be constant over the whole measurement resulting in data that is easier to interpret.

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Chapter 13

Spin Injection and Detection in Spin Valves with Integrated Tunnel Barriers

Jeannette Wulffhorst, Andreas Vogel, Nils Kuhlmann, Ulrich Merkt, and Guido Meier

Abstract In paramagnetic metals, a nonequilibrium spin polarization injected from a ferromagnetic region can be sustained. We give an overview of various possibilities to realize spin injection in all-metal spin-valve devices consisting of two ferromagnetic electrodes and an interconnecting normal metal strip. Contributions to the local and the nonlocal magnetoresistance of such lateral spin valves are discussed. The strong increase of spin-injection efficiency as a consequence of the incorporation of tunnel barriers into spin valves is understood in the framework of a diffusive theory that includes spin diffusion, spin relaxation, and spin precession.

13.1 Introduction

Various approaches to generate, manipulate, and detect spin currents are presently investigated to better understand the mechanisms employed in advanced spintronic devices. Generation and detection of spin-polarized currents can be realized with ferromagnetic materials, which provide a spin-resolved density of states at the Fermi energy. The spin Hall effect and spin-current induced magnetization switching in all-metal spin valves open new perspectives for basic physics and possible applications that use the spin of the electron in addition to its charge. Spin-dependent effects already implemented in today's devices are the giant magnetoresistance (GMR) in read heads of hard-disk drives and the tunnel magnetoresistance (TMR) in magnetic random-access memories. Mesoscopic spin valves provide the outstanding opportunity to determine spin-dependent transport properties like spin-precession and spin-diffusion length in a paramagnetic channel by all-electrical means. The integration of tunnel barriers at the interfaces between ferromagnet and nonmagnetic channel enhances the spin polarization injected into the channel. Spin-dependent properties are determined in nonlocal geometry, where the charge current and the voltage probes are spatially separated in the spin valves. We describe how charge current and spin current can be controlled separately in this measurement setup. All-electrical transport measurements are performed. In various normal metals the spin-dependent nonlocal spin-valve effect and spin precession are observed. The

experimental results are supported by a theoretical description of the spin-dependent transport including spin diffusion, spin relaxation, spin precession, and tunnel barriers. From the comparison of the experimentally observed spin precession and the theoretical description, the spin-relaxation time and the spin-relaxation length in aluminum and in copper are determined.

13.2 First Experiments

In the beginning of the 1980s, a new device called spin valve emerged. It consists of ferromagnetic elements (F) connected via a paramagnetic metal (N) or an insulator (I). One ferromagnetic element is used to generate a spin current that is injected into the paramagnetic metal either directly or through an insulating tunnel barrier. A second ferromagnetic element serves as a spin-sensitive detector. An external magnetic field can align the magnetizations of the ferromagnetic elements parallel or antiparallel. A current I_C is sent through those elements. As illustrated in Fig. 13.1a, the current can be applied perpendicular to the ferromagnets plane (CPP spin valve) or in the plane of the ferromagnets (CIP spin valve). The CPP spin valves with FNF layer sequence, see Fig. 13.1a, resulted in the discovery of the GMR by Fert and coworkers [1] and Grünberg and coworkers [2]. Their work seeded the field of research of ferromagnetic/paramagnetic multilayers [3–5] and resulted in the application of the GMR in today's read heads of hard-disk drives.

In the following, the lateral mesoscopic CIP spin valve, see Fig. 13.1b, is discussed. Here ferromagnetic elements and paramagnetic channel are arranged in one plane. Two ferromagnetic strips with a distance less than the spin-relaxation length λ_{sf} of the connecting paramagnetic strip are used as spin-polarized injector and detector electrodes. The first who presented an all-electrical measurement setup were Johnson and Silsbee in 1985 [6]. In their so-called nonlocal measurement geometry, they electrically detected the coupling between electronic charge and spin at an interface between a ferromagnetic (permalloy) and paramagnetic (aluminum)

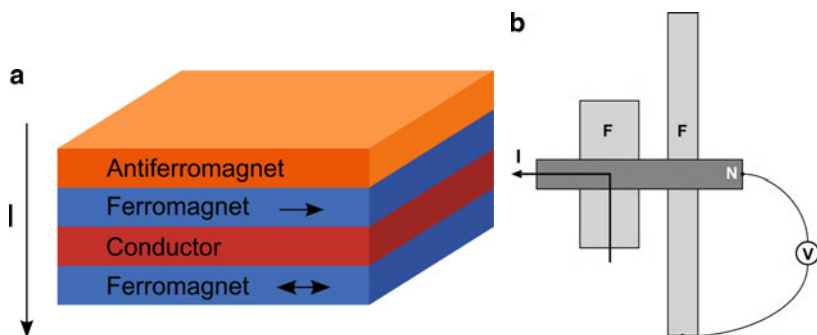


Fig. 13.1 (a) Multilayer design of a spin valve sending the current perpendicular to the ferromagnet plane and (b) lateral design with nonlocal measurement geometry

metal. At the injector interface, a spin accumulation emerges in the paramagnetic channel because of a charge current driven from the ferromagnet into the metal. This nonequilibrium magnetization in the paramagnetic metal can be detected as an electric voltage. If the charge current and the voltage probes in the spin valves are spatially separated, one denotes the setup a “nonlocal geometry”. There are two possible methods to observe the spin-relaxation length λ_{sf} with this setup. The first possibility is to measure the resistance change at the detector interface $\Delta R_{NL} = R_{\text{parallel}} - R_{\text{antiparallel}}$ between the parallel and the antiparallel orientation of the electrode magnetizations for different electrode distances. Subsequent comparison with a diffusive spin-transport theory yields the spin-relaxation length λ_{sf} of the paramagnetic channel. The other possibility is to use a perpendicular magnetic field that causes a precession of the injected magnetization in the paramagnetic metal. Depending on the magnetic field, one observes the nonlocal resistance for parallel and antiparallel orientation of the electrodes. Johnson et al. detected a voltage of some picovolts (pV) with a spin-precession measurement for parallel orientation of the electrode magnetizations [6].

Two years later, van Son and coworkers determined theoretically the coupling of charge current and spin current at a FN interface [7]. The conversion of spin-up and spin-down currents near a FN interface gives rise to an electrochemical potential difference of spin-up and spin-down electrons, the so-called spin accumulation. In 1988, Johnson and Silsbee published a theoretical description of a whole spin-valve device [8]. They integrated spin precession, spin relaxation, and spin diffusion in the two-dimensional diffusion equations. In the following years, the research focused mainly on lateral spin valves with a semiconducting channel replacing the paramagnetic channel [9–12]. In 2001, Jedema and coworkers could demonstrate spin accumulation and its detection at room temperature in an improved nanostructured spin-valve device [13]. In comparison to Johnson et al. 1985 (ca. 60 pV) [6], they enhanced the value of the voltage about three orders of magnitude (150–1,500 nV) and obtained a spin-relaxation time of 1,000 nm at 4.2 K and 350 nm at room temperature in copper. One year later, Jedema et al. presented spin-precession measurements of a permalloy/aluminum/permalloy-structure for parallel and antiparallel alignment of the electrode magnetizations [14]. They described their results with a one-dimensional diffusion theory that included a perpendicular magnetic field tilting the electrode magnetization out-of-plane [14, 15]. A spin-relaxation length of 600 nm for aluminum was obtained.

13.3 Spin Injection and Detection in Spin Valves

13.3.1 Theory

For simplification of the nonlocal concept, the theoretical description of spin-dependent effects is based on diffusive transport in one dimension. Assuming that the spin-relaxation length λ_{sf} is large in comparison to the mean free path of the

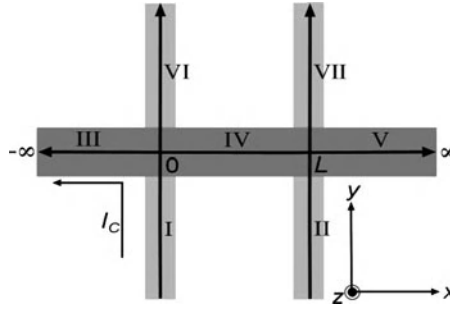


Fig. 13.2 Schematic spin-valve device subdivided into seven regions: regions I and VI denote the ferromagnetic injector electrode; regions II and VII are parts of the detector electrode; and the regions III, IV, and V belong to the interconnecting normal metal channel. The electrode spacing is L . A current I_C is driven from region I to region III and the voltage is probed at region II and V, that is, in a nonlocal measurement geometry. The space directions are defined as shown in the coordinate system

electrons, the transport of spin-up and spin-down electrons can be described independently [7]. Based on the idea of Johnson and Silsbee [8], we include spin precession, spin relaxation, spin diffusion, and tunnel barriers in the diffusion equations and solve them for the geometry depicted in Fig. 13.2. The chemical potentials μ of the electrons in the case of no charge current are derived following the approach of Kimura et al. [16]. The current in a ferromagnet exhibits the bulk spin polarization α , which yields a spin current $I_S = \alpha I_C$ when I_C is the charge current. At the boundaries of the ferromagnets, that is, at the interfaces to the normal metal, a source of spin current is assumed. The spin currents diffuse according to their conductivities partly into the ferromagnet and partly into the normal metal. As a result, the spin current at the interface within the ferromagnet is reduced in comparison to the bulk material and within the normal metal a spin current is generated. A concomitant splitting of the electrochemical potential for spin-up and spin-down electrons occurs [7, 9].

First, the derivation of the diffusion equations for the chemical potentials is described regarding spin relaxation, spin precession, and spin diffusion in a normal metal. The difference of the excess particle densities of the spin-up and spin-down electrons is defined as $\Delta n = n_\uparrow - n_\downarrow$, which we address as spin splitting in the following. In our description, no space direction is preferred, that is, spin-up and spin-down electrons can point in all three dimensions which leads to the spin splittings Δn_x , Δn_y , and Δn_z . The indices x , y , and z indicate the space direction as illustrated in Fig. 13.2. Spin precession occurs in an external magnetic field $\mathbf{H}$ that points into the z -direction. Thus, the time evolution of the spin splittings can be written as $\partial \Delta n_x / \partial t = \omega_L \Delta n_y$, $\partial \Delta n_y / \partial t = -\omega_L \Delta n_x$, and $\partial \Delta n_z / \partial t = 0$ with the Larmor frequency $\omega_L = g \mu_B \mu_0 H / \hbar$, the gyromagnetic factor g of the free electron, and the Bohr magneton μ_B . Spin relaxation is described by $\partial \Delta n / \partial t = -\Delta n / \tau_N$, where τ_N is the spin-relaxation time. Spin diffusion is given by $\partial \Delta n / \partial t = D_N \partial^2 \Delta n / \partial x^2$, where D_N is the diffusion constant. Thus, in the steady

state the diffusion equations read

$$\frac{\partial \Delta n_x}{\partial t} = \omega_L \Delta n_y - \frac{\Delta n_x}{\tau_N} + D_N \frac{\partial^2 \Delta n_x}{\partial x^2} = 0, \quad (13.1)$$

$$\frac{\partial \Delta n_y}{\partial t} = -\omega_L \Delta n_x - \frac{\Delta n_y}{\tau_N} + D_N \frac{\partial^2 \Delta n_y}{\partial x^2} = 0, \quad (13.2)$$

$$\frac{\partial \Delta n_z}{\partial t} = -\frac{\Delta n_z}{\tau_N} + D_N \frac{\partial^2 \Delta n_z}{\partial x^2} = 0. \quad (13.3)$$

These one-dimensional diffusion equations have to be solved for the nonlocal geometry depicted in Fig. 13.2. The solution is expressed in terms of the spin-splitting voltages to enable straightforward comparison with experiments. The derivation of the spin-splitting voltages ΔV_{Nx} and ΔV_{Ny} has been given in detail in [17]. The following boundary conditions are employed: the spin-splitting voltages have to be zero in the bulk far away from the interfaces, they have to be continuous at the interfaces, and the spin currents for each space direction have to be continuous. A distinction is drawn between the polarization of the tunnel current β_1 and the resulting spin polarization $P \leq \beta_1$ in the normal metal [17]. The parameter $\beta_1 \leq \alpha$ depends on the quality of the interfaces and a concomitant spin scattering. In addition, we extend the description taking into account that the spin polarization P in the normal metal drops between the electrodes because of spin relaxation [18]. An exponential decrease of the spin polarization P along the normal metal yields the spin polarization β_2 at the interface between normal metal and detector electrode [19]. Furthermore, we assume single-domain ferromagnets – implying that because of exchange coupling only one direction (here the y direction) exhibits a spin-splitting. Therefore, no spin precession occurs in the case of an undisturbed magnetization in low external magnetic fields. In experiments, the alignment of the single domain is achieved by a large shape anisotropy of the ferromagnetic electrodes, see Sect. 13.3.2. Because of its magnetization alignment, the ferromagnetic detector electrode is only sensitive to the spin-splitting voltage in y direction ΔV_{Ny} . At the detector electrode, the spin-splitting energy $e\Delta V_{Ny}$ [17] between the chemical potentials in the normal metal for electron spins pointing parallel to the magnetization of the electrodes is given via the relation

$$\begin{aligned} & \Delta V_{Ny}(L) \\ = & \frac{-Qe^{Lk_{N1}}(2\beta_1 R_{C1} + \alpha R_{F1})(2R_{C2} + R_{F2}) \left[\frac{R_{N1}}{R_{N2}} \sin(Lk_{N2}) - \cos(Lk_{N2}) \right] I_C}{2e^{2Lk_{N1}} [Q(2R_{C1} + R_{F1}) + 1] [Q(2R_{C2} + R_{F2}) + 1] - 2 \left[\frac{R_{N1}}{R_{N2}} \sin(Lk_{N2}) - \cos(Lk_{N2}) \right]^2} \end{aligned} \quad (13.4)$$

with

$$R_{C1,2} = \frac{2}{(1 - \beta_{1,2})^2 S_{C1,2} \Sigma_C},$$

$$R_{N1,2} = \frac{6}{\sigma_N S_N k_{N1,2}},$$

$$R_{F1,2} = \frac{2\lambda_F}{(1 - \alpha)^2 \sigma_F S_{C1,2}}.$$

$S_{C1,2}$ are the contact areas and Σ_C the total tunnel conductance of the interface between normal metal and electrode,¹ S_N is the cross-sectional area of the normal metal, L is the center-to-center distance between the electrodes, $\sigma_{N,F}$ is the conductivity, and $\lambda_{N,F}$ the spin-relaxation length in the normal metal and the ferromagnet, respectively. Q is the abbreviation $Q = (1 + R_{N1}^2/R_{N2}^2)/R_{N1}$. The factors k_{N1} in the exponential functions and the factor k_{N2} in the trigonometric functions are measures of the spin-relaxation and the spin-precession strength, respectively, and are defined by

$$k_{N1} = \sqrt{\frac{1}{2D_N\tau_N} \left(1 + \sqrt{1 + \omega_L^2\tau_N^2}\right)}, \quad (13.5)$$

$$k_{N2} = \frac{\omega_L\tau_N}{\sqrt{2D_N\tau_N}} \frac{1}{\sqrt{1 + \sqrt{1 + \omega_L^2\tau_N^2}}}. \quad (13.6)$$

For the calculation of the spin-splitting voltages, a typical set of parameters is used in accordance with our experiments, that is, permalloy for the ferromagnetic electrodes and aluminum for the paramagnetic channel. The conductivity of the aluminum $\sigma_N = 2.2 \times 10^7 \Omega^{-1} \text{m}^{-1}$, the conductivity of the permalloy $\sigma_F = 3.1 \times 10^6 \Omega^{-1} \text{m}^{-1}$, the average electrode spacing $L = 820 \text{ nm}$, the spin-relaxation time in aluminum $\tau_N = 7.76 \times 10^{-11} \text{ s}$, the diffusion constant in aluminum $D_N = 6.37 \times 10^{-3} \text{ m}^2 \text{ s}^{-1}$, the normalized difference in the conductances for the spin-up and spin-down electrons $\beta_1 = 0.054$, the spin-relaxation length in aluminum $\lambda_N = 703 \text{ nm}$, and the current $I_C = 50 \mu\text{A}$. We take the bulk spin polarization $\alpha = 0.35$ [20] as well as the spin-relaxation length $\lambda_F = 4.3 \text{ nm}$ [21], which cannot be deduced from our experiments. We consider tunnel barriers at the interfaces with an average total conductance per cross-sectional area of $\Sigma_C = 4.15 \times 10^{10} \Omega^{-1} \text{m}^{-2}$.

¹ Unless otherwise noted, in the following, Σ_C is the total tunnel conductance averaged over the interface between normal metal and injector electrode as well as normal metal and detector electrode.

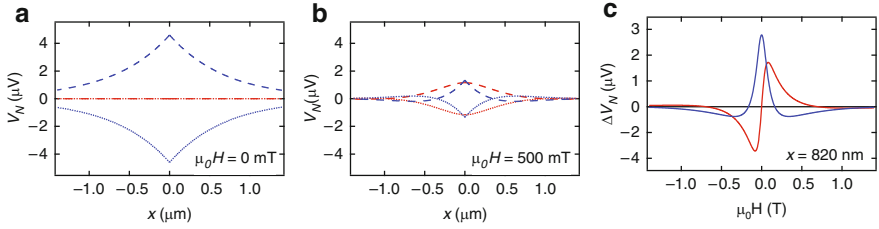


Fig. 13.3 (a), (b) Spin-resolved voltages along the normal metal, that is, aluminum. Two different external magnetic fields in z direction have been assumed: (a) 0 mT and (b) 500 mT. (c) Spin-splitting voltages ΔV_{N_x} and ΔV_{N_y} at the detector electrode in dependence of the external magnetic field applied in z direction. Dashed and dotted lines are the voltages $V_{N\uparrow}$ and $V_{N\downarrow}$ of the spin-up and the spin-down electrons. Red and blue lines correspond to the spin-splitting voltages ΔV_{N_y} and ΔV_{N_x} , respectively. The parameters are given in the text

The spin-resolved voltages are plotted along the lateral dimension of the normal metal in Figs. 13.3a and 13.3b in the absence and presence of an external magnetic field. The injector electrode is located at $x = 0$ and the detector electrode at $x = 820$ nm (see Fig. 13.2). Figure 13.3a shows the well-known exponential decrease of the spin-splitting voltages in the absence of an out-of-plane external magnetic field and therefore without spin precession. As the injected spins are parallel to the y -axis, the spin-splitting voltage ΔV_{N_x} has to be zero. With increasing external magnetic field, see Fig. 13.4b for 50 mT, the spin-splitting voltage in y direction is slightly reduced and a spin-splitting voltage in x direction occurs. In Fig. 13.3b the spin-splitting voltages at a relatively high external magnetic field of 500 mT are plotted. One observes the inversion of the spin-splitting voltages due to spin precession and a more pronounced exponential drop of the spin-splitting voltages due to the contribution of the Larmor precession (see (13.5) and (13.6)). The nonvanishing spin splitting in x direction ΔV_{N_x} at $x = 0$ might be surprising if one has a ballistic picture in mind but note that a diffusive approach in the steady state is used. The spin-splitting voltages at the detector electrode in dependence of the external magnetic field are shown in Fig. 13.3c. Without external magnetic field, the spin-splitting voltage ΔV_{N_x} is zero and ΔV_{N_y} is at its maximum. With increasing magnetic field, an oscillatory behavior of the spin-splitting voltages due to spin precession is observed. The voltages show an exponential decrease toward higher magnetic fields because of the Larmor precession, see (13.5). Hence, the spin-splitting voltages are attenuated at higher magnetic fields.

All calculations so far have been performed with tunnel barriers that have a conductance per cross-sectional area of about $4 \times 10^{10} \Omega^{-1} \text{ m}^{-2}$. The values have been obtained from measurements of the contact resistances. Tunnel barriers are known to enhance the spin-splitting voltages [22]. If the tunnel barriers are omitted, a drastic decrease of the spin-dependent effects is expected. Figure 13.4a shows the spin-splitting voltages along the lateral dimension of the normal metal in the absence of tunnel barriers at the interfaces to the ferromagnetic electrodes. An external magnetic field of 50 mT is assumed. As expected, the spin-splitting voltages are reduced

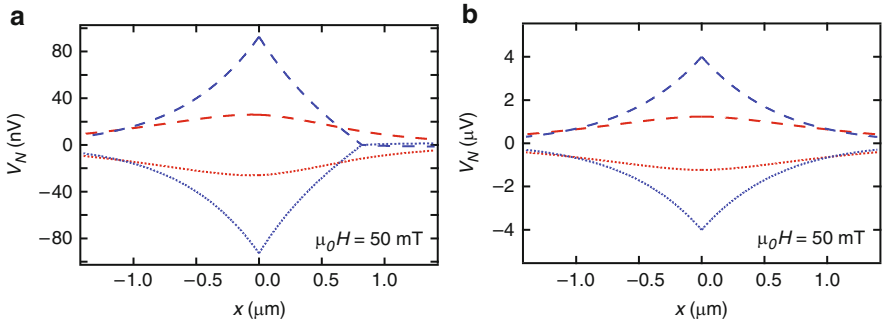


Fig. 13.4 Spin-splitting voltages in the normal metal, that is, aluminum, (a) without and (b) with tunnel barriers at the interfaces to the ferromagnetic electrodes. The external magnetic field in z direction is 50 mT. *Dashed and dotted lines* are the voltages $V_{N\uparrow}$ and $V_{N\downarrow}$ of the spin-up and the spin-down electrons. *Red and blue lines* correspond to the spin-splitting voltages ΔV_{Ny} and ΔV_{Nx} , respectively. The set of parameters is specified in the text

by two to three orders of magnitude compared to the situation with tunnel barriers, see Fig. 13.4b. The presence of tunnel barriers is crucial for the magnitude of the spin-splitting voltages for two reasons. Firstly, a tunnel barrier between the injector electrode and the normal-metal strip increases the spin-injection rate [22]. Note that the value of ΔV_{Ny} in Fig. 13.4a at $x = 0$ is smaller than in Fig. 13.4b by a factor of 50. Secondly, the tunnel barrier at the detector electrode strongly decreases the spin current into the electrode, which otherwise acts as a spin sink [8, 23, 24]. The spin splitting vanishes very fast in ferromagnetic materials because of their small spin-relaxation lengths. Without tunnel barriers, the spin current into the detector electrode intensifies the decrease of the spin-splitting voltage ΔV_{Ny} in the region of the normal metal between the injector and the detector electrode. This spin-sink effect becomes evident in the pronounced asymmetry of ΔV_{Ny} around $x = 0$ in Fig. 13.4a. The spin-splitting voltage ΔV_{Nx} is not affected in the same manner because only electrons with a spin orientation in y direction can diffuse into the detector electrode. A slight asymmetry is also observed in the shape of ΔV_{Nx} as both spin-splitting voltages are coupled via spin precession. The influence of the barrier parameters on the interface and hence on the spin-splitting voltage observed at the ferromagnetic detector electrode is described in more detail in Sect. 13.3.3.

As described in Sect. 13.2, the experimentally observed voltage is typically translated into a spin-dependent contact resistance. This nonlocal resistance

$$R_y = \pm \frac{1}{2} \frac{2\beta_2 R_{C2} + \alpha R_{F2}}{2R_{C2} + R_{F2}} \frac{\Delta V_{Ny}(L)}{I_C} \quad (13.7)$$

is the resulting voltage drop between normal metal and detector electrode normalized to the charge current I_C . The sign of the resistance R_y corresponds to the parallel and antiparallel magnetizations of the electrodes.

In a magnetic field aligned perpendicular to the sample plane, the magnetizations of the electrodes are reversibly turned away from the easy axes toward an out-of-plane state with increasing magnitude of the external magnetic field. In the limit of high magnetic fields (larger than 1.5 T), this results in an out-of-plane orientation of the magnetizations along the magnetic field in both electrodes. Therefore, no spin precession occurs anymore. Thus, the resistance saturates at the level of the parallel configuration of the magnetizations. This behavior can be described with the relation [14, 15]

$$R_H = R_y(H) \cos^2(\vartheta) + |R_y(H = 0)| \sin^2(\vartheta), \quad (13.8)$$

where ϑ is the angle between the easy axes of the electrodes and the magnetizations. This angle ϑ is zero at zero field and increases up to 90° with increasing magnitude of the external magnetic field. There are two possibilities to obtain the angle ϑ in dependence of the external magnetic field. The term $|R_y(H = 0)| \sin^2(\vartheta)$ and therewith ϑ can be obtained by a polynomial fit to the arithmetic average of the experimental curves for the parallel and antiparallel magnetization configurations. Alternatively, measurements of the anisotropic magnetoresistance (AMR) of the ferromagnetic electrodes can give direct access to the field dependence $\vartheta(H)$.

13.3.2 Permalloy Electrodes for Spin-Valve Devices

To comprehensively determine spin injection, spin diffusion, and spin precession within spin-valve devices, it is crucial to characterize the constituting materials for the electrodes and the interconnecting normal metal strip in detail. This section deals with the properties of the ferromagnetic injector and detector electrodes. The most common materials used as ferromagnetic electrodes are iron, nickel, cobalt, and their alloys [13, 14, 17, 18, 25]. The sample design and the primarily studied property determine which ferromagnetic material is to be preferred. Given that the coercive fields of injector and detector should be different for the spin-valve measurements, one can either use two different materials or different electrode shapes. We focused on the optimization of electrodes made of permalloy, a ferromagnetic alloy with the stoichiometry $\text{Ni}_{80}\text{Fe}_{20}$.

To determine the domain structure of the electrodes, first the micromagnetic behavior of the electrodes in an external magnetic field is briefly reviewed. The injector and detector should be quasi-single-domain to get an unequivocal result. Thereafter, the effect of substrate temperature during material deposition on the specific resistance is described. In the last part of this section, the dependence of spin polarization on the thickness of the deposited ferromagnet is discussed.

13.3.2.1 Magnetic Characterization of Permalloy Electrodes

For the observation of the spin-valve effect as well as the AMR of spin valves, an external magnetic field is applied to vary the magnetization of the electrodes between their two saturated states. Since the shapes of the electrodes are different, their coercive fields differ. By changing the external field to intermediate field strengths, two relative magnetic configurations are possible: a parallel and an antiparallel state.

At the edges of domains, magnetic stray fields emerge either at domain walls or at the edge of the ferromagnetic element. These stray fields can be visualized by magnetic-force microscopy (MFM). For the interpretation of transport measurements on spin-valve devices the knowledge of the micromagnetic behavior of the electrodes is important. Therefore the electrodes' magnetizations were determined at different external field strengths via MFM measurements.

Two differently prepared samples have been investigated via MFM and AMR measurements [26–28]. For the sample displayed in Figs. 13.5a–c first the paramagnetic channel, in this case made of aluminum, is deposited onto the substrate, followed by the deposition of the ferromagnetic electrodes. In Figs. 13.5d–f the electrodes have been deposited first.

Figures 13.5b,c,e,f show MFM images of the parallel and the antiparallel alignment of the magnetizations of the electrodes. For the case of the electrodes superimposed on the normal metal strip, see Figs. 13.5b,c, it is clearly visible that the electrodes consist of at least three domains. There is a small domain in the middle of each electrode and large domains at both ends of each electrode. In particular, the micromagnetic behavior of the small domain is very complex.

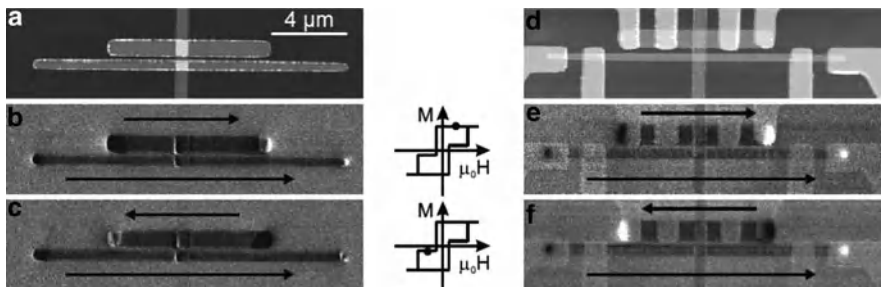


Fig. 13.5 Stray fields of permalloy electrodes recorded with a magnetic-force microscope at room temperature. (a) Scanning-electron micrograph showing the topography of the spin-valve device with a planar aluminum strip that runs from the bottom of the image to the top. The long axes of the ferromagnetic electrodes are directed horizontally. (b), (c) The magnetic configurations obtained at positive and negative external magnetic fields. The dots in the schematic hysteresis loops indicate the state of the corresponding image. The arrows illustrate the directions of the magnetizations of the electrodes. (d) Topography of the spin-valve device in the design using planar electrodes. (e), (f) The magnetizations of this device at positive and negative external magnetic fields. Adopted from [28]

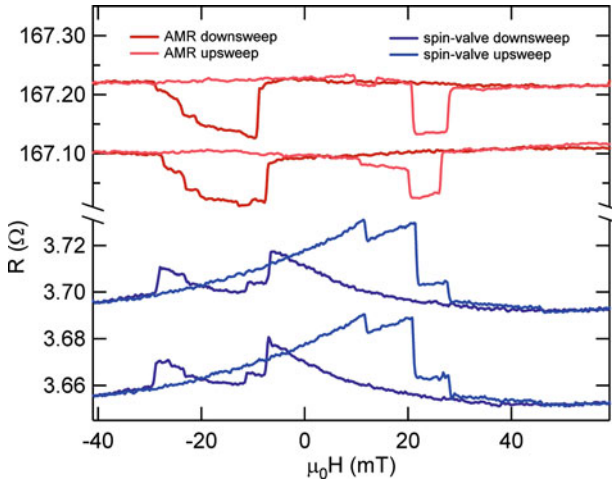


Fig. 13.6 Measurements on spin-valve devices with a planar aluminum strip and permalloy electrodes on top of this strip. The *lower half* of the picture shows data recorded in local spin-valve geometry, the *upper half* shows AMR traces of the longer electrode. Adopted from [28]

For a convincing interpretation of the transport measurements it is important that the micromagnetic behavior of the electrodes is quasi-single-domain. Additionally, in more complex micromagnetic structures conduction electron spins are scattered when a current passes a domain wall, reducing the local spin polarization and hence decreasing the spin-valve effect. To achieve simpler micromagnetic behavior, the fabrication process was adjusted as exemplified in the following.

In the second design, the fabrication of the electrodes is the initial step. Permalloy is deposited directly on the flat GaAs substrate. Again, MFM measurements [27,28], presented in Figs. 13.5e,f, were recorded. The images show that the electrodes are now quasi-single-domain. Thus, their micromagnetic structure and their hysteresis is as desired.

The complex magnetic switching behavior of the electrodes deposited on top of the aluminum strip also becomes obvious in AMR measurements [26,28] presented in Fig. 13.6. For direct comparison, AMR traces of the long electrode are shown as well as measurements of the entire device in local spin-valve geometry. In the regions of the AMR traces with reduced resistance, the long electrode changes its magnetization step by step from antiparallel to parallel with regard to the applied field. Following the spin-valve trace in Fig. 13.6, the resistance slowly rises to a maximum resulting from the reversible magnetization changes of the shorter electrode and then drops back to the initial value in three sharp flanks. These flanks coincide with the ones in the AMR traces and so are caused by the magnetization switchings of the multiple domains contained in the long electrode. AMR measurements [27,28] of flat electrodes are presented in Figs. 13.7a,b. The spin-valve signal of the entire device coincides with the AMR trace of the measurement of the long

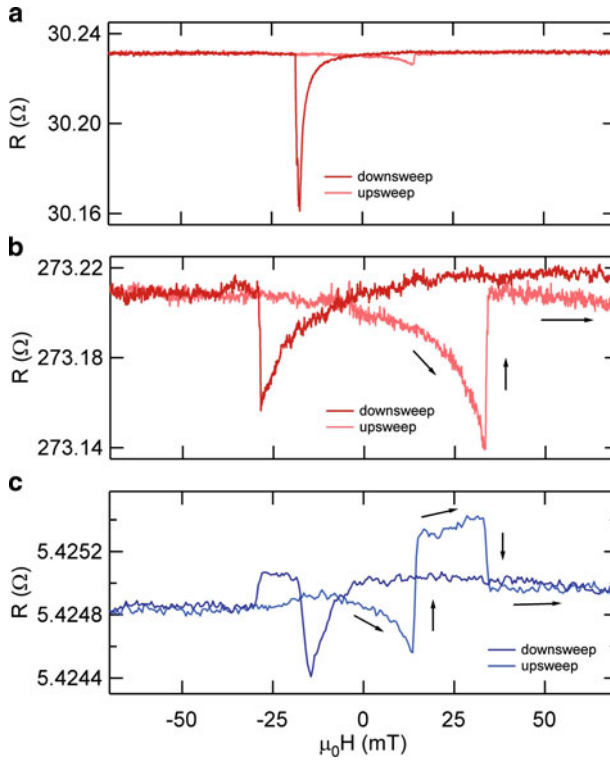


Fig. 13.7 Measurements on spin-valve devices with planar permalloy electrodes. Anisotropic magnetoresistance of (a) the shorter and (b) the longer electrodes. (c) Spin-valve signal recorded in local spin-valve geometry [28]

electrode: at a specific field strength the entire electrode switches, confirming the single-domain structure found in the MFM images.

13.3.2.2 Dependence of Specific Resistance on Substrate Temperature

In spin-valve devices, spin-polarized currents are generated in ferromagnetic electrodes. The extent of the observed spin-valve effect depends on the bulk spin polarization α . A way to increase the amplitude of the spin-valve effect is to generate a higher bulk spin polarization α . For the devices described here, permalloy is used because it reliably creates spin-polarized currents while its deposition is well controllable and its micromagnetic behavior can be tailored. The reduction of intrinsic impurities can improve the spin polarization and reduce the specific resistance. At impurities domain walls are pinned, creating spin scattering and thus reducing the spin polarization available for spin valves.

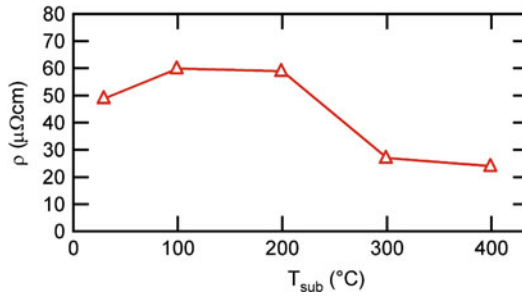


Fig. 13.8 Specific resistance ρ of permalloy thin films measured at room temperature in dependence on substrate temperature [29]

The electrodes of spin valves described so far were created by thermal evaporation. Current efforts [29, 30] deal with the reduction of the specific resistance of permalloy, because at low temperatures the resistance is dominated by intrinsic impurities. Hence a lowering would increase the spin polarization. DC-magnetron sputter deposition is used to fabricate the electrodes. The dependence of specific resistance on the substrate temperature during deposition is investigated. Measurements prove that the specific resistance of permalloy can be reduced by a factor of approximately 3 with this procedure, see Fig. 13.8. This yields specific resistances of the permalloy nanostructures of $20 \mu\Omega \text{ cm}$, which is close to the lowest value for permalloy reported in the literature [31].

13.3.2.3 Dependence of Spin Polarization on Layer Thickness

An important parameter for the characterization of spin-valve devices is the spin polarization α of a current flowing through the ferromagnet. In a design of spin valves presented by Yang et al. [32], the permalloy electrodes were adjusted to achieve a complete in situ fabrication of the samples. In this layout the current is, unlike in the typical spin-valve design, only polarized along the short path perpendicular to the ferromagnetic film. Since the layer has to be thin so as to obtain switchable electrodes, the assumption that the current obtains the bulk polarization is not a priori given. In the following paragraph, measurements are presented, where the spin polarization of a current flowing perpendicular to the plane of a ferromagnetic layer is investigated.

For reduced layer thickness d of the ferromagnetic film, one has to keep in mind that the alignment process of spins depends on the length of the path the electrons take through the material, following a Lambert law with a characteristic spin-scattering length λ_F . If the current flows perpendicular to the layer, the maximum length in which the current can be polarized is the film thickness d . A simple description of the dependence of spin polarization on layer thickness has to include

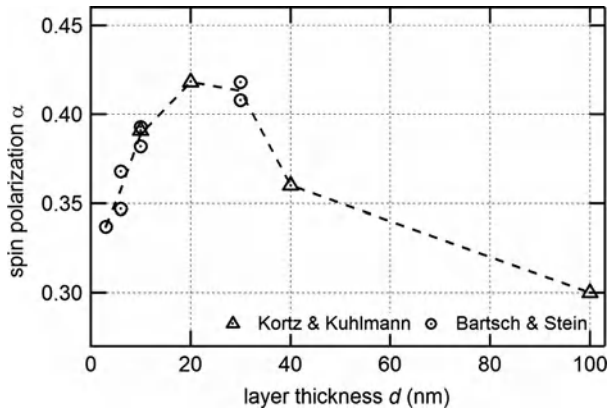


Fig. 13.9 Measured spin polarization versus thickness of the permalloy layer. Results of two series of measurements depicted by *triangles* (first series) and *circles* (second series). The *dashed line* is a guide to the eyes

the two limiting cases: the polarization should be at maximum for bulk material and obviously should fall to zero for vanishing thickness. In between, the polarization should be reduced significantly for layer thicknesses below the spin-relaxation length λ_F .

There are only few experimental ways to independently determine the spin polarization of a current. A comparatively simple technique is point-contact Andreev-reflection (PCAR) spectroscopy. The well-known and intensively used technique [33–35] utilizes spin-dependent Andreev-reflection [36] of an electron into a hole at a metal–superconductor interface. A deeper insight into the method and the theoretical background can be found in Chap. 14² as well as in [20, 33–36].

To systematically investigate the dependence of the spin polarization α on the layer thickness d , various samples with different thicknesses of permalloy were fabricated and their spin polarization was measured via PCAR spectroscopy. To ensure that the current flows only perpendicular to the layer plane, the permalloy films were deposited on 100 nm of gold, which provides a specific resistance one order of magnitude smaller than the value of permalloy. A more detailed description of the sample design and the current paths are given in [20].

The dependency of the spin polarization α on thickness d is presented in Fig. 13.9. There is indeed a correlation between layer thickness and spin polarization. Both are linked in a nontrivial way. From a simple image, an exponential decay could have been expected. In the data, a maximum is found for a thickness $d \approx 20$ nm and not for bulk as initially expected.

The physical origin of this behavior is currently under discussion. In a first approach, different spin-relaxation lengths $\lambda_{F,\uparrow(\downarrow)}$ for spin-up and spin-down charge

² The spin polarization α is addressed as P in Chap. 14.

carriers [21] are taken into account. At a certain distance x from the metal–ferromagnet interface, the charge carriers with the shorter spin-diffusion length are already fully aligned yet the others are not. In this regime, the polarization would by definition be higher than in the bulk. The expected layer thickness to observe this maximum should lie between the different spin-relaxation lengths for spin-up and spin-down electrons and hence at about 5 nm [21, 37]. Although the value belonging to the measured maximum differs from this expectation by a factor of 4, the idea is promising. Since a small variation in the specific resistance results in a significant change of λ_F , the values in the publication by Dubois et al. [21] may differ from those in the present experiment.

The publication by Yang et al. [32] reveals an interesting analogy to the results presented here. The thickness of the permalloy layer in the proposed design for the spin-valve devices of Yang et al. equals 20 nm. This is in accordance with the layer thickness where maximum spin polarization was found in Fig. 13.9. Although no reason is given why this layer thickness is used by Yang et al., it is possible that the value resulted from an optimization process for the spin-valve effect. There is a strong demand for more detailed studies of optimized layer thicknesses for the injection electrodes in spin-valve devices.

13.3.3 Spin Valves with Insulating Barriers

The quality of the interface between the ferromagnetic electrodes and the normal metal is crucial for high spin injection and its successful detection both with and without tunnel barriers. In this section, the focus will be on spin valves with FIN interfaces.

In 2000, Rashba found a theoretical solution for the conductivity mismatch by inserting a tunnel barrier in-between ferromagnetic metal and semiconductor [22]. For efficient spin injection, the ability of tunnel contacts to support a considerable difference in electrochemical potentials under the conditions of slow spin relaxation is of importance. Fisher and Giaever have proven with their pioneering experiments that electrons can tunnel from one metallic electrode through a thin tunnel barrier into a second metallic electrode [38]. To calculate the total current density through the tunnel barrier, one has to include the current density $j_{a \rightarrow b}$ from electrode a to electrode b and for the inverse current density $j_{b \rightarrow a}$. The difference leads to the total current density through the tunnel barrier:

$$j(V) = \frac{4\pi e}{\hbar} \sum_{k_t} \int_{-\infty}^{\infty} |M_{ab}(E)|^2 \rho_a(E) \rho_b(E - eV) [f_a(E) - f_b(E - eV)] dE. \quad (13.9)$$

In this integral k_t is the transverse momentum, $M_{ab}(E)$ is the matrix element for the transition, $\rho_{a,b}(E)$ is the density of states in electrode a,b, $f(E)$ is the Fermi distribution function, and V is the bias voltage across the tunnel barrier [39]. Solving the

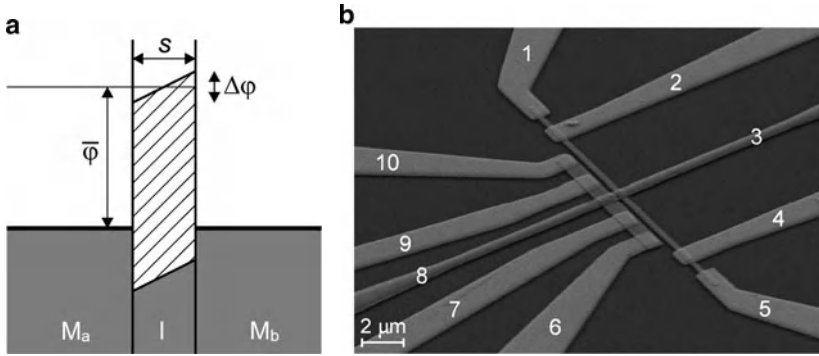


Fig. 13.10 (a) Sketch of a trapezoidal barrier at zero bias with the average barrier height $\bar{\varphi}$ and the barrier asymmetry $\Delta\varphi$. Trapezoidal tunnel barriers are described by Brinkman's theory [40]. (b) Scanning-electron micrograph of a spin-valve device. Two permalloy electrodes are contacted via eight gold leads, numbered 1–2, 4–7, and 9–10. The aluminum strip running from the *left* to the *right* is contacted via the leads 8 and 3. Adopted from [19]

integral is the main obstacle to find a handy expression for the description of the current through tunnel barriers [59]. In the following, a solution derived by Brinkman et al. [40] is presented. When trapezoidal barriers are assumed, see Fig. 13.10a, beside the thickness s the asymmetry of the barrier $\Delta\varphi$ and the average barrier height $\bar{\varphi}$ is needed. Brinkman et al. have calculated the tunneling current numerically and have found a parabolic dependency between the differential conductance $dG(V)$ and the applied bias voltage for low voltages ($\lesssim 0.4$ V):

$$\frac{dG(V)}{G(0)} = 1 - \left(\frac{C \Delta\varphi}{16\bar{\varphi}^{\frac{3}{2}}} \right) eV + \left(\frac{9}{128} \frac{C^2}{\bar{\varphi}} \right) (eV)^2 \quad (13.10)$$

with $G(0) = \frac{e^2 A \sqrt{2m\bar{\varphi}}}{h^2 s} \exp\left(-\frac{2s}{\hbar} \sqrt{2m\bar{\varphi}}\right).$

$G(0)$ is the differential conductance at zero bias and C is the abbreviation $C = 4s\sqrt{2m}/(3\hbar)$ with the electron mass m and the cross-sectional area of the tunnel contact A . Characteristic parameters of a tunnel barrier can be obtained from the coefficients of a fit of the measured differential conductance as a function of the bias voltage. A parabolic fit yields the coefficients $K_0(\bar{\varphi}, s)$, $K_1(\bar{\varphi}, \Delta\varphi, s)$, and $K_2(\bar{\varphi}, s)$:

$$dG(V) = \frac{dI}{dV} = K_0(\bar{\varphi}, s) + K_1(\bar{\varphi}, \Delta\varphi, s)V + K_2(\bar{\varphi}, s)V^2. \quad (13.11)$$

From these coefficients, the average height, the thickness, and the asymmetry of the tunnel barrier result:

$$\begin{aligned}
\bar{\varphi} &= \frac{e}{4} \sqrt{\frac{K_0}{2K_2}} \left| \ln \left(\sqrt{K_0 K_2} \frac{h^3}{\sqrt{2} e^3 m \pi A} \right) \right|, \\
s &= \frac{2\hbar}{e} \sqrt{\frac{K_2}{K_0} \bar{\varphi}}, \\
\Delta\varphi &= -\frac{K_1}{K_0} \frac{12\hbar \bar{\varphi}^{\frac{3}{2}}}{\sqrt{2} m e s}.
\end{aligned} \tag{13.12}$$

A detailed experimental investigation of tunnel barriers and comparison with theory is important to understand the barriers' influence on the spin polarization injected into the normal metal. In the following, the influence of aluminum oxide tunnel barriers and their parameters, that is, average barrier height $\bar{\varphi}$, barrier asymmetry $\Delta\varphi$, barrier thickness s , and average total conductance Σ_C per cross-sectional area of the tunnel barriers, on the injection and detection of spin-polarized currents in lateral spin valves is reported. Valenzuela and Tinkham [25] observed a linear increase of the spin polarization as the barrier transparency decreased. We included AlO_x barriers with different thicknesses s and average heights $\bar{\varphi}$ into spin-valve devices by varying the oxygen pressure, the oxidation time, and the thickness of the oxidized aluminum strip systematically. The particular properties of the tunnel barriers are ascertained via measurements of the current–voltage characteristic and the differential conductance as functions of the bias voltage [19].

Spin-valve devices are fabricated in three steps using electron-beam lithography and lift-off processing. First, two permalloy electrodes with lateral dimensions of $8\,\mu\text{m} \times 0.81\,\mu\text{m}$ and $16\,\mu\text{m} \times 0.27\,\mu\text{m}$ are thermally evaporated onto a Si/SiO_2 substrate. For details, see [17, 27] and Sect. 13.3.2. The center-to-center distance between the electrodes is $L = 820\,\text{nm}$ and the thickness is $30\,\text{nm}$. The surface is cleaned by RF argon-plasma etching to improve the interface quality. Subsequently, an aluminum strip with a nominal thickness d_{Al} between 1 and 3 nm is deposited on top of the electrodes using DC-magnetron sputtering. A tunnel barrier is formed via oxidation in pure oxygen for $t = 5\,\text{min}$ up to $t = 30\,\text{min}$ at a pressure p between 0.01 and 200 mbar. After the oxidation process, an aluminum strip with a width of 550 nm and a thickness of 50 nm is deposited. The average total conductance Σ_C per cross-sectional area of the tunnel barriers is determined by its thickness s and the average barrier height $\bar{\varphi}$. A characterization of the barrier formation is required. Spin-valve devices with nine different sets of process parameters (see Fig. 13.11a–c) have been fabricated to investigate their influence on the properties of the aluminum oxide barriers. The specific values of d_{Al} , p , and t are modeled via the method experimental design described in [41]. Measurements of the current–voltage characteristic and the differential conductance as a function of the bias voltage have been performed at temperatures of liquid helium. The data are consistent with the characteristic shape for tunnel barriers as described by the theory of Brinkman et al. [40]. We observe an increasing tunnel conductance between $T = 2\,\text{K}$ and room temperature, which indicates pinhole-free tunnel

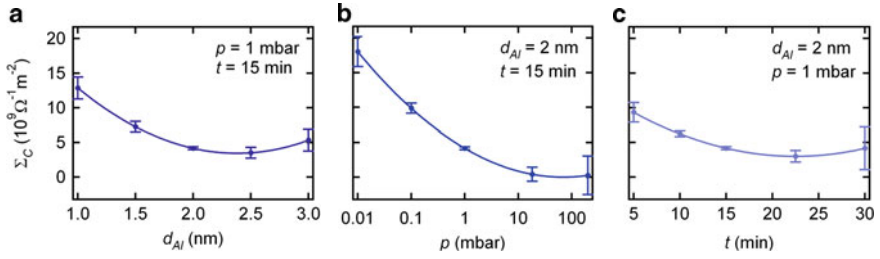


Fig. 13.11 Dependence of the average total conductance Σ_C per cross-sectional area of the tunnel barriers on (a) the nominal thickness of the aluminum strip d_{Al} , (b) the oxygen pressure p , and (c) the oxidation time t

barriers [42, 43]. Depending on the process parameters, we obtain a barrier thickness between $s = 1.05 \text{ nm}$ and $s = 1.45 \text{ nm}$ and an average barrier height between $\bar{\varphi} = 0.19 \text{ eV}$ and $\bar{\varphi} = 1.98 \text{ eV}$. Average total conductances per cross-sectional area from $\Sigma_C = 1.42 \times 10^9 \Omega^{-1} \text{m}^{-2}$ up to $\Sigma_C = 3.92 \times 10^{13} \Omega^{-1} \text{m}^{-2}$ are achieved. For $d_{\text{Al}} = 1 \text{ nm}$, $p = 0.01 \text{ mbar}$, and $t = 5 \text{ min}$, no verifiable tunnel barrier has been formed. In the framework of experimental design, we approximate the relation between the properties of the tunnel barriers and the process parameters via a quadratic polynomial fit. Figure 13.11a–c show the functional dependence of the average total conductance Σ_C per cross-sectional area on d_{Al} , p , and t within the analyzed range. It decreases with the nominal thickness d_{Al} of the oxidized aluminum layer. The slight increase for $d_{\text{Al}} \geq 2.5 \text{ nm}$ can be ascribed to an artifact due to the fit function. The stoichiometric portion of oxygen in the aluminum oxide barrier increases with the oxygen pressure p and the oxidation time t [44, 45]. Hence, the decrease of the tunnel conductance Σ_C in Figs. 13.11b,c can be explained by an increase of the average barrier height $\bar{\varphi}$.

In Fig. 13.12, the experimental data of the change in the nonlocal resistance $\Delta R_{\text{NL}} = R_{y,\text{parallel}} - R_{y,\text{antiparallel}}$ are compared to theory, see (13.7). Our experimental data for the nonlocal spin-valve effect follow the theoretical curve in Fig. 13.12. We observe a saturation for tunnel conductances $\Sigma_C \leq 6 \times 10^{10} \Omega^{-1} \text{m}^{-2}$. The maximum spin polarization $P = 3.7\%$ in the normal metal is in good agreement with results reported in other publications [13, 46].

13.3.4 Connecting Paramagnetic Channel

This section deals with spin-polarized carriers in the paramagnetic channel of the spin-valve device with planar permalloy electrodes (see Sect. 13.3.2). The most common metals used as paramagnetic channel are copper [13, 47, 48], aluminum [6, 14, 17, 19, 25], gold [46, 49], and silver [50, 51]. Recently, successful spin injection, detection, and precession was observed in the semiconductor GaAs [18] and graphene [52, 53]. In the following, we will focus on spin-valve and spin-precession

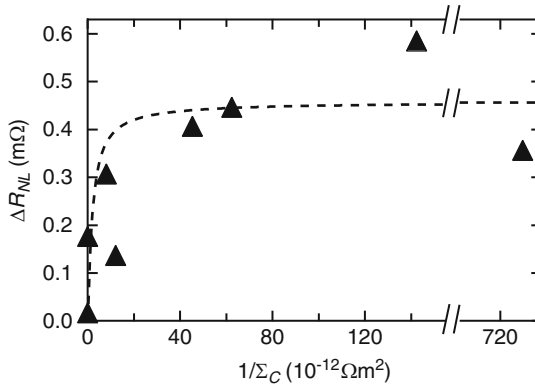


Fig. 13.12 Nonlocal spin-valve effect ΔR_{NL} versus average total tunnel conductance Σ_C of the tunnel barriers at the injector electrode and the detector electrode. The *dashed line* marks the theoretical change of ΔR_{NL} for a fixed polarization of the tunnel current $\beta_1 = 0.037$. *Black triangles* depict the experimental data

measurements in copper and aluminum. In the last part of this section, we will discuss the results reported in the literature for different paramagnetic channels. We list them in Table 13.1.

Nonlocal spin-transport measurements using lock-in techniques have been performed at temperatures of liquid helium. Typical results are shown in Figs. 13.13a–d. A current of amplitude $I_C = 50 \mu A$ and frequency $f = 67.3$ Hz is sent from the injector electrode into the aluminum strip. By applying an external magnetic field parallel to the long axes of the electrodes, the magnetization is switched between the parallel and the antiparallel orientation. The coercive fields of the two electrodes are determined via the AMR, see Sect. 13.3.2 and [17, 27]. In hysteresis loops, slightly different values are found for each of the four coercive fields. The average values for the aluminum (copper) device are -18 and 3 mT (-18 and 3 mT) for the shorter and -25.0 and 13 mT (-23.0 and 11 mT) for the longer electrode. All coercive fields are in a range of ± 2 mT around the average values. The coercive fields of both electrodes are not symmetric to zero field for two reasons: the superconducting solenoid, which produces the external magnetic fields, has a remanence of 8 mT. Secondly, during and after their preparation, the permalloy electrodes are oxidized at the surface at ambient air. This presumably produces a thin antiferromagnetic layer that shifts the hysteresis loops by a few millitelsa because of exchange-bias coupling [54].

Spin-valve and spin-precession experiments probe the voltage between the metal strip and the detector electrode. This voltage normalized with the charge current I_C is the nonlocal resistance R_y , see (13.7). Its sign corresponds to the parallel or to the antiparallel orientation of the magnetizations of the electrodes. In the measurements, the nonlocal resistance is not symmetric around zero. For clarity, we distinguish between the theoretical nonlocal resistance R_y and the observed nonlocal resistance R_{NL} with an offset. The spin-valve experiments have been performed

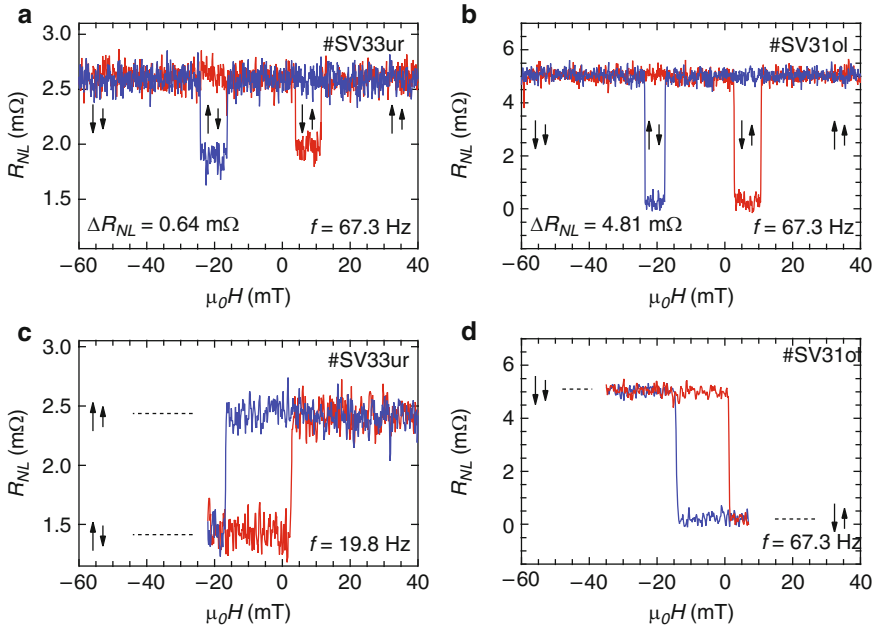


Fig. 13.13 Magnetoresistance of a spin valve recorded in the nonlocal geometry with an (a) aluminum and (b) copper channel, $I_C = 50 \mu\text{A}$. Minor loops of the magnetoresistance are depicted starting at (c) positive saturation fields for the aluminum and (d) negative saturation fields for the copper channel. Red and blue lines denote the positive and negative sweep direction of the external magnetic field. Arrows depict the orientation of the electrodes' magnetization

with the external magnetic field applied parallel to the long axes of the electrodes in y direction. In this case, no spin precession occurs because the spins already point in the y direction due to the magnetizations of the electrodes. The spin-valve effect is explained with the theoretical description by setting the external magnetic field in z direction to zero ($H_z = 0$). Only two values are possible for R_y in accordance with the parallel and the antiparallel configuration of the magnetizations of the electrodes. The external magnetic field switches the magnetizations between these two states. The nonlocal magnetoresistance of a spin valve measured with lock-in technique at a current amplitude of $I_C = 50 \mu\text{A}$ at a temperature of 1.6 K for aluminum and copper is shown in Figs. 13.13a,b. Red and blue lines denote the positive and the negative sweep direction of the external magnetic field. Following the magnetoresistance in Fig. 13.13a in the positive sweep direction of the external magnetic field, the signal remains on the same level as at negative saturation fields until the coercive field of the shorter electrode at $(3 \pm 2) \text{ mT}$ is reached. Then the resistance drops to a lower level and remains the same up to the coercive field of the longer electrode at $(13 \pm 2) \text{ mT}$. Finally, the resistance increases back to the initial level. The regions with increased resistance correspond to the parallel configurations of the magnetizations and the regions with decreased resistance

correspond to the antiparallel configurations. Note that in the nonlocal measurement, one obtains a pure spin-valve signal because of magnetization changes of the electrodes without contributions of parasitic effects like the AMR or the local Hall effect [27].

Minor loops have been recorded to support our interpretation and are displayed in Fig. 13.13c for the aluminum channel starting at positive saturation fields and in Fig. 13.13d for the copper channel starting at negative saturation fields.³ Following, for example, the curves in Fig. 13.13d with a starting field at negative saturation, the resistance remains at the same level up to the positive coercive field of the shorter electrode and then drops to the resistance level of the antiparallel configuration. The turning point of the sweep of the external magnetic field is between the coercive fields of both electrodes. While the external magnetic field is swept in the negative direction, the resistance remains at the decreased level until the negative coercive field of the shorter electrode is reached. Then the resistance increases back to its initial value at negative saturation fields. Thus, only two parallel and two antiparallel alignments of the magnetizations are observed resembling the genuine spin-valve behavior.

The comparison of the spin-valve measurements for copper with those for aluminum shows an eight times higher value of ΔR_{NL} for the spin-valve device with copper channel. The Al_xO_y tunnel barriers for both materials are formed with the same set of parameters (oxygen pressure $p \approx 1$ mbar, the oxidation time $t \approx 15$ min, thickness of the oxidized aluminum strip $d \approx 2$ nm). Calculations yield a spin polarization of the tunnel current of $\beta_1 = 0.054 \pm 0.003$ for the device with aluminum channel and $\beta_1 = 0.156 \pm 0.013$ for the device with copper channel.⁴

Next, the experiments on spin precession are presented. The external magnetic field is applied perpendicular to the sample plane in the z direction (see Fig. 13.2). Measurements are shown in Figs. 13.14a,b. Dark and light blue lines correspond to the parallel and the antiparallel configuration of the magnetizations of the electrodes at zero field, respectively. Spin precession is observed in aluminum (Fig. 13.14a) and copper (Fig. 13.14b). In these graphs, the solid lines are fits to the measured data based on the theoretical description in Sect. 13.3.1. In the limit of high magnetic fields (larger than 1.5 T), the magnetizations are out-of-plane along the magnetic field in both electrodes. Therefore, no spin precession occurs anymore and the resistance saturates at the level of the parallel configuration of the magnetizations. This behavior is described with (13.8) as introduced in Sect. 13.3.1. The angle ϑ between the easy axes of the electrodes and the magnetizations is zero at zero field and increases up to 90° with increasing magnitude of the external magnetic field. The term $|R_y(H = 0)| \sin^2(\vartheta)$ in (13.8) and therewith the angle ϑ can be obtained

³ The different amplitudes in Figs. 13.13a and 13.13c are caused by different lock-in frequencies of 67.3 and 19.8 Hz, respectively.

⁴ These calculations were performed assuming that the total conductances of the injector and detector interface are different. For the spin valve with aluminum channel the conductances have the value $\Sigma_{\text{C1}} = 3.7 \times 10^9 \Omega^{-1} \text{ m}^{-2}$ and $\Sigma_{\text{C2}} = 4.6 \times 10^{10} \Omega^{-1} \text{ m}^{-2}$ and for the one with the copper channel $\Sigma_{\text{C1}} = 8.95 \times 10^9 \Omega^{-1} \text{ m}^{-2}$ and $\Sigma_{\text{C2}} = 2.95 \times 10^{10} \Omega^{-1} \text{ m}^{-2}$.

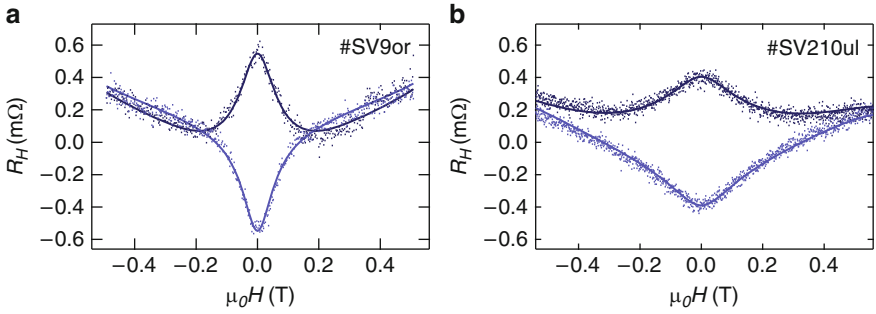


Fig. 13.14 Magnetoresistance of the spin-valve device with (a) aluminum and (b) copper channel recorded in the nonlocal geometry with the external magnetic field pointing out-of-plane (z direction). Dark blue lines correspond to the parallel orientation of the magnetizations of the electrodes at zero field, light blue lines to the antiparallel orientation. Offsets in the magnetic field of -8 mT and in the resistance have been subtracted from the experimental data. Solid lines are fits according to (13.8)

by a polynomial fit to the arithmetic average of the two experimental curves as $R_y(H)$ changes its sign when the magnetization configuration is switched from parallel to antiparallel. Thus, the term $R_y(H) \cos^2(\vartheta)$ is eliminated in the arithmetic average. The polynomial fit has to be mirror symmetric to $H = 0$ and was applied up to the sixth order. The results of the polynomial fit for $\vartheta(H)$ are used in the fit procedure of the measured resistances with (13.8). The following material parameters have been used for the fits: the bulk spin polarization $\alpha = 0.35$ and the spin-relaxation length in permalloy $\lambda_F = 4.3$ nm have been taken from the literature [20,21]. The conductivities $\sigma_{Al} = 2.2 \times 10^7 \Omega^{-1} \text{m}^{-1}$, $\sigma_{Cu} = 10.67 \times 10^7 \Omega^{-1} \text{m}^{-1}$, $\sigma_F = 3.1 \times 10^6 \Omega^{-1} \text{m}^{-1}$, as well as the total conductance⁵ per cross-sectional area of the tunnel barrier have been determined from the sample. All cross-sectional areas have been deduced from the device geometry and an average electrode spacing of $L = 820$ nm from the center of one electrode to the center of the other has been taken. Fit parameters are $\tau_N = 7.76 \times 10^{-11}$ s, $D_N = 6.37 \times 10^{-2} \text{m}^2 \text{s}^{-1}$, and $\beta_1 = 0.054$ for the aluminum channel and $\tau_N = 6.83 \times 10^{-11}$ s, $D_N = 9.67 \times 10^{-2} \text{m}^2 \text{s}^{-1}$, and $\beta_1 = 0.043$ for the copper channel. This leads to spin-relaxation lengths of $\lambda_{Al} = 703$ nm and $\lambda_{Cu} = 2,571$ nm.

As a summary in Table 13.1, results of different publications for the spin-relaxation length in different paramagnetic channels are listed. The method used to determine λ_{sf} , that is, spin-valve measurements with different average electrode spacings (SV) or spin-precession measurement (SP), the temperature, and the conductivity of the paramagnetic channel are quoted if available.

⁵ $\Sigma_{Cl,Al} = 4.6 \times 10^{10} \Omega^{-1} \text{m}^{-2}$, $\Sigma_{C2,Al} = 3.7 \times 10^{10} \Omega^{-1} \text{m}^{-2}$, $\Sigma_{Cl,Cu} = 1.77 \times 10^{12} \Omega^{-1} \text{m}^{-2}$, and $\Sigma_{C2,Cu} = 2.05 \times 10^{12} \Omega^{-1} \text{m}^{-2}$.

Table 13.1 Overview of results for different paramagnetic channels

Material	Method	Temperature (K)	λ_{sf} (nm)	Conductivity ($1/\Omega$ m)	Publ.
Ag	SV	300	700	5.0×10^7	[50]
	SV	77	3,000	9.1×10^7	[50]
	SP	40	564	5.9×10^7	[51]
Al	SV	300	350	1.1×10^7	[14]
	SV	4.2	650	1.7×10^7	[14]
	SP	4.2	600	1.7×10^7	[14]
	SP	4.2	703	2.2×10^7	[19]
Au	SV	10	63	5.0×10^7	[46]
	SV	15	168	2.5×10^7	[49]
Cu	SV	300	350	3.5×10^7	[13]
	SV	300	400	4.3×10^7	[48]
	SV	4.2	1,000	7.0×10^7	[13]
	SV	10	1,000	14.5×10^7	[48]
	SP	4.2	2,571	10.7×10^7	
GaAs	SP	50	6,000		[18]
Graphene	SP	300	1,300–2,000	$1.1\text{--}4.2 \times 10^6$	[52]

13.4 Outlook

In spintronics, injection, transport, and all-electrical detection of highly spin-polarized currents in paramagnetic channels are the main challenges. Recent experiments with semiconducting materials as connecting channels are also promising [18, 52, 53]. Experiments concerning the high-frequency properties and potential applications of lateral spin-valve devices and their components are also in the focus of interest. In the literature, the precessing magnetization of a ferromagnet is discussed as an injector of a spin current into adjacent conductors via Ohmic contacts [55]. This so-called spin pumping has been measured by converting the spin accumulation into a voltage using the precessing magnetization as its own detector [56]. Recently, the ferromagnetic resonance of a single submicron ferromagnetic strip has been detected in an on-chip microwave transmission line [57]. In first attempts, spin-valve devices and their components are used to detect the spin Hall effect [58] and to reversibly induce a magnetization switching of a ferromagnetic particle with pure spin current [32].

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Chapter 14

Growth and Characterization of Ferromagnetic Alloys for Spin Injection

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Abstract Spin electronics with semiconductor/ferromagnet hybrids is a topic of ongoing interest. We review developments in hybrid spintronics and give an overview of achievements in efficient spin injection from ferromagnetic metals into semiconductors. The focus of this work is on thin Heusler films grown on semiconductor substrates. Ni_2MnIn films are deposited on a variety of substrates by coevaporation of nickel and the alloy MnIn . The almost perfect lattice match between Ni_2MnIn and InAs qualifies this alloy for basic research in spintronics. Point-contact Andreev spectroscopy serves to quantify the spin polarization relevant to transport. Nanopatterning of Ni_2MnIn electrodes with electron-beam lithography and lift-off processing is examined. In this context, the influence of post-growth annealing on the film's morphology and crystal structure is studied in situ using transmission-electron microscopy. The electrodes are completed by a copper strip to form a lateral spin-valve. In first measurements in local geometry we have detected the spin-valve effect.

14.1 Introduction

In mainstream semiconductor electronics, the spin of the electron is ignored. A field called spintronics has emerged where the electron spin carries information beside its charge. This offers opportunities for a new generation of devices combining standard semiconductor electronics based on charge with spin-dependent effects.

Use of the spin in digital information processing is based on its alignment, up or down relative to an axis of reference. This axis can be defined by an applied magnetic field, the magnetization direction of a ferromagnetic microstructure, or the crystal direction of a crystal with non-vanishing spin-orbit interaction. Adding the spin degree of freedom to semiconductor electronics will enhance its capability and performance. Expected merits of such new devices are non-volatility, increased processing speed, decreased power consumption, and increased integration density compared with conventional semiconductor devices. Major challenges in the field of spintronics that are addressed by experiment and theory include the optimization

of spin lifetimes, the detection of spin coherence in nanostructures, transport of spin-polarized carriers over relevant length scales and across heterointerfaces, and the manipulation of spins on sufficient fast timescales. Optical methods for spin injection, detection, and manipulation have shown the ability to determine the above-mentioned quantities precisely. They are also successfully applied to determine the interaction of the electron spin with nuclear spins, photons, and magnetic fields.

Magnetism has always been important for information storage and it is therefore no surprise that this field provided the initial success in the applications of spin-based electronics. High-capacity hard drives nowadays rely on a spintronic effect, the giant magnetoresistance (GMR), to read data. More sophisticated storage technologies making use of the spin are in an advanced state. Magnetic random access memories (MRAM) are non-volatile memories with high switching rates and rewritability challenging semiconductor-based RAM. The unique feature that semiconductors bring into spintronics is their tunability. Only semiconductors provide bandstructure engineering, modulation doping in heterostructures, and tuning carrier concentration with gates. The ability of semiconductors to amplify optical and electrical signals is a consequence of their tunability. Achieving practical spintronic devices based on semiconductors would allow a wealth of new types of devices and improved functionalities. It is envisioned that the merging of semiconductor-based electronics, photonics, and magnetics will ultimately lead to spin-based devices, such as spin field-effect transistors, spin light-emitting diodes, spin resonant-tunneling devices, fast optical switches, modulators, encoders, decoders, and quantum bits for quantum computation and communication [1]. The success of the envisioned devices depends on a deeper understanding of the fundamental spin interactions in solids, in particular the roles of dimensionality, defects, and spin-orbit interaction. To summarize, the prospects of the control of the spin degree of freedom in semiconductors, semiconductor heterostructures, and ferromagnets will offer a potential for high-performance spin-based semiconductor electronics.

The focus of this work is the development of components for semiconductor/ferromagnet hybrid devices and their integration [1–7]. The idea of a spin transistor [7] has created a new branch of research in solid-state physics, combining semiconductors with ferromagnetic metals or utilizing ferromagnetic semiconductors in all-semiconductor devices [8]. Currently, injection, transport, and detection of spin-polarized electrons in semiconductors are investigated. On the way to a possible spin transistor and related devices, a multitude of issues must be addressed. First of all, the injection of spin-polarized carriers from a ferromagnetic metal into a semiconductor has to be demonstrated. This problem has been discussed controversially but now a consensus on the basic principles has been reached. At least this holds for the limiting cases of diffusive [9] and ballistic [10] spin transport in the semiconductor. However, up to now in semiconductor/ferromagnet hybrid devices the suppression of spin scattering at a heterointerface remains a major challenge.

In epitaxially grown diluted magnetic II–VI semiconductors, the electron spin could be aligned in external magnetic fields and the spin polarization was detected

via the circular polarization of light emitted from an integrated light-emitting diode (LED) [11]. Such spin-LEDs have been built with ferromagnetic metals as spin injectors [6, 12]. These experiments exhibit spin injection rates of up to 30% at liquid helium temperatures [6]. Even at room temperature, a spin-injection rate of 2% has been achieved in such a device [12]. In other optical experiments, up to 9% of spin-injection efficiency has been demonstrated at a temperature of 80 K with cobalt as ferromagnetic injector [13]. Schottky barriers [6, 12] or tunneling barriers [13] play an important role in all these optical devices.

Efficient spin injection from ferromagnets into semiconductors is much more difficult to detect in pure transport experiments and was demonstrated as late as in 2007 [14], almost two decades after the proposal of the spin transistor by Datta and Das [7]. The experimental difficulties gave rise to theoretical works on the transport processes at the ferromagnet/semiconductor interface. In case of diffusive transport significant spin injection rates are only obtained for a spin polarization close to 100% in the injecting contacts [9]. Half-metallic magnets could provide such high spin polarizations at the Fermi energy [15, 16]. In particular, the Heusler alloy Ni_2MnIn , whose lattice constant almost perfectly matches the one of InAs [17], is predicted to exhibit 100% spin polarization at epitaxial interfaces to this semiconductor [18].

Pioneering experiments on quasi-ballistic ferromagnet/semiconductor hybrid devices without gate electrodes exhibited spin-dependent transport with resistance changes in the range of 0.1% [19]. In hybrid transistors with ferromagnetic contacts on InAs, we could observe a spin-related magnetoresistance in the order of 1%, which could be tuned by the gate voltage already in 2002 [20]. However, possible parasitic magnetoresistance effects such as anisotropic magnetoresistance in the ferromagnetic contacts or local Hall effects in the two-dimensional electron system require a detailed analysis of the dependency on temperature, gate voltage, and magnetic-field strength to prove spin-dependent transport.

As in the optical investigations tunneling barriers should improve the spin-injection rate and the spin-related magnetoresistance. In fact, calculations on the influence of barriers predict an improvement in the diffusive [21] as well as in the ballistic [10] limit. Transport experiments on hybrid structures, which comprise the ferromagnet MnAs, the ferromagnetic semiconductor $\text{Ga}_{1-x}\text{Mn}_x\text{As}$, and an AlAs tunneling barrier yielded magnetoresistance changes of up to 30% at a temperature of 5 K [22]. Devices with Schottky barriers also show encouraging results for spin-polarized transport in ferromagnet/semiconductor hybrids [23]. Consequently, the focus of ongoing work in the field of semiconductor-based spintronics lies on the improvement of growth conditions of the injector material on the semiconductor, the use of Schottky or tunneling barriers, and the optimization of injector materials with a high degree of spin polarization.

In the early 1980s, de Groot discovered a new type of magnetic material, the half-metallic ferromagnets, in which the majority spin electrons are metallic, whereas the minority spin electrons are semiconducting [15]. A simplified sketch of the densities of states of a non-magnetic metal, a conventional ferromagnetic metal, and a half-metallic ferromagnet is given in Fig. 14.1. Two decades later, the properties of these

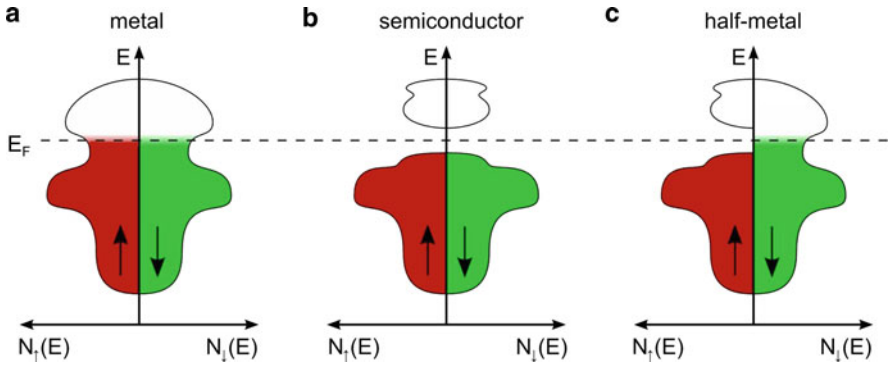


Fig. 14.1 Simplified densities of states for (a) a paramagnetic metal with degenerate spin-up and spin-down states, (b) a semiconductor, and (c) a half-metallic ferromagnet, where for the majority spins the density of states is zero at the Fermi energy

compounds predicted by theory have become real, which is especially important for the field of magnetoelectronics and for the field of spintronics with semiconductors.

Some Heusler alloys based on the $L2_1$ crystallographic phase fulfill the condition that the conduction electrons at the Fermi energy are 100% spin polarized. Such alloys have remained of interest to both theorists and experimentalists since they were first considered by Heusler in 1903 [24]. Historically the interest focused on the unexpected result that some of these materials are strongly ferromagnetic, although they are made by combining elements which are considered to be non-magnetic. With respect to spinelectronics the high spin polarization at the Fermi energy is most important.

The static spin polarization P_S

$$P_S = \frac{N_{\uparrow}(E_F) - N_{\downarrow}(E_F)}{N_{\uparrow}(E_F) + N_{\downarrow}(E_F)} \quad (14.1)$$

is derived from the densities of states $N_{\uparrow}(E_F)$ and $N_{\downarrow}(E_F)$ at the Fermi energy for spin-up and spin-down electrons. For transport experiments in the ballistic regime, apart from the densities of states at the Fermi energy, the Fermi velocities $v_{F\uparrow}$ and $v_{F\downarrow}$ are important as shown theoretically by Mazin [25]. Therefore, the spin polarization

$$P = \frac{N_{\uparrow}(E_F)v_{F\uparrow} - N_{\downarrow}(E_F)v_{F\downarrow}}{N_{\uparrow}(E_F)v_{F\uparrow} + N_{\downarrow}(E_F)v_{F\downarrow}} \quad (14.2)$$

is defined. In the following, we use this definition of the spin polarization.

It is intended to inject spin-polarized electrons from thin half metallic films into semiconductors because the high degree of spin polarization is expected to be transferred into the semiconductor. In this work, we focus on the ternary intermetallic compound Ni_2MnIn , which belongs to the class of Heusler alloys with general composition X_2MnY ($\text{X} = \text{Cu}, \text{Co}, \text{Ni}, \dots$; $\text{Y} = \text{Al}, \text{Ge}, \text{Si}, \text{In}, \dots$) [15, 24]. Figure 14.2 shows a sketch of the conventional cell of Ni_2MnIn .

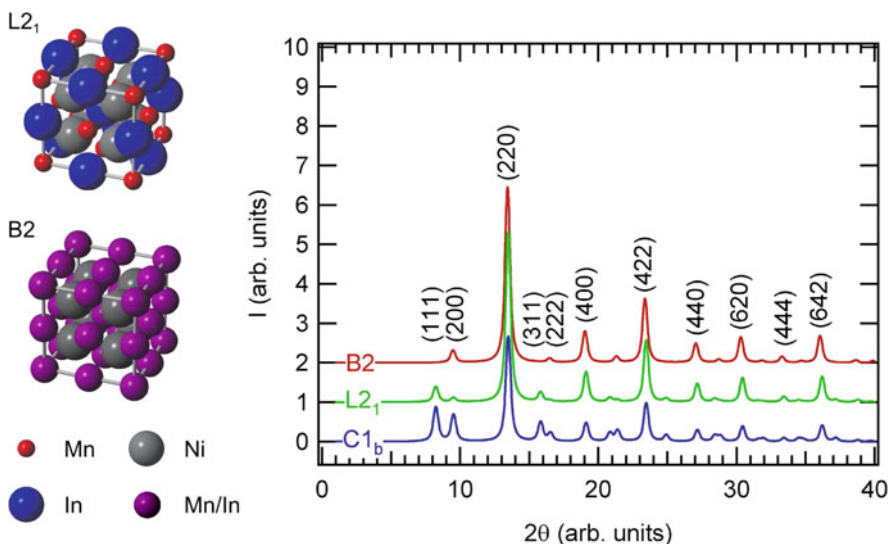


Fig. 14.2 Conventional cells of the competing L_{21} and $B2$ structure of the full Heusler alloy Ni_2MnIn . For the ordered L_{21} structure, the cell contains eight nickel, four manganese, and four indium atoms. They are arranged as four face-centered cubic lattices aligned along the space diagonal of the conventional cell. The lattice constant for both is 0.6022 nm [26]. Calculated diffraction patterns of the full Heusler (X_2YZ) and the half Heusler (XYZ) crystal structures $B2$, L_{21} , and $C1_b$. Note the absence of the (111) reflex for the disordered $B2$ structure

The ordered L_{21} phase has a cubic structure (L_{21} , $Fm\bar{3}m$). Lattice constants of (0.605 ± 0.003) nm are reported in the literature [27–33]. They perfectly match that of InAs $a_{InAs} = 0.606$ nm. Indiumarsenide is the semiconductor of choice also because of its strong and tuneable spin–orbit interaction [34]. It has been shown by Dong et al. [35] and Xie et al. [17] that epitaxial growth of Ni_2MnIn on InAs(001) can be achieved with molecular-beam epitaxy. However, the crystallographic structure reported there deviates from the expected ordered L_{21} phase. Complete disorder of the manganese and indium sublattices in the L_{21} crystal structure would result in a $B2$ simple cubic crystal. The identification and distinction of the two crystal structures can be obtained from electron diffractometry. Figure 14.2 shows calculated diffraction patterns¹ of the ordered L_{21} structure and the disordered $B2$ structure. Important for the identification of the respective phase is the (111) reflex of the L_{21} structure, which is not present in the $B2$ structure. An important criterium for the quality of the alloy is the Curie temperature. Literature values for bulk crystals of

¹ Computer code ‘PowderCell for Windows v1.0’ W. Kraus and G. Nolze, Federal Institute for Materials Research and Testing, Rudower Chaussee 5, D-12489 Berlin, Germany. This software is intended to simulate x-ray powder diffraction. A correction of the diffraction angles allows the determination of the positions of reflexes of electron diffraction. However, the intensities in electron diffraction may vary from the ones observed in X-ray diffraction patterns.

the phase $L2_1$ vary from 314 to 323 K [28, 31, 33] and for thin films from 170 to 318 K [17, 35]. The formation of the B2 structure is supposed to cause the reduced Curie temperature [17, 35].

14.2 Experimental

14.2.1 Growth and Structure Investigations

We grow thin Ni_2MnIn films by coevaporation of the element nickel and the alloy $MnIn$. Using only two sources is possible due to the similar vapor pressures of manganese and indium at temperatures above 1,000 K [36, 37]. The evaporated materials are contained in Al_2O_3 crucibles, which are enclosed in molybdenum furnaces heated by electron impact. A cross-beam ion-source mass spectrometer and a feedback loop enable us to keep the rate of evaporation constant for an hour and longer. A heatable substrate holder allows to adjust the temperature of the substrate up to $T_{sub} = 600^\circ C$ [38]. The thicknesses of the films are calculated from the deposition time and the deposition rate calibrated by atomic-force microscopy. The stoichiometry of the films is determined by energy-dispersive X-ray spectroscopy (EDX). Their spin polarization is determined by point-contact Andreev reflection spectroscopy (PCAR). Details of this technique can be found in [36, 39, 40] and in Sect. 14.2.2.

We deposit nickel and $MnIn$ simultaneously on a variety of substrates including $InAs(100)$, in situ cleaved $InAs(110)$, $Si(100)$ with native oxides, Si_3N_4 membranes supported by silicon frames, and amorphous carbon films supported by copper grids. To achieve smooth surfaces free from contaminations and oxides, $InAs$ substrates are cleaved immediately prior to the deposition of the Heusler alloy. The crystal structure of the films was determined by reflection high energy electron diffraction (RHEED). The difference in the lattice constants of silicon ($a_{Si} = 0.5431$ nm) [41] and Ni_2MnIn ($a_{Ni_2MnIn} = 0.6022$ nm) does not allow oriented growth. Therefore, we assume the crystal structure and the morphology of the films grown on silicon to be comparable to the ones grown on amorphous carbon films and on Si_3N_4 membranes. For amorphous carbon, this assumption was proven by comparative investigations [42]. The Heusler films on Si_3N_4 membranes and amorphous carbon are used for investigations of the morphology, the crystal structure, and the stoichiometry by transmission-electron microscopy (TEM), transmission-electron diffraction and energy-dispersive X-ray spectroscopy (EDX). The investigations are carried out in a Philips CM12 TEM with a resolution of 3×10^{-10} m at an acceleration voltage of 120 kV. The attached EDX-spectrometer LINK-ISIS 300 from Oxford Instruments uses an $Si(Li)$ -detector and allows a resolution of the quantitative analysis of one atomic percent. The Si_3N_4 membranes are chemically more stable than amorphous carbon that reacts with nickel at high temperatures [43]. The stability is important for the post-growth annealing described in Sect. 14.3.2.

To pave the way for hybrid devices that incorporate nanostructures of Heusler alloys a preparation process is required that is compatible with the demanding growth conditions of half-metallic ferromagnets. Once such a process is established, Ni_2MnIn could replace conventional ferromagnets such as iron, cobalt, nickel, and permalloy ($\text{Ni}_{0.8}\text{Fe}_{0.2}$) to enhance the spin-injection rate that spintronic devices rely on. Heusler electrodes are patterned using the lift-off technique. High substrate temperatures are incompatible with electron-beam resist [26] as the organic resist hardens and cannot be removed in a lift-off step. To meet the process parameters of the resist, Heusler films were grown at low substrate temperatures and annealed at higher temperatures after the lift-off step.

14.2.2 Electrical Characterization

Only few techniques provide quantitative access to the spin polarization at the Fermi energy of a metal. Among them are tunneling spectroscopy with ferromagnet/insulator/superconductor contacts, spin resolved photoelectron emission spectroscopy, and point-contact Andreev reflection spectroscopy to name a few.

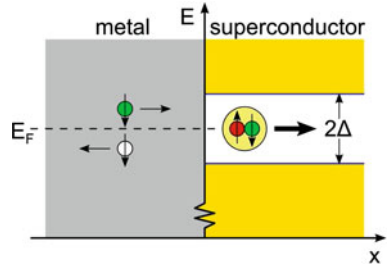
The preparation of homogeneous tunneling barriers in ferromagnet/insulator/superconductor tunneling contacts, which are typically made of Al_2O_3 , is a great challenge [44]. Attempts in this direction are promising [16], but the experimental effort is immense.

Photoelectron emission spectroscopy requires an elaborate preparation because the method is very sensitive to the quality of the surface. The information is recorded from a thin surface layer with a thickness in the nanometer range. A surface oxide would deteriorate the measured polarization, i.e., this technique requires an ultra-high vacuum environment [45].

Point-contact Andreev reflection spectroscopy is a method to determine the spin polarization of ferromagnetic metals [39]. In this technique, the normal current is converted into a supercurrent via Andreev reflection at a ferromagnet/superconductor interface, a process that strongly depends on the availability of spin states at the Fermi level. Soulen and coworkers have investigated half metallicity with this technique. For NiMnSb , $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$, and CrO_2 , polarization values have been observed between 60% and 90% [39]. We have established this technique in Hamburg as a tool for the characterization of ferromagnetic thin films. We shortly discuss some aspects of the method that are important in the context of our experiments.

In the energy range of the superconducting energy gap electron transport from a normal metal into a superconductor is only possible by generation of Cooper pairs at the interface. This process is illustrated in Fig. 14.3. At the superconductor/normal metal interface, the incoming electron needs another electron with reversed spin to form a Cooper pair. At the same time, a hole is generated and retroreflected into the metal. In reality, the onset of the superconducting energy gap is not abrupt but increases rather smooth from zero in the normal metal to the full gap size of 2Δ on the length scale of the superconducting coherence length [38].

Fig. 14.3 Simplified sketch of the Andreev reflection at an abrupt metal–superconductor interface



A quantum mechanical description of the processes at a normal metal–superconductor interface was given by Blonder, Tinkham, and Klapwijk by the so-called BTK model [46]. This model takes into account electron- and hole-like states in the superconductor and uses the Bogoliubov–de Gennes equation [47] for the description of the quasi particles. Scattering at the interface is described by a delta-shaped potential $W(x)$ of height V :

$$W(x) = \delta(x) \cdot V \quad \text{with} \quad Z = \frac{mV}{\hbar k_F} \quad (14.3)$$

where m is the electron mass. The dimensionless parameter Z describes the quality of the interface, i.e., interface roughness and contamination as well as the influence of different Fermi velocities $v_F = \frac{\hbar k_F}{m}$ in the normal metal and the superconductor. For a ballistic contact with a perfect interface $Z = 0$, whereas $Z \gg 1$ for a tunneling barrier which represents the other limit.

The solution of the Bogoliubov–de Gennes equations yields a set of reflection and transmission probabilities $A(E)$, $B(E)$, $C(E)$, and $D(E)$ for an incident electron with energy E . The probability of Andreev reflection is given by $A(E)$, the probability for normal reflection by $B(E)$. The probabilities $C(E)$ and $D(E)$ describe electron-like and hole-like transmission. With the help of the probability coefficients, which include the above-mentioned Z parameter, the current–voltage relation

$$I_{\text{NS}}(U) = \kappa \cdot \int_{-\infty}^{\infty} [f(E - eU) - f(E)][1 + A(E) - B(E)] dE \quad (14.4)$$

can be calculated [46]. The constant κ includes the actual size of the contact area and $f(E)$ is the Fermi–Dirac distribution function.

The BTK model is valid for normal metals without spin polarization. Soulen and collaborators have extended the BTK model to ferromagnetic materials [39]. In this case, the total current is divided into a completely polarized current and a completely unpolarized current

$$I = (1 - P) \cdot I_{\text{unpol}} + P \cdot I_{\text{pol}} \quad (14.5)$$

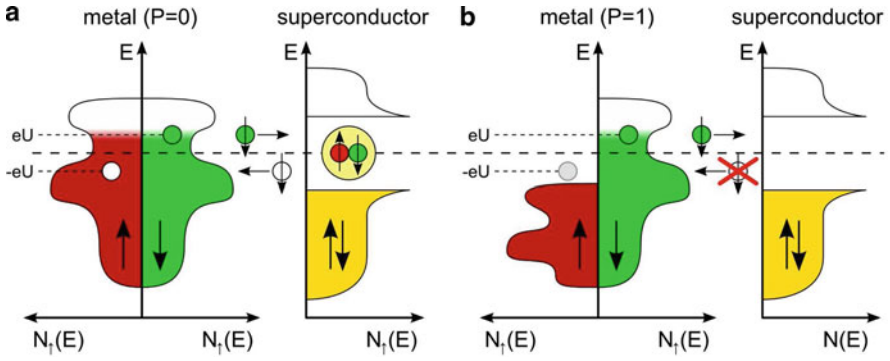


Fig. 14.4 (a) Sketch of the transport process at the interface between a normal metal and a superconductor and (b) at the interface between a ferromagnet with full spin polarization $P = 1$ and a superconductor. The voltage drop across the contact is U . The solid (open) circles represent electrons (holes). In (a) charge transport by generation of a Cooper pair is possible, whereas in (b) Andreev reflection is suppressed due to the lack of spin-up states

weighted with the polarization P . The unpolarized current is calculated with the BTK model. The polarized part is calculated for vanishing Andreev-reflection probability $A(E)$. The unpolarized case is sketched in Fig. 14.4a. In case of completely polarized electrons, the Andreev reflection is suppressed as illustrated in the sketch of Fig. 14.4b. Because of the lack of spin-up states the retroreflection of the corresponding hole is forbidden. Note that in principle the Andreev reflection probability in ballistic ferromagnet/superconductor junctions depends on the spin orientation of the incident quasi particle [48]. This explicit spin dependence is neglected in the widely accepted approach of Soulen et al. [39, 49].

From the BTK model and (14.5), current–voltage curves can be calculated for temperatures T , polarizations P , and barrier heights Z . The derivative $dG = \frac{dI}{dU}$ easily converts the current–voltage curves into conductance–voltage curves.

The current–voltage curves for the Andreev spectroscopy and temperature-dependent resistivity of the samples are measured using a current-driven four-terminal setup shown in Fig. 14.5a and b. To avoid losing the spin information due to scattering events, it is crucial to perform the point-contact measurements at a contact in the ballistic transport regime [25]. This is ensured by restricting the geometrical dimensions of the contact area to lengths below the electron mean free path, in form of a point contact. However, also contacts in the diffusive regime can be evaluated using the diffusive extension [49, 50]. We use a measurement setup similar to the one described by Soulen and coworkers [39]. At liquid helium temperatures, a superconducting niobium tip is lowered onto the sample. The conductance versus voltage dependency of the contact is measured in a current driven four-terminal setup using lock-in technique as sketched in Fig. 14.5a and c. The obtained curves are fitted using the BTK model. A high resistivity of the sample appears as a resistor in series to the point contact, as shown in Fig. 14.5d, and can lead to a falsification of the measured curves. Besides the contribution to the measured resistor $R = R_K + R_S$

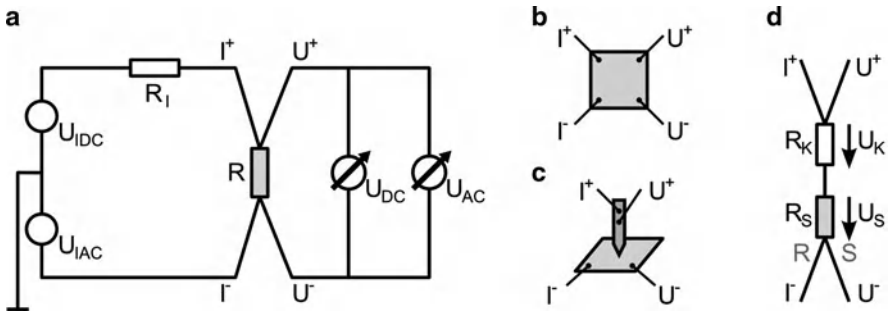


Fig. 14.5 (a) Circuit diagram of a current driven four-terminal setup ($R_I \gg R$). (b) Wiring of a sample for resistivity measurements in van der Pauw geometry. (c) Wiring for the point-contact Andreev reflection spectroscopy. (d) Consideration of the resistor in (a) as a combination of the point-contact resistor R_K and a series resistor R_S

the voltage drop U_S over the series resistor leads to a broadening in the measured contact voltage. By estimating a value for the series resistor, the effect can be eliminated by applying Ohm's law. To minimize the series resistor, a highly conductive underlayer can be deposited prior to the deposition of the ferromagnetic layer. The conductive layer guides the current paths to the point contact [40]. This approach is only suitable if the growth of the thin-film samples does not crucially depend on the substrate as it does for epitaxial growth.

14.3 Results and Discussions

14.3.1 Thin Films

Figure 14.6 shows a transmission-electron micrograph and a transmission-electron diffractogram of a (54 ± 3) nm thin $\text{Ni}_{42}\text{Mn}_{29}\text{In}_{29}$ Heusler film grown on an amorphous carbon film at a temperature of 300°C . This temperature was found to be necessary to generate polycrystalline films of the desired L_{21} crystal structure [26]. The image in Fig. 14.6a shows a granular film consisting of accumulations of crystallites. Single crystallites are clearly visible because of their Bragg contrast. While the crystallites exhibit a mean diameter of 20 nm the accumulations are 100 to 200 nm wide. The accumulations are separated from each other by canyons visible as dark areas due to their high transparency for electrons. Figure 14.6b shows a sector of the transmission-electron diffractogram in comparison with the calculated diffraction patterns of Ni_2MnIn in the B2 structure and L_{21} structure. The presence of the (111) reflex proves that the film possesses at least partially the L_{21} structure. However, the complete absence of the B2 structure cannot be guaranteed because both structures have the other reflexes visible in Fig. 14.6b in common.

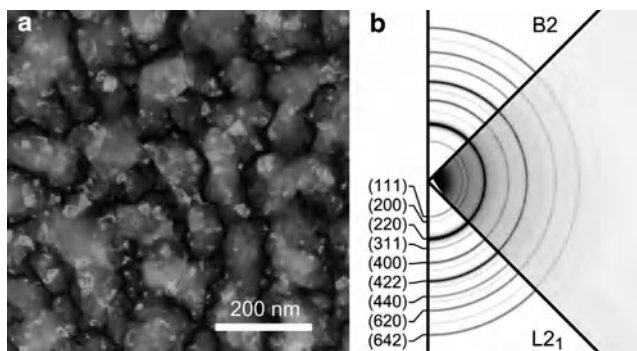


Fig. 14.6 (a) Negative transmission-electron micrograph of a Heusler film grown on amorphous carbon at a substrate temperature of 300°C. (b) Transmission-electron diffractogram in comparison with diffraction patterns calculated for Ni_2MnIn in the B2 structure and L2_1 structure. From [51]

It was not possible to perform sensible point-contact Andreev reflection spectroscopy measurements on the sample, which has been simultaneously grown on silicon. This can be understood with the help of the micrograph in Fig. 14.6a. In the region of the canyons, the current paths must proceed in the silicon substrate. At low temperatures, this leads to a high additional resistance in series to the point contact. To avoid this series resistance, a Heusler film was deposited on a silicon substrate covered by a thin gold film in the same evaporation process. The current paths are guided by the high-conductivity gold layer underneath the Heusler layer toward the point contact, virtually eliminating a series resistance [40]. This sample layout is possible because the growth on silicon does not crucially depend on the substrate as it does for InAs. Andreev reflection measurements performed on the Ni_2MnIn film grown on Si/Au substrates resulted in a spin polarization of $(30 \pm 1)\%$ [38].

The canyons visible in Fig. 14.6a have been observed earlier [42, 52]. To answer the question whether the canyons can be avoided by increasing the film thickness, samples of different thicknesses have been grown under the growth conditions described above. Figure 14.7 shows scanning-electron micrographs of Ni_2MnIn films grown on Si(100) at a substrate temperature of 300°C. The different thicknesses of the samples were obtained by varying the evaporation time, while keeping all other parameters constant. The first sample shown in Figs. 14.7a1 and a2 with an average thickness of (40 ± 2) nm consists of 100 nm to 200 nm wide accumulations of crystallites separated by canyons. At a thickness of (76 ± 4) nm, as shown in Figs. 14.7b1 and b2, the layer consists of 300–400 nm wide accumulations. Not until a thickness of (155 ± 8) nm is reached the films show a continuous surface as visible in Figs. 14.7c1 and c2. The closing of the surface is due to merging of the accumulations. Nanometer wide holes in the film support this assumption.

Figure 14.8 shows scanning-electron micrographs of films grown on InAs(100) during the same evaporation as the samples displayed in Fig. 14.7. Like the films deposited on silicon, the layers are disjointed consisting of particles separated by canyons. In this case, the particles are small, single crystals or consist of only a

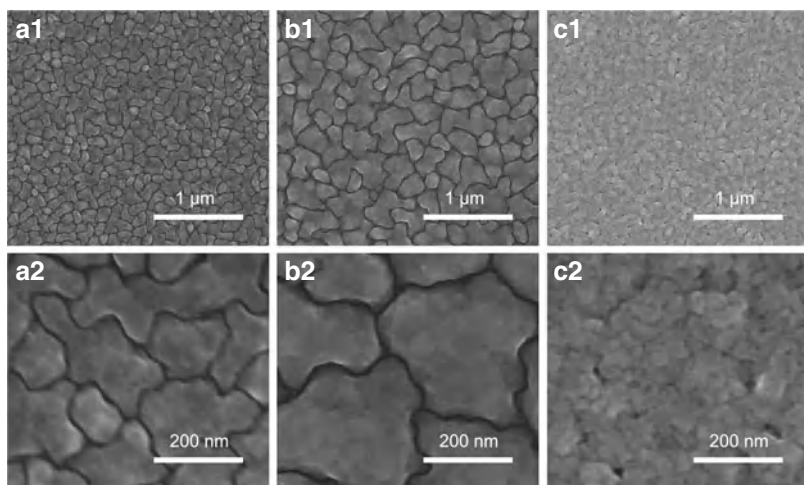


Fig. 14.7 Scanning-electron micrographs of Ni_2MnIn layers on $\text{Si}(100)$ at a substrate temperature of 300°C . The mean thickness of the Heusler layer shown in (a1) and (a2) at different magnifications is (40 ± 2) nm, in (b1) and (b2) it is (76 ± 4) nm, and in (c1) and (c2) it is (155 ± 8) nm. From [51]

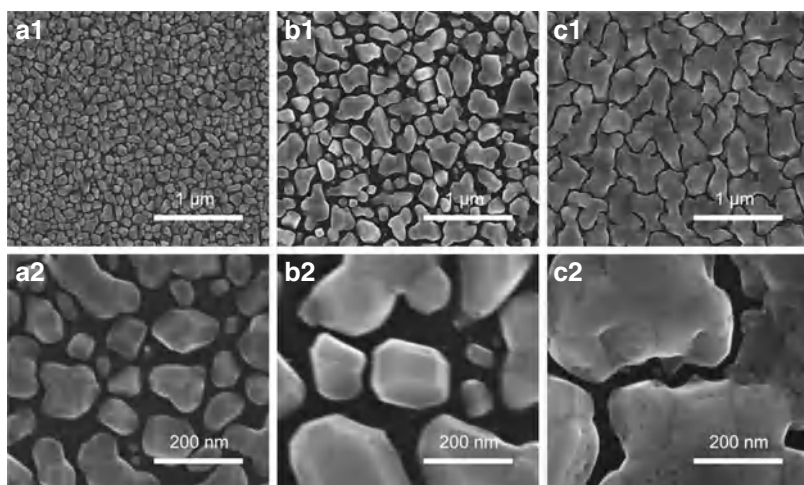


Fig. 14.8 Scanning-electron micrographs of Ni_2MnIn layers grown on $\text{InAs}(100)$ at a substrate temperature of 300°C in the same evaporation as the layers on silicon presented in Fig. 14.7. The mean thickness of the Heusler layer shown in (a1) and (a2) is (40 ± 2) nm, in (b1) and (b2) it is (76 ± 4) nm, and in (c1) and (c2) it is (155 ± 8) nm. The listed thicknesses have been determined from simultaneously grown films on silicon and have to be treated with care in view of the morphology of the layers. From [51]

few single crystals. This can be deduced from the faceting of the particles that is clearly visible in Fig. 14.8b2. While the average size of the crystallites on Si(100) stays the same, the crystallites on InAs grow with increasing evaporation time. In the films displayed in Figs. 14.8c1 and c2 the single crystals merge to accumulations of a few crystallites as evidenced by the faint gray boundary lines. In contrast to the film on Si(100) shown in Figs. 14.7c1 and c2, the layer on InAs(100) displayed in Figs. 14.8c1 and c2 does not close.

Despite the canyons, it is possible to perform point-contact Andreev reflection spectroscopy measurements on the samples grown on InAs because of the comparatively low resistivity of this semiconductor [53]. Figure 14.9 shows normalized conductance–voltage curves of a point-contact measurement on the sample shown in Figs. 14.8(c1) and (c2). The measurements were performed at temperatures between 2 and 9 K. The evaluation is carried out by fitting the experimental curves with the diffusive BTK model [39, 49, 54] that yields the solid black lines. The dependency of the polarization P on the BTK parameter Z is plotted in the inset. A parabolic extrapolation to $Z = 0$ results in a comparatively low spin polarization of $(17 \pm 2)\%$. Possibly, the observed crystallites do not exhibit the desired $L2_1$ structure but the undesired B2 structure.

Layer thicknesses of 155 nm and more to obtain closed films are not desirable in view of nanopatterning as the film thickness limits the sizes of lateral nanostructures. Nanostructured electrodes for spin injection are, in general, 10–40 nm thick

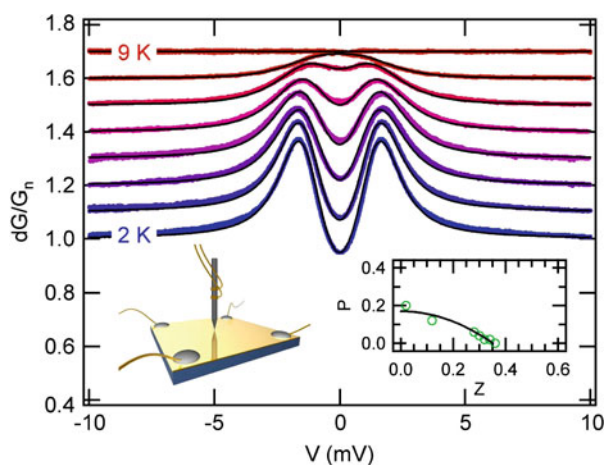


Fig. 14.9 Normalized conductance–voltage curves of a point-contact Andreev reflection spectroscopy measurement taken at temperatures between 2 and 9 K. The measured data were corrected assuming a series resistor of $R_s = 1.0 \, \Omega$ and normalized with resistances between $R_n = 12.7$ and $13.8 \, \Omega$. The curves for temperatures above 2 K are plotted with an offset. The insets show a sketch of the measurement setup and the dependence of the spin polarization on the Z parameter. From [51]

[4–6, 55]. The deposition of the layers at lower substrate temperatures may avoid the formation of canyons.

Diffraction patterns of an NiMnIn and an Ni₂MnIn film grown at a substrate temperature of 200°C on cleaved InAs surfaces are shown in Figs. 14.10a and b. They exhibit point reflexes which remain when moving the electron beam across the sample stepwise. The diffraction patterns prove that the layers are monocrystalline or consist of equally aligned crystallites. The lines added in Fig. 14.10a are guides to the eye showing a cubic lattice. Together with the reflexes in the center of the quadrangle the points form a base-centered type of pattern which belongs to a face-centered cubic lattice. The stoichiometry of 1:1:1 of the sample suggests the C1_b structure of a half-Heusler alloy. The oblique angles are due to the restricted geometry of the diffraction setup in the scanning-electron microscope. Figure 14.10b shows the straightened diffraction pattern of an Ni₂MnIn film grown at a substrate temperature of 200°C on an in situ cleaved (110) surface of InAs. The lines added are again guides to the eye, the displayed reciprocal lengths were calibrated using a Au(100) sample in transmission. The obtained lengths of 3.1 and 2.2 Å correspond to the lattice distances $d_{(200)} = 3.03 \text{ Å}$ and $d_{(220)} = 2.15 \text{ Å}$ in Ni₂MnIn [33]. The reflexes (200) and (220) suggest the expected (220) orientation of the film on the InAs(110) substrate. However, the absence of (111)-type reflexes points to the

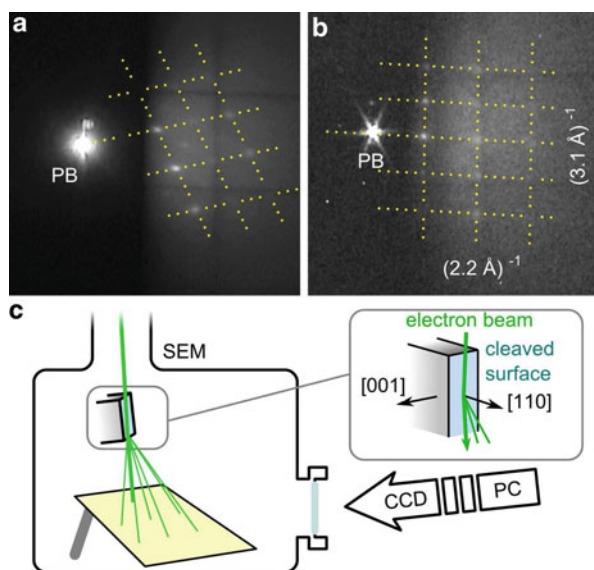


Fig. 14.10 Electron diffraction under grazing incidence. Diffraction patterns of (a) a NiMnIn film and (b) of an Ni₂MnIn film on cleaved (110) surfaces of InAs. (c) Sketch of the measurement setup in a modified scanning-electron microscope (SEM). The primary electron beam (PB) is diffracted at the sample that is approximately tilted by 90°. The diffraction pattern is recorded by a camera (CCD) and a computer (PC). The use of an SEM simplifies the control of the electron beam and the positioning of the sample. From [51]

presence of the B2 structure in this film. The determination of the spin polarization yields a value of $(28 \pm 1)\%$ [36]. Like in the previously presented film on InAs, the comparatively low value of the spin polarization is presumably due to the presence of the B2 structure. A resistivity measurement in van der Pauw geometry [56] on a simultaneously grown film on silicon yielded a resistivity of $103 \mu\Omega \text{ cm}$. This value strongly differs from the bulk resistivity $10 \mu\Omega \text{ cm}$ of Ni_2MnIn [57], but lies in the range of full Heusler Ni_2MnZ ($Z = \text{Al}, \text{Ga}, \text{Ge}$) thin films grown with molecular-beam epitaxy [58].

14.3.2 Nanopatterning

Because of the temperature sensitivity of the photoresists, the films were grown at low substrate temperatures and annealed at higher temperatures after the lift-off step. Figure 14.11 shows transmission-electron diffractograms taken in situ during annealing in comparison with calculated diffraction patterns (see Footnote 1) of the B2 and the L_{21} structure. The film was deposited on an Si_3N_4 membrane at a low substrate temperature of 50°C . The temperature profile of the post-growth annealing is shown in the inset. Figure 14.12a and b show transmission-electron micrographs before and after annealing.

The as-grown film depicted in Fig. 14.12a exhibits a nanocrystalline to amorphous morphology presumably due to the low substrate temperature during its deposition. Crystallites usually are easily visible because of their Bragg contrast but cannot be observed here. The diffractogram of the as-grown film in sector A of Fig. 14.11 supports this assumption. Only one very diffusive diffraction ring is visible. The annealing already causes a granular B2 phase at a temperature of 300°C as can be deduced from the separated point reflexes in sector B. Further annealing for several hours at temperatures of 400°C leads to the formation of the L_{21} structure evidenced by the (111) reflex in sector F. The morphology of the annealed film becomes apparent in the transmission-electron micrograph in Fig. 14.12b. The average crystal size is approximately 100 nm. Figure 14.13a–c shows transmission-electron micrographs of a pair of Ni_2MnIn electrodes patterned on an Si_3N_4 membrane using the lift-off technique. Figure 14.13a was taken before, b and c after the annealing depicted in the inset in Fig. 14.11. Figure 14.13c is a close-up of Fig. 14.13b. The sequence shows the effects of the post-growth annealing on a nanopatterned Ni_2MnIn electrode. The annealing generates the desired crystal structure in an originally amorphous film without affecting the lithographically defined shape of the structure. No blurring or fraying is observed. The thinning of the upper border of the left electrode is caused by the shading due to the resist mask and the angle between the vapor beams from the nickel and MnIn sources. The composition of the sample before and after annealing was determined by EDX. Within the accuracy of the analysis, the composition remained the same. This means post-growth annealing only affects the morphology and the crystal structure. Using this nanopatterning and annealing process, it is possible to integrate the Heusler alloy Ni_2MnIn into hybrid ferromagnet/semiconductor nanostructures.

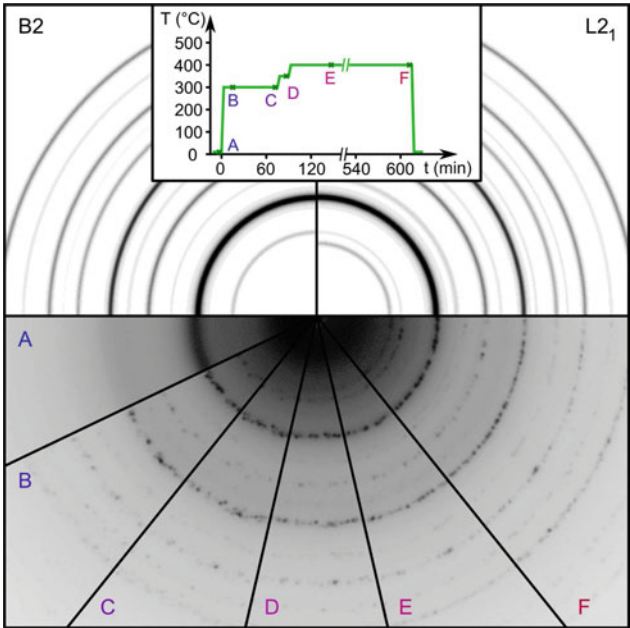


Fig. 14.11 Transmission-electron diffractograms of an Ni_2MnIn film on an Si_3N_4 membrane in comparison with calculated diffraction patterns of the B2 and the L_{21} structure. The *inset* shows the temperature profile of the annealing process and the times A–F at which the diffractograms were taken. From [51]

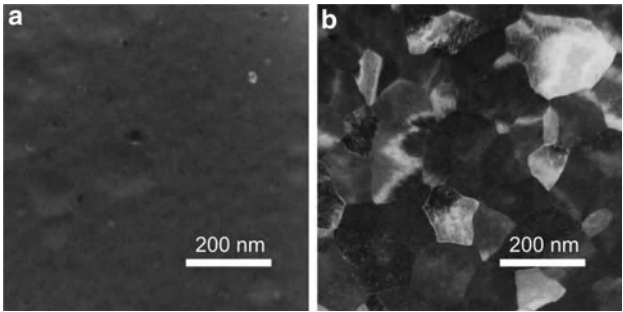


Fig. 14.12 Negative transmission-electron micrographs of the film depicted in Fig. 14.11. (a) Before and (b) after annealing

14.3.3 Heusler-Based Spin-Valves

We have prepared a spin valve with Ni_2MnIn electrodes for measurements of the local spin-valve effect [4, 55]. Figure 14.14a shows a scanning-electron micrograph of this device, its geometry is presented in Fig. 14.14b. Details about lateral spin

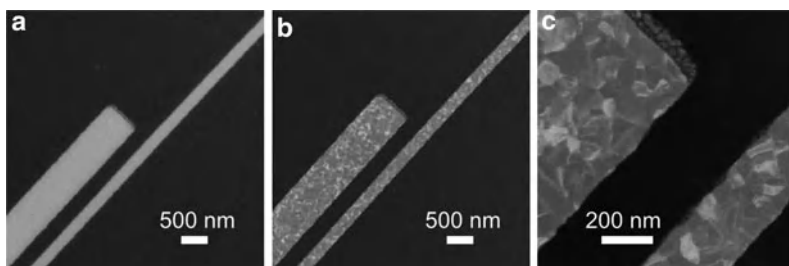


Fig. 14.13 Negative transmission-electron micrographs of a pair of Ni_2MnIn electrodes. (a) Before, (b) and (c) after annealing

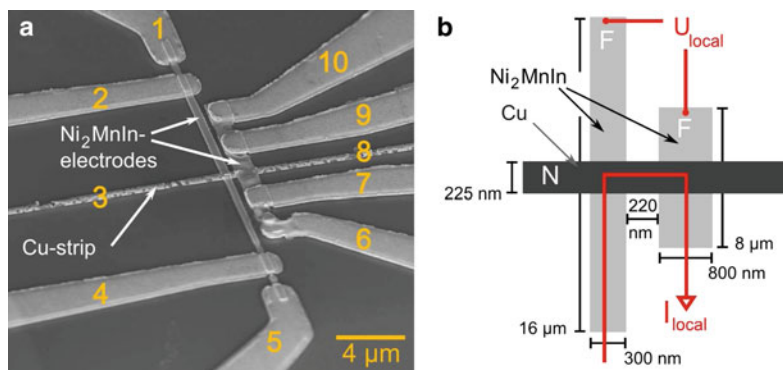


Fig. 14.14 (a) Scanning-electron micrograph of a spin-valve structure with Ni_2MnIn electrodes. The image was taken after completing the transport measurements. Between the contacts 4 and 5 the Heusler electrode is tapered which was caused by high current densities. (b) Geometry of the lateral spin-valve structure

valves with conventional ferromagnetic electrodes can be found in the article by Wulforst, Vogel, Kuhlmann, Merkt, and Meier in this book.

The nanostructured Ni_2MnIn electrodes are created by lift-off processing. A spin polarization of $(33 \pm 1)\%$ was found for an Ni_2MnIn film deposited on silicon in the same coevaporation step. The scanning-electron micrograph in Fig. 14.14 shows electrodes with smooth edges in the desired geometry after post-growth annealing of 6 h at 400°C . The copper strip of this demonstrator exhibits fissures and is rather inhomogeneous, which might be caused by residual resist of a non-optimized lift-off process. Nonetheless, we observed a clear spin-valve signal as exemplified in Fig. 14.15. For these measurements in local geometry, the current was applied at contacts 1 and 10 shown in Fig. 14.14a. The voltage has been measured between contacts 5 and 6. The external magnetic field is aligned parallel to the long axes of the electrodes. At magnetic field strengths above 500 mT, the magnetizations of both electrodes are adjusted parallel. External fields between ± 150 mT are enough to record all irreversible switching events in the magnetoresistance. Figure 14.15 shows two full loops exhibiting almost the same spin-valve signal. The numerous

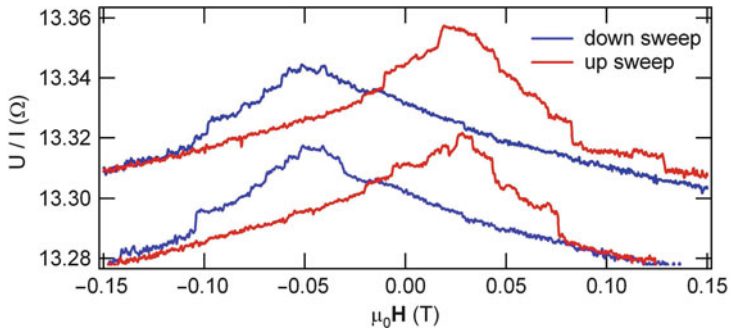


Fig. 14.15 Spin-valve effect in local geometry. The electrodes consist of the Heusler-alloy Ni_2MnIn . The interconnecting metal channel is made of copper. The applied current is $50\text{ }\mu\text{A}$. Shown are two full sweeps of the external magnetic field between $\pm 150\text{ mT}$. The *upper curves* are offset by $0.003\text{ }\Omega$ for clarity

small jumps in the magnetoresistance indicate that the Ni_2MnIn electrodes reverse their magnetization in a complicated multiple-domain process rather than by single-domain switching as it is known for optimized permalloy electrodes [4, 59]. The reversible increase of the resistance for vanishing external magnetic fields is a contribution of the anisotropic magnetoresistance to the total resistance of the device and is well known from permalloy/aluminum spin valves measured in local geometry [55].

14.4 Conclusions

The growth of the full Heusler alloy Ni_2MnIn has been studied on various substrates. High substrate temperatures of 300°C that are required for the growth of the L_{21} crystal structure with a high spin polarization lead to the formation of canyons in the deposited layers. The growth of Ni_2MnIn on InAs seems to favor the disordered B2 structure and is thus a demanding choice for nanopatterning. Films deposited on $\text{Si}(100)$ surfaces at least partially exhibit the desired L_{21} structure. Point-contact Andreev reflection spectroscopy yields spin polarizations lower than the spin polarizations of the ferromagnetic elements iron, cobalt, nickel, or permalloy. The low values of the spin polarization are presumably caused by the coexistence of the L_{21} and the B2 phase. Using a lift-off and a post-growth annealing process, a nanofabrication is demonstrated that is compatible with the requirements for the highly oriented growth of Heusler alloys and with the limited temperature range tolerable for common organic resist masks. Nanopatterned and annealed Ni_2MnIn spin valves can be used to investigate the spin injection into semiconducting or metallic channels in prospective spintronic devices. We have already demonstrated the local spin-valve effect in a spin valve with Ni_2MnIn electrodes.

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Chapter 15

Charge and Spin Noise in Magnetic Tunnel Junctions

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Abstract Manipulation of magnetization by electric current lies in the mainstream of the rapidly developing field of spintronics. The electric current influences the magnetization through the spin-torque effect. Entering a magnet, spin-polarized current exerts a torque on the magnetization, which aligns the magnetization parallel or antiparallel to the spin polarization of the current. The spin-torque effect can be used for fast magnetization switching in magnetic tunnel junctions (MTJ) that consist of two magnetic layers separated by a tunnel barrier. Moreover, applying external magnetic field and passing electric current simultaneously, one can induce a wide variety of nonequilibrium dynamical regimes, ranging from hysteretic switching between two static orientations of magnetization to steady nonequilibrium magnetization precession. Theoretical description of nonlinear nonequilibrium magnetization dynamics is given by the Landau–Lifshitz–Gilbert (LLG) equation. In this approach, the magnetization is treated on a classical level, resulting in a deterministic dynamics, which can exhibit crossover from periodic to chaotic orbits. In presence of spin-polarized current, there are nonequilibrium fluctuations of magnetization – the *spin shot noise* – that distort the classical dynamics of magnetization. Those fluctuations originate from the discrete nature of spin and, in this respect, they are similar to the well-known shot noise in the charge transport that stems from the discreteness of charge.

A particular feature of the nonequilibrium spin noise is its dependence on the angle between the magnetizations of the magnetic layers forming the junction. This peculiarity leads to the appearance of so-called “hot” and “cold” spots with different noise strengths in the deterministic trajectory of magnetization. Due to the tunnel magnetoresistance effect, the distortion of deterministic magnetization dynamics by the spin shot noise transforms into fluctuations of electric current that are registered experimentally. Peculiar features of the spin shot noise are thereby reflected in the frequency spectrum of electric current fluctuations.

At present time, there are two theoretical approaches to the treatment of the nonequilibrium spin shot noise and the complementary charge shot noise in MTJs. One is based on the extension of Landauer–Büttiker formalism to magnetic junctions, the other one uses the introduction of stochastic Langevin terms into the LLG equation with subsequent derivation of the Fokker–Planck equation for the

distribution function of magnetization. In this review, we discuss both approaches with an emphasis on the second one. In addition, a general review of theoretical and experimental works concerning equilibrium and nonequilibrium noise in magnetization dynamics is given. In particular, we discuss the effects of noise in different regimes of magnetization dynamics, such as switching of magnetization between two static orientations and steady state nonequilibrium magnetization precession.

15.1 Introduction

The impact of noise on a given physical system is of fundamental interest in any use-oriented consideration. In particular, in the field of nanotechnology, where device extensions have reached the nanometer scale, the role of noise may be significant and may lead to a reduction or to an enhancement of essential properties of corresponding devices. Consequently, the study of (*classical* and *quantum*) fluctuations in nanosystem has attracted considerable amount of attention in recent years, though, of course, the concept of noise is not new to physics and it has been studied very intensively throughout the last century in connection with diverse phenomena.

Random fluctuation of observable quantities – or simply noise – may be of different origin. The indeterministic nature of quantum mechanics is the reason for quantum fluctuations. However, already the “classical” noise on its own displays a broad variety of appearances. Thermal noise, for example, – which is due to thermal agitations that cause the occupation number of a state of a system to fluctuate – occurs at any finite temperature T , and is thus present in any system even when it is in equilibrium. Apart from this equilibrium noise, there are also nonequilibrium sources of fluctuations. The shot noise, for instance, can be observed in electrical circuits and is traced back to the random nature of quantum mechanical tunneling processes for the individual charge carriers, or in other words to the discreteness of charge.

The following review is devoted to nonequilibrium noise in magnetic nanodevices. By magnetic nanodevices, we understand systems in which spin-torque driven magnetization dynamics can be observed, as, for example, spin valves or magnetic tunnel junctions (MTJ). Investigations of noise in such systems concentrate to a large extent on thermal fluctuations. This will be reflected in the large fraction of the corresponding theoretical and experimental works reviewed here. Apart from their unquestionable relevance, such studies provide a link to the investigation of nonequilibrium sources of noise and are thus a suitable starting point when studying the latter.

At low temperatures, the nonequilibrium noise may become dominant. In particular, the spin shot noise plays a crucial role in the magnetization dynamics at low temperatures. In this sense, the central aim of our discussion is to depict the underlying mechanisms that lead to the occurrence of shot noise in magnetic nanodevices and to present their mathematical description. The emphasis is on what we call the Langevin approach, based on the introduction of stochastic terms into the Landau–Lifshitz–Gilbert (LLG) equation. A main ingredient is the Keldysh path integral formalism.

Of great interest, not only in the context of possible applications, is the estimation of switching rates. Spin-torque switching exhibits in general a large sensitivity to fluctuations. We address this topic by means of a generalized Fokker–Planck approach. Within this approach, the alteration of switching rates due to spin torque is described by an Arrhenius law with an effective temperature T_{eff} . The latter differs from the real temperature T , as it incorporates the effects of the damping, the spin torque, and – as we will show – the nonequilibrium noise.

The paper is organized as follows. In Sect. 15.2, we review some of the relevant literature connected to theoretical and experimental investigations of noise in magnetization dynamics. The Langevin approach is explained in Sect. 15.3 on the example of MTJs. In Sect. 15.4, we introduce the Fokker–Planck approach and show that taking into account the nonequilibrium noise leads to a renormalization of the effective temperature. Section 15.6 gives our final conclusions.

15.2 Noise and Magnetization Dynamics

Noise was introduced into the description of magnetization dynamics in 1963 in the pioneering work by Brown [1], who considered the effect of thermal fluctuations on the dynamics of a mono-domain particle. Brown modeled temperature by a random component of the effective magnetic field entering the LLG, hence assuming a constant absolute value of the magnetization vector at all temperatures. From the stochastic LLG, he was able to derive a Fokker–Planck equation for the probability distribution, depending on the two spherical angles. A very important point in Brown’s work was that he established a fluctuation–dissipation theorem (FDT) for magnetic systems. An FDT is a general relation between the equilibrium fluctuations of a physical quantity and the out-of-equilibrium dissipation of energy. In the case of a mono-domain magnetic particle, as Brown showed [1], the FDT states that the correlator of the random field is proportional to the friction parameter, which in this case is the Gilbert damping parameter α_0 , that is,

$$\langle h_i(t) h_j(t') \rangle \propto \alpha_0 k_B T \delta_{ij} \delta(t - t'), \quad (15.1)$$

where $h_i(t)$ denotes the i -th Cartesian component of the random field at time t , k_B is the Boltzmann constant, and T the temperature.

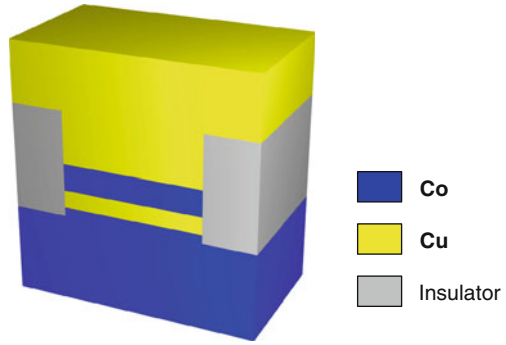
Here, we are mainly interested in the influence of noise on magnetization dynamics in presence of spin-polarized currents. A spin-polarized current interacts with the magnetization of a free magnetic layer and may transfer angular momentum to it, resulting in the spin-transfer torque (STT) phenomenon [2]. STT may in particular lead to the reversal of magnetization and to a steady state precession. Corresponding to these two dynamical regimes, studying noise in STT dynamics, one often concentrates on its influence on either the switching rates or the precession spectrum.

As far as the first point is concerned, temperature effects on the LLG were theoretically considered, for example, in [3–7]. On the other hand, a number of

experiments on noise induced switching (with and without STT) have been carried out [8–14]. Concerning the LLG without STT, Wernsdorfer et al. [8] argue that the magnetization reversal of ferromagnetic nanoparticles can be well described by the Neel–Brown model [1, 15], where the probability for the magnetization to switch decays exponentially with time over a characteristic relaxation time τ . The latter obeys the Arrhenius law $\tau \sim e^{U/k_B T}$, with U being the potential barrier height. An implicit assumption in the Neel–Brown theory is that magnetization dynamics is governed by a torque from an effective magnetic field, which is derivable from the free energy of the system. However, the spin-torque term is nonconservative and the concept of a corresponding potential barrier is not well defined, which complicates the situation considerably. For thermally activated switching in presence of STT, Urazhdin et al. [11, 12] found that the activation energy strongly depends on the magnitude and the direction of the current. To capture the experimentally observed features, they introduced an effective temperature unrelated to the true temperature in the Neel–Brown formula. Its current directional dependence indicated that the heating is not the ordinary Joule heating. Based on a stationary solution of the Fokker–Planck equation Apalkov and Visscher [5, 6], and in a less general framework Li and Zhang [3], linked this effective temperature to the spin torque. In these models, the alteration of switching rates is caused by a change of the elevated effective temperature in the Arrhenius factor. This leads to a probability distribution that, in general, is not a Boltzmann distribution. Another approach to noise in magnetic structures was discussed by Foros et al. in [7, 16]. The authors studied effects of temperature in magnetization dynamics in the context of normal metal/ferromagnet/normal metal (NFM) structures [7] and in spin valves [16] using the Landauer–Büttiker formalism [17, 18]. In [7], it is shown that there are two sources of thermal noise in magnetization dynamics: Apart from the thermal agitation of the magnetization due to intrinsic processes as encapsulated in Brown’s description [1], one has to consider thermal fluctuation of the spin current outside the ferromagnet. These fluctuations affect the magnetization by means of the STT. As a consequence of the FDT, this leads to a renormalization of the Gilbert damping parameter. Vice versa, one can include this second type of thermal noise into the dynamical description by using the renormalized instead of the bare Gilbert damping in the random field correlator of (15.1). Finally, as investigated by the same authors in detail in [19], the magnetization noise in spin valves induces resistance noise (due to GMR) that can be measured when converted to voltage noise. The contribution from (thermal) spin current noise to resistance noise was shown to be significant.

Let us now come to the second dynamical regime identified with STT, the steady state precession of magnetization, and shortly review its theoretical description. The precession becomes possible as a consequence of the interplay between the damping and the spin torque. The damping tends to align the magnetization vector in the direction of the effective magnetic field. The spin torque, on the other hand, pushes the magnetization in the direction of the spin-polarized current. These two competing contributions can lead to an undamped precession if the direction and magnitude of the spin torque are tuned in such a way that it compensates the damping. During

Fig. 15.1 Spin valve device. Two ferromagnetic layers (blue) are separated by a nonmagnetic spacer (yellow). The magnetization of the thick ferromagnet is fixed, whereas that of the thin layer is free to move



the motion, the absolute value of the magnetization is conserved. Hence, the tip of the magnetization vector precesses along a closed trajectory on the surface of a sphere, at some angle θ from the equilibrium position. While increasing the current, θ changes – as the motion takes place on a curved surface, the corresponding dependence of the angle on the current is a nonlinear one. Slonczewski identified the steady state precession with the excitation of spin waves in the free magnetic layer [2, 20]. On the other hand, within Berger's approach, a uniform precession was assumed to take place [21]. In [22], Slavin and Kabos developed an approximate theory of microwave generation in a current driven magnetic nanocontact, such as the spin valve of Fig. 15.1 when the current carrying area is restricted to a point contact (e.g., in the upper Cu layer) and the magnetic layers are laterally extended. For the steady state case, they showed that when a spin-polarized current flows through a nanocontact magnetized by an external magnetic field, a nonlinear quasihomogeneous precession will be induced in the free layer. The nonlinearity is, as described above, of geometric origin and results in a nonlinear shift of the precession frequency with the current, as reported, for example, by Rippard et al. in [23, 24]. The theory is based on the assumption that the magnetic oscillations excited by spin-polarized current lead to propagating spin waves in the free magnetic layer, however it is assumed that only one spin-wave mode is excited in the multilayer [25]. In [26], it is shown that taking into account the spatial structure of the spin wave results merely in a renormalization of the parameters of the nonlinear oscillator model [25]. Hence, even a macrospin model, in which spatial uniformity of the excited spin wave is assumed, can yield good qualitative results.

In the context of microwave oscillations, effects of thermal noise have been discussed as well [27–30]. Experimental data of Sankey et al. [27] indicated that the coherence time of a STT-driven nano-oscillator is limited by thermal effects. In particular, at low temperatures, thermal deflections about the equilibrium magnetic trajectory were associated with the broadening of the linewidth, while at high temperatures thermally activated transitions between different modes were suspected to influence the dynamics. Theoretical calculations of Kim et al. [28] and Tiberkevich et al. [29, 30] indeed showed that the equilibrium noise can at least partially account for the observed spectral linewidth. Very recently, interesting data from noise measurements on nano-oscillators based on MgO tunnel junctions was reported by Georges et al. [31]. It was observed that the noise is not dominated by

thermal fluctuations. Measurements of the spectral linewidth over a broad temperature range revealed only minor changes, whereas variation with the current was significant. The observed features can be summarized as follows. Firstly, the spectral linewidth as a function of current displays a nonmonotonic behavior, an initial decline is followed by a subsequent increase. Secondly, the background noise level is asymmetric in current. In [31], these features are attributed to the excitations of incoherent magnetic modes and/or the presence of hot spots.

Indeed, as the spin-torque experiments [23, 27, 32–34] are performed under clear nonequilibrium conditions, it is natural to address other than thermal sources of noise as well. A possible source of nonequilibrium noise is the spin shot noise. By analogy with the charge shot noise, the quantization of the angular momentum transfer leads to spin shot noise. The effect is a random torque acting on the free ferromagnet. It was shown by Foros et al. [7] that the spin shot noise is the dominant contribution to magnetization noise at low temperatures. Their study was restricted to NFN structures and they did not discuss the problem when two ferromagnetic leads are present. Spin-dependent shot noise was studied also in [35–38]. The authors of these works mainly concentrated on the relationship between the noise and intrinsic properties of the materials, such as the spin-flip scattering rate, spin–orbit coupling, magnetic impurities, spin and charge relaxation times, and so on. In [39], some of the present authors investigated spin shot noise in MTJs by means of a Keldysh approach. It was shown that inclusion of the nonequilibrium noise in the LLG can explain the experimentally observable nonmonotonic dependence of the microwave power spectrum on the voltage, as well as its saturation at low temperatures. We will discuss this approach in Sect. 15.3.

Finally, it is worth mentioning that effects of noise are also considered in connection with nonuniform magnetic structures [40, 41].

15.3 Langevin-Approach

There are at least two different approaches to noise in magnetic system. In the magnetoelectronic circuit theory [42–44], which is an extension of the Landauer–Büttiker (LB) approach [18, 45], spin current fluctuations are calculated from a scattering problem [19]. The absorption of their transverse component allows to identify the spin current fluctuations with a corresponding random torque. The latter can be used to enlarge the equations of motion by a stochastic spin-torque term. An alternative approach is the following: Starting from a microscopic model, one can directly derive the equations of motion for the magnetization of a free magnetic layer in contact with a spin-polarized current. Fluctuations will naturally arise due to the nonequilibrium situation, and will comprise the random part of the stochastic LLG

$$\frac{d\mathbf{m}}{dt} = -\gamma_0 \mathbf{m} \times \mathbf{H}_{\text{eff}} + \alpha_0 \mathbf{m} \times \frac{d\mathbf{m}}{dt} + \frac{\gamma_0}{M_s \mathcal{V}} \mathbf{m} \times [\mathbf{m} \times (\mathbf{I}_s + \mathbf{I}_s^R)], \quad (15.2)$$

where $\mathbf{m}$ is a unit vector in the free layer's magnetization direction, $\mathbf{H}_{\text{eff}}$ the effective magnetic field, $\mathcal{V}$ the volume of the switching element, M_s the absolute value of the free layer's magnetization, γ_0 the gyromagnetic ratio, α_0 the Gilbert damping parameter, and $\mathbf{I}_s + \mathbf{I}_s^R$ the spin-polarized current with random part $\mathbf{I}_s^R$. Since the result of this approach will be the derivation of the stochastic LLG (15.2), which is a Langevin-type equation, we will call it the Langevin approach. Following [39], we demonstrate the method on the exemplary model of a MTJ consisting of a free and a fixed ferromagnetic layer separated by a tunnel barrier.

Let us introduce our model Hamiltonian, allowing for an external magnetic field $\mathbf{H}$, tunneling of itinerant electrons through the barrier and exchange coupling between the itinerant electrons and the free layer's magnetization. It reads

$$H_0 = \sum_{k,\sigma} \epsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{l\sigma} \epsilon_l d_{l\sigma}^\dagger d_{l\sigma} - \gamma \mathbf{S} \cdot \mathbf{H} - 2J \mathbf{S} \cdot \mathbf{s} + \left[\sum_{kl\sigma} W_{kl} c_{k\sigma}^\dagger d_{l\sigma} + h.c. \right]. \quad (15.3)$$

The notation is as follows: The creation (annihilation) operators $c_{k\sigma}^\dagger$ ($c_{k\sigma}$) and $d_{l\sigma}^\dagger$ ($d_{l\sigma}$) describe the itinerant electrons of the fixed and the free magnetic layer, respectively. $\sigma = +$ corresponds to the respective majority and $\sigma = -$ to the minority spin band, and the indices k and l label momentum. The operator $\mathbf{S}$ describes the total spin of the free layer. It is connected to the free layer's magnetization via $\mathbf{S} = \mathbf{M}\mathcal{V}/\gamma$. $\mathbf{s} = \frac{1}{2} \sum_{l\sigma\sigma'} d_{l\sigma}^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} d_{l\sigma'}$ is the quantum operator associated with the spin of itinerant electrons, where $\boldsymbol{\sigma}$ denotes the vector of Pauli matrices. J is the exchange coupling constant and W_{kl} are tunneling matrix elements.

We make the following assumption, which is essential in the subsequent considerations: We assume that the time between two tunneling processes is much larger than the relaxation time in the free ferromagnet, or in other words: We have a complete spin relaxation in the free magnetic layer. This assumption, valid in the sequential tunneling regime, allows us to introduce an *instantaneous* reference frame with spin quantization axis directed along the free layer's magnetization direction. To account for this situation, we can apply a unitary transformation $U_{\theta,\phi}$ that rotates the reference frame from the laboratory (z axis in fixed layer's magnetization direction) to the instantaneous one (z' axis in free layer's magnetization direction). The polar angle θ and the azimuth angle ϕ characterize the position of the free layer in the laboratory coordinate system. To render the free layer's magnetization a dynamical variable, we make use of the Holstein–Primakoff parametrization [46]

$$S_z = S - b^\dagger b, \quad S_- = b^\dagger \sqrt{2S - b^\dagger b}, \quad S_+ = \sqrt{2S - b^\dagger b} b, \quad (15.4)$$

where $b^\dagger, b$ are usual bosonic operators and $S_\pm = S_x \pm iS_y$. At low temperatures, we can assume that the expectation value of $b^\dagger b$ is much smaller than $2S$ allowing

to treat the square root to zeroth order in $b^\dagger b$. Taking all of the above mentioned into account, we can write for the Hamiltonian (15.3) in the instantaneous reference frame

$$\begin{aligned}
 H_0 = & \sum_{k,\sigma} \epsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{l\sigma} (\epsilon_l - JS\sigma) d_{l\sigma}^\dagger d_{l\sigma} \\
 & + Jb^\dagger b \sum_{l\sigma} \sigma d_{l\sigma}^\dagger d_{l\sigma} - \gamma SH_z + \gamma b^\dagger b H_z \\
 & + \left[\sum_{kl,\sigma\sigma'} W_{kl}^{\sigma\sigma'} c_{k\sigma}^\dagger d_{l\sigma'} - b\sqrt{2S} \left(J \sum_l d_{l\downarrow}^\dagger d_{l\uparrow} + \frac{\gamma}{2} H_- \right) + h.c. \right], \quad (15.5)
 \end{aligned}$$

where we used the notation $H_\pm = H_x \pm iH_y$. The unitary transformation to the instantaneous reference frame, results in spin-dependent tunneling matrix elements

$$W_{kl}^{\sigma\sigma'} = \langle \sigma | \sigma' \rangle W_{kl}, \quad \langle \sigma | \sigma \rangle = \cos \frac{\theta}{2} e^{-\frac{i}{2}\sigma\phi}, \quad \langle \sigma | \sigma' \rangle = \sigma' \sin \frac{\theta}{2} e^{\frac{i}{2}\sigma\phi} \quad (15.6)$$

appearing in the Hamiltonian (15.5).

How to proceed? Due to our parametrization (15.4), the dynamics of magnetization is encoded in the time dependence of the bosonic operators b and $b^\dagger$. Hence, we would like to derive the respective equations of motion, which – once translated back into the laboratory coordinate system – we expect to reproduce the LLG equation along with possible higher-order corrections. A suitable route to this end is to apply the Keldysh formalism [47], which enables us to cope with the nonequilibrium situation. In any case, we are left to a perturbative expansion of Hamiltonian (15.5) respective of the resulting Keldysh action. Processes relevant for the spin torque should be those in nonzero order in both the tunneling and the spin flips, reflecting the coupling to the reservoirs (finite bias) and the underlying mechanism of spin transfer. The corresponding diagrammatic contributions are hence easy to guess. They are sketched in Fig. 15.2. However, we still have to be careful: Since we are working in the instantaneous reference frame, we have to keep in mind that in this coordinate system we have for $m_\pm = m_x \pm im_y$

$$\langle m_\pm \rangle = 0, \quad \langle \partial_t m_\pm \rangle \neq 0, \quad (15.7)$$

whereas

$$\langle m_z \rangle = 1, \quad \langle \partial_t m_z \rangle = 0. \quad (15.8)$$

Finally, we must not forget that there is a relative shift of the chemical potentials in the free and fixed magnetic layer, corresponding to the applied bias voltage.

The calculation goes now as follows. In compliance with the general scheme of the Keldysh approach, we switch to symmetric (“ c ”) and antisymmetric (“ q ”) linear combinations of the field operators. The former correspond to the dynamical variables, and in accordance with parametrization (15.4) they are connected to the

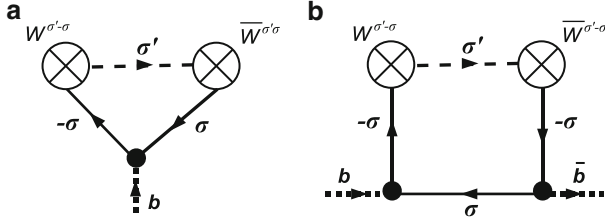


Fig. 15.2 Diagrams for spin-flip processes: **a** first order, **b** second order. Solid (dashed) lines denote electronic propagators in the free (fixed) layer. Bold dashed lines are propagators of HP bosons. Tunneling vertices are denoted by circles with crosses

$m_{\pm}$ components of the free layer's magnetization in the instantaneous reference frame via

$$b_{cl}(t) = \sqrt{\frac{M_s \mathcal{V}}{2\gamma}} m_+(t), \quad \bar{b}_{cl}(t) = \sqrt{\frac{M_s \mathcal{V}}{2\gamma}} m_-(t). \quad (15.9)$$

We obtain the corresponding equations of motion when varying the action $\mathcal{A}$ with respect to the quantum component

$$\frac{\delta \mathcal{A}}{\delta b_q} = 0, \quad \frac{\delta \mathcal{A}}{\delta \bar{b}_q} = 0. \quad (15.10)$$

On the other hand, the (effective) bosonic action can be calculated from the above-mentioned perturbative expansion leading to the diagrams, shown in Fig. 15.2, for the first- and second-order spin-flip processes.

Let us see how the corresponding analytical expressions look like. To this end, we introduce the fermionic Green functions for the itinerant electrons of the free and fixed layer. The Keldysh formalism involves a matrix structure of the Green functions, with a retarded (R), advanced (A), and Keldysh (K) component [47]. For the retarded and advanced components in the energy domain, we obtain

$$G_{l\sigma}^{\text{R/A}} = \frac{1}{\epsilon - \epsilon_{l\sigma} \pm i0}, \quad G_{k\sigma}^{\text{R/A}} = \frac{1}{\epsilon - \epsilon_{k\sigma} \pm i0}, \quad (15.11)$$

where $\epsilon_{l\sigma} = \epsilon_l - \sigma JS$ are the energies of the itinerant electrons with momentum l and spin σ in the free ferromagnet, and $\epsilon_{k\sigma}$ the corresponding energies for the fixed layer. The Keldysh components are

$$G_{l\sigma}^K = (1 - 2n_F^d(\epsilon))\delta(\epsilon - \epsilon_{l\sigma}), \quad G_{k\sigma}^K = (1 - 2n_F^c(\epsilon))\delta(\epsilon - \epsilon_{k\sigma}), \quad (15.12)$$

where chemical potentials $\mu_{d/c}$ for the free and fixed layer are included in the fermionic distribution functions $n_F^{c/d}$. Finally, for future use, we define the matrices

in Keldysh space

$$\gamma^{cl} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \quad \gamma^q = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \quad (15.13)$$

We may now translate the diagrams shown in Figs. 15.2a,b into the analytical expressions. However, let us start with the contribution of zeroth order (in spin flips and in tunneling). It reads

$$\mathcal{A}_0 = \int dt \bar{b}_q(t) \left(i \partial_t b_{cl}(t) + \gamma \sqrt{S/2} H_+ \right) + \text{c.c.} \quad (15.14)$$

The resulting equations of motion are

$$i \partial_t b_{cl} + \gamma \sqrt{S/2} H_+ = 0 \quad (15.15)$$

and a corresponding complex conjugate equation for $\bar{b}_{cl}$. Equation (15.15) describes the precession of the magnetization around the magnetic field $\mathbf{H}$ and forms the first term of the LLG equation (15.2).

Let us come to the diagram shown in Fig. 15.2a. To extract its contribution to the action, we have to calculate

$$-J\sqrt{S} \sum_{kl\sigma\sigma'} W_{kl}^{\sigma'-\sigma} \bar{W}_{kl}^{\sigma'\sigma} b_\sigma \text{Tr} \left\{ G_{l\sigma}^d \gamma^q G_{l-\sigma}^d G_{k\sigma'}^c \right\}, \quad (15.16)$$

where for brevity the symbolic notation b_σ with $b_\uparrow = b_q$ and $b_\downarrow = \bar{b}_q$ was introduced. The resulting action reads

$$\mathcal{A}_1 = \frac{i}{2\sqrt{S}} I_s \int dt \left\{ \bar{b}_q(t) \sin \theta e^{-i\phi} - b_q(t) \sin \theta e^{i\phi} \right\}. \quad (15.17)$$

Variation of (15.17) with respect to b_q and $\bar{b}_q$ gives the following contribution to the equations of motion

$$\frac{\delta \mathcal{A}_1}{\delta b_q(t)} = -i \frac{I_s}{\sqrt{2S}} \sin \theta e^{i\phi}, \quad \frac{\delta \mathcal{A}_1}{\delta \bar{b}_q(t)} = i \frac{I_s}{\sqrt{2S}} \sin \theta e^{-i\phi}. \quad (15.18)$$

Again, using the HP parametrization (15.4) and the relation between $\mathbf{S}$ and $\mathbf{m}$, (15.18) can be readily translated into the corresponding equation of motion for the magnetization. The result is the spin-torque term of (15.2)

$$\partial_t \mathbf{m} = \frac{\gamma}{M_s \mathcal{V}} \mathbf{m} \times (\mathbf{I}_s \times \mathbf{m}). \quad (15.19)$$

As far as the remaining diagram (Fig. 15.2b) is concerned, we have to distinguish two contributions: One with two quantum components and one with a quantum and a classical component, respectively.¹ In the first case, we obtain

$$J^2 S b_q \bar{b}_q \sum_{kl\sigma\sigma'} |W_{kl}^{\sigma'\sigma}|^2 \text{Tr} \left\{ G_{l-\sigma}^d(\varepsilon) \gamma^q G_{l\sigma}^d(\varepsilon - \omega) \gamma^q G_{l-\sigma}^d(\varepsilon) G_{k\sigma'}^c(\varepsilon) \right\}. \quad (15.20)$$

In the second case we have²

$$J^2 S b_{cl} \bar{b}_q \sum_{kl\sigma\sigma'} |W_{kl}^{\sigma'\sigma}|^2 \text{Tr} \left\{ G_{l-\sigma}^d(\varepsilon) \gamma^q G_{l\sigma}^d(\varepsilon - \omega) \gamma^{cl} G_{l-\sigma}^d(\varepsilon) G_{k\sigma'}^c(\varepsilon) \right\}. \quad (15.21)$$

The resulting action is

$$\mathcal{A}_2 = \int dt \left[\bar{\alpha}(\theta) (\bar{b}_q \partial_t b_{cl} - \bar{b}_{cl} \partial_t b_q) + \frac{2i}{S} \mathcal{D}(\theta) \bar{b}_q b_q \right], \quad (15.22)$$

where

$$\bar{\alpha}(\theta) = \frac{\hbar \gamma}{e M \mathcal{V}} \left(\frac{dI_{\text{sf}}(\theta)}{dV} \right), \quad (15.23)$$

$$\mathcal{D}(\theta) = \frac{M_s \mathcal{V}}{\gamma} \alpha_0 k_B T + \frac{\hbar}{2} I_{\text{sf}}(\theta) \coth \left(\frac{eV}{2k_B T} \right). \quad (15.24)$$

The spin-flip current I_{sf} can be calculated from the electric conductances $G_{\text{P(AP)}}$ in the parallel (antiparallel) configuration as follows

$$\frac{dI_{\text{sf}}(\theta)}{dV} = \frac{\hbar}{4e} \left[G_{\text{P}} \sin^2 \left(\frac{\theta}{2} \right) + G_{\text{AP}} \cos^2 \left(\frac{\theta}{2} \right) \right]. \quad (15.25)$$

Prior to discussing the meaning of this quantity let us inspect the action (15.22) and the resulting equations of motion more closely. The actions contains two parts. The first term is a damping term. In the LLG equation, it will result in a renormalization of the Gilbert damping parameter. The renormalization is due to the coupling to the reservoirs. The enhancement of the damping, (15.23), is closely related to the spin pumping enhanced damping as discussed in [48, 49] in the framework of the LB formalism. We thus recover the same result as was obtained within the LB approach: The nonequilibrium situation leads to dissipation and therefore to a modified FDT. What about the second term of (15.22)? As one can see the term is quadratic in the quantum component. The usual procedure in such a case [47] is to introduce a Hubbard–Stratonovich auxiliary field, which decouples the action. Let us denote this

¹ The $cl-cl$ component vanishes by virtue of the fundamental properties of the Keldysh formalism.

² In addition there is a contribution with $q \leftrightarrow cl$.

(complex) field by $I_+^R = I_{s,x}^R + iI_{s,y}^R$. Demanding $\int dI_+^R d\bar{I}_+^R \exp\{-\frac{1}{4\mathcal{D}} I_+^R \bar{I}_+^R\} = 1$, we can write

$$\begin{aligned} & \int dI_+^R d\bar{I}_+^R e^{-\frac{1}{4\mathcal{D}} I_+^R \bar{I}_+^R} e^{i\mathcal{A}_{21}} \\ &= \int dI_+^R d\bar{I}_+^R e^{-\frac{1}{4\mathcal{D}} I_+^R \bar{I}_+^R} \exp\left\{-i\frac{1}{\sqrt{2S}} (I_+^R \bar{b}_q + \bar{I}_+^R b_q)\right\}, \end{aligned} \quad (15.26)$$

where we abbreviated the second term of (15.22) by $\mathcal{A}_{22}$. As one can see the result is a noise-averaged term that is linear in the quantum component. The linear action constitutes a resolution of functional δ -functions of the Langevin equations on $b_{cl}(t)$ and its complex conjugate. The stochastic properties are encoded in the auxiliary field I_+^R , precisely in the correlator (15.24). For b_{cl} , the Langevin equations read $i\partial_t b_{cl} = \frac{1}{\sqrt{2S}} I_+^R$. This corresponds to $i\partial_t m_+ = \frac{\gamma}{M\mathcal{V}} I_+^R$ leading to the random term of the stochastic LLG equation. Adopting the notation $\mathbf{I}_s^R \equiv \delta\mathbf{I}_s$, in conclusion, we have found

$$\partial_t \mathbf{m} = \frac{\gamma}{M_s \mathcal{V}} \mathbf{m} \times (\delta\mathbf{I}_s \times \mathbf{m}), \quad (15.27)$$

where the stochastic field is characterized by

$$\langle \delta I_{s,i}(t) \delta I_{s,j}(t') \rangle = 2\mathcal{D}(\theta) \delta_{ij} \delta(t - t') \quad (15.28)$$

with the correlator $\mathcal{D}(\theta)$ given by (15.24).

To complete our discussion, we add some comments concerning the correlator (15.24). To start with, we note that $\mathcal{D}$ contains two parts, an equilibrium part (which is phenomenological, and in compliance with the FDT proportional to α_0 taking into account intrinsic damping processes) and a nonequilibrium part. The nonequilibrium part exhibits a dependence on the mutual orientation of the fixed and free layer's magnetizations. This angle dependence enters the correlator through the spin-flip current I_{sf} . The physical meaning behind this quantity is the following: I_{sf} counts the total number of spin-flip events, irrespective of their direction. Hence, even if there is no contribution to the spin current I_s , the spin-flip current I_{sf} may acquire a nonzero value. The discreteness of angular momentum transfer in each spin-flip event leads to the occurrence of the nonequilibrium noise. In this sense, the nonequilibrium part of (15.24) can be identified with the spin shot noise.

15.4 Fokker–Planck Approach to Spin-Torque Switching

Spin-torque switching is observable in two different regimes. On the one hand, the spin torque can switch the magnetization of a free ferromagnet when the current exceeds a critical value I_c . On the other hand, switching is also observed for currents below I_c . In the second case, the actual switching procedure is mainly noise induced.

A suitable description of switching times in this regime can be obtained from the Fokker–Planck approach, which was recently introduced by Apalkov and Visscher in the context of thermal fluctuations [5, 6]. Within this approach, switching rates are specified by an Arrhenius like law with an *effective* temperature T_{eff} . The latter differs from the real temperature T , as it is influenced by the damping and the spin torque. In the sequel, we present a generalization of the method to nonequilibrium noise and show that the spin shot noise alters the effective temperature.

Let us start our consideration with the Fokker–Planck equation as introduced by Brown [1]. We denote the probability density for the magnetization of a mono-domain particle by $\rho(\mathbf{m}, t)$. The corresponding Fokker–Planck equation can be written in the form of a continuity equation

$$\frac{\partial \rho(\mathbf{m}, t)}{\partial t} = -\nabla \cdot \mathbf{j}(\mathbf{m}, t) \quad (15.29)$$

with probability current [1]

$$\mathbf{j}(\mathbf{m}, t) = \rho(\mathbf{m}, t) \dot{\mathbf{m}}_{\text{det}}(\mathbf{m}) - D \nabla \rho(\mathbf{m}, t). \quad (15.30)$$

Here $\dot{\mathbf{m}}_{\text{det}}$ denotes the deterministic part of the stochastic LLG (15.2) and D is the random field correlator. We recall that the dynamics governed by (15.2) conserves the absolute value of $\mathbf{m}$. As a consequence, the movement of the tip of $\mathbf{m}$ is restricted to the surface of a sphere, which we will call the m -sphere. The gradient and the divergence in (15.29) and (15.30) are two-dimensional objects, both living on the m -sphere.

We now observe that in presence of anisotropy the phase space will be in general separated. The potential landscape will exhibit different minima referring to stable and metastable states of the magnetization. Precession of the magnetization takes place around one (or more) of these equilibrium positions. We refer to orbits of constant energy as Stoner–Wohlfarth (SW) orbits. Now, considering the dynamics of the magnetization vector one can distinguish two different time scales. The time scale for the angular movement, on the one hand, is characterized by the precession frequency. On the other hand, there is also a time scale for a possible change in energy. In the following, we will require that the time scale for the change in energy is much longer than the time scale for constant energy precession. In other words: we assume that the magnetization vector stays rather long on a SW orbit before changing to higher/lower energies. In this low damping and small current limit, we can introduce an energy-dependent probability density by identifying $\rho'_i(E(\mathbf{m}), t) \equiv \rho(\mathbf{m}, t)$, where the index i takes into account that the energy dependence may be different in different regions of the m -sphere. The above-mentioned time scale separation allows us to average out the movement along the SW orbit and to be concerned with only the long time dynamics.

The idea of the FP approach is to translate (15.29) into a corresponding equation for $\rho'_i(E)$. For thermal noise, this has been done in [5]. We now give a generalization of the method to the angle-dependent spin shot noise of Sect. 15.3. To this end, we

write the correlator (15.24) in the form

$$D(\theta) = D_{\text{th}} + D_0 [1 - P \cos \theta], \quad (15.31)$$

where D_{th} is the thermal part (first term of (15.24)) and $D_0 [1 - P \cos \theta]$ the nonequilibrium part (second term of (15.24)) of the correlator. We abbreviated the angle-independent part of the spin shot noise by D_0 . We also used

$$P = \frac{G_{\text{P}} - G_{\text{AP}}}{G_{\text{P}} + G_{\text{AP}}}. \quad (15.32)$$

In general, we can write the Fokker–Planck equation for the distribution $\rho'_i(E(\mathbf{m}), t)$ in the form

$$\frac{\gamma P_i(E)}{M_s \mu_0} \frac{\partial \rho'_i(E, t)}{\partial t} = -\frac{\partial}{\partial E} j_i^{\text{E}}(E, t), \quad (15.33)$$

where $P_i(E)$ is the period of the orbit with energy E . j_i^{E} is the probability current in energy. It is given by

$$\begin{aligned} j_i^{\text{E}}(E, t) &= \oint [\mathbf{j}(\mathbf{m}, t) \times d\mathbf{m}] \cdot \mathbf{m} \\ &= -\gamma \alpha \rho'_i(E, t) I_i^{\text{E}}(E) + \gamma J \rho'_i(E, t) \mathbf{m}_p \cdot \mathbf{I}_i^{\text{M}} - \frac{\partial \rho'(E)}{\partial E} M_s D_{\text{th}} I_{\theta, i}^{\text{E}}. \end{aligned} \quad (15.34)$$

The constant J is defined in such a way that $J \mathbf{m}_p = \gamma / (M \mathcal{V}) \mathbf{I}_s$ if $\mathbf{m}_p$ is a unit vector in direction of $\mathbf{I}_s$. Furthermore, we have introduced the following integrals along the SW orbit

$$I_{\theta, i}^{\text{E}} = I_i^{\text{E}} + \frac{D_0}{D_{\text{th}}} \left(I_i^{\text{E}} - P \oint \cos \theta H_{\text{eff}} d\mathbf{m} \right), \quad (15.35)$$

$$I_i^{\text{E}}(E) = \oint H_{\text{eff}} d\mathbf{m}, \quad (15.36)$$

$$\mathbf{I}_i^{\text{M}}(E, t) = \oint d\mathbf{m} \times \mathbf{m}. \quad (15.37)$$

A steady state solution of the FP equation is obtained by setting $j_i^{\text{E}} = 0$. From (15.34), we get the following differential equation for the probability density ρ' :

$$\frac{\partial \ln \rho'(E)}{\partial E} = \frac{\gamma}{D_{\text{th}} M_s} \lambda(E) [-\alpha + \eta(E) J] \equiv -\mathcal{V} \beta'(E), \quad (15.38)$$

where the right-hand side serves as a definition of an inverse effective temperature $\beta'(E)$. From (15.38), one can see that, depending on the sign of the spin current,

the spin torque may either enhance or diminish the damping, leading to a lower or higher effective temperature, respectively. In (15.38), we have defined

$$\eta_i(E) = \frac{\mathbf{m}_p \cdot \mathbf{I}_i^M(E)}{I_i^E(E)} \quad (15.39)$$

and

$$\lambda(E) = \frac{I^E}{I_\theta^E}. \quad (15.40)$$

η_i can be viewed of as the ratio of the work of the Slonczewski torque to that of the damping [5]. The quantity λ gives the renormalization of the effective temperature as compared to the pure thermal case. We can write for λ

$$\lambda(E) = \frac{T_{\text{eff}}}{T'_{\text{eff}}}, \quad (15.41)$$

where T_{eff} is the effective temperature when only equilibrium noise is present and T'_{eff} the effective temperature when both equilibrium and nonequilibrium noise are included. It should be observed from (15.38) that the effective temperature is in general energy dependent. The corresponding probability distribution will thus, in general, differ from the Boltzmann distribution. However, when we turn off the nonequilibrium, $\lambda(E) \equiv 1$ and $J = 0$. In this case, the solution of (15.38) is exactly a Boltzmann distribution.

In the remainder of this section, we evaluate λ for an exemplary system with easy axis and easy plane anisotropy. The easy axis is chosen to be the z axis and the easy plane is the y - z plane. The magnetization direction of the fixed layer, $\mathbf{m}_p$, is taken to be antiparallel to the z axis. Let us use the following convention for the spherical coordinates: $m_x = \cos \vartheta$, $m_y = \sin \vartheta \sin \varphi$, $m_z = \sin \vartheta \cos \varphi$. The SW condition defines the orbits of constant energy. For our system, it reads

$$\frac{E(\mathbf{M})}{\mu_0} = -\frac{1}{2} H_K M_S (\mathbf{m} e_z)^2 + \frac{1}{2} M_S^2 (\mathbf{m} e_x)^2. \quad (15.42)$$

We abbreviate $\kappa_1 = H_K M_S$ (characterizing the strength of easy-axis anisotropy), $\kappa_2 = M_S^2$ (characterizing the strength of easy-plane anisotropy) and $d = \frac{\kappa_1}{\kappa_2}$, being the ratio of easy-axis to easy-plane anisotropy, so that (taking the magnetic constant $\mu_0 = 1$) we can obtain from (15.42) the dimensionless energy c

$$c \equiv \frac{2E}{\kappa_2} = -dm_z^2 + m_x^2. \quad (15.43)$$

This relation defines the “potential landscape” of our system. We can distinguish three regions: Two potential wells, one around $\varphi = 0$ (well 1) and one around $\varphi = \pi$ (well 2), and a third region (region 3) with energies above the saddle point energy, separating the two wells. Switching takes place if the magnetization vector changes from some orbit in the one well to an orbit in the other well. Equation (15.43)

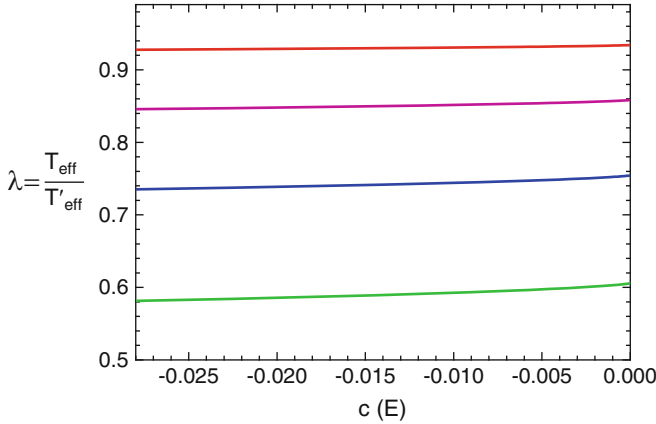


Fig. 15.3 $\lambda = T_{\text{eff}}/T'_{\text{eff}}$ as a function of c in the case $\mathbf{m}_p \uparrow \downarrow \mathbf{e}_z$ for $eV = k_B T$ (red), $eV = 5k_B T$ (magenta), $eV = 10k_B T$ (blue), $eV = 20k_B T$ (green)

defines the orbits of integration for the evaluation of (15.40). Let us concentrate on orbits lying in the potential well around $\varphi = 0$ with energies $c \leq 0$.³ In addition, we assume a strong easy plane anisotropy, allowing to consider small deviations of ϑ around $\frac{\pi}{2}$.

We fix the Gilbert damping to $\alpha = 0.01$, the ratio of anisotropies to $d = 0.028$, the polarization to $P = 0.81$, and $M_s \mathcal{V} / \gamma = 10\hbar$. These values define the ratio D_0/D_{th} as a function of $eV/k_B T$. The results for $\lambda = T_{\text{eff}}/T'_{\text{eff}}$ are plotted in Fig. 15.3. As can be seen from the plot taking into account the nonequilibrium noise results in a renormalization of the effective temperature. This renormalization is proportional to the applied voltage V and can be very strong for sufficiently large values of V . The deviation from the purely thermal case ($\lambda = 1$) approaches 15% for $eV = 5k_B T$ and is thus experimentally not negligible! For $eV = 10k_B T$, the deviation is even in the order of 25% and grows further with the voltage. The variation of λ with energy is on the other hand very weak. This indicates that the influence of the angle dependence is rather small or in other words: The angle dependence of the correlator does not lead to a significant variation of T_{eff} with precession orbit.

Let us continue our discussion of the renormalized effective temperature by considering the limit where the equilibrium part of the correlator is much smaller than its nonequilibrium part and thus may be neglected. In this case, we define the following quantity of interest

$$\lambda'(E) = \frac{D_0}{D_{\text{th}}} \lambda(E). \quad (15.44)$$

³ Note that due to the symmetry of (15.42) this can be done without loss of generality.

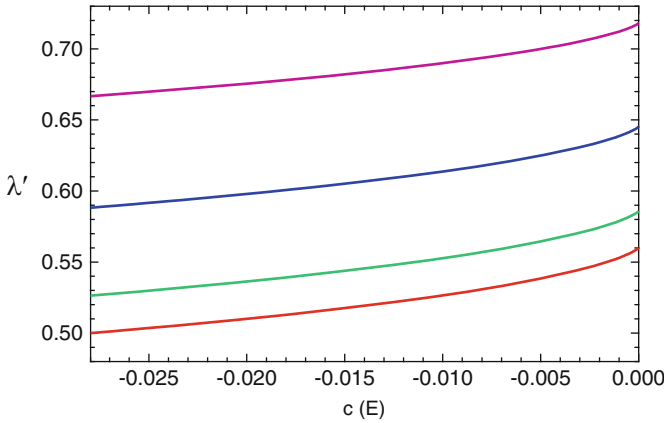


Fig. 15.4 λ' as a function of c in the case $\mathbf{m}_p \uparrow \downarrow \mathbf{e}_z$ for $P = 1$ (red), $P = 0.9$ (green), $P = 0.7$ (blue), $P = 0.5$ (magenta). $d = 0.028$

One should note the difference between λ and λ' . From (15.40), we see that λ is the ratio of the effective temperatures T_{eff} and T'_{eff} for systems without and with nonequilibrium noise respectively. On the other, from the definition (15.44), it is clear that λ' is a measure for the influence of the angle dependence of the correlator. The stronger λ' deviates from $\lambda' = 1$ the stronger is the influence of the angle dependence.

In Fig. 15.4, we plot λ' for our model system for $d = 0.028$ and different values of P . As one can see from Fig. 15.4 the largest deviation from $\lambda'(E) = 1$ (corresponding to the strongest influence of the angle dependence) is present at the minimum of the well ($c = -d = -0.028$). The smallest deviation from $\lambda'(E) = 1$ is observed for orbits which lie near the separatrix. The overall change of $\lambda'(E)$ for $P = 1$ is of the order of 10%.

These results provide a good insight into the influence of the angle dependence. As $\lambda' \sim 1/T'_{\text{eff}}$, a small value of λ' indicates a “hot” spot whereas large values of λ' correspond to “cold” spots on the m -sphere. For the particular system under consideration, cf. (15.42), the equilibrium position of the magnetization is roughly along the z axis. SW orbits of precession are symmetric with respect to this axis. At the bottom of the well $\theta = \pi$ and the spin shot noise has its maximal value. We hence expect a hot spot at the minimum of the well. With increasing energy the orbits will become larger. The angle θ will vary along these orbits. However, as the orbit energy grows the trajectories increasingly go through regions of smaller θ , so that the average value of θ will diminish with orbit energy. As a consequence, the nonequilibrium noise will become smaller as well. Cold orbits should be hence those that are in the vicinity of the separatrix. This is exactly what can be read off from Fig. 15.4. Our findings are thus in agreement with the geometrical situation. Cold spots and hot spots on the m -sphere are shown in Fig. 15.5.

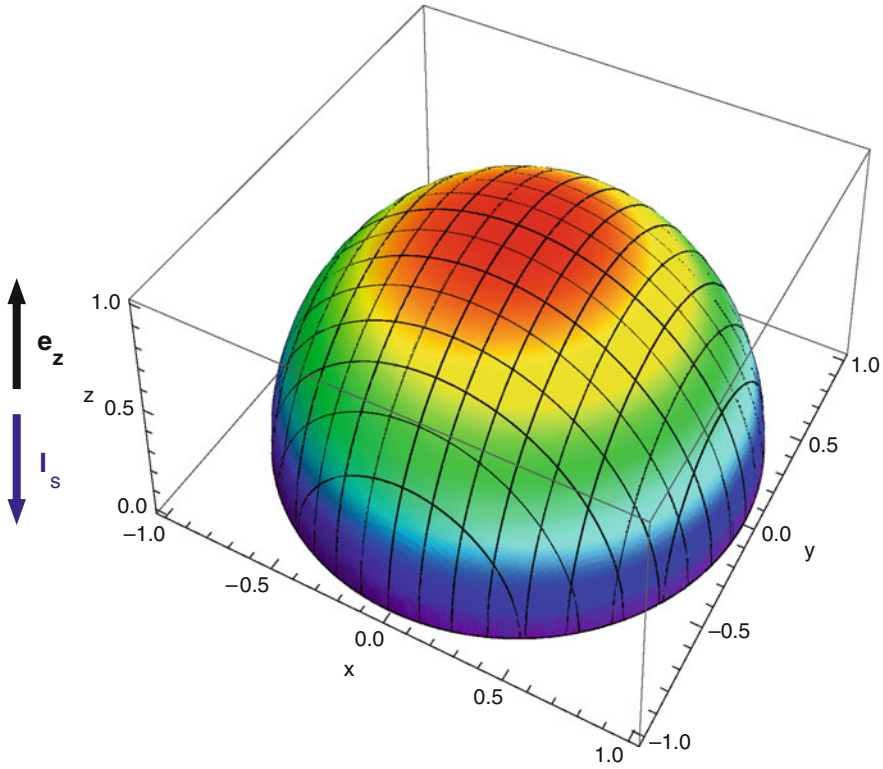


Fig. 15.5 Hot spots (red) and cold spots (blue) on the M -(half-) sphere in case of $\mathbf{m}_p \uparrow \downarrow \mathbf{e}_z$. The noise intensity is highest at the bottom of the well

15.5 Switching Time of Spin-Torque Structures

The switching process can be analyzed by performing numerical simulations of the Langevin equations of motion with the inclusion of temperature and shot noise via the random field term. In this section, we present such simulations for Gilbert damping of $\alpha = 0.01$, an anisotropy ratio of $d = 0.028$, and with a spin-torque current characterized by J and polarized in the $\mathbf{m}_p = -\mathbf{e}_z$ direction.

Before going further, it would be useful to consider how the system acts in the absence of the noise. In such a case, the switching occurs when the energy current (15.34) is positive for all values of energy between the starting position (say positive z direction) and the saddle point. Since the probability function $\rho_i^E(E, t)$ is always positive it stand to reason that a switch will only happen if $0 \leq -\alpha I_i^E(E) + J \mathbf{m}_p \cdot \mathbf{I}_i^M(E)$. We plot this quantity as a function of energy for various values of the spin-current J in Fig. 15.6. From this, we also gain a useful reference value for the critical current, which is $J_c = \frac{\alpha I_i^E(E_{\text{sad}})}{\mathbf{m} \cdot \mathbf{I}_i^M(E_{\text{sad}})} = 0.00645 M_s$. The positive value signifies the tendency toward the switching. In the first example with

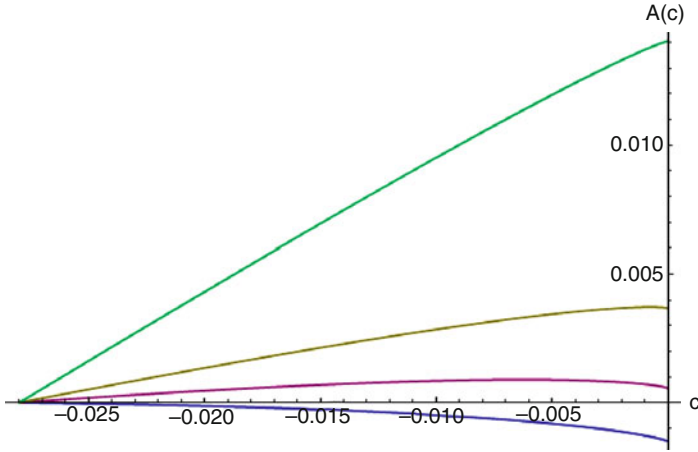


Fig. 15.6 Plots $A(E) = -\frac{\alpha}{M_s} I_i^E(E) + \frac{J}{M_s} \mathbf{m}_p \cdot \mathbf{I}_i^M(E)$ as a function of energy $c = 2E/\kappa_2$ over a range of spin-torque current. From bottom to top: $J = 0.77J_c$ (Blue), $J = J_c$ (Yellow), $J = 1.08J_c$ (Purple), $J = 1.55J_c$ (Green)

$J = 0.77J_c$, the noiseless system, being driven by the dissipation toward the stable position, does not switch. It is worth noticing that in the presence of the noise the switching nevertheless does occur, but it takes exponentially long time. In the three other examples $J \geq J_c$ and the magnetization current is always directed toward the saddle. Therefore, even the noiseless system does switch and the noise serves to introduce an uncertainty in the switching time.

Putting the thermal noise back into the system, we set the noise strength parameter to $D_{\text{th}} = 0.00001\gamma M_s$. Simulations are then run by starting each particle at $\theta = 0$, allowing it to come into thermal equilibrium with the system, turning the current on and calculating how long it takes for it to go past the saddle point into the second well. This is done for many particles for a given current value and over several different current values. A typical trajectory of the system is represented by the graph θ as a function of time in Fig. 15.7 for $J = 1.08J_c$. It may be seen that it takes many revolutions before the system finally switches to the basin of attraction of the true stationary points at $t \approx 200$. Moreover, even after the switching, the damped oscillations persist for quite a while.

Since the initial condition is taken out of a stationary distribution (without the spin current) and subsequent evolution is subject to the Langevin noise, the time of the switching is a random quantity. The percentage of trial systems that have switched as a function of time is shown in the left-panel of Fig. 15.8 for four different values of the spin-current. The time derivatives of these graphs provide probability distribution functions of the switching time. One may then evaluate the first moment of these distributions which gives the mean switching time for a given value of the spin current. The right-panel of Fig. 15.8 shows such a mean switching time as a function of J/J_c . One may notice that for $J \leq J_c$ the switching time grows exponentially, while for $J \geq J_c$ the switching time becomes relatively short (although it is still substantially longer than the inverse precession frequency).

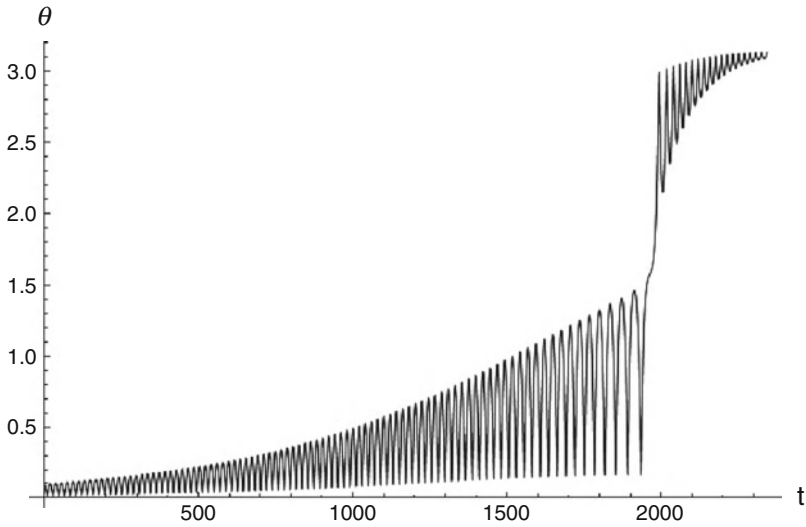


Fig. 15.7 A typical realization of θ as a function of time (in units of $(\gamma M_s)^{-1}$) for $J = 1.08J_c$

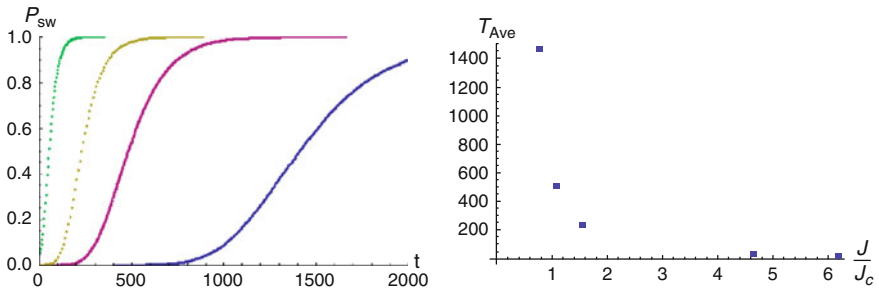


Fig. 15.8 *Left:* The switching probability as a function of time (in units of $(\gamma M_s)^{-1}$) for various current values. (Green = $3.10J_c$, Yellow = $1.55J_c$, Purple = $1.08J_c$, Blue = $0.77J_c$.) *Right:* The average switching time (in units of $(\gamma M_s)^{-1}$) as a function of J/J_c

15.6 Conclusions

We conclude with the following remarks. The study of noise in dynamical magnetic systems is a broad and fascinating field. In particular, in view of potential applications of magnetic nanodevices both equilibrium and nonequilibrium noise may play an important role. For example, the stability of magnetic storage devices is strongly influenced by thermal fluctuations. The functionality of new generation technologies (such as the magnetic random access memory (MRAM) [50] with STT writing or the racetrack memory [51]) is largely based on the spin-torque phenomenon. The latter is a nonequilibrium effect and, thus, nonequilibrium sources of noise may also play an important role beside the temperature.

A way to introduce fluctuations into magnetization dynamics is to add a random component to the effective field or to the current in the phenomenological LLG equation. The noise is then defined by the value of its correlator (and its higher-order cumulants). The determination of the noise correlator is of great importance as it defines the noise properties. In addition, it may give insight into the physical context.

A powerful and very flexible tool is the Langevin approach based on the Keldysh path integral formalism. Starting from a microscopic model, one derives the equations of motion for the magnetic system. Fluctuations naturally arise as a generic feature of the Keldysh approach. We have demonstrated the applicability of this method to magnetic systems on the example of spin shot noise in MTJs. The spin shot noise correlator arose naturally, as a consequence of the sequential tunneling approximation, in second order in spin-flip processes.

The Keldysh formalism is however not restricted to the system described above. In particular, it may be used in the context of nonuniform magnetic textures (as domain walls for instance). Promising advances in this direction have already been reported [41] and demonstrate the versatility of the method as well as inspire us with curiosity about future developments.

Finally, to investigate the influence of the spin shot noise on spin-torque switching rates, we have generalized the Fokker–Planck approach of [5]. We have shown that the nonequilibrium noise manifests itself in a renormalized effective temperature. In particular, at low temperatures we could observe a significant variation of the noise with orbit energy, reflecting “cold” and “hot” trajectories of the magnetization vector with respect to the noise intensity.

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Chapter 16

Nanostructured Ferromagnetic Systems for the Fabrication of Short-Period Magnetic Superlattices

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Abstract A new method to fabricate arrays of ferromagnetic nanostructures is presented which is based on copying the morphology of self-assembled organic layers via ion milling into ferromagnetic Co/Pt multilayers. The self-assembly of diblock copolymer micelles is used. A very flexible tuning of the magnetic properties is possible via the variation of the multilayer composition. The impact of the growth method on the magnetic properties of the multilayer is described and the spin reorientation in Co/Pt discussed. It is demonstrated that arrays of ferromagnetic and superparamagnetic particles can be fabricated with particle sizes <20 nm. The proposed method gives direct access to the tailoring of magnetic properties of nanosized objects.

16.1 Introduction

The electronic transport in a two-dimensional electron gas (2DEG) can be manipulated via laterally varying potentials. In our approach, we wanted to influence the 2DEG's electronic states via varying magnetic potentials. This should be generated by a nanoscaled magnetic dot array, fabricated on the surface of the 2DEG containing semiconductor. If the 2DEG in a periodic potential is subject to a magnetic field, the density of states is predicted to reveal fractal substructure [1]. The interplay between the period of the potential and the magnetic length leads to an intriguing energy spectrum as a function of the magnetic flux per supercell, which is called Hofstadter butterfly spectrum [2]. To obtain a reasonable energy splitting in such modulated potentials, a periodicity well below 100 nm has to be generated. Recently, first indications of such energy splitting could be experimentally verified. Most of the experiments have used electric superlattice potentials [3–11]. To achieve high amplitudes of magnetic field modulation, dot/antidot arrays with magnetization perpendicular to the supporting surface are the best choice, which additionally should be positioned as close as possible to the 2DEG.

The fabrication of ferromagnetic nanostructures and periodic arrays of nanosized ferromagnets is still a challenge in today's research, in particular when

structure sizes below 100 nm are addressed. The straightforward top down approach is to utilize artificial structuring like lithography using electron or ion beams [12,13]. Structuring via electron-beam lithography was successfully realized before, addressing the same issue of modulating a 2DEG on larger length scales [3, 9, 11]. Nowadays, electron-beam lithography is on the way to sub-20-nm structures [14], which would meet the requirements for our experiments. This ansatz, however, causes a tremendous amount of technical experience and high-end expensive instruments, which are generally not at hand of researchers involved in basic research activities.

An alternative competitive approach is to follow a bottom-up route, that is, use self-organization phenomena for structuring. These methods use e-beam evaporation to create very small nanostructures on single crystal surfaces [15–18] or wet chemical processes [19–22]. Both methods are limited to a small span in size and do not give a direct access to varying the magnetic properties. Hence, it is still a challenge to fabricate magnetic nanostructures with tunable properties. So, to pursue the way of using self organization, it is necessary to overcome that limitation. The most promising method is to use self-assembled structures as masks for copying the morphology into a magnetic system [23,24]. To fabricate dot arrays, structures like polystyrene spheres [25], oxide particles [26] and templates from block copolymers [24,27,28] act as masks. On the basis of self-organized phenomena, antidot arrays are mostly fabricated by growth of a magnetic film on top of structured material like porous anodic alumina [29–33] and more recently utilizing polystyrene spheres as shadow masks for magnetic film deposition [34,35].

In this chapter, a method that utilizes a layer of self-organized diblock copolymer micelles, which are filled with SiO_2 to generate both dot and antidot arrays, is reviewed. The nanoscaled morphology of a monomicellar layer is transferred into an ultrathin ferromagnetic multilayer via ion milling. The advantage of this fabrication process is that the magnetic properties can be tuned independently from the morphology. This gives high flexibility to adopt the magnetic properties of the ferromagnetic film to certain prerequisites, like special substrates or magnetic properties. Diblock copolymer micelles can be produced in a wide diameter range between 10 and 100 nm, which allows to vary the nearest neighbor distance in the self-assembled layer on a surface. The micelles build a locally hexagonal pattern. In a very successful cooperation with the group of S. Förster at the Institute of Physical Chemistry of the University of Hamburg, different approaches were tested. The results are given in Sect. 16.3.

As mentioned above, the nanoparticles should have an easy axis of magnetization perpendicular to the plane of the periodic arrangement of the particles. For that purpose, a magnetic anisotropy has to be generated in the particles. One possibility is to use the shape of the particle, like in a needle, to create a spontaneous magnetization orientation. This idea can be realized using elongated particles that stand upright. However, this arrangement is unstable and will be easily destroyed. A more stable solution is to use particles with larger dimensions in the plane of the arrangement than their height. Hence, dots based on ultrathin layered systems seem to be more appropriate. In particular, thin film systems can easily be structured by ion

milling and their magnetic properties can be tuned in a wide range. The problem of ultrathin ferromagnets, however, is that generally the perpendicular magnetization is energetically not favorable due to the magnetostatic self energy. This obstacle can be overcome in ultrathin systems when the surface/interface anisotropy favors a perpendicular easy axis. In such systems, the thickness of the ferromagnetic layer can be used as a tuning parameter that determines the strength of the perpendicular anisotropy.

As the perpendicular anisotropy is due to an interface effect, the quality of the latter has to be carefully controlled and almost perfect interfaces are needed. Indeed, the highest perpendicular anisotropies are found in the growth of ultrathin films on perfect single crystal surfaces. However, many investigations have shown that multilayers, grown by magnetron sputtering, can give the desired perpendicular anisotropy as well [36–45]. To optimize the anisotropy, however, is an important issue as the stray field, which has to be maximized, is the counterpart to the interface-determined perpendicular anisotropy. Hence, a buffer or seed layer is needed that mimics the required texture of the template, which should be close to an ideal single crystal. As we intended to deposit the nanomagnets directly on the sample containing the 2DEG such seed layers are essential. The buffer layer, however, causes a separation of the nanomagnets from the 2DEG, which will reduce the field modulation in the free electron gas. To keep that reduction small, the seed layer should be as thin as possible. The solution of these problems and the achieved anisotropies are discussed in the next section. In the last section, we discuss some magnetical issues that came up during the course of this study.

16.2 Multilayer Films with Perpendicular Anisotropy

In ferromagnets, the magnetic poles located in the interior experience a magnetic field that can be described by the magnetic surface charges of the spontaneously magnetized system. To prevent surface charges, the ferromagnet generally creates so-called flux-closure structures that consist of domains which are magnetized parallel to the surface. In ultrathin films where the thickness is smaller than a characteristic length (i.e., approximately the domain wall width), no domains can be built at the surface and the magnetization is in unison throughout the film. In that case, the perpendicular alignment of the magnetization creates magnetic poles at the surface and a strong field inside the film. When the magnetization is in the film plane, the demagnetizing field is vanishingly small. This is the reason for a strong self-energy contribution, the shape anisotropy, or demagnetizing energy, in thin films that tries to align the magnetization in the film plane. The total energy difference between these two states is $E_{\text{sh}} = \frac{1}{2}\mu_0 M_s^2$ with M_s the saturation magnetization and μ_0 the vacuum permeability. This value is huge compared to common volume values of magnetic anisotropy, K_V (Co: $E_{\text{sh}} \sim 1.28 \times 10^6 \text{ J m}^{-3}$, $K_V \sim 5.0 \times 10^5 \text{ J m}^{-3}$ [46]). It follows for ultrathin films that, in general, the magnetization prefers an in-plane alignment. The interfaces, however, may cause

contributions to the magnetic anisotropy that can occasionally support a perpendicular orientation. In some systems, the contribution is fairly large and can surmount the shape anisotropy. The surface/interface anisotropy stems from the symmetry breaking at the surface and was first proposed by Néel [47]. The free energy density of the film as a function of the magnetization orientation can be written as

$$f_{\text{film}} = \{2K_S/d + K_V - E_{\text{sh}}\} \sin^2 \theta \quad (16.1)$$

with K_S/K_V the interface/bulk anisotropy, d the film thickness, and θ the angle with respect to the normal. In case the number in the curly braces, often called effective anisotropy, is positive the magnetization will be aligned along the surface normal. To achieve a positive effective anisotropy, the surface contribution must exceed the second contribution, which is generally negative as the shape anisotropy is very high. If the surface anisotropy is positive, a perpendicular magnetization will be achieved in the thin-film limit. With increasing thickness, the magnetization orientation will flip into the film plane. This magnetization flip is called spin reorientation. A high perpendicular surface anisotropy is only possible when the interface is smooth and sharp [48].

As ferromagnetic thin film system Co/Pt multilayers have been chosen, which are well known for strong perpendicular anisotropy [37, 49]. In many cases, the films are fabricated via magnetron sputtering, which is most convenient and allows for a free selection of the substrate. It was found that epitaxial growth by electron-beam evaporation gives the highest values for the surface anisotropy, in particular, when Co is grown on Pt(111) single crystals [42, 50, 51]. This indicates that one important ingredient to achieve a surface anisotropy that favors perpendicular magnetization orientation is a (111) orientation of the Pt layers. This was theoretically predicted [52] and experimentally proven [53]. Vice versa, one can infer that the magnetron sputtering must give some amount of (111) texture in the Pt film, which is deposited first as a kind of seed layer [54]. Actually, thick seed layers have been used to increase the anisotropy [55, 56]. Interestingly, it was found that the ion bombardment of Pt seed layers prior to the deposition of Co enhances the perpendicular magnetic anisotropy. This was explained by the improvement of the texture and the smoothness of the seed layer [57]. Thus, the goal is to improve the texture and the seed layer surface quality when aiming at a seed layer thickness decrease.

We have investigated the potential of ion beam sputter deposition, in which an ion beam is used to remove the material from the target. The removed material then condensates on the substrate [58]. To achieve high ion flux, we use a plasma as ion source, which is generated by electron cyclotron resonance (ECR). In contrast to conventional magnetron sputter deposition, the pressure of the sputtering gas can be low for a given deposition rate while the energy of the primary ions is in the range of 1 keV. Both working conditions are responsible for somewhat higher energies (~ 20 eV) of the deposited material [58]. The higher impact energy helps to create many defects and local intermixing as nucleation points for further growth. A large number of nucleation centers leads to two-dimensional growth, that is, layer-by-layer growth, with smooth surfaces. On the other hand, the high impact energy can

be expected to stimulate the rearrangement of atoms in the growing film to achieve the most stable lattice orientation, that is, a (111) texture.

In our studies, we found that the Pt seed layer exhibits a reasonable high texture even in thin Pt films [58]. Utilizing the magnetism of a Co layer of constant thickness as a sensitive probe, we checked the seed layer quality. While a Co film that was deposited directly onto the substrate (oxidized Si) did not exhibit any magnetic response, a seed layer of 2 nm thickness resulted in a ferromagnetic response of the Co. The films on 2 and 4 nm seed layer were in-plane magnetized, the film on 6 nm Pt causes a perpendicular anisotropy. In these studies, we were able to demonstrate that perpendicular magnetized films can be grown on nearly any material using very thin seed layers. In first studies, both Pt (seed and cap layer) and Co layers were deposited via ECR ion beam sputtering utilizing 5.3×10^{-4} mbar working pressure, a deposition rate of $0.15\text{--}0.4 \text{ \AA s}^{-1}$ for Co, and $0.25\text{--}0.65 \text{ \AA s}^{-1}$ for Pt. The base pressure was in the low 10^{-9} mbar range.

When multilayers were grown, the magnetization flipped into the film plane after two to three repetitions, which made further optimizing necessary. One route is to optimize the growth condition of the ECR sputtering, which was partially successful. As an alternative, we have evaluated the achievable magnetic properties when using magnetron and ECR ion beam sputtering and the combination of both. The result is presented in the following.

At first, single Co layers sandwiched by Pt were studied. The magnetization behavior was investigated by magneto-optical Kerr effect (MOKE). Figures 16.1a and b display magnetization curves obtained for fields applied perpendicular to and in the film plane, respectively. The result for systems fabricated by combining both sputtering techniques is given in the same plot. Combining the techniques means that the seed layer is created via ECR sputtering and the Co and Pt cap layers are

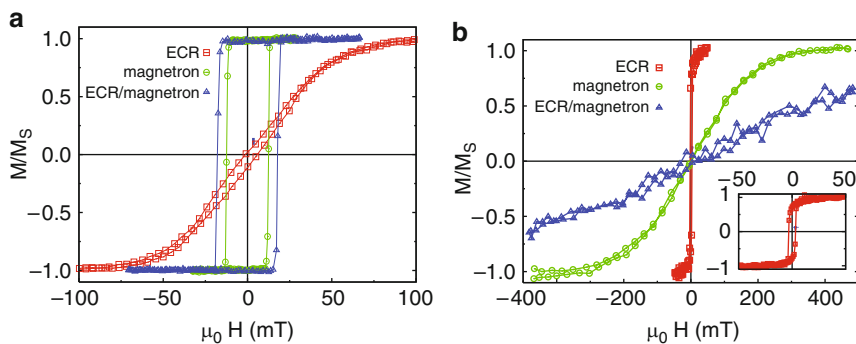


Fig. 16.1 MOKE hysteresis loops of Pt/Co/Pt single layer films grown by ECR, magnetron sputtering and combining ECR, and magnetron sputtering. The Co/Pt cap layer thickness was kept constant 0.7 nm/2 nm. For the Pt seed layer the thickness is 7 nm for magnetron sputtered films and 4 nm for the two other film systems. The polar Kerr rotation θ /longitudinal Kerr ellipticity ε was taken as a function (a) of the perpendicular and (b) of the in-plane field, respectively. The inset of (b) shows a low field scan of the ECR grown film

grown by magnetron sputtering. The film composition is the following: The Co thickness is 0.7 nm and the Pt cap layer has a thickness of 2 nm while the seed layer is 7 nm for magnetron sputtered films and 4 nm for seed layers deposited via ECR sputtering. The magnetization curves in perpendicular fields show hysteresis for the films that were made either partially or completely via magnetron sputtering. The magnetization in remanence is the same as in saturation, which indicates a bi-stable system that can be totally magnetized in the direction perpendicular to the film plane, that is, the films reveal a perpendicular easy axis of magnetization. The ECR made film shows almost no remanence, which indicates that the perpendicular direction is not an easy axis of magnetization. The relatively large slope demonstrates that the effective anisotropy is close to zero and negative. In in-plane fields, a hysteresis for the ECR made film is found (a low field scan is given as inset in Fig. 16.1b) while the other two films exhibit hard axis behavior. The hard axis curve of the magnetron sputtered film shows a larger slope than the magnetization curve for the film made by combining ECR and magnetron sputtering (ECR/magnetron). This result indicates that the effective anisotropy of the ECR/magnetron film is higher than that of the magnetron film although the seed layer is thicker in case of the magnetron made film. From Fig. 16.1, we conclude that the perpendicular magnetic anisotropy is smallest for the ECR-film and increases going from magnetron to ECR/magnetron sputtered films. From a more quantitative investigation, we obtain numbers for the surface/interface anisotropy in the above-discussed sandwiched Co single layer systems. For the ECR made film, we find $K_S = 0.29(\pm 0.02) \text{ mJ m}^{-2}$ and for the magnetron sputtered system $K_S = 0.38(\pm 0.02) \text{ mJ m}^{-2}$. The film that was made by magnetron sputtering on an ECR seed layer has the highest surface/interface anisotropy $K_S = 0.55(\pm 0.07) \text{ mJ m}^{-2}$, which fits well into the range of published values for magnetron sputtered multilayers mostly utilizing thicker seed layers. The growth conditions were: working pressure $1.6 \times 10^{-4} / 3.3 \times 10^{-3}$ mbar at acceleration voltages of 1.2/0.4 – 0.5 kV for ECR and magnetron sputtering, respectively.

For multilayers, the same trend is found. The magnetization curves for multilayers with composition $[0.5 \text{ nm Co} / 2.0 \text{ nm Pt}]_8$ grown by ECR and magnetron on an ECR sputtered Pt seed layer are shown in Fig. 16.2. The Kerr signals in saturation are almost the same in both films (Fig. 16.2a), which proves that they have the same composition. Small deviations are most likely caused by slight changes of the magneto-optic properties [59]. In perpendicular fields, the magnetron sputtered multilayer shows a square hysteresis with coercive fields of approximately 20 mT while the ECR sputtered multilayer is reversible around zero field and exhibits an irreversible behavior for higher magnetic fields. The latter shape of the hysteresis curve has been systematically investigated and correlated to domain wall propagation in a maze pattern and nucleation/annihilation of domains [60]. The appearance of these two different types of hysteresis in perpendicular fields is straightforwardly explained by different magnetic anisotropies. From that, one can deduce that the perpendicular magnetic anisotropy is higher in the ECR/magnetron sputtered film. The in-plane Kerr hysteresis allows for a quantitative estimation of the perpendicular magnetic anisotropy. While the ECR/magnetron sputtered multilayer cannot be saturated in the available in-plane field of 500 mT, the ECR sputtered film is saturated

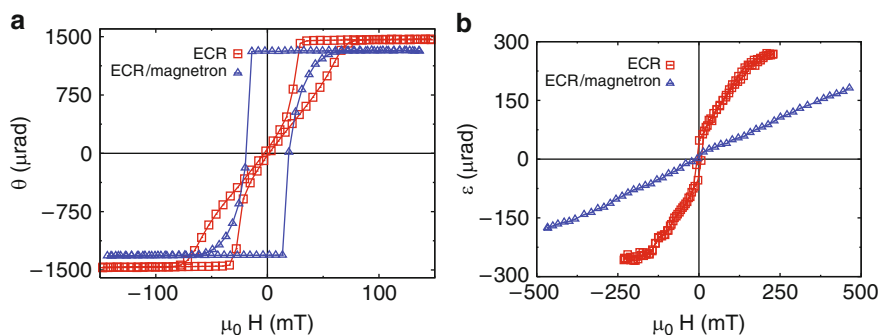


Fig. 16.2 Kerr Hysteresis loops of a $[0.5 \text{ nm Co}/2.0 \text{ nm Pt}]_8$ multilayer films grown by ECR and magnetron sputtering on a 4 nm ECR-sputtered seed layer. In (a) the polar Kerr rotation and in (b) the longitudinal Kerr ellipticity are plotted, showing the magnetization behavior in perpendicular/in-plane fields, respectively

at about 200 mT. From this result, we can deduce that the effective anisotropy of the latter multilayer is lower than that of the ECR/magnetron film, at least by a factor of 2.5. Additionally, a partial switching in in-plane fields is observed for the ECR sputtered film. The partial switching was identified as a fingerprint for a canted magnetization [61]. We will come back to that point when we discuss the magnetic properties of the multilayer films in Sect. 16.4.1.

The increase of the interface anisotropy in the series going from ECR to ECR/magnetron is due to sharper interfaces that are created when magnetron sputtering is used to grow the multilayers. Two different mechanisms can cause the decrease of interface quality. On the one hand, the interface between chemically well separated phases can be rough or the intermixing of the different materials gives a chemical roughness at the interface. Besides roughness, the intermixing or alloying at the interface can dramatically reduce the interface anisotropy [62, 63]. An intermixing of Co and Pt at each interface happens most likely in case of ECR sputtered multilayers. The higher energy of the material that is impinging on the sample is responsible for material exchange and embedding of the incoming material.

Additionally, the growing film is bombarded by Ar^+ ions with higher energies for ECR made films than in case of magnetron sputtering, which can also cause an intermixing at the interface [64, 65]. These effects are reduced for magnetron sputter deposition because of higher Ar pressure and the lower ion energy. The higher magnetic anisotropy of the ECR/magnetron sputtered multilayers compared to solely magnetron made systems indicates that the (111) texture of the seed layer is better for ECR than for magnetron sputtered films. The better crystal orientation of the seed layer represents a better template for the epitaxial growth of hcp [0001] Co. Another possibility is the roughness of the seed layer surfaces, which is apparently superior in case of ECR made buffer. In general, the finding is in agreement with the previous finding of improved magnetic quality on sputtering the seed layer prior to deposition of the Co layers [57].

In conclusion, we have been able to produce multilayers with high perpendicular magnetic surface anisotropy utilizing ECR sputtering for deposition of highly textured seed layers and magnetron sputtering to grow the Co/Pt multilayers with sharp interfaces. A further improvement of the multilayer properties is still under progress. The promising approach is to turn to noble gases with higher atomic weight, which was already demonstrated to give further enhancement of anisotropy [64]. Furthermore, it has to be ensured that the employed deposition techniques do not deteriorate the 2DEG.

16.3 Nanostructuring

The nanostructuring of the Co/Pt multilayers is performed via SiO₂-filled diblock copolymer micelles, which were fabricated in cooperation with the group of S. Förster of the University of Hamburg [68]. In the beginning of this section, the micelle fabrication is briefly presented. Next, the formation of monomicellar layers on substrates is discussed and, finally, the fabrication of arrays of dots and antidots is explained in detail utilizing the monomicellar layers.

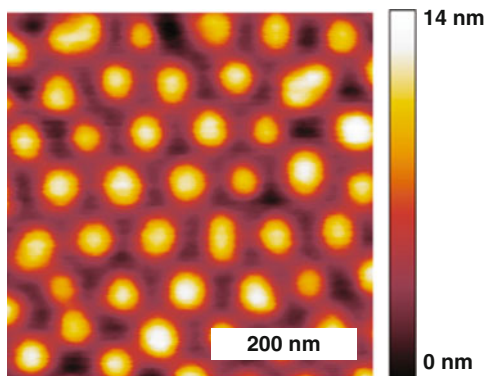
16.3.1 *Fabrication of Diblock Copolymer Micelles Filled with SiO₂*

Diblock copolymer micelles are synthesized from polystyrene and poly-2-vinylpyridine polymers following standard procedures [66]. Spherical micelles are formed in toluene. By adding tetramethylene orthosilicate (TMOS) a sol-gel process is induced inside the micelle core, which leads to the precipitation of amorphous SiO₂ [67, 68]. The block lengths of the polymers used in this publication are $N_{\text{PS}} = 750$ for polystyrene (PS) and $N_{\text{P2VP}} = 1,950$ for poly-2-vinylpyridine (P2VP). The maximum radius of the SiO₂ cores in toluene is 19.2 nm which was determined by small-angle X-ray scattering (SAXS) [68].

16.3.2 *Monomicellar Layers on Substrates*

Monomicellar layers on substrates like Si are fabricated by dip coating [68]. This method allows the coverage of large substrate areas in the range of square centimeters. Figure 16.3 shows an example of a monomicellar layer on silicon taken by atomic force microscopy (AFM). The micrograph reveals that the micelles form a local hexagonal close-packed structure by self-assembly [68, 69]. The same formation is obtained on other substrates, like, for example, Co/Pt multilayers.

Fig. 16.3 AFM micrograph of one layer of diblock copolymer micelles on silicon



The monomicellar layer is characterized by a mean nearest neighbor distance of $d_{\text{NN}} = 82(\pm 2)$ nm, a peak-to-peak (ptp) height modulation of $8.4(\pm 0.5)$ nm and a full width at half maximum (FWHM) of the micelles of $43(\pm 2)$ nm.

The height modulation is considerably smaller than the mean distance, which indicates that the micelles cannot be regarded as hard spheres [70]. The micelle spheres become prolate when deposited on the surface which is attributed to a flexible polymer shell and a rigid core [68]. The SiO_2 -filled cores are stable and keep their spherical shape [28]. As a result, they form humps, which are barely covered by polymer material, while in the region between the cores a continuous organic coating is created as the polymer shells bent sidewise and interpenetrate each other. This self-organized monomicellar layer represents the starting point for the fabrication of antidot and dot arrays.

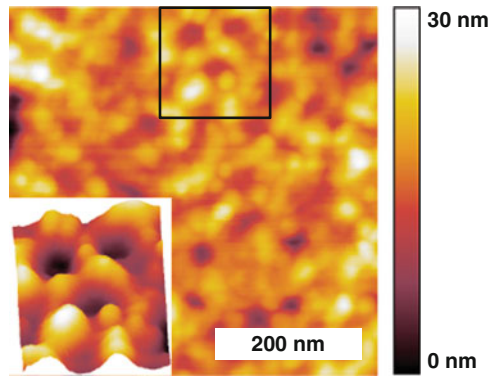
16.3.3 Fabrication of Antidot Arrays Utilizing Monomicellar Layers

The nanostructure arrays are obtained by transferring the morphology of the monomicellar layer into the magnetic film via Ar^+ ion milling at normal incidence. As the layer consists of two spatially separated species, the different sputter yield generates a copy of the morphology. Hence, depending on the sputter yields of the organic matrix and the core material, a negative or positive reproduction will result [71]. As the sputter yield of the polymer matrix, mainly carbon, is lower than that of SiO_2 , an antidot array will be created.¹

We have applied that procedure to create an antidot array in an ECR fabricated $[2.5 \text{ nm Pt}/0.5 \text{ nm Co}]_8$ film on a 4.1 nm Pt seed layer [70]. The arrangement

¹ For 2 keV Ar^+ ion bombardment at normal incidence, the sputter yield of C is 0.8 [72] while that of SiO_2 is 1.7 as determined by the simulation software “Stopping and Range of Ions in Matter” [73].

Fig. 16.4 AFM micrograph of a monomicellar layer on a Co/Pt multilayer after sputtering at normal incidence with $1.6 \times 10^{17} \text{Ar}^+/\text{cm}^2$. The *black square* gives the position of the three-dimensional detail (*inset*). The lateral size of the cutout is $150 \text{ nm} \times 150 \text{ nm}$ and the maximum height amounts to 18 nm



of the micelles was found to be similar to the arrangement shown in Fig. 16.3. After ion milling with 2 keV Ar^+ ions at normal incidence and applying a dose of $1.6 \times 10^{17} \text{Ar}^+/\text{cm}^2$, we obtain the morphology shown in Fig. 16.4. The ptp height modulation is $8.2(\pm 0.7) \text{ nm}$ with a diameter of the dips (FWHM) of about $40(\pm 7) \text{ nm}$. Although the value of the height modulation is not changed significantly, the topography is drastically altered on sputtering. The former humps have been transformed into dips (inset in Fig. 16.4) [70]. The dips are separated by height modulated ridges with highest points at the former three-fold coordinated hole sites. This formation can be appointed to the higher polymer (carbon) concentration at the former three-fold coordinated hole sites than between neighboring micelles. In summary, the dot array of the micellar layer is transformed into an antidot array by ion milling.

An antidot morphology does not necessarily mean that a magnetic antidot is created. For checking the magnetic behavior, we have performed magneto-optical Kerr effect studies. In a first step, we have studied the influence of ion bombardment on the magnetic properties of a bare multilayer. It turned out that the magnetization switches from vertical to in-plane on applying very small doses ($< 10^{15} \text{Ar}^+/\text{cm}^2$). Such a change of the behavior is known for high-energy ion bombardment and was attributed to ion-beam induced atomic disordering of Co and Pt [75, 76]. We assume that the same happens here at even lower energy (2 keV). On increasing the dose, the ferromagnetism was very quickly destroyed. The dose where the Kerr signal was no longer detectable was determined as $0.75 \times 10^{17} \text{Ar}^+/\text{cm}^2$. For doses in that range, a considerable amount of the material is removed while the remains are strongly intermixed, which eliminates any ferromagnetic long-range order. The influence of ion bombardment is different when the multilayer is covered with a monomicellar layer. Figure 16.5 gives the MOKE loops obtained for magnetic fields perpendicular (polar) and parallel (longitudinal) to the film plane. For the sake of direct comparison, hysteresis curves for the nonsputtered micelle covered multilayer, with composition $[\text{2.5 nm Pt}/\text{0.5 nm Co}]_8$, have been taken prior to milling.

As mentioned before, for the nonsputtered sample, a canted magnetization is found (see below) (Fig. 16.5). The hysteresis loop in perpendicular field exhibits zero remanence due to a multidomain state while the longitudinal hysteresis shows

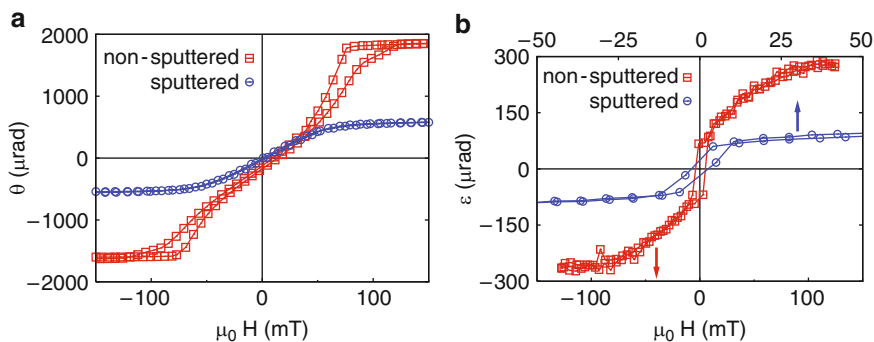


Fig. 16.5 MOKE hysteresis loops of $[2.5 \text{ nm Pt}/0.5 \text{ nm Co}]_8$ multilayers decorated with a monomicellar layer before and after sputtering with $1.6 \times 10^{17} \text{ Ar}^+/\text{cm}^2$ at normal incidence. In (a) the polar Kerr rotation and in (b) the longitudinal Kerr ellipticity are given, showing the magnetization behavior in perpendicular/in-plane fields, respectively. Some asymmetry of the Kerr loops in (a) with respect to the origin are caused by nonlinear effects [74]

a hard axis behavior [60]. After the exposure to 2 keV Ar^+ ions with an ion dose of $1.6 \times 10^{17} \text{ Ar}^+/\text{cm}^2$, which is more than twice the value that destroys ferromagnetism in the noncovered film, the film is still ferromagnetic with an easy axis in the film plane. A remanence is found for in-plane fields while in perpendicular fields the system shows a hard axis response. Obviously, the micellar layer acts as a protection layer for the Co/Pt multilayer.

A detailed analysis of the magnetic signals obtained in saturation and the area filling factor of the holes/dips obtained from scanning electron microscopy (SEM) micrographs reveal unambiguously that a magnetic antidot array has been generated by ion milling. It turns out that the holes are no longer magnetic while the film below the ridges is still ferromagnetic. We have estimated that at least two Co layers contribute to the signal in the hysteresis loops [70].

16.3.4 Fabrication of Dot Arrays Utilizing Monomicellar Layers

The same kind of SiO_2 -filled micelles was also successfully utilized for the fabrication of ferromagnetic dot arrays [28]. In order to overcome the problem that is caused by the low sputter yield of carbon, the polymer shell of the micelles was removed in an oxygen plasma before ion milling. The remaining SiO_2 cores were then used as masks for ion milling.

Figure 16.6 shows an SEM micrograph of the SiO_2 cores on a Co/Pt multilayer after the treatment in an oxygen plasma. A quantitative analysis yields an average diameter of 19.2 nm with a standard deviation of 5.5 nm , which perfectly agrees with the radius determined in toluene by SAXS [68]. The SiO_2 particles cover 16.1% of the surface. From an AFM investigation, the average dot height has been determined as 21.9 nm with a standard deviation of 5.7 nm . These values let

Fig. 16.6 SEM image of SiO_2 particles on a Co/Pt multilayer. Adopted from [28]

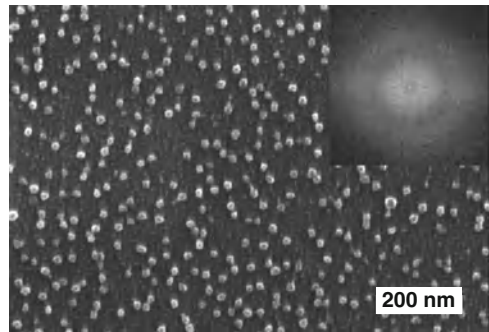
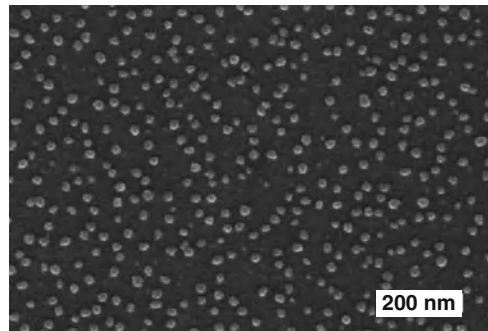


Fig. 16.7 SEM image of Co/Pt dots after ion milling, dose $5.3 \times 10^{16} \text{ Ar}^+/\text{cm}^2$. Adopted from [28]



us conclude that the SiO_2 particles are spheres [28]. The arrangement of the SiO_2 particles (Fig. 16.6) shows quite uniform nearest neighbor distances but does not show a high degree of hexagonal order, similar to the arrangement of the micelles (Fig. 16.3). The fast Fourier transform (FFT) reveals a broad ring-shaped distribution (see inset Fig. 16.6). The ring diameter represents the average distance of the particles while the broadening indicates the variation of distances around the mean separation. The broadening is mainly due to the imperfection of the self-organized ordering.

The multilayer/core system was sputtered by 500 eV Ar^+ ions at normal incidence. The multilayer composition was 3 nm Pt/0.7 nm Co/2 nm Pt/0.77 nm Co grown on a 4.1 nm platinum seed layer. An SEM image of the dot array, which was obtained by applying an ion dose of $5.3 \times 10^{16} \text{ Ar}^+/\text{cm}^2$, is shown in Fig. 16.7. Obviously, the structure of the SiO_2 particles (see Fig. 16.6) is reproduced in the multilayer film [28]. The analysis yields an average diameter of 18.3 nm with standard deviation of 5.3 nm. With respect to the SiO_2 particles, the average dot diameter is reduced by 0.9 nm while the standard deviation of the dot diameter is slightly increased. The area covered by the dots is slightly reduced from 16.1% to 14.5% of the total area. The reduction of the dot diameter is responsible for the changes of the area coverage. This indicates that the number of particles has been maintained and the transformation works as a one by one copy. From an AFM investigation, the average dot height has been determined to be 17.2 nm with standard deviation of 3.2 nm.

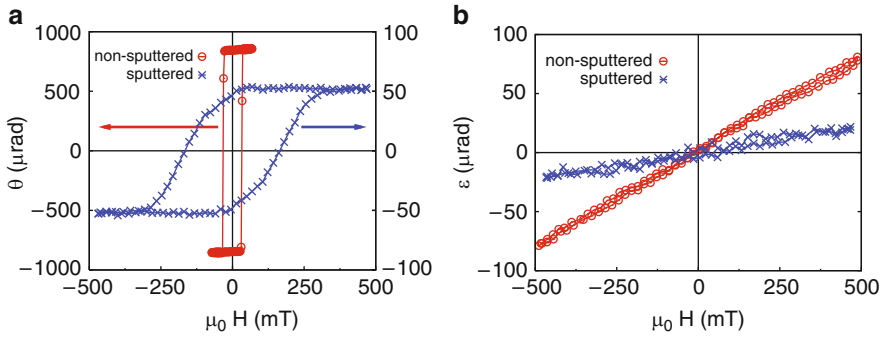


Fig. 16.8 MOKE hysteresis loops of multilayers decorated with SiO_2 particles. The multilayer composition was [3 nm Pt/0.7 nm Co/2 nm Pt/0.7 nm Co] (magnetron) on a 4.1 nm Pt buffer layer (ECR). The curves show the magnetization behavior before and after ion milling with $5.1 \times 10^{16} \text{ Ar}^+/\text{cm}^2$ at normal incidence. The polar Kerr rotation θ /longitudinal Kerr ellipticity ε was taken as a function (a) of the perpendicular field and (b) of the in-plane field, respectively

MOKE hysteresis loops obtained before and after sputtering are plotted in Fig. 16.8. In a perpendicular magnetic field, almost square hysteresis is found with hard axis loops for fields applied in-plane. This indicates that the dot array is ferromagnetic and the perpendicular easy axis of magnetization is conserved. In milling experiments on bare multilayers, the magnetism was already destroyed when applying far lower doses. Similarly to the fabrication of antidot arrays, here, the SiO_2 cores protect the multilayer from being affected by ions whereas the magnetic material between the cores is removed or at least the ferromagnetism is erased.

On sputtering, the polar signal decreases by more than a factor of 10 in the polar hysteresis (Fig. 16.8). Comparing this value with the area filling factor derived from the SEM analysis (14.5%), it is obvious that the Kerr signal is smaller than the signal that one would expect from the area filling factor. Therefore, we must conclude that the magnetic active volume is smaller than the dot volume observed with AFM and SEM. From the numbers, we can calculate the magnetic active volume of the dots [28]. Vice versa it is a strong hint that the magnetic response stems solely from the dots. In Sect. 16.4.2, we will present another proof for the magnetism being a property of the dots only. On sputtering, the coercive field increases, which is due to a change of the reversal mechanisms. In films, the magnetization is generally reversed via domain nucleation and domain wall propagation, which can circumvent high anisotropy determined barriers. In the small structures, we obtain coherent rotation of all moments of the whole particle, which is directly depending on the anisotropy and in general the coercivities are larger. The coercivity increase is by no means due to the change of magnetic anisotropy. Assuming the same signal reduction for the longitudinal signal as for the polar signal, we can estimate from the in-plane magnetization curve that the anisotropy in the dot system is four times smaller than in the multilayer system before sputtering [28].

To conclude, a magnetic dot array with perpendicular easy axis has been created successfully.

16.4 Magnetic Behavior of Multilayers and Nanostructures

16.4.1 Multilayers

As already explained in the foregoing sections, a perpendicular orientation of magnetization becomes possible when the surface or interface anisotropy overcomes the shape anisotropy in the ultrathin layer limit. On thickness increase, the magnetization will flip into the film plane. Within the spin reorientation transition (SRT) in a very small thickness range, the two competing contributions can cancel and the effective first-order anisotropy constant becomes very small and eventually will be zero. This situation has caused some speculation about the behavior of the system at that peculiar point in the 1990s [77, 78]. The zero anisotropy apparently indicates that there is no longer an aligning force for the magnetization, which means that the magnetization orientation is unstable against smallest perturbations. The latter means that no net magnetization can be observed, as even thermal excitation causes a fluctuation of the magnetization orientation in time. This peculiar situation is immediately lifted when a more accurate description of the angle-dependent energy is used. From general symmetry considerations, the angle-dependent energy can be given as a series expansion of the orientation of magnetization with respect to particular directions in space, that is, in case of uniaxial behavior, with respect to the easy axis. In general, the lowest order is dominant and higher contributions are neglected. The representation given in (16.1) is such an expansion in first-order approximation. When certain effects lead to a decrease of the first-order expansion coefficient, the description has to be expanded to next higher order. The higher order contribution will determine the behavior of the magnetic system. Hence, (16.1) has to be expanded to the second-order approximation

$$f_{\text{film}} = K_{\text{1eff}} \sin^2 \theta + K_2 \sin^4 \theta. \quad (16.2)$$

The second anisotropy constant K_2 will dominate if the effective first-order contribution becomes vanishing small. The stability analysis of the above formula leads to a cubic expression. The resulting phase diagram is published in [79, 80]. The peculiarity is that the transition from vertical to in-plane orientation of magnetization can proceed via two different states that are connected to certain alignment of the magnetization. In one case ($K_2 > 0$) the magnetization starts to tilt and the magnetization has a fixed angle to the surface normal that is determined by the canting angle

$$\sin \theta = \sqrt{-\frac{K_{\text{1eff}}}{2K_2}}. \quad (16.3)$$

In the second situation ($K_2 < 0$), the SRT proceeds via a state of coexisting phases. The phases involved are those with vertical and in-plane orientation of magnetization. The depth of the local minima of the free energy changes when crossing that range in the anisotropy space. The population of the individual phases

is determined by statistics [81] although the magnetic microstructure will strongly influence the occupation strength. A spin reorientation via the state of coexisting phases has been identified in case of ultrathin Co films on Au(111) [82–84]. In case of Co/Pt multilayers, we were able to verify that a magnetization canting appears in the transition from vertical to in-plane orientation of magnetization [61, 85]. On increase of the Co thickness, the shape of the hysteresis changes from rectangular with full remanence to a shape with small or even no remanence (Fig. 16.9). This change of the magnetization curve is due to the decomposition of the single-domain state that is found at higher fields into a multidomain state. The latter is stable in vanishing fields. The multidomain state is created as it allows to reduce the magnetostatic energy. The reduction is on expense of domain wall energy as the number and length of domain walls increase with number of domains. The fact that the system can insert domain walls indicates that the anisotropy constant has been lowered as the domain wall energy scales with $\sqrt{K}$. As long as K is large, it costs too much energy to incorporate enough domain walls to obtain a reasonable reduction of the magneto-static self-energy. The magnetization switches between single-domain states on field sweep, which gives a rectangular hysteresis loop. When anisotropy becomes smaller, the domain wall energy drops and total energy can be gained by creating a multidomain state. Figure 16.9b displays the hysteresis curves obtained in in-plane fields. While a curved reversible behavior is found in case of higher anisotropy, a stronger response (smaller saturation fields) is found in the second case. This behavior is another indication of a reduced magnetic anisotropy. Most surprising, however, is the hysteresis found in small fields. A continuous magnetization change is found for vertical fields in this field range that indicates some domain wall displacements. In-plane irreversible changes are identified, which indicate some in-plane components of magnetization to exist and switch. Actually, this peculiar form of hysteresis is identified as a fingerprint for the canting of magnetization. The small hysteresis loop can be used to determine the canting angle and/or

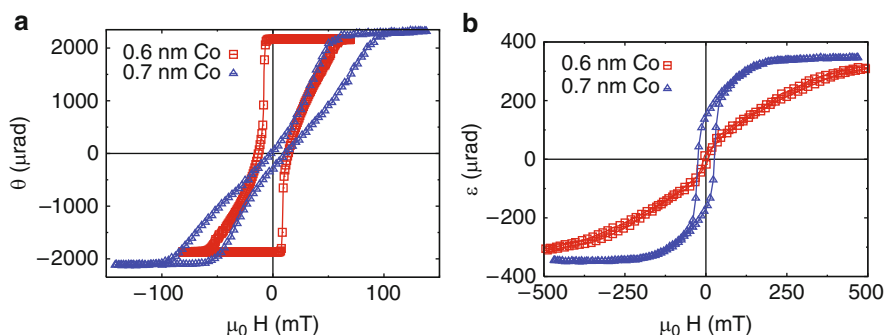


Fig. 16.9 MOKE hysteresis loops for 0.6/0.7 nm Co layers in $[\text{Co}/1.0 \text{ nm Pt}]_8$ multilayers (magnetron) on a 4-nm Pt buffer layer (ECR). The polar Kerr rotation θ /longitudinal Kerr ellipticity ε was taken as a function (a) of the perpendicular field and (b) of the in-plane field, respectively. Some asymmetry of the Kerr loops in (a) with respect to the origin are caused by nonlinear effects [74]

K_2 [85, 86]. In literature, numerous examples for the same pair of hysteresis loops can be found, the meaning of the in-plane hysteresis, however, has never been commented [37, 39, 43]. The unambiguous proof for canting, however, was gained by imaging the magnetic microstructure in the Scanning Electron Microscope with Polarization Analysis (SEMPA or spin-SEM) [61].

For the phase of magnetization canting, it is important whether there is any anisotropy existing in the plane of the film or not. If anisotropy is effective, the in-plane direction with lowest energy will determine the plane in which the magnetization cants. If the in-plane magnetic property is isotropic, the magnetization is free to take any direction of the in-plane component. The angle with respect to the normal, however, is fixed, determined by the first- and second-order anisotropy constants (16.3), and the magnetization is apparently free to rotate on a cone. This is the reason why that state is often called cone state. Commonly, it has been assumed that the magnetization is unstable against small perturbations that let the magnetization rotate about the surface normal in time. The consequence is that no remanent in-plane magnetization can be expected. Surprisingly, the magnetic microstructure analysis revealed a wavy domain pattern in the in-plane magnetization component that is stable in time. A detailed analysis has demonstrated that all in-plane components of magnetization are populated with the same probability, while the major part of the magnetization is oriented perpendicular to the surface [61]. This means that the magnetization orientation of the total domain pattern is characterized by magnetization distribution that is actually lying on a cone. The main and important conclusion is that the system realizes the often proposed cone state in the space domain. This alternative realization of the cone state was not considered before our magnetic microstructure investigation.

In the foregoing discussions, the change of magnetization orientation was driven by the Co film thickness. In Fig. 16.10, the dependence of the first- and second-order anisotropy on Pt interlayer thickness is shown. It is amazing that the anisotropies change at all. The first-order anisotropy constant increases continuously with Pt thickness. As the total change is larger than the volume anisotropy of Co and as Pt does not contribute considerably to the magnetic moments, it is evident that the finding hints to the interface anisotropy. Obviously, the interface quality of the second and higher Co layers in the multilayer stack is worse than that of the first (or a single) layer when the Pt interlayer thickness is too small. The roughness adds up so that the interface anisotropy of the higher Co layers does not give a sufficiently high interface anisotropy. So the total amount of ferromagnetic material causes the in-plane orientation of magnetization via the dominance of shape anisotropy. With interlayer thicknesses above 1.5 nm, the total interface anisotropy can balance the shape anisotropy. With Pt thicknesses beyond 3 nm, the interface quality is almost reestablished and the anisotropy of the total stack becomes as large as that of the single layer on the almost ideal buffer layer. In contrast to the effective first-order anisotropy, the second-order anisotropy is vanishingly small below 1 nm Pt interlayer thickness and reveals a step like increase to the value found for the single layer, which is almost the Co bulk value of $1.25(\pm 0.25) \times 10^5 \text{ J m}^{-3}$ [46]. This result indicates that an interlayer thickness of $>1 \text{ nm}$ is of sufficient quality as template to initiate a predominant

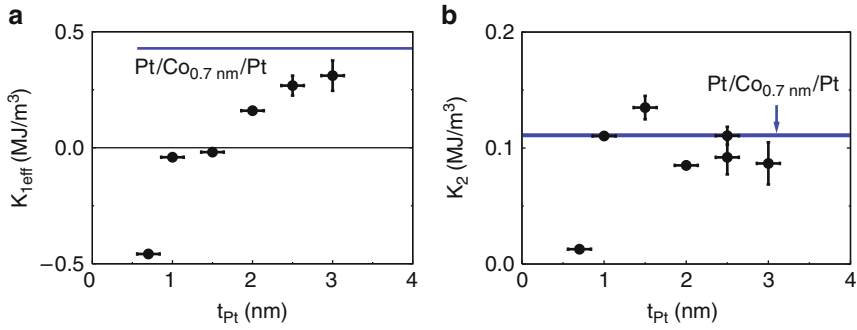


Fig. 16.10 Evolution of the anisotropy constants as a function of the Pt interlayer thickness. (a) First order effective anisotropy $K_{1\text{eff}}$ and (b) second-order anisotropy K_2 . The multilayer consists of magnetron sputtered $[0.7 \text{ nm Co}/x \text{ nm Pt}]_8$ on a 4 nm Pt seed layer (ECR). For comparison the anisotropies of a single Pt/0.7 nm Co/Pt layer are given

hcp growth of Co. For smaller thicknesses, the Co either is disordered or grows as a mixture of hcp and fcc structure (K_2 is negative for fcc) [28].

In conclusion, we may say that the very accurate investigation of the magnetic properties of the multilayers allows to extract information about the interface and film structure, which is very sensitively fixed by the Pt interlayer.

16.4.2 Dots

In small ferromagnets, multidomain states can be prevented when the size of the ferromagnet is below some characteristic length, which roughly scales with the domain wall width of the system (see above). In this case, single-domain particles are obtained. For our multilayers, we estimate the critical length to be ≥ 20 nm. Hence the ferromagnetic dots discussed in Sect. 16.3.4 will be single-domain particles. Single-domain particles are commonly believed to reverse via coherent rotation of all magnetic moments in the particle. To reverse the magnetization, the Zeeman energy has to overcome the total anisotropy energy of the particle, which is the anisotropy energy times the particle volume

$$E_{\text{tot}} = K_{1\text{eff}} V_{\text{Dot}}. \quad (16.4)$$

On size reduction of the particle, the total anisotropy energy decreases. At very small sizes, this energy gets comparable to the thermal energy. In this case, the thermal energy will cause a random switching of the magnetization between the two directions along the easy axis in time. The particle behaves like a paramagnet. This size-dependent behavior is called superparamagnetism as all of the exchange coupled moments of the dot switch simultaneously [87–89]. Such systems reveal a distinct temperature behavior. On cooling the particles can be forced again in a

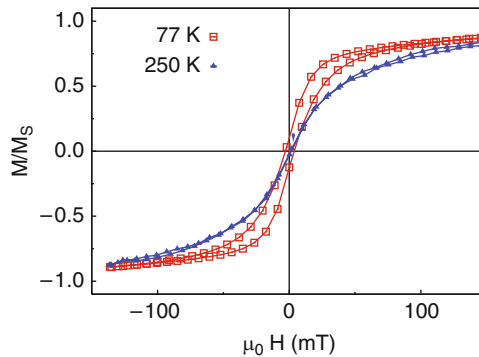


Fig. 16.11 Magnetization of an array of magnetic dots of diameter <12 nm as a function of the magnetic field for different temperatures. The dots have been prepared by ion bombardment of SiO_2 particles on $[3 \text{ nm Pt}/0.7 \text{ nm Co}/1 \text{ nm Pt}]$ with a 4 nm Pt buffer layer (ECR sputtering). The magnetization was normalized to the value obtained at 425 mT

bi-stable state that is similar to a ferromagnetic behavior. We have fabricated a dot array with particle diameters <12 nm that exhibits superparamagnetic behavior. The hysteresis curves for different temperatures are shown in Fig. 16.11. At room temperature, the magnetization behavior is “S” shaped as expected for a paramagnet. When cooling the magnetization curve starts to show hysteresis and remanence is found. Due to the fact that the dots have a certain size distribution, the transition to a ferromagnetic state is smeared out over a large temperature interval. The appearance of superparamagnetism on decrease of the dot size demonstrates that the ferromagnetism discussed above for the ≈ 19 nm particles is really a property of the dots. In conclusion, the appearance of a superparamagnetic behavior proves that an ensemble of ferromagnetic nanodots has been generated by the technique introduced in this review.

16.5 Summary

In this review, we have presented a very convenient method to fabricate arrays of magnetic nanostructures with particle sizes below 20 nm. For structuring, we have used self-assembly of single layers of diblock copolymer micelles containing an amorphous SiO_2 core that are deposited on ferromagnetic multilayers. The morphology of the micellar layer is transferred into the multilayer via ion milling. It has been shown that ferromagnetic antidot and nanostructure arrays could be successfully generated.

Simultaneously, the multilayer properties were optimized to achieve an easy axis of magnetization perpendicular to the supporting plane. We have shown that multilayer properties can be improved by combining two different deposition techniques, that is, ion beam (ECR) and magnetron sputtering. The optimization of the

multilayers came along with very interesting new findings about the properties of Co/Pt multilayers. We could demonstrate that the spin reorientation proceeds via the phase of canted magnetization. A new realization of the so-called cone state, not considered before, was found when the magnetic domain pattern was imaged with high resolution. A realization in the spatial domain was identified.

A great advantage of our method is that the tuning of the magnetic properties is separated from the nanostructuring. This offers a high flexibility compared to other techniques as it opens the way to create nanodots of given size but with varying magnetic properties. Here, we have shown how arrays of ferromagnetic and superparamagnetic particles can be produced by changing the lateral size of the SiO₂ cores acting as masks.

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Chapter 17

How X-Ray Methods Probe Chemically Prepared Nanoparticles from the Atomic- to the Nano-Scale

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Abstract Chemically prepared nanoparticles are an exceptional class of materials that owe their properties to both the three-dimensional confinement and their large surface area. To characterize their electronic and geometric structural properties, X-ray methods offer unique capabilities. In this chapter, we demonstrate how a combination of X-ray spectroscopy and X-ray diffraction techniques allows one to characterize these nanoparticles with respect to their local atomic properties, their long-range crystalline order and their nanoscale core–shell structure.

17.1 Local Atomic Structure: Chemical State and Coordination

The influence of nanometer length scales in matter is reflected by the widening of the electronic band gap due to quantum confinement, by the modification of the chemical state of the atomic constituents and by changes of chemical bonding and local atomic coordination within the cluster. We thus want to determine the local coordination and chemical bonding arrangement of the atomic constituents in a nanoparticle and determine possible deviations from the bulk material. With near-edge X-ray absorption fine structure (NEXAFS), we can determine the local electronic structure and the oxidation state of selected atomic centres within the cluster [1]. Using highly monodisperse lanthanide phosphates nanoparticles, synthesized by the method of Lehmann et al. [2], we demonstrate how NEXAFS determines the chemical state of the rare earth element within these clusters.

In Fig. 17.1, the schematic overview of a typical X-ray spectroscopy system with synchrotron radiation is given. In particular, the combination of NEXAFS with photoelectron spectroscopy and resonant inelastic X-ray scattering gives deep insight into the electronic structure properties of matter. In Fig. 17.2, we illustrate how NEXAFS a powerful method is for studying the 4f electron occupancy and hybridization state of a rare earth ion in a chemically prepared nanoparticle. In X-ray absorption, we monitor the $3d^{10}4f^n \rightarrow 3d^94f^{n+1}$ lanthanide transitions. Due to

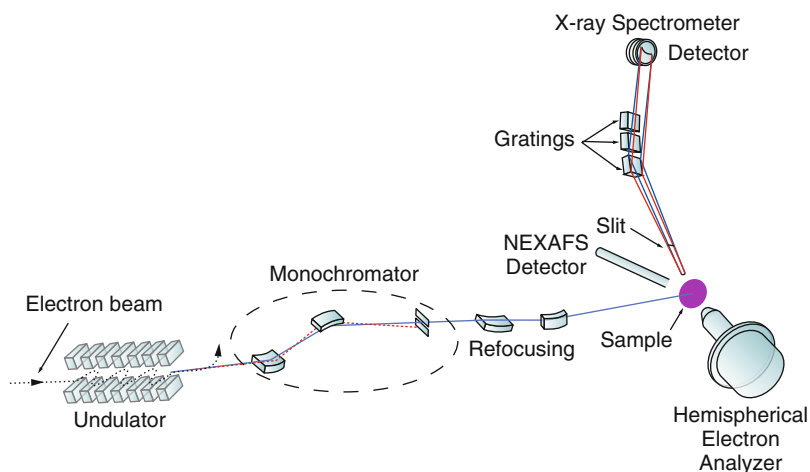


Fig. 17.1 Schematic representation of an X-ray spectroscopy experiment at a synchrotron radiation source. Synchrotron radiation is generated in an undulator, monochromatized and focused onto the sample. Using electron spectrometers, X-ray spectrometers and partial yield detectors for near-edge X-ray absorption spectroscopy the electronic structure of the sample is determined

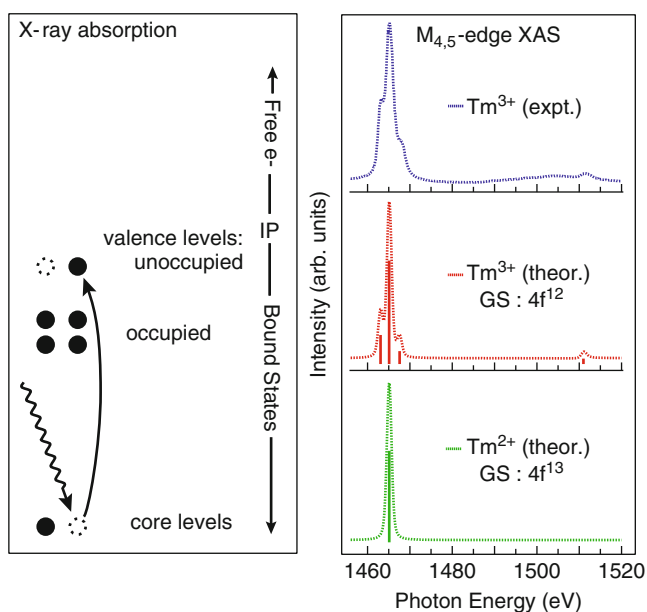


Fig. 17.2 *Left panel:* Cartoon of X-ray absorption spectroscopy or near-edge X-ray absorption fine structure (NEXAFS). *Right panel:* Determination of the Tm chemical state comparing experimental and atomic Hartree-Fock calculated spectra for ground state configuration 4f¹² and 4f¹³, respectively. (From [1])

sufficiently deep 3d shell, the spin–orbit interaction of the $3d^9$ hole in the final state is much larger than the $3d^9 4f^{n+1}$ exchange interaction. Thus, the final states are split into two group of lines: $3d_{3/2}(M_4)$ and $3d_{5/2}(M_5)$. Each individual M_4 and M_5 absorption line shows additional multiplet structure, which is governed by the direct- and exchange-Coulomb interaction between the $3d^9$ core hole and the 4f electrons (which is strong due to the strong 4f shell localization), the direct Coulomb interaction between 4f electrons, and the small spin-orbit interaction of the 4f electrons, that is, the fine structure is due to the intermediate coupling of the $3d^9$ hole and 4f electrons in the lanthanide ions. Since the exchange interaction between the 3d hole and 4f electrons will vary as a function of the number of electrons in the 4f shell, we can use this fine structure as a very precise measure of the valence electronic structure of the rare earth ions.

This is shown for the Tm-orthophosphate nanoparticles, where we compared the fine structure of our lanthanide X-ray absorption spectra with Hartree–Fock calculations using Cowan’s atomic multiplet approach with relativistic corrections [3], in the intermediate coupling, that is, including all the states of the $4f^n$ ground state configuration and $3d^9 4f^{n+1}$ final state configuration, for all the lanthanide ions and for all ionization states known in the solid state. The calculated spectra are compared with our measured NEXAFS curves. In the case of thulium ions, an electronic ground state configuration of $4f^{12}$ is revealed, corresponding to a valency of III of the thulium ions in the orthophosphate nanoparticles.

In Fig. 17.3, we can now determine the chemical state of the rare earth ions for the whole series of rare earth orthophosphate nanoparticles. In comparison to computation, we find that all lanthanide ions in the orthophosphate nanoparticles are in the triply ionized ground state, establishing a 4f occupancy in the ground state of $4f^0$ for La up to $4f^{13}$ for Yb. In analyzing the rare earth orthophosphate nanoparticle series, we observe how the total integrated intensity of the M_4 and M_5 absorption lines decreases going from light lanthanides towards the heavier ones. This is due to the fact that the summed cross section of the two spin–orbit absorption bands is proportional to the number of valence holes [5] and this number reduces going from lighter to the heavier lanthanides. However, the branching ratio of the two spin–orbit split lines $I(M_5)/I(M_4) + I(M_5)$ is not given by its statistical value $6/4 + 6 = 0.4$.

There are two effects that cause the change in the branching ratio [6]. The first effect is the presence of spin–orbit coupling in the initial state (due to the strong 4f localization) that splits the 4f valence electrons into bands of different values of the total atomic angular momentum quantum number J ($j-j$ coupling limit), and further the J selection rules set preferences on the transitions to the two different manifolds. For this reason, the Yb^{3+} absorption spectrum shows only one line, which is due to the fact that the only empty hole in the ground state is in the $^2F_{7/2}$ state so that only the $3d_{5/2} \rightarrow 4f_{7/2}$ transition is allowed. The second effect is the presence of the 3d hole–4f electrons electrostatic interactions ($L-S$ coupling) in the final state that couple the core hole to the valence holes differently for the two manifolds [6]. (In the $L-S$ coupling limit, the spin selection rule will play a role in the transitions to the two different manifolds.)

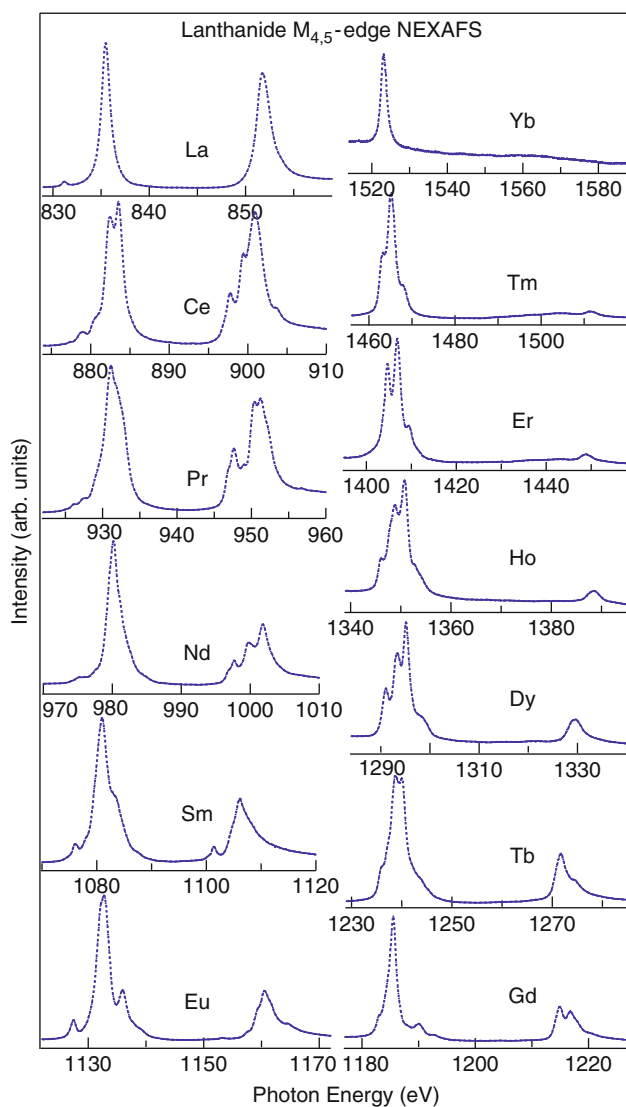


Fig. 17.3 $M_{4,5}$ -edge NEXAFS spectra of lanthanide ions measured by monitoring MNN Auger lines. Comparison of the characteristic multiplet structure of the spectra to the calculated one by Thole [4] reveals that all rare earth ions are in a triply ionized ground state. (From [1])

In conclusion, NEXAFS allows us to determine the local coordination of specific atoms in the nanoparticles. For lanthanide nanoparticles, we find that the local coordination of the rare earth ions is the same throughout the particle. In particular, the valency of the rare earth ions does not change at the surface of the nanoparticles.

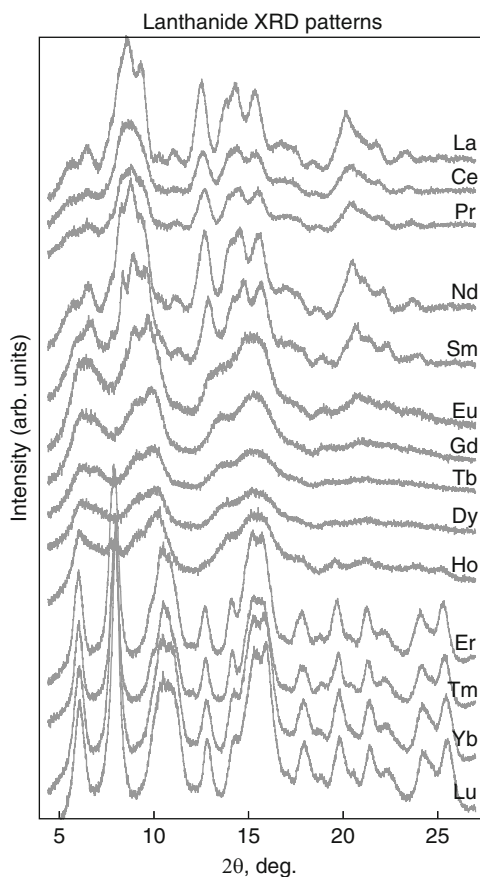


Fig. 17.4 X-ray powder diffraction patterns of lanthanide phosphate nanoparticles. Broad diffraction peaks indicate the small crystal size of 2–5 nm. La–Sm crystals are in monoclinic phase, Er–Lu crystals are in tetragonal phase, and Eu–Ho crystals are in “mixed phase” between monoclinic and tetragonal phase and are even smaller in size as seen from the peak width. (From [1])

17.2 Crystallinity and Cluster Structure

An important parameter of nanoparticles is their crystal structure, which we can determine with X-ray diffraction. In Fig. 17.4, we show X-ray powder diffraction for the rare earth orthophosphate nanoparticles. All diffraction patterns are characterized by considerably broad reflection peaks owing to the small crystallite size of the nanoparticles. In the case of intermediate lanthanide ions (Eu, Gd, Tb, Dy, Ho), the diffraction patterns are extremely broad, indicating a smaller crystallite size.

In general, the observed trend of the crystal-phase and the crystal-size evolution through the lanthanide series is closely related to the decreasing effective cation radii with increasing atomic number, called “lanthanide contraction” also known

from the lanthanide orthophosphate bulk crystals [7]. Ni et al. have shown that distinct similarities exist between the monoclinic and tetragonal structures [7]. Both atomic arrangements are based on [001] chains of intersecting phosphate tetrahedra and lanthanide (Ln) polyhedra. As the structure “transforms” from monoclinic to tetragonal, the lanthanide polyhedron transforms from LnO_9 to LnO_8 . Projected along the [001] direction, the tetrahedra–polyhedra chains exist in the (100) planes, with two planes per unit cell in both structures. In the monoclinic phase, the planes are offset by 2.2 Å along [010] relative to those in the tetragonal phase, in order to accommodate the larger lanthanide atoms. The shift of the planes in the monoclinic phase allows the Ln atom to bond to an additional oxygen atom to complete the LnPO_9 polyhedron. Additionally, Ni et al. showed that the unit cell of TbPO_4 tetragonal is larger than that of GdPO_4 monoclinic, despite the fact that Tb is smaller than Gd [7]. This fact demonstrates that the void space in the tetragonal phase is larger compared to that in the monoclinic phase, thus, creating an energetically unfavorable situation. As the intermediate lanthanide particles have a very small size of ≤ 3 nm, which corresponds to a surface to volume contribution of $\sim 70\%$, a rearrangement of the atomic positions occurs so that the total energy of the system is minimized. Furthermore, because the lattice energies of the monoclinic and tetragonal phases close to the phase transition point are very similar, the atoms occupy surface sites that are in between the monoclinic and tetragonal phases. This leads to a missing short-range order of the atomic arrangements that is fingerprinted in the very broad diffraction peaks.

A detailed analysis of the diffraction patterns can be done by full-profile Rietveld refinements [8] that revealed in the present case that the structures are isotropic with no preferred direction. The mean particle size was roughly determined from the broadening of the reflection peaks using the Scherrer formula [9, 10]. The calculated diffraction patterns of the nanoparticles using the Rietveld structural refinement showed that the larger lanthanide ions (La, Ce, Pr, Nd, Sm) adopted the same monoclinic phase of monazite as their respective bulk crystals and the smaller ones (Er, Tm, Yb) adopted the same tetragonal structure of xenotime as their respective bulk crystals. In Fig. 17.5, we have shown the crystallographic calculations compared with the measured diffraction patterns for LaPO_4 and LuPO_4 nanoparticles.

In contrast, the lanthanide nanoparticles from the middle of the series (Eu, Gd, Tb, Dy, Ho) deviate from the crystal phase of the respective bulks. Their diffractograms exhibit a crystal phase that is neither monoclinic nor tetragonal, but a “mixed phase” of monoclinic and tetragonal. In addition, our measurements revealed a slight phase transition from monoclinic to tetragonal, where Eu showed a crystal phase more similar to monoclinic and Ho showed a crystal phase more similar to tetragonal. In particular, the diffraction patterns of the intermediate lanthanides are characterized by very broad peaks at large scattering angles (above 20°), corresponding to diffraction from atomic planes at small interatomic distances of 1.5–2 Å. These blurred diffraction patterns are a fingerprint for a missing short-range order of the atomic arrangements in these nanoparticles. So the lanthanide nanoparticles from the middle of the series (Eu, Gd, Tb, Dy, Ho) are much smaller in size than the nanoparticles containing either larger lanthanide ions (La, Ce, Pr,

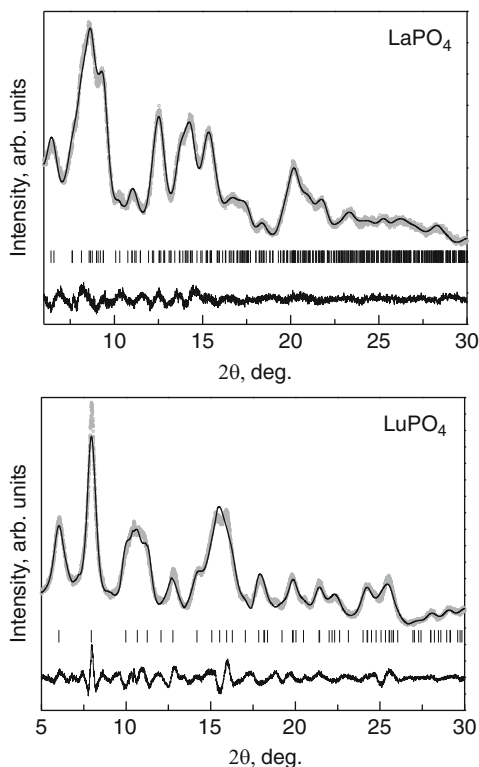


Fig. 17.5 Measured and calculated powder diffraction patterns for LaPO_4 and LuPO_4 nanoparticles. Crystallographic calculations were based on the monazite and xenotime structures [7] for LaPO_4 and LuPO_4 , respectively. Experimental data (circles), calculated profiles (solid line through the circles) are presented together with the calculated Bragg positions (vertical ticks). The difference curve between the measured and calculated profiles is presented as a solid line below. (From [1])

Nd, Sm) or smaller ones (Er, Tm, Yb, Lu), which have particle sizes of $\sim 3\text{--}5$ nm diameter according to the X-ray diffraction peak shapes.

17.3 Core–Shell Structures on the Nanoscale

To stabilize nanoparticles, a commonly used procedure is to grow protective shell materials covering the inner particle core. A central aspect is often to prevent oxidation of the nanoparticle that influences the luminescence properties. Since semiconductor nanoparticles allow to tailor the band gap and luminescence properties through cluster size, these materials are very promising for application. Unfortunately, these systems are very sensitive to surface oxidation, reducing the luminescence quantum yield in semiconductor nanoparticles. Furthermore, growing

a thin shell of a wide band gap semiconductor on the outside of the emitting particle allows substantial improvement of their quantum efficiency as the relevant wave functions are confined to the interior of the particle [11]. Especially colloidal CdSe/CdS and CdSe/ZnS core/shell nanoparticles are promising emitters for quantum information technology, biological labelling [12], and realization of devices such as thin film LEDs or nanoparticle quantum dot lasers [13, 14], even as single photon sources [15]. Introducing a middle shell (CdS or ZnSe) between the CdSe core and the ZnS outer shell allows to reduce the strain inside the nanoparticle considerably and stability is further improved [16].

Thus for core-shell systems, it is crucial to determine the core and shell thickness, which control optical properties such as emission wavelength and photostability. Techniques such as transmission electron microscopy (TEM) and powder X-ray diffraction (XRD) yield information about the shape of the particles and the long-range order of the atoms [17, 18]. But the information content is limited at aperiodic parts at the surface or at interfaces. To ensure photostability of the nanoparticle, the shell materials are also chosen so as their lattice constant differs only little from the core material. This makes it difficult to obtain detailed structural and geometrical information from TEM and XRD.

With NEXAFS, we can selectively determine the contributions of the core and shell materials and determine the relative core and shell thickness [19]. For CdSe clusters covered with a CdS shell, we demonstrate this approach in Figs. 17.6 and 17.7. Here, the Cd M-edge NEXAFS of a CdS bulk sample and CdSe nanocrystals are shown together with CdSe/CdS nanoparticles of 4.4 ± 0.4 nm and

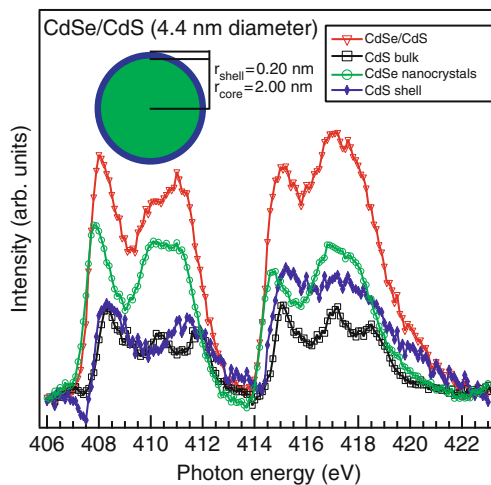


Fig. 17.6 Cd M-edge spectra of CdSe/CdS core/shell nanoparticles with diameters of 4.4 ± 0.4 nm (after subtraction of a polynomial background). Subtracting the CdSe nanocrystal signal (*circles*) from the total spectrum (*triangles*), we obtain the contribution from the CdS shell (*filled diamonds*). The *inset* shows a schematic to scale drawing of the core and shell with the radii obtained from the measurement. (From [19])

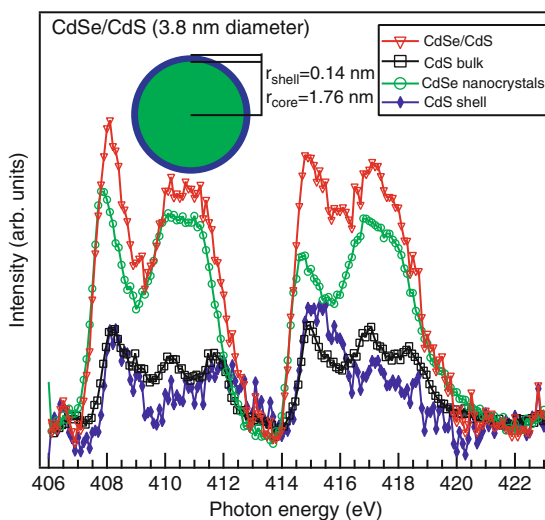


Fig. 17.7 Cd M-edge spectra of CdSe/CdS core/shell nanoparticles with diameters of 3.8 ± 0.3 nm (after subtraction of a polynomial background). Subtracting the CdSe nanocrystal signal (circles) from the total spectrum (triangles), we obtain the contribution from the CdS shell (filled diamonds). The inset shows a schematic to scale drawing of the core and shell with the radii obtained from the measurement. (From [19])

3.8 ± 0.3 nm diameter. The two main features correspond to transitions from the Cd $3d_{5/2}$ and $3d_{3/2}$ to the unoccupied 5p and 4f levels as has also been observed earlier [20]. They are thus a measure of the local partial density of states of those unoccupied levels. We observe a shift of 0.2 eV between the spectra for pure CdSe nanocrystals and CdS bulk, which is due to the different chemical environment of the Cd atoms.

We are now able to separate the contributions from the two different Cd species in a NEXAFS measurement of CdSe/CdS core/shell nanoparticles (triangles). We compare those with the spectra of pure CdS (squares) and CdSe (circles). By scaling the intensity of the CdSe spectrum and then subtracting it from the core/shell spectrum, we obtain the contribution from CdS in the nanoparticle shell (filled diamonds). When comparing this result with the CdS spectrum, we find a good agreement for the Cd $3d_{5/2}$ feature (left part). However, the Cd $3d_{3/2}$ signal differs in intensity and shape from the CdS bulk spectrum. This is attributed to the fact that the detection window of the electron analyzer is too small to contain fully the Auger features of both the Cd $3d_{5/2}$ and $3d_{3/2}$. The window has therefore been chosen to fully include the Cd $3d_{5/2}$ signal, resulting in a slightly distorted signal from the $3d_{3/2}$ features.

Now, we can use the intensity ratio of the spectral contribution of CdSe and CdS to determine the ratio of core radius to shell radius. The measured intensity is proportional to the amount of excited material, that is, the volume of the core V_{core} or shell V_{shell} , respectively.

Table 17.1 Thicknesses of core and shell from CdSe/CdS core/shell nanoparticles determined from the NEXAFS spectra

Diameter (nm)	$V_{\text{core}}/V_{\text{shell}}$	$r_{\text{core}}/r_{\text{tot}}$	r_{core} (nm)	r_{shell} (nm)
3.8 ± 0.3	4.0 ± 0.15	0.93 ± 0.16	1.76 ± 0.20	0.14 ± 0.13
4.4 ± 0.4	3.2 ± 0.07	0.91 ± 0.09	2.00 ± 0.20	0.20 ± 0.15

The electron mean free path in the nanoparticles is determined by the universal curve to be ~ 1 nm for 400 eV electrons [21]. The inverse of the mean free path then is the absorption coefficient $\sigma = 1,000 \mu\text{m}^{-1}$. The proportionality of σ and the total electron yield measurement (TEY) of nanoparticles has been studied in detail by Fauth [22] for several particle sizes. There, a linear regime is presented where the TEY depends linearly on σ and loss mechanisms can be neglected. The absorption coefficient in our case is however outside this linear regime. Thus, the measured intensities were corrected for electron absorption inside the particle.

From the spectra, we then get a values for the ratio of core volume to shell volume, see Table 17.1. The ratio of core radius r_{core} to total radius r_{tot} can then be calculated with

$$\frac{r_{\text{core}}}{r_{\text{tot}}} = \sqrt[3]{\frac{V_{\text{core}}}{V_{\text{core}} + V_{\text{shell}}}} = \sqrt[3]{\frac{V_{\text{core}}/V_{\text{shell}}}{1 + V_{\text{core}}/V_{\text{shell}}}}. \quad (17.1)$$

Applying (17.1), we then obtain the core and shell radii, see Table 17.1. The core radii of the two particles differ by 0.2 nm with the larger particle also having the larger core. The shell thicknesses show less variation and again the larger particles is covered by a thicker shell.

We observe that the capping CdS shells are very thin; especially that of the 3.8 ± 0.3 nm particle suggests that it is a single closed monolayer. It has been observed earlier that the growth of CdS shells on CdSe nanoparticles can be controlled precisely [23]. The difference in core size can also be determined using optical absorption and emission [24]. However, the non-luminescent capping shell cannot be detected with this method. In this case, it is also difficult to identify the shell thickness using HRTEM, as these thin shells grow without lattice defects on the cores and hence the core-shell boundary cannot be easily determined. Thus, we can here apply NEXAFS as a tool to investigate the structure of layered systems.

17.4 Summary

The combination of X-ray spectroscopy and X-ray diffraction methods is uniquely suited to characterize chemically prepared nanoparticles with respect to their different length scales. These are their local atomic properties – namely the chemical state and valency as well as the local bonding arrangement and coordination to neighbouring atoms. Here, NEXAFS spectroscopy has been crucial, since it is sensitive

to the atom specific electronic structure of selected atomic centres and allows to extract information on the local coordination. Next to the local, atomic properties the crystalline order within a nanoparticle has been determined with X-ray diffraction. Here, both the crystal structure and the size of the nanoparticles is measured through a detailed analysis of peak positions and peak shapes. Finally, the important aspect of the composition of core-shell nanoparticles has been determined using the chemical state selective properties of NEXAFS spectroscopy, where the core and shell thickness within nanoparticles has been determined. Thus X-ray methods are uniquely powerful to fully characterize nanoparticles on their relevant length scales.

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