Magneto-optical properties of self-assembled
III-V semiconductor nanostructures

PROEFSCHRIFT

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Front cover image: A contour plot of the photoluminescence spectra as function of the gate voltage of a charge tunable self-assembled InAs/GaAs quantum dot in strong interaction with electrons in the back contact.

Back cover image: A contour plot of the magnetoluminescence spectra of a self-assembled InAs/GaAs quantum ring, which displays a quadruplet splitting.

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Trefwoorden: III-V halfgeleiders, nanostructuren, fotoluminescentie, magnetoluminescentie, kwantum punten, kwantum ringen, g-factor, diamagnetische verschuiving.
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Chapter 1

Introduction

In this introductory chapter a brief historical overview on semiconductor nanostructures is given. The general properties of zero-dimensional nanostructures are explained by discussing InAs/GaAs quantum dots. Quantum dots (QDs) consist typically out of a cluster of \(\sim 10^5\) atoms of a semiconductor compound surrounded by a matrix of a different semiconductor compound. The difference in semiconductor material results into a difference of the energy band gap, which gives rise to the confinement of charge carriers in the quantum dots. As the emphasis in this thesis is on the optical properties of different nanostructures studied by photoluminescence experiments, the basics of these measurements are explained. To motivate the study of semiconductor nanostructures, future applications and prospects of self-assembled quantum dots are discussed. The chapter is concluded by the scope of this thesis.

1.1 History

It was in 1982 that Y. Arakawa and H. Sakaki first proposed to use zero-dimensional semiconductor nanostructures in order to improve the lasing properties of the conventional quantum well lasers [1]. As the density of states of a zero-dimensional system consists of discrete peaks, as compared to the constant density of states of a two-dimensional system (see Fig. 1.1), several advantages are obtained by introducing a laser based on zero-dimensional nanostructures. For one, the laser threshold current is lower as compared to the quantum well laser threshold current as the electron-hole overlap in a zero-dimensional structure is larger, resulting in a larger oscillator strength. Moreover, the temperature dependence of the laser threshold current of lasers based on zero-dimensional nanostructures is negligible. This is easily understood as the discrete density of states gives rise to only a small thermal redistribution of charge carriers to other discrete energy levels. In 1982 it was not yet possible to grow zero-dimensional semiconductor nanostructures. Therefore Y. Arakawa and H. Sakaki used a quantum well structure in a high magnetic field (\(\sim 30\,\text{T}\)) to give a proof of principle; the magnetic field provided the confinement in the lateral plane whereas the quantum well confined the charge carriers in the growth direction. The improvements in semiconductor growth in the mid-80’s provided the opportunity to grow intrinsic zero-dimensional semiconductor nanostructures in Stranski-
Figure 1.1: The density of states (DOS) of a three-, two-, one- and zero-dimensional structure as function of the energy $E$. Whereas the two-dimensional quantum well structure has a constant density of states, the zero-dimensional nanostructure (such as quantum dots) has a discrete distribution of states.

Krastanow mode [2, 3]. These nanostructures are nowadays generally known as quantum dots and, in fact, quantum dot lasers have been realized. A comparison between quantum well and quantum dot laser devices is given in Refs. [4, 5]. At the moment the aim is to grow as high density and optical quality quantum dot samples as possible in order to improve further the gain of the quantum dot lasers.

At the same time, another driving force for the study of the properties of zero-dimensional systems is coming from the electronics industry. From the early 1970's onward there has been a strong increase of the number of transistors on a chip, which is well described by Moore’s law. Figure 1.2 shows that whereas in 1970 there were 1000 transistors on a single chip, this number has increased towards over 1 billion transistors on the same chip nowadays. This corresponds to a size reduction of a single transistor going down from $\sim \text{cm}$ to $\sim 65 \text{nm}$. The desire to fabricate faster and smaller processors is governed by the need to decrease the production costs of a single chip as well as the need to increase the computing power. To satisfy this demand it is necessary to integrate the data processing, transport and storage on a single chip. This means that we have to combine electronic, photonic and magnetic properties on the scale of a few thousand atoms preferably compatible with the currently used materials and techniques. For these nano-sized dimensions the physical laws are no longer governed by classical mechanics, but by quantum mechanics. As quantum dots have similar dimensions, the fundamental properties of these nanostructures can give more insight on the physical behavior of transistors at the nanoscale. In the next section a short introduction to quantum dots is given.
Figure 1.2: The increase of the number of transistors per chip over time is described by Moore’s law. Moore’s law states that every two years the number of transistors on a chip doubles [6, 7].

1.2 Quantum dots

Semiconductor engineering allows for the tuning of electric, magnetic and optical properties of semiconductors structures. In Fig. 1.3 various semiconductor compounds are shown. Their lattice constants $a_0$ are plotted against the band gap energy $E_g$ and the corresponding wavelength $\lambda$ at room temperature. Heterostructures consist of layers of different semiconductor compounds, which are grown atomic layer by atomic layer using for instance Molecular Beam Epitaxy (MBE), Metal-Organic Vapor-Phase Epitaxy (MOVPE) and Chemical Beam Epitaxy (CBE) (see chapter 2). As different semiconductor compounds have different lattice constants, heterostructures in general consist of strained layers. These layers have different conduction and valence bands and consequently a different band gap energy $E_g$. Quantum dots are formed as a consequence of the compressive strain between two different semiconductor compounds. An extended overview of different quantum dot structures is given in Ref. [9] and references therein. As an example InAs quantum dots in a matrix of GaAs are treated here. The quantum dots are grown in the Stranski-Krastanow growth mode [2] in which a thin layer of InAs is grown on top of GaAs. The lattice constant of InAs is larger as compared to GaAs. Consequently, when the thickness of the InAs layer reaches a certain critical thickness (typically $\sim 1.7$ monolayers (ML)), a strain induced transition from two-dimensional layer-by-layer growth to three-dimensional island growth occurs. The result is the formation of self-assembled quantum dots on top of a thin wetting layer. An Atomic Force
CHAPTER 1.

<table>
<thead>
<tr>
<th>Compound</th>
<th>Bandgap Energy (eV)</th>
<th>Lattice Constant (Å)</th>
<th>Wavelength (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AlP</td>
<td>1.6</td>
<td>0.4</td>
<td>5.4</td>
</tr>
<tr>
<td>GaP</td>
<td>2.3</td>
<td>0.8</td>
<td>6.2</td>
</tr>
<tr>
<td>AlAs</td>
<td>1.8</td>
<td>0.6</td>
<td>6.6</td>
</tr>
<tr>
<td>GaAs</td>
<td>2.0</td>
<td>0.9</td>
<td>5.8</td>
</tr>
<tr>
<td>AlSb</td>
<td>1.5</td>
<td>0.7</td>
<td>6.1</td>
</tr>
<tr>
<td>InP</td>
<td>1.9</td>
<td>0.6</td>
<td>6.4</td>
</tr>
<tr>
<td>GaSb</td>
<td>1.4</td>
<td>0.8</td>
<td>5.6</td>
</tr>
<tr>
<td>InAs</td>
<td>1.7</td>
<td>0.9</td>
<td>5.9</td>
</tr>
<tr>
<td>InSb</td>
<td>1.6</td>
<td>1.0</td>
<td>5.5</td>
</tr>
</tbody>
</table>

Figure 1.3: The band gap energy $E_g$ and the corresponding wavelength as function of the lattice constant $a_0$ for different semiconductor compounds at room temperature [8].

Microscopy (AFM) image of these quantum dots is shown in Fig. 1.4. The lateral sizes of these dots are $\sim 70\,$nm in diameter and the dots have a height of $\sim 5\,$nm. However, the size, shape and composition of the dots can be tuned by varying the growth conditions.

For applications it is desired to have the quantum dots embedded in a matrix as surface oxidation and defects deteriorate the optical properties of the quantum dots at the surface. Moreover, the implementation of the quantum dots in a $p-i-n$ junction or Schottky device allows to control the electronic properties. Therefore the quantum dots are in general capped by a thick layer of GaAs. However, the capping process itself alters the structural properties of the dots due to segregation and diffusion of atoms out and into the quantum dots during the capping process. In order to analyze the influence of the capping layer it is necessary to cleave the sample and analyze the cross-section of the dots. Such analyzes can either be performed by doing Cross-sectional Transmission Electron Microscopy (X-TEM) or Cross-sectional Scanning Tunneling Microscopy (X-STM). In this thesis X-STM is the method used to analyze the individual dot properties (see chapter 2). An X-STM image of an InAs/GaAs quantum dot is shown in Fig. 1.5. The bright contrast corresponds to the indium rich areas and the dark contrast corresponds to the GaAs. By performing X-STM on a large number of dots, the average quantum dot properties are determined. In the case of Fig. 1.5 the quantum dot resembles a pyramidal quantum dot with an indium rich top and an indium poor bottom.

Due to the smaller energy band gap in InAs quantum dots as compared to...
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Figure 1.4: An AFM image of $1 \times 1 \mu m^2$ of self-assembled InAs/GaAs quantum dots. The lateral sizes of these dots are 70 nm in diameter and the dots have a height of 5 nm. The atomic steps of the wetting layer are resolved. Image taken from Ref. [10].

the GaAs matrix, the quantum dots confine charge carriers in all three dimensions, which gives rise to discrete energy levels inside the dots. The discrete energy levels resemble the atomic energy level structure and therefore the quantum dots are often called 'artificial' atoms. Since the height of the dot is much smaller than the lateral sizes, the major contribution in the confinement energy is determined by the height of the dot. This can be seen directly by approximating the quantum dot with a square box of dimensions $a$, $b$ and $c$ in the $x$-, $y$- and $z$-direction. The energy $E$ of a particle in a box is given by [11]:

$$E_{n,k,l} = \frac{\hbar^2 \pi^2}{2m} \left( \frac{n^2}{a^2} + \frac{k^2}{b^2} + \frac{l^2}{c^2} \right), \quad (1.1)$$

where $n$, $k$ and $l$ are integers corresponding to the quantum numbers of the different energy levels inside the dot, and $m$ is the mass of the particle confined

Figure 1.5: Cross-sectional Scanning Tunneling Microscopy (X-STM) image of a pyramidal shaped InGaAs/GaAs quantum dot. The black arrow indicates the wetting layer. The bright (dark) area corresponds to the indium (gallium) rich area. Image adapted from Ref. [10].
in the box. For \( c < a, b \), which is the case for example the quantum dots shown in Figs. 1.4 and 1.5, the main contribution to the energy is due to the quantization energy in the growth direction. Nevertheless, it is obvious that treating a quantum dot as a box of dimensions \( a, b \) and \( c \) gives a too crude approximation. However, it has been shown that the quantum dot confinement potential can already be described quite reasonably by a parabolic potential [12], as is discussed in chapter 3.

1.3 Photoluminescence

![Diagram of photoluminescence](image)

Figure 1.6: A schematic overview of a photoluminescence experiment. (a) Electrons from the valence band are excited to an energy above the conduction band leaving behind a hole in the valence band. (b) The electron and hole relax towards to lowest available energy state (in this case the lowest discrete energy level in the quantum dot) on the time scale of \( \sim \) ps. The Coulomb interaction between the electron and hole results in the formation of an exciton (a bound electron-hole pair). (c) On the time scale of \( \sim \) ns the exciton recombines and its energy is carried away by a photon. This light is known as the photoluminescence of the quantum dot and the emission energy is determined by the size, shape, composition and environment of the quantum dot.

The main focus in this thesis is on the optical properties of zero-dimensional nanostructures. In order to study the optical properties, photoluminescence (PL) experiments are performed. A schematic overview of a PL experiment is shown in Fig. 1.6. In PL experiments a light source (usually a laser) is used to excite electrons from the valence band to the conduction band leaving behind a hole in the valence band. Different relaxation processes such as phonon-mediated processes in the bulk semiconductor material and Auger processes in semiconductor QDs result into the relaxation of the electron and hole towards the lowest available energy state. Typically for a QD the captures times are in the order of \( \sim \) ps, depending on the excitation conditions and the material properties. The electron and hole in the quantum dot experience the Coulomb
interaction resulting in the formation of a bound electron-hole pair, which is the neutral exciton $X^0$. These Coulomb energies are typically in the order of $\sim 10$ meV, which is relatively small compared to the QD confinement energy (for the InAs/GaAs QDs studied in this thesis $\sim 80$ meV). On the time scale of $\sim$ ns the electron and hole recombine. The capture times and relaxation times can be measured in time-resolved PL measurements. The energy of the recombining exciton is carried away by a photon. The energy of the photon is therefore a direct measure of the confinement energy of the exciton in the QD, which is determined by the size, shape and composition of the QD and the surrounding material properties. Higher excitation density conditions increase the number of captured carriers in the quantum dots, resulting in the creation of biexcitons in the dots. The biexciton ($2X^0$) constitute of two electron-hole pairs. Finally, in several experiments described in this thesis an external voltage could be used to provide additional electrons (holes) to the quantum dots and thereby creating charged excitons. This allows us to extract additional information about the Coulomb interaction between the carriers present in the dot. In summary, photoluminescence is able to reveal the electronic, optical and even magnetic properties of individual quantum dots. Unraveling these properties is crucial in order to utilize these nanostructures in future applications.

1.4 Possible applications

As was already mentioned in the first section, many efforts have been put in the realization of the semiconductor quantum dot laser. Twenty-five years after the first proposal [1] the first commercial QD lasers are available. One of the current issues is the enhancement of the output power of these lasers [13]. Therefore many studies are performed to increase the QD density in a single layer and different approaches are used to stack a large number of QD layers on top of each other in order to increase the gain. Quantum dot laser sources are developed to operate at the wavelengths of 1.3 and 1.55 $\mu$m, compatible with the telecommunication wavelengths. An important advantage of these laser sources is that they are GaAs and InP based and therefore allow easy on-chip integration with future III-V photonic chips, where they can for instance be used as optical modulators and optical switches. Another major advantage of the QD based devices is the low energy consumption as compared to the present devices due to their high oscillator strength. Besides acting as a source for optical output, the QD layers can also be implemented as an infrared detector [14] and there have been proposals to use them in solar cells [15, 16, 17].

Instead of using high density QD layers ($> 10^{11} \text{cm}^{-2}$) as mentioned for the previous applications, much research is also done in order to achieve extremely low QD densities ($< 10^9 \text{cm}^{-2}$). In the case of low QD densities it is possible to optically address individual quantum dots. Individual quantum dots are considered as important candidates for single photon laser sources, equivalent to the single molecule laser [18]. In order to achieve lasing operation the gain of a single quantum dot needs to be enhanced. Several approaches are investigated, such as implementing the QD in microcavity pillars [19, 20] and photonic cavities [21]. One of the major obstacles is the request for the site control of the quantum dots. In the case of the photonic cavity it is for example necessary to position the quantum dot preferably in the center of the cavity. Moreover, the quantum
Figure 1.7: (a) The level diagram of a radiative decay of the biexciton state $2X^0$ via the intermediate exciton state $X^0$ to the ground state $G$ in a quantum dot. The competing two photon decay paths are distinguished only by the circular polarization of the photons, as indicated by the colored arrows (red and blue arrows have opposite circular polarization). (b) The fine structure splitting $\delta_1$ in the exciton state $X^0$ allows to discern the two optical decay paths and the emitted photons (with linear polarization) are not longer entangled.

Quantum entanglement is a property of a quantum mechanical state of a system of two or more objects in which the quantum states of the constituting objects are linked together so that one object can no longer be adequately described without full mention of its counterpart. Entangled photon pairs are essential for quantum information applications such as quantum key distribution and controlled quantum logic operations. Figure 1.7 shows the level diagram of the radiative decay of the biexciton, via the intermediate $X^0$ state. For ideal quantum dots the two competing photon decay paths are only distinguished by the polarization of the emitted photons (Fig. 1.7(a)). Unfortunately, the exciton state $X^0$ in general exhibits a fine structure splitting $\delta_1$ (Fig. 1.7(b)), which prevent the entanglement of the photons in the biexciton decay scheme. This splitting is discussed in more detail in chapter 3 and arises from the breaking of the in-plane quantum dot symmetry. As will be shown in chapter 6, this splitting is dependent on the quantum dot size. Furthermore, Stevenson et al. [26] showed that the application of an in-plane magnetic field can tune this fine structure splitting to zero, opening the way for triggered entangled photon pairs based on individual quantum dots.

As a last example, single quantum dots are also proposed to be used as qubits in the field of quantum information processing. A qubit consists of a two level quantum system. Contrary to the regular bit, which adapt 1 or 0, the qubit can be described by a superposition of the states $|0\rangle$ and $|1\rangle$ as $\Psi = \alpha|0\rangle + \beta|1\rangle$. A possible two level system is the two polarization state of the photon or the spin-up and spin-down states of the electron. The spin-up and spin-down electron state are splitted in a magnetic field by the Zeeman energy. The Zeeman energy is proportional to the $g$-factor, which will be introduced in detail.
in chapter 3. The confinement of electrons in semiconductor nanostructures like quantum dots allows for better control and isolation of the electron spin from its environment. Control and isolation are important issues to consider for the design of a quantum computer [33, 34]. Initialization of the quantum computer can be achieved by allowing all spins to reach their thermodynamic ground state at low temperature $T$ in an applied magnetic field $B$. Virtually all spins will be aligned when the Zeeman energy is much larger than the thermal energy. One way of performing single qubit operations can be performed by changing the local effective Zeeman interaction. One possible route to achieve this is using tunable $g$-factors on single quantum dot level, allowing to change the ground state from spin-up to spin-down or vice versa and thereby writing a "1" or a "0". In order to understand better the influences of the quantum dot size, shape and composition and the quantum dot environment on the $g$-factor several studies have been performed recently [35, 36, 37, 38]. In fact, there have been proposals to use quantum dots with a $g$-factor of zero. In that case an external electric field can be used to tune the sign of the $g$-factor [39, 40, 41, 42]. Another way of performing single qubit operations is by the Electron Spin Resonance (ESR) technique [43, 44].

1.5 Scope of the thesis

In this thesis the optical properties of several zero-dimensional semiconductor nanostructures are investigated both in the presence and absence of an external magnetic field. An overview of the different experimental techniques used to grow, prepare, characterize and analyze the nanostructures studied throughout this thesis is given in chapter 2. Special emphasize is put on the confocal microscopy setup, which is used to study the individual magneto-optical properties of the different nanostructures.

The main goal of chapter 3 is to provide a description of the magneto-optical properties of individual quantum dots. For this purpose the quantum dot potential is described using the harmonic oscillator model, which in most cases is a good approximation of the quantum dot confinement potential. This model is used to introduce the concepts of the exciton $g$-factor and the exciton diamagnetic coefficient. The provided description allows for the analysis of the micro-photoluminescence experiments on a sample containing a low density of InAs/GaAs quantum dots. From these experiments it is inferred that the $g$-factor is dependent on the emission energy of the quantum dot, which is determined by the size of the dots.

In chapter 4 the measurements on charge-tunable InAs/GaAs quantum dots are discussed, which are similar to the ones studied in chapter 3. In a charge-tunable device the quantum dots are embedded in a Schottky structure. By applying a gate voltage additional charges can be introduced in the quantum dots. This allows for the study of different charged exciton complexes as function of gate voltage. It will be shown that the strong interaction of the charged carriers inside the quantum dot with the electron reservoir at the back contact of the Schottky device, gives rise to new many-body signatures observed in the quantum dot photoluminescence. The many-body signatures are understood using the Mahan and Anderson exciton model.

In chapter 5 the same charge-tunable quantum dots are investigated in a
magnetic field. Along with the positive diamagnetic shift observed for the majority of the quantum dots, two different types of negative diamagnetic shift are reported. This is the first time that such a negative diamagnetic behavior is reported for quantum dots. We will show that the shallow character of our quantum dots causes a negative diamagnetic shift for the highly negatively charged exciton complexes. The second type of negative diamagnetic shift is observed even for the neutral exciton and is linearly dependent on the magnetic field, resembling the shakeup lines in quantum wells.

InAs/InP quantum dots, which emission wavelength is compatible with the telecommunication wavelength, are studied in chapter 6 using AFM, X-STM, macro-PL and micro-PL. By measuring the magneto-optical properties of a large number of individual quantum dots it is shown that both the height and the diameter of the quantum dots determine the value of the $g$-factor. Moreover, it will be shown that the exciton $g$-factor can be engineered to zero by growing quantum dots of appropriate size. We also demonstrate that the fine structure splitting is reduced by growing relatively large quantum dots. A theoretical model is used to calculate the effect of the quantum dot size on the $g$-factor. The model is both qualitatively and quantitatively in agreement with the experimental obtained results.

In chapter 7 InAs/GaAs quantum rings are studied. These nanostructures have gained a lot of attention in the last decade as they are a candidate to display the Aharonov-Bohm effect. First the Aharonov-Bohm effect will be described, after which the magnetization measurements on a large ensemble of quantum rings are discussed. These measurements demonstrate the presence of the Aharonov-Bohm effect for single electrons in these rings. Using a model based on the X-STM measurements on these nanostructures we reproduce the magnetic field position of this oscillation.

The optical properties of the self-assembled InAs/GaAs quantum rings have been investigated for magnetic fields up to 30 T for both a large ensemble of quantum rings as well as individual rings in chapter 8. The introduced model of chapter 7 is extended and used to interpret the macro magnetoluminescence data. Importantly, both in the ensemble and single ring magnetoluminescence characteristic features of the ring-like geometry have been found.

In the last chapter 9 the experiments on type II InP/GaAs quantum dots are discussed. For these dots the hole is located outside the quantum dot, whereas the electron is confined inside the quantum dot creating a type II exciton. In the magnetoluminescence of the ensemble of dots we did not resolve any oscillatory behavior related to the AB effect in contrast to experiments reported on the same sample [45]. Decisive magnetoluminescence measurements on individual dots did not reveal any AB related phenomena within the experimental resolution of 40 $\mu$eV. Our results show the necessity to study subtle magnetoluminescence properties of nanostructures on an individual dot level, rather than on a large ensemble of dots.
Chapter 2

Experimental methods

2.1 Abstract

This chapter gives an overview of the experimental techniques to grow, prepare, characterize and analyze the nanostructures studied throughout this thesis. The first section gives a short introduction to the two methods used for growing the nanostructures. In order to study individual nanostructure properties additional sample preparation steps may be required and are described in the second section. The third section gives an introduction to Atomic Force Microscopy (AFM) and Cross-Sectional Tunneling Microscopy (X-STM). These techniques are used to analyse the structural properties, i.e. size, shape and composition, of the nanostructures. The macro-photoluminescence setup utilized to analyse the optical properties of a large ensemble of nanostructures is discussed in the fourth section. The main goal of this chapter is to describe the confocal microscope setup used to study the magnetoluminescence properties of single nanostructures. The working principle, the actual confocal microscope setup, the dispersion and detection system and the alignment procedure are discussed extensively in fifth section of this chapter. Finally, the advantages and practical implementation of the solid immersion lens (SIL) are discussed.

2.2 Sample growth

The semiconductor nanostructures studied in this thesis are grown using two different growth techniques: Molecular Beam Epitaxy and Metal-Organic Vapor Phase Epitaxy. In this section these two types of epitaxial growth techniques are briefly described.

2.2.1 Molecular Beam Epitaxy

In the Molecular Beam Epitaxy (MBE) growth technique, molecular beams are directed onto a heated substrate, where the atoms can be incorporated into the crystal lattice. The source materials are evaporated from effusion cells. The molecular flux from the cells is controlled through the cell temperature. The control of the flux provides a handle to determine the composition of the grown structures. Moreover, each cell can be shielded by a cell shutter allowing
growth of different semiconductor compounds on top of one another. During the growth process the sample is heated in order to increase the mobility of the deposited atoms. The sample temperature is a crucial parameter to determine the nanostructure density. In general a low (high) sample temperature of about $420 - 480 \degree C$ ($480 - 530 \degree C$) allows to achieve a high (low) quantum dot density of $10^{10} - 10^{11}$ cm$^{-2}$ ($10^8 - 10^9$ cm$^{-2}$). The growth speed and the amount of deposited material are also crucial parameters determining the quantum dot density. For the lowest QD densities of $\sim 10^8$ cm$^{-2}$ the typical growth temperature is $530 \degree C$, the growth speed is 0.13ML/s, and the amount of deposited InAs is 2.1 ML. The sample can be rotated during growth in order to obtain better uniformity in the layer structures; all cells face the sample under slightly different angles, and the flux of the molecular beam is not homogenous. The structural quality of the sample is monitored during growth by the Reflection High Energy Electron Diffraction (RHEED) signal. In RHEED a high energy electron beam hits the sample under a grazing angle, generating a diffraction pattern on a phosphorus screen. This pattern provides information about the growth and the sample quality. Moreover, the RHEED pattern is crucial in order to determine the transition from layer-by-layer growth to three-dimensional island growth. For in-depth information about the MBE technique, see for example Ref. [46].

2.2.2 Metal-Organic Vapor Phase Epitaxy

In Metal-Organic Vapor Phase Epitaxy (MOVPE) gaseous metal-organic molecules (precursors) are flowing together with a background gas over a heated sample substrate. The heat causes the precursors to decompose. The organic part of the molecule flows away, whereas the metal part diffuses and chemisorbs on the substrate. The growth can be controlled by the gas composition, gas flux and substrate temperature. By controlling these growth conditions, it is possible to tune the size, shape, composition and density of the quantum dots. An extended review on this growth technique is for example given in Ref. [47].

2.3 Sample preparation

In the case of a high quantum dot density a mask can be created on top of the sample in order to study the optical properties of individual quantum dots. An electron beam lithography process is applied to produce small apertures ranging from $\sim 0.4 \mu m$ to $\sim 1.5 \mu m$. The different steps of the masking process are shown schematically in Fig. 2.1(a). First a $\sim 400$ nm thick layer of photoresist coating poly-methylmethacrylate (PMMA) with 7% anisole is spun on top of the sample. The blue pattern shown in Fig. 2.1(b) is written by exposing the photo-resist to the e-beam. The pattern contains 19 by 19 apertures and each row and column is aligned with respect to the triangular marker, which serves as an alignment marker in the confocal microscope. The distance between the apertures is $\sim 20 \mu m$. The e-beam acceleration voltage is set to 20 kV with an aperture of 30 $\mu m$. In general 10 patterns are written each with a higher dose ranging from $250 - 340 \mu C/cm^2$. By increasing the dose the apertures become smaller as there is more electron back scattering resulting in the exposure of larger areas of the photo-resist. Table 2.1 lists the aperture size as function of
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Figure 2.1: (a) The masking process schematically shown in several steps: (1) the unprocessed sample, (2) applying the photo-resist, (3) writing the pattern with the e-beam, (4) removal of the photo-resist which was exposed to the e-beam, (5) evaporating a layer of aluminium on top and (6) lift-off the non-exposed photo-resist together with the aluminium on top creating the apertures. (b) The mask as used for the samples. The triangular markers indicate the different rows and columns of apertures. (c) An example of an aperture as obtained by the electron beam lithography process. The diameter is $1 \times 1 \mu m^2$ by using a dose of $240 \mu C/cm^2$.

the dose. After the patterning the PMMA is developed in a solution of methyl isobutyl ketone and isopropyl alcohol in the ratio 1:3 for 60 s and rinsed in alcohol for another 60 s. The part of the photo-resist that was exposed to the e-beam is removed. In the following step a 100 nm thick layer of aluminium is evaporated on top. The aluminium that is deposited directly on top of the sample acts as the mask, whereas the aluminium on top of the unexposed PMMA layer is removed by placing the sample in a solution of acetone for about 30 minutes. The sample is further cleaned in an ultrasonic bath of isopropyl alcohol. An example of an aperture created by the above described process is shown in Fig. 2.1(c).

Table 2.1: The aperture size versus the dose obtained with the above described procedure as determined by scanning electron microscopy.

<table>
<thead>
<tr>
<th>aperture size (µm)</th>
<th>dose (µC/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.34 − 1.40</td>
<td>210</td>
</tr>
<tr>
<td>1.25 − 1.28</td>
<td>220</td>
</tr>
<tr>
<td>1.11 − 1.14</td>
<td>230</td>
</tr>
<tr>
<td>1.00 − 0.983</td>
<td>240</td>
</tr>
<tr>
<td>0.889 − 0.851</td>
<td>250</td>
</tr>
<tr>
<td>0.775 − 0.753</td>
<td>260</td>
</tr>
<tr>
<td>0.686 − 0.649</td>
<td>270</td>
</tr>
<tr>
<td>0.584 − 0.568</td>
<td>280</td>
</tr>
<tr>
<td>0.522 − 0.490</td>
<td>290</td>
</tr>
<tr>
<td>0.437 − 0.423</td>
<td>300</td>
</tr>
</tbody>
</table>
CHAPTER 2.

2.4 Structural characterization

Scanning Probe Microscopy (SPM) is an essential tool for the study of the structural properties of semiconductor nanostructures. By scanning surfaces with a nanosized probe, it is possible to gain information about structures at the surface with dimensions in the nanometer range. Throughout this thesis two different SPM techniques are used to address the structural properties of the samples: Atomic Force Microscopy (AFM) and Cross-sectional Scanning Tunneling Microscopy (X-STM). The former is normally used to quickly assess the quality of the growth, the nanostructure density and their rough sizes. The latter is far more time consuming as it demands special sample preparation, but it yields more detailed and accurate information on the grown structures.

2.4.1 Atomic Force Microscopy

An AFM probe consists out of a tip, with a typical radius of $\sim 20$ nm, attached to a cantilever. By bringing the tip in close proximity to the surface ($\sim$ nm), different forces act between the tip and surface. Depending on the tip-surface distance, these forces cause the cantilever to deflect. A constant deflection can be maintained, by monitoring the deflection and adjusting the tip-surface distance accordingly with piezo crystals using a feedback loop. By scanning the probe across the surface and measuring the voltage applied to the piezo a topographic image of the surface can be made.

The samples studied in this thesis often have an uncapped quantum dot layer grown on top, enabling the assessment of the quantum dot density and size. However, the acquired images are convoluted with the tip shape as the AFM tip has a finite radius. This is shown schematically in Fig. 2.2. Quantum dots typically have lateral dimensions similar to the AFM tip size. Measuring the dimensions of such quantum dots leads to an overestimation of the lateral size. More importantly, the uncapped quantum dots at the surface will not have the same dimensions as the capped quantum dots, since the capping process
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will lead to segregation and diffusion of atoms in and out of the quantum dot. Nevertheless, in general the quantum dot density and overall quality of the sample can be inferred from AFM. Further benefits of an AFM are the ambient operational conditions, stability and relative large scan speeds, all contributing to the ease and speed up of the characterization process.

2.4.2 Cross-sectional Scanning Tunneling Microscopy

In STM the probe consists out of a sharp conductive metal tip with a radius of a few nm. By biasing the tip with respect to the surface and maintaining a tip-surface distance of ~ 5Å, a tunneling current can be established between the tip and surface. Generally the tunneling process takes place through a single atom, leading to atomic resolution. The tunneling current depends exponentially on the tip-surface distance and a feedback loop controlling the tip-surface distance by a piezo element can maintain a fixed tunneling current. By measuring the voltage applied to the piezo, the topography of the surface can be imaged. However, the tunneling does not solely depend on the tip-surface distance, but also on the local electronic environment of the surface underneath the tip. The acquired data is thus a convolution of the topography and electronic contrast. By measuring at either large negative or large positive tip-surface bias, the topographic contrast dominates over the electronic contrast [48, 49]. By imaging the electronic contrast, it is possible to identify chemically different atoms.

In order to assess capped quantum dots, a technique called Cross-sectional STM (X-STM) has been developed. An extended review of this technique can be found in Refs. [49, 50]. In X-STM measurements a sample is cleaved in Ultra High Vacuum in order to prevent oxidation and contamination of the cleaved surface. The capped quantum dots are probed by X-STM and the dimensions and compositions are accurately determined. However, the cleavage is performed through a random plane of the quantum dot and a large number of quantum dots need to be measured to allow the determination of the shape, average size and composition profile of the quantum dots.

2.5 The macro photoluminescence setup

The macro-photoluminescence setup is schematically shown in Fig. 2.3. This setup is used to analyse the PL quality and wavelength of a large ensemble of nanostructures. The excitation is provided by a Nd:YAG continuous wave laser, emitting at 532 nm (~ 2.33 eV) with a power of 25 mW. A low pass filter is positioned in front of the laser to ensure that light with higher harmonic frequencies is blocked. Via multiple mirrors the laser light is directed onto the sample. The sample is placed inside a helium flow cryostat, which allows control of the sample temperature between 4.5 K and room temperature. The spot size of the laser beam is ~ 4 mm², which corresponds to an excitation density of ~ 500 mW/cm². In order to decrease (increase) the excitation density, neutral density filters (a focussing lens) are introduced in the excitation path. The PL from the sample is collected and collimated using the collection lens. A mirror directs the collected PL towards a monochromator for spectral analysis. In front of the monochromator a filter blocks the reflected laser light, and a lens focuses the PL onto the entrance slit of the monochromator. The monochroma-
CHAPTER 2.

The macro-photoluminescence setup. A green laser excites a sample mounted in the helium flow cryostat. The PL of the sample is collected and directed towards the monochromator. In the monochromator the light is dispersed and detected by either the InGaAs or InSb detector.

The confocal microscope

The majority of PL experiments on individual nanostructures have been performed with the confocal microscope attoCFM-I from Attocube Systems AG. In this section the confocal working principle, the setup properties and the alignment procedure are described.

The confocal principle

To study the photoluminescence of individual nanostructures a high spatial resolution of the optical setup is required. For example, a relatively low quantum dot density of $10^9$ QDs/cm$^2$ results already to an inter dot distance of $\sim 300$ nm. To achieve high spatial resolutions in the order of the inter dot distance, diffraction limited optics is necessary. To understand why diffraction is actually limiting the spatial resolution, consider the following. Assume that a single quantum dot is a point light source, illuminating the objective. The spherical waves from the quantum dot converge via the objective to the focal point. However, this will not result in a single point in the image. Due to the finite aperture of the objective, a Fraunhofer diffraction pattern is formed, analogous to the Airy diffraction pattern caused by a circular aperture. The radius of the central Airy
Figure 2.4: The working principle of a confocal microscope. A light source illuminates with a focused beam the sample in the focal plane of the objective. The photoluminescence is collected using the same objective and focused onto a pinhole. Any collection light which is out of focus (indicated by the blue color), both vertically and laterally, will be partially blocked by the pinhole.

The typical resolution of the used confocal microscope system with a NA = 0.65 objective and an excitation wavelength of 635 nm is $d = 508\,\text{nm}$.

The confocal microscope setup uses diffraction limited optics to achieve these high spatial resolutions. The working principle of a confocal microscope is depicted in Fig. 2.4. A light source illuminates the sample with a diffraction limited spot in the focal plane of the objective. The photoluminescence is collected using the same objective and focused onto a pinhole. Any collection light which is out of focus, both vertically and laterally, is blocked by the pinhole. The pinhole ensures that both the illumination and collection originate from the same diffraction limited spot, hence the name confocal microscope. The spatial resolution can be improved further if the pinhole has a diameter smaller than the Airy disk. In that case it is possible to improve the resolution by a factor of 1.36 [53]. By scanning the illumination/collection spot over the sample, a lateral two-dimensional image of the sample can be formed.

\[ r_{\text{Airy}} = 0.61 \frac{\lambda}{NA} \]  \hspace{1cm} (2.1)

where $\lambda$ is the wavelength of the light and NA is the numerical aperture of the objective. A larger numerical aperture means that light from a larger light cone is focused by the objective: the object in front of the objective will have more resolvable details, since the diffraction pattern will be of smaller influence. As a measure for the resolution, often Sparrows criterion [52] is used, which describes the FWHM $d$ of the central Airy disk by:

\[ d = 0.52\frac{\lambda}{NA} \]  \hspace{1cm} (2.2)
In general a spot size of $\sim 1 \mu m^2$ contains on average several quantum dots. However, the spot size alone does not determine the number of quantum dots contributing to the photoluminescence. The diffusion of electrons and holes outwards from the spot to neighboring dots results in the observation of a higher amount of photoluminescence peaks as expected by considering the spot size only. Although these dots are laterally out of focus, they still weakly contribute to the PL. More importantly, the spot size can be considerably larger for experiments where the excitation wavelength is very different from the collection wavelength. This is due to the chromatic objective used for both the excitation and collection: to optimize the collection efficiency the excitation spot is moved out of focus. Nevertheless individual quantum dots can still be studied as the different quantum dots will have spectrally different properties, due to the inhomogeneous size distribution of the quantum dot ensemble.

2.6.2 The confocal microscopy setup

A schematic overview of the setup for measuring the micro-photoluminescence is shown in Fig. 2.5. The setup consists out of four different parts: the cryostat containing the superconducting magnet, the microscope stick with the sample stage, the optical head and the dispersion and detection system. These parts are described and characterized in the following sections.

(A) The cryostat

The Cryovac cryostat consists out of four compartments. The outer vacuum shield is used to thermally isolate the inner chambers from the environment. This vacuum contains a super-isolation foil and is evacuated by a turbo-pump to a typical pressure of $\sim 10^{-5} - 10^{-6}$ mbar. The magnet bath contains a superconducting magnet, capable of producing magnetic fields up to 10 T along the optical axis (i.e. Faraday-geometry). In order to operate this magnet the bath needs to be filled with liquid helium ($\pm 40$ l). The procedure to cool down the magnet is described in Ref. [54]. Two temperature sensors are present in the magnet bath: one at the bottom of the cryostat and one at the lambda plate (not shown in Fig. 2.5). The resistance values for room temperature, liquid nitrogen temperature (77 K), and liquid helium temperature (4.2 K) are $\sim 30$, 7 and 2 $\Omega$, respectively. It is possible to pump the helium bath at the lambda plate in order to decrease the temperature to $\sim 2$ K, which allows for magnetic fields up to 12.5 T. The insert is separated from the magnet bath by the inner vacuum shield. Depending on the preferred experiment, this shield can be evacuated or filled with helium contact gas.\(^1\) The insert is used to accommodate for the microscope stick and can be filled with liquid helium ($\pm 7$ l) independently from the magnet bath. The length of the microscope stick is matched to the cryostat such that the sample is at the center of the magnet.

(B) The microscope stick

The microscope stick consists out of two parts: an optical stick and a vacuum tube, together forming a closed vacuum system. The sample is mounted at the

\(^1\)In case that the temperature of the insert is different as compared to the magnet bath, thermal isolation is needed.
Figure 2.5: A schematic overview of the micro-photoluminescence setup positioned in the helium bath cryostat. The inset shows the objective, the sample stage, and the piezo-positioners, which are attached to the microscope stick.
Figure 2.6: (a) Schematic drawing of the interference setup used to calibrate the piezo step size and to determine the spot size of the confocal microscope. A laser is coupled into a fiber and focused on a sample using an objective. The reflected light from that sample is collected by the same fiber and coupled into a detector. By scanning the sample in the x- and y-direction an interference image of the sample surface is measured. (b) Step size of the piezos as function of the driving voltage (0 − 20 V) at room temperature for positive and (c) negative voltages. The filled squares are the measurements and the open circles are taken from the calibration data provided by Attocube [55].

The optical interference signal is measured on a calibration sample in order to verify the step size of the x and y positioners versus the applied piezo voltage data provided by Attocube. Figure 2.6(a) shows a schematic drawing of the interference experiment. The actual interference is between the light reflected by the fiber end (1) and the light reflected by the sample (2). The optical path difference is thus twice the distance from the fiber end to the sample. A calibration grating is used to determine the number of steps needed to move over a certain number of step edges. For an applied voltage of −20 V (+20 V) the piezo moved 850 ± 50 nm (800 ± 50 nm). The step size increases linear with the applied voltage from 0 − 20 V. The calibration results (filled squares) are shown in Fig. 2.6(b) and (c), and are in agreement with the calibration data provided by Attocube (empty circles) [55]. At 4 K the step size decreases to 4–5 nm/V [55]. Therefore the maximum voltage of 30 V at room temperature is increased to 50 V at 4 K. The maximum operation frequency of the piezo positioners is 1 kHz. The capacity of the piezos serve as an indication of the temperature and the corresponding values for different temperatures are given in table 2.2. Details about the principle of operation of the piezos can be found elsewhere.

Details about the principle of operation of the piezos can be found elsewhere.
Table 2.2: The piezo capacity at different temperatures. These values can vary by $\pm 10\ nF$.

<table>
<thead>
<tr>
<th>T (K)</th>
<th>Cap. x-piezo (nF)</th>
<th>Cap. y-piezo (nF)</th>
<th>Cap. z-piezo (nF)</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>458</td>
<td>470</td>
<td>516</td>
</tr>
<tr>
<td>77</td>
<td>178</td>
<td>172</td>
<td>168</td>
</tr>
<tr>
<td>4</td>
<td>110</td>
<td>116</td>
<td>119</td>
</tr>
</tbody>
</table>

For the measurements the optical stick is inserted in the vacuum tube and the system is flushed several times with helium gas to prevent moisture and dirt on the optics and/or the sample stage before it is inserted in the insert of the cryostat. To allow for heat exchange between the sample and the helium bath, $\sim 10\ mbar$ of helium contact gas is introduced in the microscope stick.

(C) The optical head

On top of the microscope stick the optical head for the excitation and collection of the PL can be mounted. A schematic drawing of the optical head is shown in Fig. 2.7. The optical head consists out of three arms: the excitation arm, collection arm and the imaging arm. A fiber coupled laser source provides the excitation light. The laser light is coupled out of the fiber at the excitation arm of the optical head and is collimated by lens L1. Half of the light is reflected down by the $50:50$ beam splitter to the microscope objective for excitation of the sample. The other half passes the beam splitter and is detected by a power meter.

![Figure 2.7: A schematic overview of the different optical elements and the optical paths in the optical head.](image-url)
The microscope objective collects the PL and the reflected laser light. As the objective is chromatic, the wavelength collimated by the objective depends strongly on the focus adjusted by the $z$-positioner. The collimated beam passes the lower beam splitter, where half of the light is lost. The beam passes a second beam splitter, which is used for alignment purposes and which is taken out after the alignment steps. The light passes a quarter lambda plate and a linear polarizer. The quarter lambda plate and linear polarizer allow for the determination of the polarization state of the light (see Refs. [51, 53, 56] for in-depth reading on the polarization of light). A high pass filter blocks the laser light and the remainder of the light is focused onto the single mode collection fiber using lens L3. In fact the collection fiber with a diameter of $\sim 5 \, \mu m$ acts as the pinhole of the confocal microscope. The collected light is analyzed using a dispersion and detection system. The details of the optical components used in the optical head and the microscope objective are given in table 2.3.

(D) The dispersion and detection system

![Schematic drawing of the triple monochromator system. Note that the fiber head can be placed on slits S2, S4 and S6 for triple, double and single stage measurements, respectively. The turrets T1-T3 contain 3 different gratings.](image)

The light coupled into the collection fiber travels through the fiber to a dispersive system for spectral analysis. The light is dispersed using a triple stage monochromator of Princeton Instruments and detected with either a OMA V InGaAs array or a Si CCD camera. A schematic drawing of the triple stage monochromator is depicted in Fig. 2.8. The first stage has a focal length of 750 mm and the second and third stage have a focal length of 500 mm each. In this thesis the triple monochromator is used in three different modes: the single, double additive, and triple additive mode. In additive mode the stages of the triple monochromator disperse the light in order to obtain the highest spectral resolution. The triple monochromator can also be used in the subtractive mode as is explained in Ref. [57]. Triple stage measurements are performed by connecting the fiber head to slit S1 or S2, double stage measurements...
Table 2.3: The specifications of the used components in the optical head.

<table>
<thead>
<tr>
<th>Component</th>
<th>Specifications</th>
</tr>
</thead>
<tbody>
<tr>
<td>Illumination fiber</td>
<td>Single Mode 630 nm</td>
</tr>
<tr>
<td></td>
<td>Numerical aperture NA: 0.12</td>
</tr>
<tr>
<td></td>
<td>Mode field diameter: 4.3 $\mu$m</td>
</tr>
<tr>
<td>Illumination lens (L1)</td>
<td>Spectral range: 350 – 1550 nm</td>
</tr>
<tr>
<td></td>
<td>Numerical aperture NA: 0.25</td>
</tr>
<tr>
<td></td>
<td>Focal distance: 11 mm</td>
</tr>
<tr>
<td></td>
<td>AR-coated range: 600 – 1050 nm</td>
</tr>
<tr>
<td></td>
<td>Clear aperture: 5.7 mm</td>
</tr>
<tr>
<td>CCD lens (L2)</td>
<td>Spectral range: 350 – 1550 nm</td>
</tr>
<tr>
<td></td>
<td>Focal distance: 50 mm</td>
</tr>
<tr>
<td></td>
<td>AR-coated range: 400 – 700 nm</td>
</tr>
<tr>
<td>Collection lens (L3)</td>
<td>Spectral range: 350 – 1550 nm</td>
</tr>
<tr>
<td></td>
<td>Numerical aperture NA: 0.25</td>
</tr>
<tr>
<td></td>
<td>Focal distance: 11 mm</td>
</tr>
<tr>
<td></td>
<td>AR-coated range: 1050 – 1600 nm</td>
</tr>
<tr>
<td></td>
<td>Clear aperture: 5.7 mm</td>
</tr>
<tr>
<td>Beam splitter</td>
<td>Spectral range: 350 – 1550 nm</td>
</tr>
<tr>
<td></td>
<td>Beam splitter cubes: 50 : 50</td>
</tr>
<tr>
<td></td>
<td>non-polarizing</td>
</tr>
<tr>
<td></td>
<td>AR-coated range: 700 – 1100 nm</td>
</tr>
<tr>
<td></td>
<td>BK7 glass</td>
</tr>
<tr>
<td>Vacuum window</td>
<td>Diameter: 25.4 mm</td>
</tr>
<tr>
<td></td>
<td>Thickness: 4.0 mm</td>
</tr>
<tr>
<td></td>
<td>BK7 glass</td>
</tr>
<tr>
<td></td>
<td>No AR coating</td>
</tr>
<tr>
<td></td>
<td>Angle 4°</td>
</tr>
<tr>
<td>Objective</td>
<td>Spectral range: 350 – 1500 nm</td>
</tr>
<tr>
<td></td>
<td>Numerical aperture NA: 0.65</td>
</tr>
<tr>
<td></td>
<td>Focal distance: 2.75 mm</td>
</tr>
<tr>
<td></td>
<td>Working distance: 1.6 mm</td>
</tr>
<tr>
<td></td>
<td>Clear aperture: 3.6 mm</td>
</tr>
<tr>
<td>Collection fiber</td>
<td>Single Mode 830 nm</td>
</tr>
<tr>
<td></td>
<td>Numerical aperture NA: 0.12</td>
</tr>
<tr>
<td></td>
<td>Mode field diameter: 5.6 $\mu$m</td>
</tr>
</tbody>
</table>
CHAPTER 2.

Figure 2.9: Spectra of the Ar calibration light as measured by the Si CCD camera (black line) and the InGaAs array detector (red line). The central wavelength was set to 960 nm, the exposure time is 5 ms and the 750 grooves/mm grating in single mode was used for both detectors. The offset between both spectra is due to a small misalignment between both detectors and can be corrected with the software.

by connecting the fiber head to S4 and single stage experiments are done by connecting the fiber head to slit S6. Each of the turrets (T1-T3) shown in Fig. 2.8 contain three gratings. The first stage has gratings of 750, 1100 and 1800 grooves/mm and the second and third stage have gratings of 750, 900 and 1800 grooves/mm. Operating in single mode with 750 grooves/mm and a focal length of 750 mm gives a 62 μeV/pixel, whereas in triple additive mode with 3 gratings of 1800 grooves/mm results in 6 μeV/pixel (in both cases assuming a pixel size of 25 μm and a wavelength of 900 nm [57]).

The grating images the spectrum onto one of two liquid nitrogen cooled detectors, a 2D Si CCD camera or an InGaAs array, which have detection ranges of 0.35 – 1.1 μm and 0.8 – 1.7 μm, respectively. A comparison of the detection efficiency of both detectors has been made. The measurements were done using the same Ar calibration lamp and the area underneath each of the two peaks in Fig. 2.9 was determined. The ratio of these two areas and the quantum efficiency (QE) of both detectors at 960 nm gives the relative detection efficiency DE(%) = ∫\text{InGaAs} \times \text{QE(CCD)} / ∫\text{CCD} \times \text{QE(InGaAs)} \times 100%. The detection efficiency is the relative number of photons needed for 1 detector count. The Si CCD detector has a quantum efficiency of 55% at 965 nm, while the InGaAs detector has a quantum efficiency of 80% at the same wavelength [58]. The intensities determined from Fig. 2.9 are 2.46 and 25.2 for the InGaAs and CCD, respectively. The relative detection efficiency therefore yields 6.7%. This means that the detection efficiency at 960 nm of the InGaAs array is much lower as compared to the CCD camera, although the QE of the InGaAs is higher at this wavelength. This is a direct consequence of the sensitivity of both detectors. The CCD camera only needs 6 electrons per count whereas the InGaAs detector needs 65 electron per count. Taking this sensitivity into account the detection efficiency is calculated to be
9.2%, slightly higher than the measured value of 6.7%.

### 2.6.3 The solid immersion lens

In order to enhance both the collection efficiency and the spatial resolution a solid immersion lens (SIL) can be mounted on top of the sample. This is a solid hemispherical lens made out of high refractive index material, which is attached to the sample. The high refractive index produces a smaller spot size on the sample surface, schematically shown by Figs. 2.10(a) and 2.10(b). The SILs used in this thesis are made of a glass material (LaSFN9) and have a refractive index of 1.83. The collection efficiency is given by:

$$\eta = \frac{1}{2} \left( 1 - \sqrt{1 - \left( \frac{N_A n_{mat}}{n_s} \right)^2} \right),$$  \hspace{1cm} (2.3)

where $n_{mat}$ is the refractive index of the sample material ($n_{GaAs} = 3.5$) and $n_s$ is the refractive index of the material directly above the host surface. From Eq. 2.3 it follows that the collection efficiency is increased by roughly a factor of $n_{SIL}^2$ [59, 60]. The spot size is reduced by a factor of $n_{SIL}$ as can be deduced from Eq. 2.2 by replacing NA with the effective numerical aperture $\frac{N_A n_{mat}}{n_s}$.

![Figure 2.10: The excitation light (red solid line) from the objective is focussed on the sample and emission from the sample (green dotted line) is collected by the same objective (a) without and (b) with SIL. The spot size with SIL is smaller whereas the collection efficiency is higher.](image)

The mounting of the SIL on the sample is a critical step in the successful use of a SIL. This has to be done carefully to avoid damage, dust and contaminations from affecting the SIL or sample surface. The SIL is attached using a small amount of vacuum grease. The vacuum grease is applied via a fiber end on the side of the flat part of the lens.

Experimentally the spot size with and without SIL is determined on the calibration grating in the interference experiment (Fig. 2.6(a)). The spot size of the system is experimentally best determined by scanning over a step edge of the sample. In Fig. 2.11(a) a line profile of such a scan is shown for a wavelength of 635 nm. The derivative is shown in Fig. 2.11(b) and fitted with a Gaussian beam profile. The Full Width Half Maximum (FWHM) of the spot size without the SIL is 498 ± 30 nm, which compares well to the calculated value of 508 nm using Eq. 2.2. With a SIL having $n_{SIL} = 1.83$, Eq. 2.2 predicts an improvement in resolution from 508 to 278 nm. The experiment with SIL gives the line profile...
CHAPTER 2.

and derivative of the line profile shown in Fig. 2.11(c), (d). The horizontal axis of Fig. 2.11(a) is corrected with the magnification of the SIL and a FWHM of 260 ± 20 nm is found, which is again in good correspondence with the predicted value. The magnification was determined from Fig. 2.10(e) and (f), which shows two images of the grating without and with SIL, respectively. The magnification is a factor of 3.25 with SIL. Note that the images do not show a perfect $2 \times 2 \mu m^2$ chess-pattern due to the non-linearity of the piezos.

2.6.4 The alignment

An accurate alignment of the confocal microscope is crucial in order to perform PL measurements. First a rough alignment of the optical head is performed. This is done by connecting a 635 nm laser via the fiber to the excitation and the detection arm successively. Both beams need to be collimated and aligned such that they are projected on the same position on a screen placed several meters from the optical head. In order to make sure that the beam is on the optical axis a pinhole is used for the alignment of the excitation arm. The adjustment screws of the excitation arm are used to align the beam through the pinhole. After this alignment the pinhole is removed and the collection arm is aligned such that the spot is at the same position as the aligned spot of the excitation arm. The spot size should be $\sim 3.6 \text{ mm}$ in order to match the clear aperture of the microscope objective.

After this first alignment step the optical head is mounted on the microscope stick. The microscope stick is not in the vacuum tube yet as the first rough alignment is done by observing the laser spot directly on the sample. For this purpose the laser is again connected to the excitation arm, and the CCD imaging camera is connected to a monitor. By using the adjustments screws to tilt the complete head (and not the separate arms which are already aligned) the spot is aligned on the objective. Using the $x$- and $y$-positioners the sample is aligned with respect to the spot. By moving the $z$-piezo such that the distance between objective and sample is $\sim 1.56 \text{ mm}$ a focus is observed on the CCD camera.\footnote{When a SIL is mounted on top of the sample two focusses are found. One is from the top of the SIL and one of the sample itself. The SIL needs not to be higher than 1.1 mm to prevent the collision between the SIL and the objective before the second focus is obtained.} The CCD camera is used to align the excitation spot more precisely on the objective. After this step the optical head is detached and the microscope stick is placed in the vacuum tube and prepared for cooling down. Before cooling down the alignment is checked again by mounting the optical head and using the adjustments screws to obtain a nice spot on the CCD camera.

When the system is cooled down the optical head is mounted again and the final alignment is performed. The alignment of the detection arm can be done by connecting the detection fiber to the collection arm and achieving a nice spot on the CCD camera. When the detection arm is aligned the excitation laser is connected to excitation arm and the collection fiber is connected to collection arm. The reflected laser light is then collected, dispersed by the monochromator and detected by either the Si CCD or InGaAs detector. After this step the second beam splitter is removed. The fine tuning of the alignment is done by optimizing the laser intensity by using the adjusting screws on the collection arm, the focus of lens L3 and the $z$-positioner. When the laser intensity is optimized the next step is to optimize the PL intensity. A high-pass filter is
Figure 2.11: Scan of the edge of the sample. (a) The line scan and (b) the derivative of the line scan and Gaussian fit without SIL. The FWHM is $\sim 500 \text{ nm}$ and comparable with the diffraction limited spot size. (c) The line scan and (d) the derivative of the scan and a Gaussian fit with SIL. The FWHM is decreased to $\sim 260 \text{ nm}$. (e) A 2 $\mu\text{m}$ ”chess” grating imaged by scanning the surface without the SIL and (f) with SIL, which reflects the magnification by the SIL.
placed in front of the collection fiber in order to block out laser light. As the objective is chromatic the PL signal is optimized by adjusting the z-piezo. Moreover, the signal is further optimized by adjusting lens L3, which is also chromatic. In some cases there is no PL signal to optimize as the PL wavelength is quite different from the wavelength of the alignment laser. In this case a 1315 nm wavelength laser can be connected to the excitation arm after which this signal is optimized by adjusting both the detection arm, lens L3 and the z-piezo. In general this alignment is sufficient to observe some PL intensity, and after further optimization the experiments can be commenced.

2.7 Summary

A brief overview is given of the two different growth techniques used to grow the samples studied in this thesis. A short overview of the AFM and X-STM techniques for structural analysis are given after which the macro-PL setup is discussed. The emphasis in this chapter is on the confocal microscope used to investigate the magneto-optical properties of single nanostructures. The FWHM of the excitation spot size is determined on 260 nm using a solid immersion lens. The triple monochromator and both detectors are characterized and the alignment procedure of the setup is discussed.
Chapter 3

Quantum dot magnetoluminescence: Zeeman splitting and diamagnetic shift

3.1 Abstract

The main goal of this chapter is to understand the optical properties of individual quantum dots in a magnetic field. For this purpose the quantum dot potential is described by a harmonic oscillator potential. This model is used to introduce the concepts of the exciton $g$-factor $g_{ex}$ and the exciton diamagnetic coefficient $\alpha_d$. This description allows for the analysis of the micro-photoluminescence experiments on a sample containing a low density of InAs/GaAs quantum dots. The experiments show a dependence of $g_{ex}$ on the emission energy, which can be related to both the height and lateral size of the quantum dot.\(^1\)

3.2 The harmonic oscillator model

3.2.1 A single particle picture

In order to gain more insight in the important parameters governing quantum dot physics, a model of the quantum dot potential is needed. A realistic model of a quantum dot potential has to take into account the effects of the composition profile and strain on the band structure of the dot. For this purpose a complicated model is needed. However, most of the QD physics can already be understood by modeling the QD potential by a harmonic oscillator potential, given by:

\(^1\)These results are submitted as: Size dependent exciton $g$-factor in self-assembled InAs/GaAs quantum dots, N. A. J. M. Kleemans, J. van Bree, R. van Voornveld, G. J. Hamhuis, R. Nötzle, A. Yu. Silov, and P. M. Koenraad, (2009).
\[ V_{QD} = \frac{1}{2} m^* (\omega_x^2 x^2 + \omega_y^2 y^2 + \omega_z^2 z^2), \tag{3.1} \]

where \( m^* \) is the effective electron or heavy hole mass and \( \omega_x, \omega_y \) and \( \omega_z \) are the corresponding oscillator eigenfrequencies in the lateral (\( x, y \)) and growth (\( z \)) direction. In reality there is a smooth and continuous transition of the quantum dot potential to the band structure of the surrounding barrier material. Especially in quantum dots having a shallow QD potential the higher energy states can deviate from the ones obtained by the harmonic oscillator model.

The Hamiltonian describing the harmonic oscillator in a magnetic field is given by:
\[ \mathcal{H} = \frac{1}{2m^*} (p - qA)^2 + V_{QD}, \tag{3.2} \]
where \( p \) is the momentum operator, \( q \) is the electric charge, and \( A \) is the vector potential corresponding to a magnetic field \( B = \nabla \times A \). Let us first consider the solution of the Schrödinger equation \( \mathcal{H} \psi_i = E_i \psi_i \) for \( B = 0 \). The eigenenergies \( E_i \) and corresponding wave function \( \psi_i \) are given by [11]:
\[ E_i = (n_x + \frac{1}{2}) \hbar \omega_x + (n_y + \frac{1}{2}) \hbar \omega_y + (n_z + \frac{1}{2}) \hbar \omega_z, \tag{3.3} \]
\[ \psi_i(r) = \phi^{n_x}_x(x) \phi^{n_y}_y(y) \phi^{n_z}_z(z), \tag{3.4} \]
where \( i \) is a label containing the quantum numbers \( n_x, n_y \) and \( n_z \), \( r \) is the coordinate vector, and where
\[ \phi^{n_\alpha}_\alpha(\alpha) = \left( \frac{m^* \omega_\alpha}{\hbar} \right)^{\frac{1}{4}} \frac{1}{\sqrt{2^{n_\alpha} n_\alpha !}} e^{-\frac{m^* \omega_\alpha}{2} \sqrt{\frac{m^* \omega_\alpha}{\hbar}}} H_{n_\alpha}(\alpha \sqrt{\frac{m^* \omega_\alpha}{\hbar}}), \tag{3.5} \]
\[ H_{n_\alpha}(\xi) = (-1)^{n_\alpha} e^{\xi^2} \frac{d^n}{d\xi^n} e^{-\xi^2}, \tag{3.6} \]
where \( \alpha = x, y, \) or \( z \), and \( H_{n_\alpha}(\xi) \) Hermite polynomials of order \( n_\alpha \). A one-dimensional Harmonic potential with oscillator frequency \( \omega \) and its corresponding eigenenergies and wave functions are shown in Fig. 3.1(a). The energies are equidistant with a splitting of \( \hbar \omega \).

For a realistic dot the height is typically \( \sim 5 \text{ nm} \), which is several times smaller compared to the typical lateral extension \( \sim 30 \text{ nm} \). Consequently, the quantization energy in a quantum dot is mainly determined by \( \omega_z \). Therefore the dot can be seen as quasi two-dimensional, with \( \omega_z \gg \omega_{x,y} \). In general QDs with smaller sizes will lead to larger oscillator frequencies and therefore to larger confinement energies. This can be understood by analyzing the typical decay length. The wave function for \( n = 0 \), which is the ground state, has a Gaussian shape and decays in the host material. The typical lateral extension length \( l_x \) is given by:
\[ l_x = \sqrt{2 \hbar / m^* \omega_x}, \tag{3.7} \]

One can directly see that when \( l_x \) decreases, \( \omega_x \) increases. Figure 3.1(b) shows schematically the typical extension lengths: \( l_z \approx l_z \approx 30 \text{ nm} \). Assuming an effective mass of the electron (hole) of \( m^*_e = 0.052 \) (\( m^*_h = 0.0654 \)), corresponding to
Figure 3.1: (a) A one-dimensional harmonic potential with oscillator frequency $\omega$, eigenenergies $E_n = \frac{1}{2} \hbar \omega + n \hbar \omega$ and the corresponding wave functions. (b) As the typical length scale in the $z$-direction $l_z$ is smaller compared to the lateral length scales $l_{x,y}$, the confinement energy is mainly determined by $\hbar \omega_z$.

bulk In$_{0.7}$Ga$_{0.3}$As, the typical confinement energies for the electron (hole) in the growth direction and lateral directions are $\hbar \omega_{e}^z = 116$ meV ($\hbar \omega_{h}^z = 94$ meV) and $\hbar \omega_{e}^{x,y} \sim 3$ meV ($\hbar \omega_{h}^{x,y} \sim 3$ meV), respectively.

3.2.2 A single particle picture in the presence of a magnetic field

In classical mechanics a current $I$ traversing a trajectory with area $A$ gives rise to a magnetic moment $\mu = I \times A$ [61]. In the presence of an external magnetic field $B$, the magnetic moment couples to the magnetic field. In order to minimize the total magnetic energy of the system $E_{\text{Magnetic}} = -\mu \cdot B$, the magnetic moment tends to align parallel to the direction of the magnetic field. In a similar manner, an electron orbiting in a crystal or atom couples to the magnetic field by its orbital momentum. The orbital motion of the electron results in the diamagnetic behavior as discussed in the following subparagraph. Moreover, the spin of the electron is the intrinsic magnetic moment of the electron and couples also to the external magnetic field resulting in the Zeeman splitting between spin up and spin down electrons as is discussed in the second subparagraph.

(A) The diamagnetic shift

The solution for the energy levels in Eq. 3.2 for an harmonic oscillator in the presence of a magnetic field (along the $z$-direction) is given by the Fock-Darwin energies [62, 63]:

$$E_{\ell,m_z,n} = (2\ell + 1 + |m_z|)\hbar \sqrt{\left(\frac{\omega_c}{2}\right)^2 + \omega_{x,y}^2} + \frac{m_z}{2} \hbar \omega_c + (n + \frac{1}{2})\hbar \omega_z, \quad (3.8)$$

where $\omega_c = \frac{qB}{m^*}$ is the cyclotron frequency, $\ell$ is the angular momentum quantum number, $m_z$ is its projection along the $z$-axis and $n$ corresponds to the quantization in the harmonic oscillator potential in the $z$-direction. Effectively, the magnetic field couples to the angular momentum of the charge carrier. Throughout this thesis the quantum dot states with different $\ell$ are labeled analogous to the atomic orbital states as shown in table 3.1.
Table 3.1: The quantum dot states with different angular momentum $\ell$ are labeled analogous to the atomic orbital states. Note that due to the large quantization energy along the $z$-axis the $p_0$ state in a quantum dot is not considered.

<table>
<thead>
<tr>
<th>shell</th>
<th>$\ell$</th>
<th>$m_z$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s$</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$p_{\pm}$</td>
<td>1</td>
<td>$\pm 1$</td>
</tr>
<tr>
<td>$d_0$</td>
<td>2</td>
<td>0</td>
</tr>
<tr>
<td>$d_{\pm}$</td>
<td>2</td>
<td>$\pm 2$</td>
</tr>
</tbody>
</table>

It is instructive to analyse the ground state $E_{0,0,0} = \hbar \sqrt{\left(\frac{\omega_c}{2}\right)^2 + \omega_{x,y}^2} + \frac{1}{2} \hbar \omega_z$. In general, for small $B$ we have $\omega_c < \omega_{x,y}$. Using a Taylor expansion we find:

$$E_{0,0,0} = \hbar \omega_{x,y} + \frac{\hbar \omega_c^2}{4 \omega_{x,y}} + \frac{1}{2} \hbar \omega_z. \quad (3.9)$$

From this equation we deduce that the major contribution of a magnetic field on the energy levels is an additional parabolic confinement in the lateral plane, resulting in an increase of the ground state energy proportional to $\omega_c^2 \sim B^2$. This quadratic dependence on the magnetic field is known as the diamagnetic shift. Using Eq. 3.7 the ground state energy as function of $B$ can be rewritten as:

$$E_{0,0,0}(H) = E_0 + \frac{e^2 B^2 l_y^2}{8m^*} = E_0 + \alpha_d B^2, \quad (3.10)$$

where $E_0$ is the ground state energy at $B=0 \, \text{T}$ and $\alpha_d$ is the diamagnetic coefficient, which is proportional to $l_y^2$. Quantum dots having a large lateral size $l_{x,y}$, and thus small $\omega_{x,y}$, will have a larger increase in energy as compared to quantum dots having a smaller lateral extend. Assuming an In$_{0.7}$Ga$_{0.3}$As QD of 30 nm diameter, we obtain for the diamagnetic coefficient of the electron $\alpha_d \sim 31 \, \mu eV/\text{T}^2$. However, as we will show later on, typically we find for the diamagnetic shift of an exciton $\alpha_d \sim 8 \, \mu eV/\text{T}^2$. The difference is due to the different mass of the exciton as well as due to the fact that we ignore electron-hole interaction. Clearly, the lateral extend of the exciton is smaller than the lateral extend of the quantum dot.

The magnetic length is introduced [64] in order to compare the lateral confinement energy to the magnetic confinement energy. The magnetic length is given by $l_B = \frac{\hbar}{\sqrt{|eB|}}$, which corresponds to $\sim 26 \, \text{nm}$ at $B = 1 \, \text{T}$. As a result of the additional magnetic confinement the effective length $l_{x,y}$ is decreased. The effective length in a magnetic field can be obtained by replacing $\omega_{x,y}$ in Eq. 3.7 by $\omega' = \sqrt{\omega_{x,y}^2 + \left(\frac{\omega_c}{2}\right)^2}$. 

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(B) The Zeeman splitting

In the previous paragraph the coupling of $B$ to the orbital motion of the charge carriers is described. The charge carriers also posses a spin $s$, resulting into an additional magnetic moment which couples to $B$. This interaction is known as the Zeeman interaction.\(^2\) The magnetic moment $\mu_s$ of a free charge particle is related to the spin via \[65\]:

$$\mu_s = g_0 \left( \frac{q}{2m} s \right).$$ \hfill (3.11)

where $g_0$ is the gyromagnetic ($g$) factor. For a free electron $g_0 = +2.0023 \ [65]$, and we introduce the Bohr magneton by $\mu_B = \frac{e}{2m} = +0.0579 \text{ meV/T}$. The Zeeman Hamiltonian for the spin of a free electron is given by:

$$\mathcal{H}_{\text{Zeeman}} = \mu_s \cdot B = g_0 \mu_B s \cdot B.$$ \hfill (3.12)

Up to now we have treated only free electrons. For electrons in a semiconductor crystal Eq. 3.12 is still valid, but the free electron $g$-factor $g_0$ is replaced by an effective $g$-factor taking into account the band structure of the crystal. For the case of an electron confined to a quantum dot numerical calculations are required \[66\]. Nevertheless, it is important to notice that the Zeeman energy splitting evolves linear with $B$ and is given by:

$$\Delta E_{\text{Zeeman}} = g_e \mu_B B,$$ \hfill (3.13)

as electrons have spin $s = 1/2$.

For holes the situation is different. The hole states are formed from bulk valence band states, which have a $p$-character \[67\]. Therefore the valence band of any III-V semiconductor experiences spin-orbit coupling\(^3\): the orbital motion of a hole induces an effective magnetic field to which its own spin can couple. This causes the spin to be no longer a proper quantum number. Nevertheless, the total angular momentum $J = L + s$, is still a good quantum number. The valence band has $L = 1$, meaning that $J = 3/2$ or $J = 1/2$. The latter ones are the split-off states and can be neglected here; the former ones are the heavy hole $J_z = \pm 3/2$ and light hole $J_z = \pm 1/2$ states. It can be shown \[64\] that the confined state is dominantly of heavy hole character, so that the pseudospin of the hole is $J_z = \pm 3/2$ \[68\] and the hole state is then formally the same as an electron. For holes in nanostructures the situation is even more complicated \[69\]. However, it can be shown that the Zeeman Hamiltonian is still valid: every quantum dot state, both electron and hole, will experience a Zeeman shift with their own characteristic $g$-factor. Actually it can be shown that Zeeman splitting for holes is not perfectly linear, but also contains a small quadratic dependence on the magnetic field (see chapter 6), which is a result of the heavy hole-light hole mixing. However, for the nanostructures studied in this thesis the quadratic dependence is negligible.

\(^2\)In fact, the Zeeman interaction is also the origin of the splitting of the nonzero angular momentum states in a magnetic field. The nomenclature in this thesis is such that the Zeeman interaction only takes into account the spin of the particle.

\(^3\)The bulk conduction band states have an $s$-character. The $s$ and $p$ character are present in the Bloch part of the wave function. We note that although the envelope wave function of electrons (holes) can have a $p$-like ($s$-like) character the Bloch part of the wave function stays $s$-like ($p$-like). The spin-orbit coupling only works on the Bloch part of the wave function and therefore is only present for the holes.
CHAPTER 3.

In table 3.2 the electron $g$-factors $g_e$ are given for different semiconductor compounds. It is directly clear that these differ substantially among the different materials and from the free electron $g$-factor. Moreover, the confinement in nanostructures will alter the values considerably as compared to the given bulk values. For the hole $g$-factors $g_h$ there is still much ambiguity and no reliable values can be given. More details on the calculations of both electron and hole $g$-factor can be found in chapter 6.

Table 3.2: The electron $g$-factor $g_e$ for InAs, GaAs and InP. The free electron $g$-factor $g_0 = +2$. The values are taken from Ref. [67].

<table>
<thead>
<tr>
<th>material</th>
<th>$g_e$</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs</td>
<td>-0.44</td>
</tr>
<tr>
<td>InAs</td>
<td>-14.6</td>
</tr>
<tr>
<td>InP</td>
<td>+1.26</td>
</tr>
</tbody>
</table>

3.2.3 The Coulomb and exchange interaction

Up to now the situation of a single particle in a quantum dot was considered. When more than one charge carrier is present in the dot, the energy structure of the quantum dot is perturbed by the Coulomb and exchange interaction between the different charges. In order to treat these interactions as a perturbation, the quantization energies of the electrons ($\hbar\omega_e$) and holes ($\hbar\omega_h$) need to be larger as the Coulomb and exchange energies. Typical values for the electron quantization energy have been measured to be 50 meV [70], whereas from capacitance measurements the Coulomb interaction is estimated to be $\sim 10 - 20$ meV [71, 72] and exchange interactions are an order of magnitude smaller. This implies that the Coulomb and exchange effects can be treated as perturbations to the single particle energies. This model is described in Ref. [12] in detail, and only the basic results will be discussed here using a system of two electrons.

The Coulomb interaction between two particles is given by:

$$E_{ij}^C = \frac{e^2}{4\pi\epsilon_0\epsilon_r} \int \left| \frac{\psi_i^{e(h)}(\mathbf{r}_1)\psi_j^{e(h)}(\mathbf{r}_2)}{|\mathbf{r}_1 - \mathbf{r}_2|} \right|^2 d\mathbf{r}_1 d\mathbf{r}_2, \quad (3.14)$$

whereas the exchange interaction is give by:

$$E_{ij}^x = \frac{e^2}{4\pi\epsilon_0\epsilon_r} \int \left| \frac{\psi_i^{e(h)}(\mathbf{r}_1)^* \psi_j^{e(h)}(\mathbf{r}_2)^* \psi_i^{e(h)}(\mathbf{r}_2) \psi_j^{e(h)}(\mathbf{r}_1)}{|\mathbf{r}_1 - \mathbf{r}_2|} \right|^2 d\mathbf{r}_1 d\mathbf{r}_2. \quad (3.15)$$

The states $\psi_i^{e(h)}(\mathbf{r}_1)$ and $\psi_j^{e(h)}(\mathbf{r}_2)$ are the single electron (hole) states in orbitals $i$ and $j$. The Coulomb interaction comprises the electrostatic interaction between the charges. The exchange interaction is a pure quantum mechanical
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...resulting from the anti-symmetrization principle; the many-particle state must comply with the anti-symmetrization principle, demanding that under change of spatial and spin-coordinates, the total wave function is anti-symmetric [11]. To illustrate the influence of the exchange effects, consider a system of two particles 1, 2 in orbits \( \ell = i, j \):

\[
\Psi(r_1 s_1, r_2 s_2) = \frac{1}{\sqrt{2}} [\psi_i(r_1)\psi_j(r_2) \pm \psi_j(r_1)\psi_i(r_2)] \chi, \tag{3.16}
\]

where the many-particle state \( \Psi \) is a product of a linear combination of the single particle states \( \psi_i \) and \( \psi_j \) and the two particle spin wave function \( \chi \). For fermions (bosons) the minus (plus) sign is used. As electrons and holes are fermions only the minus sign in Eq. 3.16 is considered. If both particles are in the same state \( i = j \), then the two-particle orbital wave function is symmetric and the corresponding two particle spin wave function must be anti-symmetric. This gives the singlet spin wave function, with the spins anti-parallel:

\[
\chi_{\text{singlet}} = \frac{1}{\sqrt{2}} (| \uparrow, \downarrow \rangle - | \downarrow, \uparrow \rangle), \tag{3.17}
\]

using the spin states \( | s_z, 1, s_z, 2 \rangle \), where \( s_z \) is the spin-projection along the \( z \)-axis and where \( | \uparrow \rangle \) (\( | \downarrow \rangle \)) is spin up (down). For states with \( i \neq j \), the two-particle spin wave function can be either symmetric or anti-symmetric. The symmetric two-particle spin wave function is a triplet, with spins parallel:

\[
\chi_{\text{triplet}} = \begin{cases} 
| \uparrow, \uparrow \rangle & \frac{1}{\sqrt{2}} (| \uparrow, \downarrow \rangle + | \downarrow, \uparrow \rangle) \\
| \downarrow, \downarrow \rangle
\end{cases} \tag{3.18}
\]

The energy splitting between the singlet and triplet states is related to the transition probability to go from the state \( \psi_i(r_1)\psi_j(r_2) \) to \( \psi_j(r_1)\psi_i(r_2) \), e.g. (direct) exchanging the two electrons 1, 2 among the states \( i \) and \( j \). Generally, exchange is capable of lifting the spin degeneracy. This becomes even more clear by representing the exchange term between two electrons in the Heisenberg form [73]:

\[
H_{\text{Heisenberg}} = J_{i,j} s_i \cdot s_j, \tag{3.19}
\]

where \( s_i \) are the spin of the particles and \( J_{i,j} \) is the exchange coupling constant.

By evaluating the integrals of Eqs. 3.14 and 3.15, the energy of the many particle state formed from the single particle energies by the quantum dot potential, can be corrected to first order by including these terms. The energy of the state where an exciton is present in the quantum dot, consists out of the single particle energies of the electron and hole, plus the Coulomb binding energy and the electron-hole exchange energy. The Coulomb and exchange energies are listed in Ref. [12] for both electron-electron and electron-hole interactions for orbitals \( i \) up to 2.
3.3 Excitonic states and magnetophotoluminescence

As explained in chapter 1, in a PL experiment an electron is excited from the valence band to the conduction band upon optical excitation. The electron and hole form an exciton through their Coulomb interaction. The light that is emitted from the quantum dot upon recombination of an electron and hole, i.e. the annihilation of an exciton, provides information about the confinement. Applying a magnetic field allows for analyzing the spin properties of excitons in the semiconductor material by measuring the magnetoluminescence. The optical emission is governed by the optical selection rules [74]. Here we consider the neutral exciton $X^0$, which consists of one electron-hole pair. The spin of the electron (heavy hole) is $s_z = \pm \frac{1}{2}$ $(J_z = \pm \frac{3}{2})$. Therefore there are four different possible spin configurations of $X^0$, which are labeled by $|X, X_z \rangle$ with $X$ is the exciton spin ($|s_z + J_z \rangle$) and $X_z$ is the exciton spin projection along the $z$-axis:

$$
| \uparrow \rangle | \uparrow \rangle = |1, +1 \rangle \\
| \uparrow \rangle | \downarrow \rangle = |1, -1 \rangle \\
| \downarrow \rangle | \uparrow \rangle = |2, +2 \rangle \\
| \downarrow \rangle | \downarrow \rangle = |2, -2 \rangle,
$$

where $| \uparrow \rangle$, $| \downarrow \rangle$ represents the electron spin and $| \uparrow \rangle$, $| \downarrow \rangle$ represent the heavy hole pseudo-spin.\(^4\) The optical selection rules in the dipole approximation only permit the $X = 1$ states to be optically active, as a photon carries away a spin $\pm 1$. The $X = 1$ states are therefore called the bright states, and the optical inactive exciton states $X = 2$ are called dark states. Moreover, from these rules, it can be inferred that the photoluminescence of an $X = 1$ exciton is either right ($\sigma^+$) handed circular polarized or left ($\sigma^-$) handed circular polarized for the $|1, +1 \rangle$ and $|1, -1 \rangle$, respectively, as depicted in Fig. 3.2(a).

The Heisenberg Hamiltonian Eq. 3.19 shows that the exchange Hamiltonian can be written in terms of spin. However, it tacitly assumed rotational invariance, which might not be true for quantum dots. Relaxing this assumption, the exchange Hamiltonian becomes [68, 75]:

$$
\mathcal{H}_{\text{exchange}} = - \sum_{\alpha=x,y,z} (a_\alpha s_\alpha J_{h,\alpha} + b_\alpha s_\alpha J_{h,\alpha}^3) \simeq -a_z s_z J_{h,z} - \sum_{\alpha=x,y,z} b_\alpha s_\alpha J_{h,\alpha}^3,
$$

where in the second step the linear term in $J_x$ and $J_y$ are omitted, as we neglect heavy hole-light hole mixing [75]. The matrix representation of this Hamiltonian in the basis of $|1, +1 \rangle$, $|1, -1 \rangle$, $|2, +2 \rangle$, and $|2, -2 \rangle$ is:

$$
\mathcal{H}_{\text{exchange}} = \frac{1}{2} \begin{pmatrix}
\delta_0 & \delta_1 & 0 & 0 \\
\delta_1 & \delta_0 & 0 & 0 \\
0 & 0 & -\delta_0 & \delta_2 \\
0 & 0 & \delta_2 & -\delta_0 \\
\end{pmatrix}.
$$

Here the following abbreviations have been introduced:

$$
\delta_0 = 1.5(a_z + 2.25b_z)
$$

\(^4\)The valence state as given by the hole state with $J_z = \pm 3/2$ is equivalent to the representation of the valence state by electrons with $s_z = \mp 1/2$.\(\)
Figure 3.2: (a) Excitons in a single particle picture at zero magnetic field and (b) as function of magnetic field. In the single particle picture an electron (blue) and hole (green) are confined to the ground states of the quantum dot. As function of magnetic field, these states split and shift differently for the electron and hole. Here we take \( g_e(h) \), \( \mu_B > 0 \). In the exciton picture, an interacting electron-hole pair is confined by the quantum dot. (c) At zero magnetic field the fourfold degenerate ground state can be split and mixed by exchange. (d) As function of the magnetic field, these mixed states shift and split further. Only the bright excitons are luminescent and annihilate upon emission of a photon.

\[
\begin{align*}
\delta_1 &= 0.75(b_x - b_y) \\
\delta_2 &= 0.75(b_x + b_y)
\end{align*}
\]

The first term of \( H_{\text{exchange}} \) of Eq. 3.21 gives rise to the diagonal matrix elements \( \delta_0 \), while the second term gives the off-diagonal terms \( \delta_{1,2} \). The matrix has a block-diagonal form. Therefore, the bright and dark exciton states do not mix. In the absence of rotational symmetry \( (b_x \neq b_y) \), which is the case for realistic quantum dots, the off-diagonal terms in the sub-blocks give rise to the mixing of the bright states, whereas the dark states are always mixed. The splitting between the bright states is known as anisotropy splitting or fine structure splitting (FSS) and is schematically shown in Fig. 3.2(c). In general, the splitting \( \delta_0 \) is much larger than the \( \delta_{1,2} \) splitting since the latter splittings are proportional to \( J_{2\alpha} \). A more extensive description is given in Ref. [68], in which they also state the energies and eigenstates for different dot symmetries.

There has been discussion lately on the origin of the FSS. In the past it was assumed to be caused mainly by the breaking of the in-plane symmetry by for example an elongation of the QD. In the case of InAs on GaAs the indium atoms
diffuse preferentially in the (1¯10) direction [76] causing the elongation of nanostructures in this direction. However, the breaking of the in-plane symmetry is not the only contribution to the FSS. The piezo-electric effect introduced by the lattice mismatch between different materials is also considered to be of major importance for the FSS [77, 78]. Due to the strain in the lattice mismatched materials the atoms are slightly displaced from their relaxed positions creating an effective electric field. This piezo-electric field interacts with the electron and hole and can thereby enhance the QD anisotropy. Moreover it has been discussed that the crystal field inside the quantum dot contributes to the FSS considerably [79]. Lately there has been work by Abbarchi et al. [80], in which GaAs quantum dots are grown on AlGaAs using droplet epitaxy. As these dots were grown lattice matched the effect of the piezo-electrical field on the FSS could be eliminated and therefore the FSS of these dots was totally subscribed to the elongation of these dots. The absence of the FSS is of crucial importance for the production of entangled photon sources [26], and many studies are performed to discover the origin of the FSS and a way to control it.

To summarize (see Fig. 3.2), the energy of an exciton is the sum of the single particle energies of the electron $E_e$ and hole $E_h$, corrected by the Coulomb attraction. The electron-hole exchange interaction will split at zero magnetic field the fourfold degenerate exciton state into the twofold degenerate bright and dark excitons. The twofold degenerate dark exciton states are always mixed, leading to a splitting between the resulting eigenstates. The twofold degenerate bright excitons can also be mixed in quantum dots lacking axial symmetry, resulting in the fine structure splitting. Neglecting the fine structure splittings, application of a magnetic field (Fig. 3.2(c)) results in a diamagnetic shift with a diamagnetic coefficient $\alpha_d$ and a Zeeman splitting proportional to the exciton $g$-factor $g_{ex}$:

$$E_{X=1}(B) = E_{X=1} \pm \frac{1}{2} g_{X=1}\mu_B B + \alpha_d B^2$$  \hspace{1cm} (3.26)

$$E_{X=2}(B) = E_{X=2} \pm \frac{1}{2} g_{X=2}\mu_B B + \alpha_d B^2$$  \hspace{1cm} (3.27)

Note that the diamagnetic shift is assumed to be the same for the dark ($X = 2$) states and the bright ($X = 1$) states. Neglecting the Coulomb and exchange effects, the diamagnetic shift $\alpha_d$ and $g$-factors are related to the single particle states by:

$$E_{X=1} \approx E_{X=2} \equiv |E_e - E_h|,$$  \hspace{1cm} (3.28)

$$\alpha_{ex} \equiv \alpha_d = \alpha_e - \alpha_h,$$  \hspace{1cm} (3.29)

$$g_{X=1} \equiv g_{ex} = \frac{E_{\sigma^+} - E_{\sigma^-}}{\mu_B B} = g_h - g_e,$$  \hspace{1cm} (3.30)

$$g_{X=2} = \frac{E_{\uparrow\uparrow} - E_{\downarrow\downarrow}}{\mu_B B} = g_h + g_e,$$  \hspace{1cm} (3.31)

where $\alpha_e$, $\alpha_h$, $g_e$ and $g_h$ are the diamagnetic coefficients and $g$-factors of the single particle states. The energy difference between $|1, +1\rangle$ and $|1, -1\rangle$ is the exciton Zeeman energy $\Delta E_{Zeeman}$:

$$\Delta E_{Zeeman} = E(\sigma^+) - E(\sigma^-) = g_{ex}\mu_B B,$$  \hspace{1cm} (3.32)
where $E(\sigma^+)\ (E(\sigma^-))$ is the energy of the state $|1, +1\rangle\ (|1, -1\rangle)$ with right (left) handed polarization, and where we defined $\mu_B > 0$. The exciton diamagnetic shift is given by:

$$\alpha_d = \left(\frac{E_{\sigma^+} + E_{\sigma^-}}{2} - E_0\right)B^2 = \Delta E_{\text{dia}}B^2,$$

where $E_0$ is the emission energy at $B=0$ T. Using the above definitions of $\alpha_d$ and $g_{ex}$ allow for the analysis of the magnetoluminescence of single quantum dots.

As was already stated in the previous paragraphs, the heavy hole-light hole coupling is neglected. In general, the heavy hole and the light hole band are separated considerably due to the strain and confinement of the QD. Nevertheless, heavy hole and light hole mixing may still play an important role [72] as strain also introduces mixing of the heavy and light hole states. In these cases a more extensive description of the quantum dots is needed [9]. Nevertheless, the simple model as described in this section is successful in explaining the majority of the results on the magnetoluminescence of quantum dots.

### 3.4 Experimental results

In the previous paragraph the general properties of quantum dot states and the excitonic behavior in magnetic field were discussed. In this section we discuss the experiments performed on a low density InAs/GaAs quantum dot sample [38] and analyse the results using the model and definitions of the first section of this chapter.

#### 3.4.1 Sample description and characterization

The sample under investigation is sample R80 and contains a low density of InAs/GaAs quantum dots grown by MBE.\(^5\) The structure is grown on an undoped GaAs substrate. The substrate temperature was set on 580 °C after which it was overgrown by a 200 nm layer of GaAs to obtain a flat GaAs surface. The temperature was decreased to 530 °C and a layer of InAs quantum dots was grown. As the temperature of the quantum dot growth is relatively high and the amount of InAs deposited was relatively low the quantum dot density is low. In fact, right after the RHEED pattern indicated the transition from layer-by-layer growth to island formation (corresponding to 1.7 ML of InAs) the growth of the quantum dots was stopped. The quantum dots are overgrown with a 20 nm layer of GaAs after which the substrate temperature is raised again to 580 °C being followed by a 80 nm thick layer of GaAs. Finally, in order to analyze the quantum dot sizes and density by AFM a quantum dot layer has been grown at the top layer of the sample applying the same growth conditions as for the capped quantum dots.

An AFM image of the surface dots of R80 is shown in Fig. 3.3. From this AFM image it is inferred that the quantum dot density is $5 \cdot 10^9$ cm$^{-2}$. The sizes of the quantum dots are typically 70 nm in diameter and about 5 nm in

\(^5\)This sample has been grown by T. Mano, G. J. Hamhuis, and R. Nötz in the group Photonics and Semiconductor Nanophysics at the Eindhoven University of Technology.
height. The quantum dots have on average an isotropic shape as there is no preferential elongation along the crystal axes.

In order to determine the optical emission energy and quality of the quantum dots, the photoluminescence of an ensemble of these dots is analyzed with the macro-PL setup described in chapter 2. The photoluminescence at a temperature of $T=5\,\text{K}$ is shown in Fig. 3.4(a). The emission energy peak $E_1$ is around $1.281\,\text{eV}$, and the Gaussian broadening of this PL peak is due to the inhomogeneous size distribution of the quantum dots. Two shoulders are present at the high energy side of the distribution. Figure 3.4(a) shows the ensemble PL profile. The profile is fitted well using three Gaussians. From this fit the energy positions $E_{2,3}$ of the shoulders are determined: $E_2 = 1.325\,\text{eV}$ and $E_3 = 1.356\,\text{eV}$. Figure 3.4(b) shows the PL emission as function of excitation density. At $1.435\,\text{eV}$ the PL of the WL is observed. The neutral density (ND) value corresponds to a excitation density reduction of $10^{3\text{ND}}$. For a power decrease of three orders of magnitude the peak does not alter its shape. Therefore the shoulders are attributed to quantum dots having a different height $[81, 82]$. In general, samples having a low quantum dot density will display a multimodal height distribution of the quantum dots; dots positioned in a region with a large inter-dot distance will be higher due to the surface migration of the relatively more indium atoms towards these dots as compared to quantum dots in a region with a smaller inter-dot distance. The inset in Fig. 3.4(b) shows the spectra with (solid black line) and without (dashed red line) an additional focussing lens. The focus lens increases the excitation density. There are two major differences between the two spectra. Firstly, there is a larger PL contribution to the high energy side of the spectrum. This PL is resulting from the higher excited states in the dots, i.e. the transition between an electron and a hole both in the $p$-state of the QD. Secondly, the relative contribution of the QD PL as compared to the WL PL is increased drastically. This is probably due to the
Figure 3.4: (a) Two additional shoulders are present in the spectra corresponding to quantum dots of different height. The spectra at $T=5$ K can be fitted with three Gaussians from which the peak positions are extracted. (b) The photoluminescence of a large ensemble of InAs/GaAs quantum dots of sample R80 at $T=5$ K for different excitation powers. The wetting layer is luminescent at 1.435 eV. The inset shows the PL spectra for high (red dashed line) and medium excitation density (solid line). The arrow indicates the increase of the high energy shoulder due to the PL emission of excited states in the quantum dots.
Figure 3.5: Macro-PL spectra of R80 as function of temperature. For increasing temperature the WL states disappear first as a result of thermal redistribution. Further redistribution causes the QD PL peak to become more symmetric as shown in the inset by comparing the peak shape at $T = 5\, \text{K}$ (black line) and $100\, \text{K}$ (red line). The spectra are shifted on top of each other for easy comparison. The shift of the PL to higher energy is caused by the temperature dependence of the band gap.

contribution of the higher excited states to the QD PL. The WL is only a thin layer, which does not have bound $p$-states.

To further investigate the multimodal height distribution of the QD ensemble, the PL has been measured for different temperatures. For $T > 40\, \text{K}$ the PL of the WL vanishes. From the WL intensity behavior as function of temperature, a thermal activation energy of $E_a = 9\, \text{meV}$ is obtained via the Arrhenius plot in Fig. 3.6(a). The activation energy is determined from $\ln(\text{Intensity}) = -\frac{E_a}{k_B T} + \text{constant}$, where $k_B = 8.62\, \mu\text{eV}/\text{K}$ is the Boltzmann constant. The activation energy of $9\, \text{meV}$ is the energy needed for the carriers to thermally escape the potential fluctuations in the WL, after which they are captured in the more strongly confined QDs.

For temperatures up to $T = 20\, \text{K}$ the QD PL intensity increases. Charge carriers confined to the WL, shallow donors and acceptors, and defects in the GaAs matrix, thermally escape and populate the QDs resulting in an increased emission. For $T > 20\, \text{K}$ the PL intensity decreases again. For increasing temperature the excitons in the QDs are able to thermally redistribute over the
Figure 3.6: (a) The Arrhenius plot of the WL intensity (log scale) versus $\frac{1}{k_B T}$. For $T > 20$ K carriers thermally escape from the WL towards the more confined quantum dot states. From the fit the activation energy $E_a = 9$ meV is determined. (b) The temperature dependence of the band gap of GaAs (filled circles) and InAs (filled triangles) obtained from the Varshni equation [83]. The QD PL energy as function of temperature follows the band gap change for temperatures up to 100 K. The band gap of InAs is given an offset of 1 eV.
QDs of different height in a similar way as the excitons of the WL are thermally redistributed over the more strongly confined QDs at lower temperatures. QDs of smaller height will have a stronger confinement and thermally activated carriers of the more shallow quantum dots will be captured by these QDs. This is in agreement with the observations. The shoulders at higher energy, corresponding to the emission of low dots, disappear and the peak shape becomes symmetric (see inset Fig. 3.5).

The peak positions are also dependent on the temperature. This is caused by the temperature dependence of the band gap of the semiconductors: at higher temperature, the atomic vibrations are larger and the semiconductor materials expand, leading to a larger inter atomic spacing. This results in a wider effective crystal potential, causing the spacing between the energy bands to become smaller, thus decreasing the band gap. The relationship between band gap energy and temperature can be described by Varshni’s empirical expression [83]:

\[ E_g(T) = E_g(0) - \frac{\alpha T^2}{T + \beta}, \]  

(3.34)

where \( E_g(0) \) is the band gap at \( T = 0 \) K, and \( \alpha \) and \( \beta \) are material constants. The values for the different semiconductor compounds can be found in Refs. [83, 84]. For GaAs the values are \( E_g(0) = 1.5216 \text{eV}, \alpha = 8.871 \times 10^{-4} \text{eV/K}, \) and \( \beta = 572 \text{K} \) and for InAs the values are \( E_g(0) = 0.426 \text{eV}, \alpha = 3.158 \times 10^{-4} \text{eV/K}, \) and \( \beta = 93 \text{K} \). Figure 3.6(b) shows the temperature dependence of the GaAs and InAs band gap and the QD PL peak. The band gap of InAs is given an offset of 1 eV. As can be observed, the peaks of the photoluminescence follow roughly the band gap dependence of InAs and GaAs for temperatures up to 100 K. For higher \( T \) the QD confinement is decreasing relatively faster compared to the bulk band gaps. These deviations are most likely related to the strain present in the QD system, as the material constants \( \alpha \) and \( \beta \) are strain dependent. The present of strain thus prevents an one-on-one relation between the band gap and the energy levels in a quantum dot.

### 3.4.2 Single quantum dot photoluminescence

The micro-PL experiments are performed using the confocal microscope setup and a SIL was mounted on top of the sample. The excitation is provided by a 635 nm laser diode and the light is analyzed with the triple monochromator in single stage mode with a grating of 1100 grooves/mm. Three \( \mu \)-PL spectra recorded for three different powers are shown in Fig. 3.7(a), displaying the PL of the WL states and the QDs. For the highest excitation density the \( p \)-states appear, indicated by the arrow. For higher excitation powers there also appears a broad background. This background is often observed in PL measurements and is most likely caused by charging effects around the quantum dots and WL as for higher excitation densities many charge carriers are present in and around the quantum dots [85, 86].

Typical spectra at \( T=4.2 \text{K} \) and \( B=0 \text{T} \) are shown in Fig. 3.7(b) for 250 nW, 1000 nW, 5000 nW and 20000 nW excitation powers. For low powers relatively few PL peaks are visible. The FWHM of the PL lines is determined by the resolution of the setup and corresponds to 55 \( \mu \text{eV} \). For these low powers one typically find the neutral exciton X0 lines. Increasing the power results in the
Figure 3.7: (a) Three µ-PL spectra recorded for three different powers: 1000nW, 10000nW and 100000nW showing the PL of the WL states and the QDs. For the highest excitation density the $p$-states appear indicated with the arrow. (b) A typical micro-PL spectra of sample R80 for different excitation densities. The spectra have been obtained using the triple monochromator in additive mode with the three 750 grooves/mm gratings. (c) The PL intensity of the neutral exciton $X^0$ showing a linear behavior on excitation density and the biexciton $2X^0$ showing a stronger than linear dependence on excitation density.
appearance of other lines in the spectrum. These lines can be related to the biexciton complexes \((2X^0)\), charged exciton complexes or to exciton lines from other quantum dots slightly out of focus. In order to determine the origin of such a line one can perform a lateral scan over a small area of the sample. In this way it is determined whether the PL lines from the dot studied is in the focal plane. A further test is to analyse the power dependency of the lines. In an ideal experiment the \(X^0\) (\(2X^0\)) complex has a linear (bilinear) dependence on the excitation density \([87]\). Figure 3.7(c) shows the PL intensity of a \(X^0\) and a \(2X^0\) complex. As the power increases the quantum dot is more and more likely to be occupied by more than one exciton. Therefore, for increasing excitation density the intensity of the \(X^0\) first saturates and eventually will decrease, whereas the PL intensity of the \(2X^0\) is still increasing at relatively large powers. The slope of the \(X^0\) indeed increases linear with power with power (slope \(\sim 1.02\)) and the \(2X^0\) has a superlinear dependence with a slope of 1.45. Deviations from the ideal slope of 2 are often found and depend on the sample properties and the excitation wavelength. By carefully analyzing the spectra as function of the excitation density and the position with respect to the focus the \(X^0\) lines are selected.

In order to distinguish the neutral exciton complexes from the singly charged exciton complexes \(X^+(-)\) the fine structure splitting (FSS) or anisotropy splitting \(\delta_1\) is analyzed. This splitting is absent for the singly charged exciton complexes as the total spin of the electrons (or holes) is zero and results in the absence of the exchange interaction. However, for our dots we do not observe the FSS within the experimental accuracy and therefore it is not possible to distinguish between the \(X^+(-)\) and \(X^0\). In chapter 6 we report on the observation of the FSS for InAs/InP quantum dots, and for InAs/GaAs quantum dots it has been reported in several papers \([26, 68, 77, 88]\).

### 3.4.3 Single quantum dot magnetoluminescence: \(g\)-factor and diamagnetic shift

The photoluminescence spectra of a single quantum dot as function of the magnetic field \(B\) is shown in Fig. 3.8(a) for \(B\) up to 6.8 T in steps of 0.4 T. The single line at \(B=0\) T splits in two lines of opposite circular polarization. This is the exciton Zeeman splitting \(\Delta E_{\text{Zeeman}}\) defined by Eq. 3.32. The low (high) energy peak has \(\sigma^+ (\sigma^-)\) polarization resulting in a negative value of \(g_{ex}\). The peak positions are determined by fitting the PL peaks with a Lorentzian. The filled (empty) circles in Fig. 3.8(b) correspond to \(\sigma^+ (\sigma^-)\) polarization. The diamagnetic energy \(E_{\text{dia}} = \frac{E(\sigma^+)+E(\sigma^-)}{2}\) corresponds to the filled squares. Figure 3.8(c) shows \(\Delta E_{\text{Zeeman}}/\mu_B\) as function of \(B\) and displays clearly a linear behavior. From the slope the exciton \(g\)-factor \(g_{ex}\) is determined. For this QD \(g_{ex} = -2.32\). The normalized diamagnetic shift \(\Delta E_{\text{dia}}\) (see Eq. 3.33) as function of \(B^2\) shows a linear dependence. The slope is given by the diamagnetic coefficient \(\alpha_d\), for which we find \(\alpha_d = 8.5 \mu eV/T^2\).

A large number of quantum dots has been analyzed in a similar way as was done for the QD of Fig. 3.8. In Fig. 3.9 the exciton \(g\)-factor \(g_{ex}\) is shown as a function of the emission energy at \(B = 0\) T \((E_0)\) for 84 quantum dots. The dots emitting at larger energy have in general a more negative value of \(g_{ex}\) indicated by the linear fit. From the macro-PL measurements it is inferred
Figure 3.8: (a) The photoluminescence spectra of a single InAs/GaAs quantum dot as function of magnetic field $B$ up to 6.8 T in steps of 0.4 T. The single line at $B=0$ T exhibits the Zeeman splitting $\Delta E_{\text{Zeeman}}$. The low (high) energy peaks have $\sigma^+$ ($\sigma^-$) polarization resulting in a negative value of $g_{ex}$. (b) The peak positions as determined by fitting the PL peaks with a Lorentzian. The filled (empty) circles correspond to $\sigma^+$ ($\sigma^-$) polarization and the filled squares correspond to the diamagnetic energy $E_{\text{dia}}$. (c) $\Delta E_{\text{Zeeman}}/\mu_B$ as function of $B$ shows a linear behavior. The slope of the fit gives $g_{ex} = -2.32$. (d) The diamagnetic shift $\Delta E_{\text{dia}}$ as function of $B^2$ shows a linear dependence. The slope gives the diamagnetic coefficient $\alpha_d$ for this dot of $\alpha_d = 8.5 \mu$eV/T$^2$. 
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Figure 3.9: The exciton $g$-factor as a function of the emission energy at $B=0$ T, $E_0$. Quantum dots emitting at larger energies have in general a more negative value of $g_{ex}$. The dashed line is a linear fit in order to indicate the trend. The blue filled stars, the unfilled circles and the red filled circles correspond to the smallest, medium and largest values of $\alpha_d$. The unfilled squares correspond to the WL states and are not used for the linear fit.

that dots emitting at higher energy are of different height, although the strong overlap of the different sub-ensembles in the macro-PL spectra prevents a one-to-one correlation between the emission energy and height of the quantum dots. Nevertheless, the observed trend is probably related to the height of the dots.

In order to determine whether the emission energy is solely determined by the height, $E_0$ is plotted versus $\alpha_d$. Figure 3.10(a) shows that there is a weak correlation. Quantum dots emitting at higher energy tend to have a slightly larger $\alpha_d$. This is partly a result of the confinement in the height direction; the stronger confinement in the height causes an increased lateral extension of the wave function and therefore effectively increases the extension length (see Eq. 3.7). The second reason is that the physical lateral extension of the dots is larger. This is clearly the case for the four data points having a much larger $\alpha_d$ indicated with the unfilled squares. These two data points are obtained on peaks originating from WL states. These states are much more extended as compared to the strongly confined QD states. In fact, the WL states approach the (2D) quantum well-like states and will thus experience a much stronger diamagnetic shift. This is alternatively shown in Fig. 3.9, where four different intervals of diamagnetic shift are distinguished: quantum dots emitting at $E_0 > 1.37$ eV have a large $\alpha_d$.

In Fig. 3.10(b) the correlation between the $g_{ex}$ and $\alpha_d$ is investigated. Besides that the WL states with large $\alpha_d$ are outside the distribution, there is no
Figure 3.10: (a) The diamagnetic coefficient $\alpha_d$ as function of $E_0$, the emission energy at $B=0$ T. The color bar corresponds to the three different classes of $\alpha_d$ used in Fig. 3.9. (b) The exciton $g$-factor as function of the diamagnetic coefficient $\alpha_d$. The different filled symbols correspond to three different QD emission energy regimes. The open squares correspond to WL states.
observable trend. It is again clear however, that there is a strong correlation between \( g_{ex} \) and \( E_0 \) by plotting three intervals of \( E_0 \): the data points with lowest energy (black squares) are in general above the ones with medium energy (red filled circles), which are in their turn above the points with highest energy (green filled triangles).

The found tendency is in correspondence with Refs. [89, 90]. They show that depending on the indium concentration the exciton \( g \)-factor may vary a factor of three. In order to determine the effect of the indium concentration on \( g_{ex} \) in our dots we need to perform an extended X-STM analysis. However, due to the low QD density the X-STM measurements will be too time-consuming. In table 3.3 the values of \( g_{ex} \) reported in literature and the corresponding energy windows are provided as well as our experimental values. Although our values are in the range of the previous reported values, the size, shape and composition can alter the values quite dramatically and a proper comparison is therefore hard to make.\(^6\)

For the measured sample the dependence of \( g_{ex} \) on \( E_0 \) is weak. This is mainly caused by the strong overlap of the PL shoulders (dots with lower height) with the main PL peak (higher dots). Therefore for a certain energy it is hard to state whether a QD of relatively high or low height is analyzed. As will be shown in chapter 6, the \( g_{ex} \) of InAs/InP dots is more clearly related to the height and diameter of the quantum dots. The larger wavelength region in which the InAs/InP quantum dots emit as well as a more pronounced multimodal height distribution will convincingly show that indeed the overall size of the dots is determining the value of \( g_{ex} \).

3.4.4 The quadruplet splitting: determination of electron and hole \( g \)-factor separately

In general, a splitting of a single exciton line into four lines in magnetic field is observed when the magnetic field is not parallel to the optical detection axis.\(^7\) In the extreme case, the magnetic field is perpendicular to optical axis of the detection system. This measurement configuration is known as the Voigt geometry. In the Voigt geometry the optical selection rules allow for the dark states to be observed [68]. However, for some QDs a quadruplet splitting is observed by measuring in the Faraday geometry; in the Faraday geometry the magnetic field is parallel to the optical detection path. This splitting was first observed by Bayer et al. [99]. The quadruplet splitting is attributed to a symmetry reduction of the QDs. This can, for instance, be caused by an inhomogeneous indium distribution within the quantum dot. In that case the dark \( X = 2 \) states mix with the bright \( X = 1 \) states and gain oscillator strength.

Out of the 84 quantum dots, one quantum dot was found exhibiting a quadruplet splitting. Figure 3.11(a) shows a contour plot of the PL of this dot. The color scale relates to the PL intensity, where blue (white) is low (high) intensity. The two outer lines have a strong intensity, whereas the two inner lines have lower intensity. The polarization of the upper two lines is \( \sigma^- \), whereas the lower two lines have \( \sigma^+ \) polarization. The inset shows the power dependence

\(^6\)For Ref. [91] the quantum dots are enlarged by nearly one order in magnitude in comparison to conventional quantum dots using a low strain InAs nucleation layer.

\(^7\)We only consider here the case where the growth direction of the sample is parallel to the optical detection axis.
Table 3.3: The experimental values found for the exciton $g$-factor ($g_{ex}$) and diamagnetic coefficient $\alpha_d$ and the corresponding emission energy as reported in literature for InAs/GaAs quantum dots.

<table>
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<tr>
<th>Author [Ref]</th>
<th>$g_{ex}$</th>
<th>$\alpha_d$ ($\mu$eV/T$^2$)</th>
<th>Emission energy (eV)</th>
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<td>Bayer [92]</td>
<td>[2]</td>
<td>–</td>
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</tr>
<tr>
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<td>-3</td>
<td>3.3</td>
<td>1.31</td>
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<td>1.34</td>
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<td>–</td>
<td>1 – 1.15</td>
</tr>
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<td>9.22</td>
<td>1 – 1.3</td>
</tr>
<tr>
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<td>7 – 11</td>
<td>1 – 1.3</td>
</tr>
<tr>
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</tr>
<tr>
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<td>1.33</td>
</tr>
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<td>1.31</td>
</tr>
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<td>–</td>
<td>1.32 – 1.42</td>
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<td>[1.4[, [2]</td>
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<td>1.22 – 1.32</td>
</tr>
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<td>Kleemans [38]</td>
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</tbody>
</table>
Figure 3.11: (a) The presence of both the bright and dark states due to mixing of the heavy hole and light hole states results in the observation of the quadruplet splitting. The inset shows the power dependence of the quadruplet at $B = 5 \text{T}$. The relative intensity does not depend on the excitation power. (b) A fit of the peak positions as a function of magnetic field. The filled squares (circles) correspond to the bright (dark) states with $\sigma^-$ polarization and the empty squares (circles) correspond to the bright (dark) states with $\sigma^+$ polarization. The inset shows the Zeeman splitting $\Delta E_{\text{Zeeman}}/\mu_B$ for the bright and dark states. From the fit we determine $g_{X=1} = g_e + g_h$ ($g_{X=2} = g_e - g_h$).
of the quadruplet at $B = 5 \text{T}$. The relative intensities are independent on the excitation power. In fact, the outer (inner) two lines are attributed to the PL originating from the $X = 1$ ($X = 2$) states. The different peak positions are determined by fitting the peaks with Lorentzian curves and the result is shown in Fig. 3.11(b). From the fitting $\Delta E_{\text{Zeeman}}/\mu_B$ as function of $B$ is determined for both bright and dark states. The slope gives $g_{X-1} = -1.78 \pm 0.01$ and $g_{X=2} = -0.84 \pm 0.03$. Using Eqs. 3.30 and 3.31 we obtain $g_h = -1.31$ and $g_e = -0.47$. Bayer et al. reported slightly larger values $g_h = -2.21$ and $g_e = -0.81$ [99] for a QD emitting at $1.312 \text{eV}$. This higher emission energy could correspond to a quantum dot of decreased height resulting in a more negative $g$-factor giving rise to a more negative value for both $g_h$ and $g_e$.

3.5 Summary

In this chapter we discussed the harmonic oscillator model, which approximates the quantum dot potential in general rather well. In a magnetic field the excitons experience a quadratic diamagnetic shift, which is proportional to the diamagnetic coefficient $\alpha_d$. Furthermore, the spin of the exciton gives rise to a Zeeman induced splitting, which is linear in magnetic field and proportional to the exciton $g$-factor $g_{ex}$. Both macro- and micro-PL experiments are performed on a sample containing a low density of InAs/GaAs quantum dots. Over 80 individual quantum dots have been analyzed in a magnetic field. For these dots $\alpha_d$ and $g_{ex}$ have been determined. The values of $g_{ex}$ ranged from $-2.5$ to $-0.5$, whereas $5 \leq \alpha_d \leq 10$. Larger values of $\alpha_d$ are observed for energies which are related to the WL luminescence. Analyzing the quadruplet splitting found for one of the dots allowed for determining the electron and hole $g$-factor separately: $g_e = -0.47$ and $g_h = -1.31$. Importantly, there is a clear trend between $g_{ex}$ and $E_0$: for larger emission energy a more negative value of the exciton $g$-factor is observed. From the power and temperature dependence of the ensemble PL it has been shown that the sample consists of QDs with different height, where the highest dots correspond to the smallest emission energies. Moreover, QDs with larger $\alpha_d$ have a more positive $g$-factor. From this it is inferred that quantum dots with an overall larger size have a less negative value of $g_{ex}$.

3.6 Outlook

In order to analyze better the influence of the dot height on $g_{ex}$ it is proposed to analyze $g_{ex}$ on a set of samples containing quantum dots of well determined height. Two different growth methods can be applied in order to engineer the height of the dot. The first technique is the indium flush technique. After the growth of the quantum dots the dots are partially capped with a well-determined layer thickness. After this partial capping the dots are annealed. During this procedure the uncapped part of the dots gets removed (flushed away). Therefore, the capping thickness directly determines the height of the quantum dots. After this procedure the dots are capped for optical experiments.

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In the literature there is no unambiguous definition of the sign of the $g$-factor. Reference [99] uses a different definition of the exciton $g$-factor for the bright and dark states, i.e. $(g_{X-1} = g_H + g_e$ and $g_{X=2} = g_e - g_h)$. Applying our definition we obtain for their quadruplet splitting $g_e = +0.82$ and $g_h = -2.20$, which already gives a quite different electron $g$-factor.
In principle this method already provides the opportunity to tune the dot height between 2-7 nm [100]. The second technique involves the growth of columnar InAs/GaAs quantum dots [101]. The columnar InAs QDs are formed on InAs seed QDs by alternating between GaAs intermediate layers and monolayers of InAs with extended growth interrupts after each layer. The height of the columnar InAs QDs is controlled by the number of stacked GaAs/InAs layers. Using this technique it is possible to dramatically increase the height of the dots to several tens of nm such that the height can even become larger than the lateral extension.

In order to further investigate the influence of the quantum dot height and diameter on the hole and electron $g$-factor separately it is needed to study the magnetoluminescence of individual dots positioned under a small angle with respect to the magnetic field. In this case again quadruplet splittings are expected as was the case for the dot in section 3.4. The study of $g_e$ and $g_h$ separately will help to understand better the origin of the change in $g_{ex}$. 

Chapter 4

Many-body exciton states in charge tunable self-assembled InAs/GaAs quantum dots

4.1 Abstract

It is not surprising that a localized level coupled to the Fermi sea attracts so much interest, as it gives rise to rich physics involving many-body quantum states [102, 103]. Self-assembled quantum dots (QDs) offer the opportunity to study many-body exciton states in the optical spectra of zero-dimensional nanostructures located in the proximity of an electrical contact [104, 105]. In this chapter the strong coupling between a Fermi sea of electrons and a QD is investigated. Two different many-body exciton states are observed using voltage dependent PL spectroscopy. Firstly, the presence of Mahan excitons is revealed [106], which originate from the Coulomb interaction between electrons in the Fermi sea and the hole(s) in the QD. The second type of many-body exciton is the hybridized exciton [102], originating from the tunnel interaction between the continuum of states at the back contact and the localized state in the QD. Spectroscopically, this is best observed as the smooth continuous transition between the neutral and the negatively charged exciton. The observation of many-body interactions between a well-defined QD state and a Fermi sea of electrons opens the route towards the optical observation of the Kondo effect [107, 108, 109, 110, 111, 112] in these semiconductor nanostructures.¹

¹These results have been submitted as: Many-body exciton states in self-assembled quantum dots coupled to a Fermi sea, N. A. J. M. Kleemans, J. van Bree, A. O. Govorov, G. J. Hamhuis, R. Nötzel, A. Yu. Silov, and P. M. Koenraad, (2009).
4.2 Introduction

Many-body effects in electron gases have been studied extensively in the past and have attracted a lot of interest because they were challenging both experimentally and theoretically. Nowadays the aspects of many-body interactions in three-dimensional and two-dimensional electron and hole gases are well understood and have been shown to be very important. Famous examples of this type of physics include the X-ray Fermi-edge singularity in metals [113], the metallic (Mahan) excitons [114], the Kondo effect in the resistance of metals doped with magnetic impurities [115], the quantum Hall effects in two-dimensional gases [116], the revival of Kondo physics in transport of edged lateral nanostructures [117, 118], etc. Quantum dots offer a really unique opportunity - simultaneous electronic control of the number of carriers and optical readout [119]. By now we have acquired a good understanding of the exchange and correlation interactions between the finite numbers of carriers confined in zero-dimensional nanostructures [12, 120]. The next goal is to utilize these nanostructures in operational devices, where we will have to contact the zero-dimensional systems to a three-dimensional or two-dimensional Fermi sea of carriers acting as the source or drain of the nanostructures. Due to the presence of a Fermi sea of carriers in close proximity of the quantum dots, we enter a new area of many-body interactions, where the physics of the system is governed by the interaction between carriers in the nanostructures and the carriers in the two-dimensional or three-dimensional Fermi sea. Here we come again to the Kondo-type physics described by the Anderson [102] and Fano [121] Hamiltonians, but now, very importantly, involving both optical readouts and the voltage-tunability. This combination (optics and voltage control) was never realized before in the three- and two-dimensional systems.

4.3 The charge tunable device

In this section the layer structure of the grown charge tunable device is discussed. The basics of the charging of the quantum dots are explained using the Coulomb blockade model [12] and allow to calculate the Coulomb energies for a typical InAs/GaAs quantum dot.

4.3.1 Layer structure and charging

Charge tunable devices provide the possibility to load individual charges in quantum dots in a controlled manner [105]. The principle of such devices is best explained by discussing the layer structure of the MBE grown charge tunable InAs/GaAs quantum dots studied in this chapter and the corresponding band structure, as shown in Fig. 4.1. The layer structure consists of a GaAs substrate, which is overgrown by an AlAs/GaAs superlattice. This superlattice serves only to smoothen the surface which can be rough after the oxide desorption. A 80-nm thick GaAs is grown on top of the superlattice followed by 20-nm n-doped ($\sim 10^{18}$ cm$^{-3}$) GaAs layer. This highly n-doped layer will act as the

\footnote{This sample has been grown by G. J. Hamhuis and R. Nötzel in the group Photonics and Semiconductor Nanophysics at the Eindhoven University of Technology. The sample number is R544.}
Figure 4.1: (a) The layer structure of charge tunable InAs/GaAs dots. The quantum dots are separated from the back contact by a 25 nm thick tunnel barrier. (b) The corresponding band structure for two different applied voltages $V_g^a$ and $V_g^b$. For $V_g = V_g^a$, the Fermi level $\epsilon_F$ of the back contact is below the single electron level in the quantum dot $E(e)$ and no electrons are loaded into the quantum dot. For $V_g = V_g^b$, $E(e) < \epsilon_F$ and electrons are able to tunnel to the QD.

back contact and provides the electrons to the quantum dots. The quantum dots and the back contact are separated by a 25 nm thick GaAs tunnel barrier. The quantum dots are capped by 20 nm of GaAs. A superlattice of AlAs/GaAs is grown on top acting as a blocking barrier, preventing electrons and holes to reach the top gate. The described layer structure is schematically depicted in Fig. 4.1(a). In order to contact the $n$-doped back contact, small pieces of Sn are annealed on top of the sample. During the annealing process ($\pm 2$ minutes at 350°C) the Sn diffuses through the layer structure towards the back contact. The Sn droplets on the surface are bonded using gold wires. A top gate is evaporated on top using a shadow mask and consists of 2.5 nm of Ni and 2.5 nm of Cr. The top contact is also bonded by a gold wire. Effectively, the NiCr gate acts as a Schottky contact and an external gate voltage $V_g$ can be applied between the back contact and the top gate.

In the absence of an external voltage ($V_g = 0$ V) the Fermi level at the surface is pinned in the middle of the energy gap due to the presence of a large
amount of surface states. The Fermi level is also pinned at the back contact near the conduction band due to the $n$-doping. An external voltage will tilt the bands such that the Fermi level is scanned through the different energy levels in the quantum dot. In fact the device works effectively as a capacitor [71]. By applying different voltages, one can determine the number of charge carriers in the dot. For instance, for $V_g = V_b^g$ (see Fig. 4.1(b)) the Fermi energy $\epsilon_F$ is larger than the electron level in the QD $E(e)$. Therefore in this case one (or more) electrons tunnel from the back contact to the QD, thereby charging the quantum dot. Upon illumination with a laser, electrons and holes will be created and be captured in the dots. The presence of additional carriers in the dots will lead to different charged exciton complexes. In the case of $V_g = V_a^g$, we find $\epsilon_F < E(e)$ and no additional electrons are present in the dot. Upon illumination electrons can even tunnel out of the dot for sufficiently negative gate voltages, giving rise to charging of holes in the quantum dots. Moreover, near the interface with the blocking layer a two-dimension hole gas (2DHG) is present from which holes can tunnel into the quantum dots and as a result provides additional charging of the dots [122].

The macro-PL spectrum of the sample we investigated is shown in Fig. 4.2. The QD luminescence is between 1.25 eV and 1.425 eV. In fact, the QD PL is overlapping with the WL PL. Compared to conventional measured QDs, these QDs are shallow, i.e. the confinement provided by the dots is relatively low as compared to the confinement provided by the WL. From AFM measurements we estimate a QD density of $<2 \times 10^9$ cm$^{-2}$.
4.3.2 The Coulomb blockade model

To explain the Coulomb interactions within a QD, the Coulomb blockade model, based on perturbation theory, is discussed in this subsection. We will follow the approach explained in detail in Refs. [12, 123]. There are five assumptions that form the basis of the model:

(i) The quantization energies of electrons ($\hbar \omega_e$) and holes ($\hbar \omega_h$) (see Fig. 4.3) are larger than the Coulomb energies and thus the Coulomb energies are a perturbation on the quantization energies.

(ii) The quantum dots can be treated as quasi two-dimensional, i.e. having a much smaller height as compared to the lateral size (see chapter 3).

(iii) The confinement potentials are described within the harmonic oscillator model as explained in chapter 3.

(iv) There are no dot to dot interactions. In general this is a good approximation as the distance to the back contact is $\sim 25$ nm, whereas the inter dot distance is several $\sim 100$ nm.

(v) A lever arm is introduced, which is linear with voltage. The band bending effects are thus neglected.

Using the schematic representation in Fig. 4.3, we will now derive the equation for the energies of the neutral and negatively charged trion. The energy of $X^0$ is given by the single particle energy difference between a hole and an electron in the s-shell ($E_0$ in Fig. 4.3(a)) minus the correction for the Coulomb attraction of the electron and hole $E_{eh}^{ss}$, which thus gives:

$$E(X^0) = E_0 - E_{eh}^{ss}. \quad (4.1)$$

In Fig. 4.3(a) no electron is present in the dot, as $\epsilon_F < E(e)$. By applying a forward bias $E(e)$ is lowered with respect to $\epsilon_F$, and when $\epsilon_F \sim E(e)$ an electron can be loaded into the QD (see Fig. 4.3(b)). The energy of a single electron with respect to $\epsilon_F = 0$ consist of three contributions. Firstly, the electrostatic energy $E_{V_0}$ in eV of this electron in an electric field (indicated in Fig. 4.3(a)) is given by:

$$E_{V_0} = \frac{e(V_0 - V_g)}{\lambda}. \quad (4.2)$$

where $V_0 \sim 0.6$ V is the Schottky barrier height [12, 124] and $\lambda$ is the lever arm of the device. The lever arm is given by the ratio of the distance between back contact and the top gate and distance of the QDs from the back contact; in our case $\lambda = 195/25 = 7.8$. The second contribution to the charging energy is the energy the electron loses as a result of the QD confinement, $E_c$ (see also Fig. 4.3(a)). Finally, there is the effect of an image charge in the back contact, which contributes to the image charge electrostatic energy $E_i$ (for details see

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3We neglect here the exchange energy terms explained in chapter 3, which are two orders of magnitude smaller than the Coulomb terms.
Figure 4.3: (a) The energy level diagram of the charge tunable device. The energies are measured with respect to the Fermi energy $\epsilon_F = 0$. An electron must overcome the energy barrier $V_0 - V_g$ and loses the confinement energy $E_c$. In this case the s-shell of the electron in the QD is above the Fermi level (blue dotted line) and no electrons loaded in the QD. (b) By increasing $V_g$ we get the situation where $\epsilon_F$ is equal to the s-shell of the electron in the QD, one electron (black filled circle) is loaded into the QD. Although the s-shell allows for 2 electrons, the Coulomb blockade prevents this second electron to enter the QD. (c) For larger $V_g$ the energy of the s-shell is lowered even further with respect to Fermi level and a second electron can tunnel into the dot. For this to occur, the additional lowering in energy must be equal or larger than the Coulomb repulsion between the two electrons in the s-shell ($E_{ee}$).
Ref. [125, 126]) given by:

\[ E_i = -\frac{q^2}{16\pi\varepsilon_0\varepsilon_r z}, \]  

where \( q \) is the charge of the exciton, \( \varepsilon_0 \) is the electric constant and \( \varepsilon_r = 14.3 \) is the permittivity of In\(_{0.7}\)Ga\(_{0.3}\)As [127]. Equation 4.3 gives \( E_i \sim -1 \) meV for a single electron in a QD. So the total energy of a single electron in the QD is:

\[ E(e) = \frac{(V_0 - V_g)}{\lambda} - E_c - E_i, \]  

where the electrostatic energy for \( V_g = 0 \) is \( \sim 80 \) meV, and the confinement energy is typically \( \sim 85 \) meV.

Upon photo excitation an additional electron-hole pair can be captured in the quantum dot forming the negatively charged trion \( X^- \). The energy of this trion is given by the Coulomb terms between the electron already present in the quantum dot and the photo-excited electron and hole captured in the quantum dot and adding this to Eq. 4.4:

\[ E(X^-) = \frac{(V_0 - V_g)}{\lambda} - E_c - E_i - 2E_{ss} + E_{ee}. \]  

A second electron loaded in the quantum dot experiences the same single particle energy. Note that the image charge term is quadratic with the charge, so for two electrons the total energy is \( 4E_i(e) \), where \( E_i(e) \) is the single electron image charge energy. However, there is also an additional Coulomb repulsion between the two electrons in the s-shell of the quantum dot, \( E_{ee} \). Therefore the total energy of the two electrons is:

\[ E(2e) = 2\frac{(V_0 - V_g)}{\lambda} - 2E_c - 4E_i + E_{ee}. \]  

Finally, the single hole state is obtained by removing an electron from the neutral exciton:

\[ E(h) = E_0 - \frac{(V_0 - V_g)}{\lambda} + E_c - E_i. \]  

We will now apply the Coulomb blockade model on the measurement of a typical self-assembled charge tunable InAs/GaAs quantum dot studied in this chapter. For this measurement a solid immersion lens is positioned on top of the gate to allow for single quantum dot analysis. The laser excitation has been provided by a laser diode emitting at 635 nm and the PL is analyzed with a spectrometer-detector system with a resolution of 60 \( \mu \)eV. The voltage dependent PL is shown in Fig. 4.4 for two different excitation powers. Comparing the spectra with other spectra reported in literature, for example Refs. [104, 105, 120, 128, 129], we are able to identify the different charging plateaus. The different exciton complexes are indicated in Fig. 4.4. For \( 0.12 \leq V_g \leq 0.255 \) V the neutral exciton \( X^0 \) is observed. For more positive voltages the negatively charged trion \( X^- \) is observed, whereas the charging plateaus for more negative voltages correspond to the positively charged excitons \( X^{n+} \), where \( n \) corresponds to the charged state of the exciton. For increasing excitation power (cp. Fig. 4.4(a) and (b)) the biexciton states of the different charged excitons become much more pronounced.

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4Here we assume, like we did in chapter 3, an indium concentration of 70%. We note, however, that we do not know the QD composition.
Figure 4.4: A typical contour plot of the PL of an InAs/GaAs quantum dot as function of the gate voltage $V_g$ for an excitation power of (a) 500 nW and (b) 5000 nW. The color bar corresponds to the PL counts per 10 s. The different (charged) exciton complexes are indicated in (a). For increasing excitation power the biexciton lines are more pronounced than the excitonic lines.
Using Eqs. 4.1, 4.4, 4.5 and 4.7 we are able to extract the Coulomb energies between the electrons and the holes in the quantum dot of Fig. 4.4. In order to do so we determine the voltage extend of the \( X_0 \)-plateau, which is 0.135 V and the energy difference between the \( X_0 \) and \( X^- \) plateau: \( \Delta E(X_0 - X^-) = -3.87 \) meV. Moreover, at \( V_g \sim 0.3 \) V applies \( E(X_0) = E(X^-) \) and at \( V_g \sim 0.11 \) V applies \( E(h) = E(X_0) \) (see Fig. 4.4(a)). From this we finally find the direct relation between the length of the voltage plateau and \( E_{ee} \):

\[
\frac{\Delta V}{\lambda} = E_{ee}^{ss} - 2E_i,
\]

which gives for our QD a Coulomb interaction between the two electron in the s-shell of \( \sim 23 \) meV. The value of \( E_{ch}^{ss} \) can be found by considering the initial \((i)\) and final \((f)\) states of \( X_0 \) and \( X^- \), as the PL energy is due to the difference between initial and final state (\( E_{PL} = E_i - E_f \)). The final state of \( X_0 \) has an energy \( E_f(X_0) = 0 \) since there is no electron present in the final state, and therefore \( E_{PL}(X_0) = E(X_0) \). For \( X^- \) the final state is \( E_f(X^-) = E(e) \) and thus \( E_{PL}(X^-) = E(X^-) - E(e) \). The difference in PL energy between the \( X_0 \) and \( X^- \) is thus given by:

\[
\Delta E_{PL}(X_0 \rightarrow X^-) = E_{ee}^{ss} - E_{eh}^{ss},
\]

which shows that the red shift of the \( X^- \) with respect to the \( X_0 \) is purely a consequence of a larger Coulomb attraction between the electron and the hole as compared to the Coulomb repulsion between the electrons. In fact, in a similar way it can be shown that \( \Delta E_{PL}(X_0 \rightarrow X^+) = E_{hh}^{ss} - E_{eh}^{ss} \). Therefore the blue shift of the \( X^+ \) with respect to the \( X^- \) is due to the larger Coulomb repulsion between the holes as compared to the Coulomb attraction between the electron and hole. From Eq. 4.9 we obtain for our dot \( E_{ch}^{ss} \sim 27 \) meV.

Two of the assumptions in the Coulomb blockade model are that the quantization energies are much larger as compared to the Coulomb energies and that the dot has a parabolic confinement potential. In principle this allows the model to be extended towards higher charged exciton complexes [12, 123]. However, our quantum dots are relatively shallow and therefore the p-shell in these quantum dots is probably not adequately described within this model. In fact, for the quantum dot in Fig. 4.4(a) we do not even observe the charging plateaus of the higher negatively charged exciton complexes. In general, we do not observe negatively charged excitons higher than the \( X_3^- \). Apparently the negatively charged excitons are not or weakly bound in these quantum dots. The larger effective mass of the holes allows to observe the higher positively charged exciton complexes for some quantum dots.\(^7\) In chapter 5 we will show that this weak confinement regime results in a negative diamagnetic shift for the \( X_3^- \), as the Coulomb interaction dominates over the confinement energy.

Note that for the higher charged exciton complexes \( X_n^+ \) with \( n \geq 2 \), no exchange splittings are observed. This is in contrast to what has been reported in literature (for example Ref. [120]). As we will show, our quantum dots are

\(^5\)\( E(X_0)(V_g = 0.255 \) V) = 1.365430 eV and \( E(X^-)(V_g = 0.34 \) V) = 1.36556 eV.

\(^6\)Actually, later on in this chapter we will show that this voltage corresponds to the hybridized exciton \( X_H^\dagger \), which is a superposition of both initial and final states. Nevertheless, this procedure is still a good estimate.

\(^7\)In fact, for the quantum dot shown in Fig. 4.5(b) we do not even observe the \( X^+ \).
Figure 4.5: Several charge tunable quantum dots, emitting at different energies, show many similarities in the voltage dependent PL contour plots. The arrows indicate the novel spectroscopic features which will be studied in more detail throughout this chapter. For each dot the neutral exciton and the singly charged trion are indicated.

strongly coupled to the back contact, which possibly causes these splittings to be absent. More experiments are needed to determine the cause of the absence of the exchange splittings in these dots.

4.4 Experimental results

Photoluminescence from individual QDs is detected as a function of the gate voltage $V_g$ using the confocal microscope. A large number of quantum dots has been analyzed and for several QDs the voltage dependent PL spectra are shown in Fig. 4.5. All these dots show novel spectroscopic features (indicated with the arrows) that have not been reported up to now. The goal of this section is to unravel the origin of these features.

Figure 4.6(a) shows the voltage dependent PL of the QD, which we will study in detail in this section. For small negative voltages we observe three
Figure 4.6: (a) The measured PL spectrum of a QD as function of gate voltage $V_g$. The color scale relates to the detector counts. The steep PL lines at low energy correspond to the recombination of Mahan excitons (labeled as $X^m_n$). The PL was measured at $T = 4.2$ K and the excitation density was 15 nW. (b) The line shapes for the neutral Mahan exciton $X^0_M$ shown at three different gate voltages, and (c) the line shape close to the onset of the $X^0$ plateau (green empty circles) and at the center of the $X^0$ plateau (orange filled circles). The corresponding voltages are indicated by the arrows in (a).
Figure 4.7: (a) A schematic representation of the Mahan exciton. A hole in the QD has Coulomb interaction with the Fermi sea of electrons. The two possible optical recombination paths are indicated by (1) and (2). The initial states \( i \) and final states \( f \) of the Mahan-like exciton are shown on the right corresponding to the state before and after recombination, respectively. The filled (empty) circles correspond to the electron (hole). The back contact is represented by the colored area and the dotted line indicates the Fermi energy level \( \epsilon_F \). There are many final states \( f_n \) due to different shakeup processes. (b) A schematic representation of the Anderson exciton used to describe the tunnel coupling between the electron state in the QD and the continuum of states in the back contact. On the right \( i \) and \( f \) are shown of the Anderson exciton. Again there are many final states \( f_n \) due to different shakeup processes.

lines corresponding to the neutral (multi)-excitons \( (X^0, 2X^0, \text{and } 3X^0) \). For larger negative biases we find the positively charged \( X^+ \) complexes and for more positive biases we observe the negatively charged \( X^- \) complexes, where \( n \) corresponds to the charged state of the exciton. The first indication of the strong interaction of the electrons in the QD with the Fermi sea of electrons in the back contact is observed by analyzing the charging plateaus. There is no overlap between the \( X^0 \) and \( X^- \) plateaus and between the higher negatively charged plateaus due to the strong tunnel coupling of the electrons with the states in the electronic reservoir of the back contact. On the other hand the plateaus of the positively charged exciton complexes have a weak overlap as a result of the weak tunnel coupling of the holes with the two-dimensional hole sea at the interface with the superlattice [130]. We note that this process is accompanied by the accumulation of photo-excited holes in the dot at large negative voltages [120].

Remarkably, at voltages where we observe the positively charged excitons we detect broad steep lines at lower energy, labeled as \( X^+_M \). These lines are
a result of spatially indirect transitions from the Fermi sea to the localized hole state in the QD (schematically shown in Fig. 4.7(a)). These interesting many-body excitons are known as the Mahan excitons [106, 114] and have been investigated in metals and semiconductors. The neutral Mahan exciton $X^0_M$ comprises a hole residing in the QD coupled via the Coulomb interaction with a Fermi sea of electrons in the back contact. In the equivalent manner, the $X^+_M$ are formed. For example, the $X^+_M$ state consists of two holes confined in the QD having Coulomb interaction with the Fermi sea of electrons. The Mahan lines follow the lever arm of our device and correspond directly to the Fermi level in the back contact. From the voltage dependence of the lines a lever arm of $\sim 8$ is obtained, which is slightly larger as expected from the design of the layer structure ($\sim 6$). For different dots the experimental observed lever arm varies between 6 and 9. We attribute the deviation to the segregation of Si dopants from the $n^+$ doped back contact to the tunnel barrier. A variation of the tunnel barrier with 20%, giving rise to a different lever arm, is reasonable as was shown in X-STM measurement on Si doped GaAs layers. In Ref. [131] it is shown that there is almost no segregation for relatively low growth temperatures of 480°C, but strong segregation (as much as 10 nm) for a growth temperature of 580°C. We are in between these two regimes as our dots are grown at relatively high temperatures of 515°C.

There are two possible mechanisms of recombination for the Mahan-like exciton $X^0_M$, depicted in Fig. 4.7(a). The first path (1) is the direct transition from the Fermi sea to the valence band of the QD. The second path (2) is a second-order process via the virtual electron state in the QD. Therefore, the amplitude for the photon emission process can be written as a sum of terms:

$$A_{k \rightarrow h} = A_1 + A_2 = \langle \psi_h | \psi_k \rangle + \frac{\langle \psi_h | \psi_e | U_{QD}(z) | \psi_k \rangle}{\epsilon_k - \epsilon_e},$$

(4.10)

where $\psi_e(h)$ are the confined electron (hole) states in the QD, $\psi_k$ describes a 2D electron with an in-plane momentum $k$ in the Fermi sea, and $U_{QD}$ is the QD potential in the conduction band. The matrix element is written in the spirit of the tunneling Hamiltonian approach [132, 133]. As the tunnel probability scales with $\exp \left( -\sqrt{m_e(h)} \right)$ [11], with $m_e(h)$ the mass of the electron or heavy hole, we estimate that the second-order process is more probable since $m_e < m_h$.

The line shape of the $X^0_M$ for different gate voltages is shown in Fig. 4.6(b). The FWHM of the $X^0_M$ is approximately 2.7 meV and its peculiar line shape resembles the Fermi edge singularity in quantum wells [134], which is related to the Fermi energy edge of X-ray absorption in a metal [113, 135]. The low energy tails in the spectra of $X^0_M$ and the other $X^+_M$ complexes are caused by the reaction of the Fermi sea when an exciton is suddenly removed from the system. In the initial state a localized hole is screened by the Fermi sea via the Coulomb potential. When the Mahan exciton recombines, the hole is suddenly removed from the system. This causes perturbation (un-screening) of the Fermi sea, resulting in the creation of excitations, i.e. electron-hole pairs as depicted in Fig. 4.7(a). Therefore a low-energy tail appears in the PL spectrum. The excitation of the Fermi sea is strongly enhanced near the Fermi energy level [113], resulting in the Fermi edge singularity, which gives rise to a stronger PL intensity at the high energy side of the PL peaks shown in Fig. 4.6(b).

For more positive voltages the $X^0_M$ line evolves in the $X^0$ line and the line width decreases dramatically, as is shown in Figs. 4.6(b), (c). As the gate voltage
increases from $-0.425$ V to $-0.300$ V, the line width reduces from 2.7 meV to 150 µeV. The decrease in linewidth is caused by the change from the indirect $X^0_M$ at more negative voltages to the direct $X^0$ exciton in the QD at more positive voltages. The continuous transition from the Mahan exciton to the $X^0$ is due to the strong hybridization of the QD electron states with the Fermi sea. We note that this strong coupling regime was not achieved for the ground state excitons [136] in numerous studies on QDs by others [105, 120, 128, 129]. In general, the electron state is a hybridized state between the states of the Fermi sea and the QD state, of which the relative contribution depends on the gate voltage. For larger negative gate voltages the hybridized state is dominated by the states of the Fermi sea, whereas at lower negative gate voltages it is dominated by the QD state. Similar smooth transitions were predicted theoretically [107].

Amazingly, we clearly observe a weak asymmetric line shape even in the center of the $X^0$ plateau ($V_g = -0.300$ V in Fig. 4.6(c)) due to the remaining weak tunnel interaction between the QD state and the Fermi sea giving rise to a hybridized exciton state [104, 107]. Whereas the Mahan excitons results from the Coulomb interaction between the hole and Fermi sea, the hybridized exciton is solely based on the tunnel coupling and can be understood using the Anderson model depicted in Fig. 4.7(b). In the initial state the electron level in the QD is tunnel coupled to the Fermi sea. When the hybrid exciton recombines, the electron level in the QD is suddenly changed due to the removal of the intra-QD Coulomb attraction to the localized hole. Then, the hybrid electron states in the conduction band experience a shakeup and the corresponding optical line acquires a low-energy tail [104, 107]. We notice that, for the voltages corresponding to the $X^0$ plateau, the Mahan mechanism is strongly suppressed as there is a negligible Coulomb interaction between the exciton and the Fermi sea. The singularity of the Mahan line is given by:

$$I_{PL}(\omega) = I_0(\omega_T - \omega)^{-\alpha},$$  \hspace{1cm} (4.11)

where $I_0$ determines the total intensity, $\omega_T > \omega$, $\hbar\omega$ is the emitted photon energy, $\hbar\omega_T$ is the cut-off energy of the PL spectrum, and $\alpha < 1$ simulates the Fermi edge singularity [106, 137, 108]. We note that the singularity comes from intensive creation of excitations in the electron sea in the vicinity of the Fermi surface when an exciton is removed from the system. Of course, the spectrum should be broadened, and numerical studies based on the Anderson Hamiltonian and the Mahan-exciton model are needed to differentiate the many-body mechanisms in our PL spectra. Using a convolution between Eq. 4.11 and a Lorentzian with a width $\Gamma$, given by:

$$I_{PL} = I_0\left(1 - \frac{1}{\omega_T - \omega}\right)^\alpha \cdot \Gamma \frac{\Gamma}{\Gamma^2 + \omega^2},$$  \hspace{1cm} (4.12)

allows for a perfect fit of the Mahan line shape with $\alpha < 1$, as is shown in Fig. 4.8 for two different gate voltages. From the fit we obtain $\Gamma \sim 1$ meV. Clearly this is a much larger value than expected solely from radiative and phonon dephasing processes ($\sim 50$ µeV). The additional broadening is caused by the finite distribution of electrons at the Fermi sea [135, 134].

We note that even at the lowest excitation powers the Anderson hybridization and Mahan line is still clearly visible. In Fig. 4.6 the intensity ratio between the exciton and biexciton line is $3201 : 218 \sim 15 : 1$, which already indicates
Figure 4.8: The red data points are fitted using the analytical expression for the convoluted spectrum: the solid red line is the fit. (a) For $V_g = -0.42\,\text{V}$ we use the fitting parameters $I_0 = 30.89$, $\omega_T = 1344.601\,\text{meV}$, $\Gamma = 1.06\,\text{meV}$, and $\alpha = 0.718$. (b) For $V_g = -0.38\,\text{V}$ we have a good fit using $I_0 = 12.85$, $\omega_T = 1349.043\,\text{meV}$, $\Gamma = 1.21\,\text{meV}$, and $\alpha = 0.950$.

Figure 4.9: The hybridization of the $X^0$ in the initial state $i$ is strongest (weakest) for the edges (center) of the plateau, resulting in a lower (higher) energy at the edges (center) of the plateau. The resulting PL (indicated with the arrows) has a convex curvature along the voltage plateau. For the $X^-$ the hybridization of the electron in the final state $f$ in the QD with the continuum of states in the back contact is strongest at the edges of the plateau and weakest in the center of the plateau, and the resulting PL has a concave curvature along the voltage plateau.
that the excitation is provided at very low powers. These values are determined by taking the PL intensity half way the $X^0$ and $2X^0$ plateau. In contrast to the neutral biexcitons, the charged biexcitons are not visible at these powers and only appear at higher excitation powers. Figure 4.5(b) shows a different quantum dot excited at such low powers that the $2X^0$ is absent, but for which the Mahan exciton line and the "Anderson" exciton line are clearly present.

We now focus on the tunneling in and out of the QD of an electron from and towards the back contact in the initial and final states of the $X^0$ and $X^-$. It has been shown in Ref. [104] that an electron tunneling into the dot gives rise to the $X^-_f$ feature in Fig. 4.6(a). Moreover, the fast tunnel process of the electrons is causing the peculiar shape of the $X^0$ and $X^-$ plateaus observed on top of the well-known Stark shift [138, 130]. The curvature of the $X^0$ plateau (see Fig. 4.6(a)) is partly caused by the hybridization in the initial state of the $X^0$ with the continuum of states at the back contact. The hybridization of the $X^0$ state is strongest (weakest) for the edges (center) of the plateau, since the hybridization is strongest when the energy difference between the QD state and the state at the back contact is smallest. Moreover, in general the hybridization of energy levels reduces the ground state energy of the system and therefore the $X^0$ plateau is convex (depicted in Fig. 4.9). Strikingly, for the $X^-$ a concave plateau is observed. As the final state of the $X^-$ contains an electron we have to take into account both the initial and final state to explain the opposite curvature. For the $X^-$ the hybridization of the electron in the final state in the QD with the continuum of states in the back contact is strongest at the edges of the plateau and weakest in the center of the plateau. Therefore the final state of the $X^-$ is lower in energy towards the edges of the plateau as compared to the center of the plateau, resulting in the concave shaped $X^-$ plateau. The initial $X^-$ state is only hybridized in a small voltage domain, corresponding to the $X^0_{H}$ and we will now study this feature.

The $X^0_{H}$ is the most remarkable manifestation of the tunnel interaction of the QD exciton state and the continuum of states of the Fermi sea, and it is observed at the transition between the $X^0$ and $X^-$ complex. For voltages corresponding to the $X^0$ a photo-excited electron-hole pair in the QD recombines resulting in the PL of the $X^0$. If we increase the voltage and overcome the electrostatic energy, the QD becomes loaded with an additional electron, allowing to study the negatively charged trion $X^-$. The transition from the $X^0$ to $X^-$ occurs at $V_g = -0.230$ V. We observe a continuous evolution of the PL from the $X^0$ plateau to the $X^-$ plateau, labeled as the hybridized exciton state $X^0_{H}$, which may only be observed in the regime of strong tunnel interaction [107, 139]. Such smooth (continuous) transition of the plateaus may only be observed in the regime of strong tunnel interaction, because it is an indication of strong mixing between the wave functions of the discrete state of QD and the extended states of the Fermi gas [107, 139, 140]. Clearly, the presence of the Fermi sea gives rise to a complex problem and brings about peculiar many-body phenomena, which do not exist in the case of a QD coupled with an empty continuum described within the Fano model [129, 121]. We will now apply a zero bandwidth model to reproduce the $X^0_{H}$ qualitatively.
4.5 Modeling

The zero bandwidth model is a rather simple model, in which the Fermi sea is represented by a single quantum level with an energy equal to $\epsilon_F$ [141]. We note that this model does not give the shapes of the optical lines, however it allows us to look at the mixed-valance regime (the transition $X^0 \rightarrow X^-$), where the majority of analytical approaches become very challenging. To reproduce the optical line shapes, we should use the original Anderson Hamiltonian and a continuum of states using trial wave functions, which is done for the $X^-$ in Ref. [142]. This procedure is beyond the scope of this thesis. Nevertheless, the simple zero-band model already explains the presence of the hybridized exciton states and their physical properties.\footnote{The modeling is performed in close collaboration with A. O. Govorov, Department of Physics and Astronomy, Ohio University, Athens, USA.}

In our model, the initial and final states are represented by the diagrams shown in the inset of Fig. 4.12. The final state $f$ of the emission process contains only one electron (see inset of Fig. 4.12). The final state is a superposition of the state with an electron in the back contact $a|e_{bc}\rangle$ and of the state with energy $b|e_s\rangle$ and is coupled by the tunnel interaction with interaction strength $V_{\text{tun}}$. The final state is given by:

$$\Psi_f = a|k\rangle + b|e_s\rangle.$$  (4.13)

The Hamiltonian of the final state $\mathcal{H}_f$ reads then:

$$\mathcal{H}_f = \begin{pmatrix} E_{bc} & V_{\text{tun}} \\ V_{\text{tun}} & E_s \end{pmatrix},$$  (4.14)

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.10.png}
\caption{Spin configurations of excitons involved in $X^0$ and $X^-$. The exciton initial states are denoted as $i_{1(2)s}$ and $i_{1(2)t}$ and the final states are labeled as $f_{1(2)}^{\uparrow}$ and $f_{1(2)}^{\downarrow}$. The hole can be found in the two states $j_z = \pm 3/2$.}
\end{figure}
Figure 4.11: Using the zero bandwidth model the $X^0$ to $X^-$ transition can be reproduced. The upper (lower) branch corresponds to the line labeled $X^0_H$ ($X^f_I$) of Fig. 4.6(a). The intensity of the transitions is indicated by the thickness of the lines.

where $E_{bc} = \epsilon_F$ is the energy of the electron at the back contact and $E_s = -\Delta V_g / \lambda + \epsilon_F$ is the energy of the QD s-shell. For our convenience, we choose $\epsilon_F = 0$, and the QD electron energy is given by the applied voltage and the lever arm ($\lambda \approx 8$). Also, $\Delta V_g = V_g - V_{\text{crit}}$, where $V_{\text{crit}}$ is the voltage at which the first electron tunnels into the empty QD from the back contact; $V_{\text{crit}} \sim 0.35\text{ V}$ for the studied QD. The eigen energies of this Hamiltonian are:

$$E_{f,\pm} = \left( E_{bc} + E_s \pm \sqrt{(E_{bc} - E_c)^2 + 4V_t^2} \right) / 2.$$  \hfill (4.15)

The initial excitonic states unavoidably involve spins since they include two electrons in the conduction band. We ignore the coupling of spins of electrons and holes as in our experiments, the exchange splitting in $X^0$ ($\Delta_{\text{exch}}$) was never observed in PL spectra measured with the 30 $\mu$eV resolution. Mathematically, the electron-hole exchange interaction can be neglected if the QD-continuum coupling is strong enough, i.e. $V_{\text{tun}} > \Delta_{\text{exch}}$, where $\Delta_{\text{exch}} < 30 \mu\text{eV}$.\hfill (4.9)

The spin states of exciton are built as it was done in the Kondo problem within the zero-band-width model [141]. The Kondo problem involves the interaction of a localized spin with a reservoir of spins [114, 115]. Two electrons in the conduction band form singlet and triplet states and the hole with the spins $\pm 3/2$ are added to the exciton wave function. Figure 4.10 shows these configurations.

Two electrons can form singlet and triplet states with $S_{\text{tot}} = 0$ and 1, respectively. The most interesting state of two electrons is the singlet Kondo state which gives the ground state at the voltages on the $X^0$ plateau. The initial exciton state is:

$$\Psi_i = A|X^0 + e_{bc}\rangle + B|X^-\rangle.$$  \hfill (4.16)

\footnote{We note that for the fitting in Fig. 3, we used $V_{\text{tun}} = 0.4\text{ eV}$ and, therefore, the condition $V_{\text{tun}} > \Delta_{\text{exch}}$ is applicable.}
where \( |X^0 + e_e^{\pm}) = (|s_1; b_{c_1}⟩ - |s_1; b_{c_1})|s_{+,3/2}/\sqrt{2} \) and \( |X^−⟩ = |s_1; s_1⟩|s_{+,3/2} \) represent two states in which the electron form a singlet with \( S_{tot} = 0 \), where \( |s_{+,3/2} \) is the hole state with spin up. We will ignore electron-hole exchange interaction and thus the states with \( |s_{−,3/2}⟩ \) are equivalent. The coefficients \( A \) and \( B \) are found from the Schrödinger equation. At low temperature we take only the lower-energy initial state with:

\[
E_{i,−} = \left( E_{X^{0} + e_e} + E_{X^{−}} − \sqrt{\left(E_{X^{0} + e_e} − E_{X^{−}}\right)^2 + 8V_{tun}^2}\right)/2, \tag{4.17}
\]

where \( E_{X^{0} + e_e} = \hbar \omega_{X^{0}} \) and \( E_{X^{−}} = \hbar \omega_{X^{0}} − U_{Coul} − \Delta V_g/\lambda \). Here \( \hbar \omega_{X^{0}} \) is the energy of the neutral exciton in the QD and \( U_{Coul} = \tilde{U}_{eh} − \tilde{U}_{ee} \sim 3.52 \text{meV} \) is the Coulomb energy that gives the lowering of \( X^{−} \) with respect to \( X^{0} \); i.e. \( U_{Coul} = \hbar \omega_{X^{0}} − \hbar \omega_{X^{−}}, \) where \( \hbar \omega_{X^{0}/X^{−}} \) are the observed excitonic energies. We also have a triplet initial state \( S_{tot}^t = 1 \). This state does not hybridize in the zero bandwidth model at all \cite{141}. The resultant energies of a hybridized singlet exciton at low temperature are \( \hbar \omega_{upper/lower} (∆V_g) = E_{i,−} − E_{f,-}. \) The emission spectrum of the hybrid exciton in the zero bandwidth model has several lines \cite{107}, shown in Fig. 4.11.\(^{10} \) The \( X^{−}_f \) branch corresponds to the situation where the electron in the final state is tunneling out of the QD as was discussed in Ref. \cite{104}. We also observe this feature in Fig. 4.6, indicated by \( X^{−}_f \). In fact, this feature is connected to the \( X^{−} \) plateau whereas in the used model there is a anti-crossing. This is due to the zero bandwidth character of our model. When modeling the back contact with a continuum of states one readily achieves this continuous transition of \( X^{−}_f \) to \( X^{−} \) \cite{104, 142}. Nevertheless, the most intense line at low temperature is \( \hbar \omega_{upper} (∆V_g) = E_{i,−} − E_{f,-}, \) and is the main structure of the spectrum at low temperatures. Importantly, this main structure closely resembles the experimental spectrum. We observe in Fig. 4.12 that our simple calculation reproduces the main features of the hybrid excitons: (i) there is a smooth transition between \( X^0 \) and \( X^{−} \); (ii) we obtain the strong tunnel induced bending of the \( X^0 \) and \( X^{−} \) energy at the high voltage edge of the \( X^0 \) plateau and in the beginning of the \( X^{−} \) plateau. This bending is especially important for the \( X^0 \) exciton at the end of \( X^0 \) plateau since it gives the lowering of the \( X^0 \) energy due to the binding of the QD exciton and the Fermi sea; (iii) the optical strength of \( \hbar \omega_{upper} \) strongly weakens in the transition region between the \( X^0 \) and \( X^{−} \) plateaus, as is shown experimentally in Fig. 4.6(a). Of course, these calculations only provide guidance to the understanding of the many-body interactions in our system. It is known that the Kondo effect is very delicate and becomes rapidly destroyed with temperature \cite{103} and, by no means, we can consider our comparison between theory and experiment in Fig. 4.12 as a proof for the formation of the Kondo exciton. Measurements at lower temperatures and in magnetic field are clearly needed as well as exact numerical calculations for the spectral function of the exciton at finite temperatures.

### 4.6 Summary

In conclusion, we have shown that the coupling between the QD states and the continuum of states of the Fermi sea gives rise to new optical transitions,\(^{10} \) Actually there are even more levels when the initial state \( E_{i,−} \) is taken into account. However, at \( T = 4.2 \text{K} \) the energy splitting between \( E_{i,−} \) and \( E_{i,−} \) is much larger as \( k_B T \)

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manifesting the formation of many-body exciton states. At the low voltage end of the neutral and positively charged exciton complexes we observe relatively broad lines following the lever arm of the device, which result from the recombination of a hole in the QD with electrons in the the back contact. Moreover we observe the smooth continuous transition of the $X^0$ to the $X^-$ plateau and the bending of these plateaus as a result of the strong hybridization between the QD state and the Fermi sea of electrons at the back contact. The observed lines can be well explained within the Anderson and Mahan exciton models. Our study demonstrates the possibility to investigate a variety of many-body states in QDs coupled with a Fermi sea and opens the way to investigate the Kondo effect and spin related phenomena in these systems.

4.7 Outlook

The investigation the interaction of localized states in QDs with a sea of electrons/holes is a new area of research and consequently many features in the spectra are up to now not understood. Therefore it is necessary to study in more detail these many-body interactions. For this purpose, thinner tunnel barriers are needed to achieve a stronger coupling between the electrons in the dots and the back contact. Of course the same experiments can be performed with a Fermi sea of holes by growing a $p$-doped back contact. Moreover, the
density of states at the back contact is only quasi two-dimensional due to the relatively thick $n$-doped region. In order to have a better defined Fermi sea in the back contact a two dimensional electron (hole) gas can be used. This would also result in the observation of the Landau levels in the Mahan line shape in magnetic field. Furthermore, it is interesting to study the effect of the QD confinement and the width of the tunnel barrier on the coupling strength of the dots and the back contact as well as on the exchange splittings. Of course a large number of other experiments, such as time-resolved, correlation and absorption measurements are needed to study the dynamics in these systems.
CHAPTER 4.
Chapter 5

Negative diamagnetic shift for charge tunable self-assembled InAs/GaAs quantum dots

5.1 Abstract

In order to use semiconductor quantum dots for manipulating spin and charges it is crucial to understand the Coulomb and exchange interaction between charge carriers confined in a dot and how an external magnetic field is influencing these interactions. The Coulomb and exchange interactions have been investigated by measuring the photoluminescence (PL) of QDs embedded in charge tunable devices, which allow to tune the charge state of the excitons in the dots [105, 143]. It has been shown that for neutral excitons, the exciton energy increases with $\alpha d B^2$ (neglecting the Zeeman splitting), where the diamagnetic coefficient is proportional to the area of the wave function (see chapter 3). However, the behavior of charged excitons is less well-known and potentially much more interesting because of the more elaborate Coulomb interactions [144]. In this chapter we report on the observation of two types of negative diamagnetic shift observed in the photoluminescence of self-assembled InAs/GaAs quantum dots of the same sample as studied in chapter 4.

Type A negative diamagnetic shift is only present for the highly charged exciton complexes of several quantum dots. For these dots the neutral, positively and lower negatively charged excitons exhibit a positive diamagnetic shift, whereas we observe a negative quadratic diamagnetic shift for the highly negatively charged exciton complexes for magnetic fields up to 8 T. This behavior can qualitatively be understood by a larger extension of the final state as compared to the initial state [145, 146] and is of similar origin as the negatively diamagnetic shift observed for the negatively charged trion in quantum wells [147, 148].

Most surprisingly, a second type of negative diamagnetic shift of different origin is observed for a small percentage of the QDs. For these QDs we can
clearly identify the \( X^0 \) and \( X^- \) plateau. In a magnetic field both these exciton complexes show a strong linear paramagnetic behavior. Furthermore we observe a peculiar gate voltage dependence of the \( X^- \) plateau in a magnetic field. The origin of this type B paramagnetic behavior is yet to be explained.

5.2 Type A negative diamagnetic shift

In the past it was shown that QDs with intermediate confinement have a smaller diamagnetic coefficient for the negatively charged trion \( (X^-) \) as compared to the neutral exciton \( (X^0) \) [146]. For the regime of weak confinement, as is the case for our QDs, it has been predicted that the negatively charged exciton complexes can even show a negative diamagnetic dispersion [145]. Experimentally, a weak paramagnetic dispersion for the \( X^- \) has been observed in quantum wells [149, 150, 147, 148] but up to now not for quantum dots. In this section we will discuss the negative diamagnetic shift for highly negatively charged exciton complexes, which we will refer to as type A negative diamagnetic shift.

The magneto-optical properties of charge-tunable self-assembled InAs/GaAs quantum dots are investigated, using the same sample as discussed in chapter 4. With the confocal microscope setup we measure the polarization dependent photoluminescence (PL) for the different (charged) exciton complexes in magnetic fields up to 10 T in the Faraday configuration with the magnetic field parallel to the growth direction. Fig. 5.1(a) shows a voltage dependent PL spectrum at \( B = 0 \) T. The PL spectrum shows many similarities with the ones reported in chapter 4; the \( X^0_M \) and \( X^0_H \) features are present as well as the characteristic \( X^0 \), \( 2X^0 \) and \( 3X^0 \) lines around \( V_g = -0.1 \) V. For the higher negatively charged exciton complexes the line widths become wider and less intense. The increase in line width is more clearly shown in Fig. 5.1(b), which shows the normalized PL spectrum at each gate voltage.

In order to analyze the PL lines as function of gate voltage in more detail, the spectra for the different exciton complexes at the gate voltages corresponding to the dashed lines in Fig. 5.1(a) are shown in Fig. 5.1(c). The energy difference between the different exciton complexes is a result of the different Coulomb interactions within each exciton complex. In general it is observed that the energy red shift between \( X^0 \) and \( X^- \) is largest, in agreement with other reports [105, 120]. The PL line width of the different exciton complexes increases from \( \sim 183 \mu eV \), \( \sim 246 \mu eV \), and \( \sim 182 \mu eV \) for the \( X^+ \), \( X^0 \), and \( X^- \) respectively to \( \sim 486 \mu eV \) and \( \sim 850 \mu eV \) for the \( X^{2-} \) and \( X^{3-} \). So for the negatively charged exciton complexes with one or more electrons in the p-shell the line width is broadened. We attribute this broadening to the strong hybridization of these states with the Fermi sea at the back contact. We cannot exclude that the broadening is also a result of dephasing and Auger processes in the highly charged exciton complexes, as it is hard to separate this from the broadening due to hybridization. Using the Heisenberg uncertainty relation, \( \Delta E \Delta \tau \approx \hbar \), we estimate the dephasing time \( \tau \sim ps \), which is in agreement for the dephasing time in a 2DEG [151]. Moreover it is comparable to the time scale on which Auger processes [152] and shakeup processes [114] occur. Nevertheless, the asymmetric line shape, as is shown more clearly using the straight orange line in Fig. 5.1(c), is a signature of the shakeup processes in the final state at the back contact in the same way as described in chapter 4. Moreover,
Figure 5.1: (a) The measured PL spectrum of a quantum dot as function of gate voltage. The different exciton complexes are indicated. The diamagnetic shift and the Zeeman splitting are analyzed at voltages corresponding to the dotted lines. (b) The same contour plot where at each voltage the spectrum is normalized. In this representation it is clearly shown that the line width of the highly charged exciton is increasing for increasing voltage. The logarithmic scale bars relates to the detector counts. (c) The spectra of the differently charged exciton complexes. Clearly the line width of the $X^{2-}$ and $X^{3-}$ is larger as compared to the $X^+, X^0$, and $X^-$. We attribute the line width broadening to the strong hybridization of the highly negatively charged exciton states with states at the back contact. The dashed orange line is used to show the asymmetric line shape with the low energy tail for the $X^{3-}$. 
we note that the $X^{3^-}$ in Fig. 5.1(b) exhibits instead of a "stable" flat plateau, a monotonous decrease in energy with increasing voltage. We relate this behavior to the increased hybridization of the $X^{3^-}$ complex with increasing gate voltage; as the tunnel barrier height between the Fermi sea of electrons and the QD decreases with increasing gate voltage, the hybridization of the electrons at the Fermi sea and the QD electrons increases, which results in the lowering of the PL energy.

We exclude the hybridization of the states with the WL. These hybridized states would show a strong energy dependence in the magnetic field as was reported for the $X^{3^-}$ in Ref. [153], and we do not observe this. Furthermore, the WL energy is 140 meV above the ground state luminescence of these dots, for which we expect only a very weak hybridization with the QD states. Moreover, we estimate from the lever arm and the onset of the $X^0$ plateau that the WL gets populated at $V_g = +0.9 \, \text{V}$.

Figure 5.2(a) shows the contour plot of the PL as function of gate voltage at $B = 10 \, \text{T}$. Surprisingly, the shape of the $X^{3^-}$ plateau is altered considerably.

Figure 5.2: (a) The normalized contour plot of the studied QD at $B = 10 \, \text{T}$ shows that the $X^{3^-}$ plateau has changed shape as compared to Fig. 5.2(b).
(b) The FWHM of the $X^{3^-}$ PL as function of $B$ increases for increasing $B$. 
NEGATIVE DIAMAGNETIC SHIFT FOR CHARGE TUNABLE SELF-ASSEMBLED INAS/GAAS QUANTUM DOTS

Figure 5.3: (a) The PL spectra for the neutral exciton $X^0$ at $V_g = -0.1$ V for magnetic fields up to 10 T in 1 T steps. (b) The PL spectra for the neutral exciton $X^{3-}$ at $V_g = 0.36$ V for magnetic fields up to 10 T in 1 T steps. (c) The diamagnetic shift as function of the magnetic field for the $X^{3-}$ is negative for magnetic fields up to 8 T, whereas the diamagnetic shift of the $X^0$ (shown in the inset) displays a normal quadratic behavior as function of the magnetic field. (d) The exciton $g$-factor for both exciton complexes displays a linear behavior in magnetic field. For $X^{3-}$ we find $g_{ex} = -1.69 \pm 0.03$ and for $X^0$ we obtain $g_{ex} = -1.60 \pm 0.02$. 

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Instead of a monotonous decrease of the "instable" $X^{3-}$ plateau with increasing $V_g$ (see Fig. 5.1(b)) we now observe a "stable" flat plateau. Apparently the magnetic field results into a stronger confinement of the $X^{3-}$ in the quantum dot. The enhancement of the magnetic field induced localization of charge carriers to potential fluctuations is well-known. In Fig. 5.2(b) we show the FWHM of the $X^{3-}$ at $V_g = 0.36$ V as function of the magnetic field. Clearly, for an increasing magnetic field the FWHM of the PL peak increases from 0.9 meV to 2.1 meV. This could again be a result of the stronger confinement of the carriers in the dot: the stronger confinement results to an enhanced overlap of the different charge carrier wave functions and thus for example to a reduced time scale of Auger processes, giving rise to the additional broadening.

Figure 5.3(a) and (b) show the PL spectra for magnetic fields up to 10 T for the $X^0$ and $X^{3-}$ for both circular polarizations. In both cases the upper line has $\sigma^-$ polarization giving rise to the same sign of the exciton $g$-factor. Analyzing the diamagnetic shift shows the normal quadratic behavior for the $X^0$. Remarkably, for the $X^{3-}$ we observe a negative diamagnetic shift for magnetic fields up to 8 T, which is relatively well described by a quadratic dependence up to 10 T. This effect is of similar strength as observed for quantum wells ($\sim 200 \mu$eV), but survives up to much higher magnetic fields as compared to the quantum wells ($\sim 2$ T) [147].

A possible reason for the observed paramagnetic lines, is depicted in Fig. 5.4 for the $X^{3-}$. In the initial state the Coulomb interaction between the hole and the four electrons causes the exciton to have a certain lateral extend $l_{X^{3-}}$. After recombination the final state consists of three electrons. The absence of an additional hole in the final state results in a stronger repulsion between the remaining three electrons; effectively the Coulomb repulsion in the initial state was screened by the hole present in the dot. Due to the shallow character of the QDs this repulsion gives rise to a much larger lateral extend of the final state $l_{3e}$ as compared to lateral extend in the initial state $l_{X^{3-}}$: $l_{3e} > l_{X^{3-}}$. Of course,
the diamagnetic shift also scales with the inverse reciprocal mass of the exciton complexes. This reciprocal mass is different for the initial and final state as the number of particles in the initial and final state is different as well as that the particles experience a different part of the composition. Therefore the actual physical picture is more intricate, the more because there is also considerable hybridization with the back contact. The main message is that for our QDs the Coulomb interaction is dominating over the QD confinement potential giving rise to the negative diamagnetic shift. In the case of strong QD confinement, the lateral extend of the initial and final state is mainly determined by the QD confinement and hardly by the Coulomb interaction. In this confinement regime the difference in lateral extend between initial and final state is small and will effectively result in $\alpha_d > 0$. In the strong confinement regime the Coulomb interaction can be treated as a perturbation to the single particle energies $[12]$, and the diamagnetic shift of the negatively charged exciton $X^{n-}$ is proportional to $\alpha_d B^2$ for relatively weak magnetic fields, and where $\alpha_d > 0$ (see chapter 3). In the regime of weak confinement the Coulomb energies are equal to or dominate over the single particle energies. It can be shown that in this regime the diamagnetic coefficient is proportional to $\alpha_d \propto l_i^2/m_i - l_f^2/m_f$ $[145, 146]$, where $m_i (m_f)$ is the reduced mass of the exciton complex in the initial (final) state. In fact, this expression states that in the regime of weak confinement we in general expect $\alpha_d < 0$, when $m_f < m_i$. In general, our QDs are in the intermediate regime and therefore we find $\alpha_d < 0$ only for the highly charged exciton complexes and only for a few quantum dots. For the studied dot the diamagnetic shift becomes positive again for $B \sim 8$ T. Using the simplified picture sketched in Fig. 5.4 we can understand this: the additional magnetic confinement results in the decrease of the extension of the final state (and eventually also of the initial state), such that $l_i^2/m_i > l_f^2/m_f$. In fact, this result shows that for shallow quantum dots we can tune the strength of the Coulomb interaction with respect to the total confinement energy with an external magnetic field such that one of the two dominates.

Another possible origin for the observed negative diamagnetic shift at relatively low magnetic fields is related to the strong hybridization with the back contact. In this picture the applied magnetic field would influence the coupling between the QD and the back contact and thereby reducing or enhancing the hybridization. This could lead to a negative diamagnetic behavior of the strongly hybridized $X^{3-}$. Further study, both theoretically and experimentally, is needed to unambiguously determine the origin of the type A negative diamagnetic shift.

### 5.3 Type B negative diamagnetic shift

For some QDs we observe a strong linear negative diamagnetic shift for the neutral exciton and negatively charged trion. The observation of a linear negative diamagnetic shift of the neutral exciton is highly surprising, as in a two particle picture the $X^0$ cannot display a negative diamagnetic shift. We will refer to this negative diamagnetic shift as type B negative diamagnetic shift and in this section we will analyze a typical QD exhibiting this behavior and shortly discuss its possible origins.

A contour plot of several QDs is shown in Fig. 5.5. In this section we will focus on the PL lines of QD C indicated with the circle, which shows a strong
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Figure 5.5: A contour plot of the PL as function of gate voltage $V_g$ for a several QDs (A-D) at $B = 0$ T and for the $\sigma^-$ polarization. The number of counts is per 5 s. The PL lines in the dashed circle belong to one quantum dot, which exhibits a type B negative diamagnetic shift. The dashed straight line indicates the lever arm of the device; for QDs emitting at different energies the onset of the $X^0$ plateau occurs at different voltages. The $X^0$ of the QD C is shifted to higher voltages by the amount of $\Delta V \sim 0.25$ V.

resemblance with the $X^0$ and $X^-$ lines observed for the majority of QDs (see chapter 4 and Fig. 5.1). We made sure by moving the sample laterally that the PL lines of QD C and QD B are not related. Surprisingly, the gate voltage at which these plateaus occur are shifted to higher voltages as compared to neighboring QDs emitting at similar energies. In general, QDs emitting at lower (higher) energy the onset of the $X^0$ and other plateaus is determined by the lever arm $\lambda$ of the device. From the growth we expect a lever arm of $\sim 7.8$ (see chapter 4), but by analyzing the onset of the $X^0$ plateaus for the different dots emitting at different energies (see dashed line Fig. 5.5) we obtain a lever arm of $\lambda \sim 9$. Importantly, a shift in gate voltage of $\Delta V = 0.25$ V is observed for the onset of the $X^0$ line of QD C, which we are going to study in detail. In general, the PL plateaus of the dots exhibiting the type B negative diamagnetic shift are shifted to more positive gate voltages as compared to the dots which do not exhibit this behavior. The value of this shift varies from dot to dot.

Figure 5.6(a) shows a contour plot of the PL lines of QD C as function of gate voltage at $B = 0$ T. Remarkably, we do not observe any biexciton or positively
Figure 5.6: Contour plot of the PL of QD C exhibiting a type B negative diamagnetic shift for $B = 0 - 7$ T in 1 T steps (a-h) for $\sigma^-$ polarization. The PL intensity is plotted for each magnetic field on the same scale.
charged excitons for this dot. These complexes have also not been observed for the other quantum dots that exhibit the type B linear negative diamagnetic shift. Apparently these dots have a very shallow potential for the holes, which prevent more than one hole to be present in the quantum dot.

The contour plots of the PL lines of QD C in magnetic fields up to 7 T are shown in Figs. 5.6(a-h) detected for the $\sigma^-$ polarization, which is the high energy part of the Zeeman doublet. Several striking features are observed for increasing magnetic fields. Most importantly, the $X^0$ and $X^-$ display a strong negative diamagnetic shift. The diamagnetic shift $E_{\text{dia}} = \frac{E(\sigma^+) + E(\sigma^-)}{2}$ as function of $B$ is shown in Fig. 5.7(a) for both the $X^0$ and $X^-$ plateau. For both plateaus the PL energy is taken at the center of the plateau. For the studied QD the behavior of $E_{\text{dia}}$ is well described by a linear dependence on the magnetic field. We note that for all dots displaying the type B negative diamagnetic shift this shift is linear in magnetic field. For this diamagnetic shift we define the diamagnetic coefficient $\beta$ via $E_{\text{dia}}(B) = \beta B$. For the $X^0$ and $X^-$ of QD C we find $\beta \approx -38 \mu eV/T$, whereas similar values are found for the other dots. The Zeeman splitting is shown in Fig. 5.6(b) and is linear in magnetic field, as is commonly observed, with a value of the exciton $g$-factor which is typical for InAs/GaAs quantum dots (see chapter 3). We remark that both for several of the quantum dots discussed in chapter 3 and for some of the quantum rings discussed in chapter 8 we observe a similar linear paramagnetic behavior. In fact, in Fig. 8.10(b) several lines are present in the contour plot showing the type B behavior.

By comparing Figs. 5.6(a-h) it is observed that the shape of the $X^-$ plateau changes drastically: for higher magnetic fields the $X^-$ plateau resembles more the commonly observed $X^-$ plateau. For all dots exhibiting this behavior we observe the change of the plateau shape of the $X^-$ in magnetic field. Moreover, the voltage extend of the plateau is also increased with increasing $B$, as is shown in Fig. 5.7(c). Whereas the voltage extend $\Delta V_{\text{plateau}}$ is constant for the $X^0$, we observe an increase of 150% of the $X^-$ plateau. Apparently, the additional confinement provided by the external magnetic field confines the exciton stronger in the QD thereby stabilizing the $X^-$ plateau. The weak PL from the plateau indicated with the arrow in Fig. 5.6(a) disappears as the $X^-$ plateau gets more extended.

To summarize, for dots exhibiting a type B linear negative diamagnetic shift we observe a strong negative linear diamagnetic shift for the $X^0$ and $X^-$ plateaus, whereas the Zeeman splitting is comparable to normal dots. For increasing magnetic field the plateau of the $X^-$ gets extended and changes shape. As compared to dots which do not exhibit this behavior, we find a gate voltage offset for the the charging plateaus and no highly negatively charged, positively charged and biexciton complexes. In the next subsection we will shortly discuss the most probable origin of this type B negative diamagnetic shift behavior.

### 5.3.1 Discussion

A strong negative linear diamagnetic shift has been observed for shakeup lines in quantum wells [147, 154, 155, 156]. In general, the lines originating from shakeup processes are relatively weak as this process is a second order process. Instead of detecting a weak PL intensity, we are able to observe even the weak $X^0_H$ feature in Fig. 5.6(a-h). Moreover, as has been shown in chapter 4, shakeup processes result into broad lines as the life time of the final state is only short
Figure 5.7: (a) The diamagnetic shift $E_{\text{dia}} = \frac{E(\sigma^+) + E(\sigma^-)}{2}$ at the center of the $X^0$ and $X^-$ plateau of the QD in Fig. 5.6(a-h). A linear behavior in magnetic field is observed for $B > 1$ T with $E_{\text{dia}}(B) \propto \beta B$. For $X^0$ ($X^-$) we find $\beta = -38.6 \pm 0.2\mu$eV/T ($\beta = -38 \pm 2\mu$eV/T). (b) The Zeeman splitting for $X^0$ and $X^-$ shows the normal linear behavior with $g_{ex} = -1.5 \pm 0.1$ and $g_{ex} = -1.74 \pm 0.05$, respectively. (c) The voltage extend of the $X^0$ and $X^-$ as function of $B$. Whereas the plateau length of the $X^0$ remains constant, the $X^-$ plateau length increases by 150%.
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[113, 135]. Opposite to what is expected from the shakeup process we observe sharp PL lines. We therefore exclude shakeup processes as the origin of the type B negative diamagnetic shift.

Furthermore, as we observe a similar type B negative diamagnetic shift for different nanostructures not implemented in a charge tunable structure, we exclude the influence of the back contact and the 2DHG at the interface with the blocking barrier as the origin of the type B negative diamagnetic behavior.

The relative voltage shift ∆V indicates the possible presence of a charged impurity in the proximity of the quantum dot. It is known that during the growth of these structures always a low p-doped background is present in the MBE chamber. Depending on the charged state of the impurity, the PL line of the quantum dot diffuses spectrally. However, we do not observe any spectral diffusion in our PL spectra, which would signify that our impurity stays in the same charged state. The p-doping near or in the QD could explain the absence of the positively charged excitons and biexciton and could result in a shift in gate voltage. However, the influence of a charge impurity close by or even in the dot is an intricate problem, but in general does not change the energy levels in the QD this significantly. Moreover, the presence of an impurity could not explain the negative diamagnetic shift of the X0 up to now.

The origin of the strong linear negative diamagnetic shift is still not understood. Nevertheless, the model to explain this behavior should include an additional charging mechanism in the proximity of the dot to explain the relative voltage shift and the absence of several exciton complexes. The linear behavior as function of the magnetic field indicates the presence of Landau levels, which are formed in a 2DEG and 3DEG in relatively low magnetic fields [157, 64]. Therefore it is likely that the observed PL lines are a result of a localized state with, for example, a two-dimensional state in the WL. As a last remark, the proposed model should be able to explain the stabilization of the X− plateau in magnetic field. This stabilization is a result of the additional confinement by the magnetic field on top of the weak confinement by the QD potential and implies that indeed an extended state is involved.

5.4 Summary

Two different types of negative diamagnetic shift for individual charge tunable InAs/GaAs quantum dots have been reported. The type A negative diamagnetic shift occurs only for the highly negatively charged exciton complexes. The weak confinement of these dots causes the Coulomb interactions to be dominant over the single particle energies, which eventually results in a negative diamagnetic shift for relatively small magnetic fields. By applying a magnetic field we can effectively tune the proportion between the Coulomb and confinement energies. This behavior is in line with predictions for quantum dots [145] and has been reported for quantum wells [147].

The type B negative diamagnetic shift shows a strong linear paramagnetic behavior for magnetic fields up to 10 T. For increasing magnetic field the plateau of the X− gets extended and changes shape. Therefore, again the additional confinement by the magnetic field plays a crucial role to understand the underlying physics of this observed behavior. Moreover, the same behavior seems to be present for the dots analyzed in chapter 3 and for the rings in chapter 8. There-
fore it is not a sample related phenomenon, but rather a generally occurring event. The possible origin requires further investigation.

5.5 Outlook

To understand the origin of the type B diamagnetic shift, experiments have been proposed in magnetic fields up to 30 T. In these high magnetic fields the question will be resolved whether the paramagnetic behavior continues to be linear in magnetic field or whether a critical magnetic field is needed to observe the normal quadratic diamagnetic shift. Moreover, PLE measurements and time-resolved measurements on these lines can shed a light on the dynamics of these PL lines and thereby help to identify the underlying processes.
Chapter 6

Size dependent exciton $g$-factor in self-assembled InAs/InP quantum dots

6.1 Abstract

We have studied the size dependence of the exciton $g$-factor in self-assembled InAs/InP quantum dots. Photoluminescence measurements on a large ensemble of these dots indicate a multimodal height distribution. Cross-sectional Scanning Tunneling Microscopy measurements have been performed and support the interpretation of the macro photoluminescence spectra. More than 160 individual quantum dots have systematically been investigated by analyzing their magnetoluminescence between 1200 nm and 1600 nm. We demonstrate a strong dependence of the exciton $g$-factor on the height and diameter of the quantum dots, which eventually gives rise to a sign change of the exciton $g$-factor. Moreover, we find a size dependent anisotropy splitting of the exciton emission in zero magnetic field.¹

In order to provide a qualitative and quantitative understanding on the trend between $g_{ex}$ and the emission energy an eight band $\mathbf{k} \cdot \mathbf{p}$ approximation is applied to calculate the electron and hole $g$-factors separately as function of the emission energy. The observed correlation between exciton $g$-factor and the size of the dots is in good agreement with calculations [158].

6.2 Experimental results

6.2.1 Introduction

Self-assembled quantum dots are one of the most promising candidates to be used as building blocks in quantum information processing [26, 32, 159, 41].

¹These results have been published as: size-dependent exciton $g$ factor in self-assembled InAs/InP quantum dots, N. A. J. M. Kleemans, J. van Bree, M. Bozkurt, P. J. van Veldhoven, P. A. Nouwens, R. Nötzel, A. Yu. Silov, P. M. Koenraad, and M. E. Flatté, Phys. Rev. B 79, 045311 (2009).
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For instance, single-qubit operations have been proposed by changing the local effective Zeeman interaction in a quantum dot [33, 34] (see also chapter 1). Control over the exciton $g$-factor ($g_{ex}$), defined by Eq. 6.2, is thus highly desirable for the realization of individual qubits [160]. Moreover, a sign change of the exciton $g$-factor is desirable in quantum information processing and thus there is a strong interest in quantum dots having an intrinsic exciton $g$-factor of zero. To investigate the size, shape and composition dependence of the electron, hole and exciton $g$-factor, theoretical investigations using the $k\cdot p$ approximation [89, 90, 66] as well as tight binding calculations [161, 162] have been performed on InAs/GaAs dots. The self-assembly process of quantum dots gives rise to a distribution in size, shape and composition of the dots and therefore leads to a dot to dot variation of $g_{ex}$. This opens the possibility to utilize the growth conditions to engineer $g_{ex}$ [35]. Up to now experiments on InAs/GaAs QDs revealed only a weak correlation between emission energy and $g_{ex}$ [38, 90].

In this section we will study the magnetoluminescence of InAs/InP quantum dots, which are tuned to 1.5 $\mu$m [163, 164, 165, 166]. The experimental results show that the engineering of the exciton $g$-factor is feasible at telecommunication wavelengths.

6.2.2 Sample growth and characterization

(A) Growth

Our quantum dots are grown by Metal-Organic Vapor-Phase Epitaxy (MOVPE). A layer of 100 nm of InP has been grown on a $n$-doped InP (100) substrate with a two degrees miscut towards the [110] direction. Two monolayers (ML) of GaAs were deposited as an interlayer, thereby reducing the As/P exchange reaction. On top of this interlayer a 2 ML InAs layer is grown, resulting in the formation of quantum dots. The quantum dot layer is capped by 200 nm of InP. For Atomic Force Microscopy (AFM) a layer of surface quantum dots was grown under the same conditions. From the AFM measurements we find an average height of the dots of $(2\pm1)$ nm and a dot diameter of $(34\pm5)$ nm. More details about the growth of these wavelength-tunable InAs quantum dots in InP can be found in Ref. [164].

(B) Macro photoluminescence

The sample is characterized by temperature dependent PL measurements performed on a large ensemble of dots. The quantum dots are excited by a laser operating at 532 nm with a spot size of $\sim 4$ mm$^2$. The macro PL is detected by an InGaAs array up to 1550 nm and with an InSb single channel detector above 1400 nm, using the setup described in chapter 2. The spectra taken at different temperatures are shown in Fig. 6.1. These spectra are plotted by matching at 1450 nm the spectra obtained by both detectors. Instead of a single Gaussian distribution, characteristic for highly homogeneous quantum dots, a series of peaks (P1-P9) is observed. The spectrum at $T = 4.5$ K displays strong similarities with the ones reported in Refs. [81, 82]; the peaks were identified

\footnote{This sample has been grown by P. J. van Veldhoven, and R. Nötzel in the group Photonics and Semiconductor Nanophysics at the Eindhoven University of Technology. The sample number is MO12B.}
Figure 6.1: PL spectra of a large ensemble of quantum dots measured at different temperatures. A multiple peak structure is observed consisting of 9 peaks. The peak positions at $T = 4.5\,\text{K}$ are indicated by the dotted lines. We attribute the multiple peak structure to the multimodal height distribution of the dots. Quantum dots having the smallest height have luminescence around peak P1.

Figure 6.2: X-STM characterization of InAs/InP quantum dots of (a) 3 BL (6±1 ML), 5 BL (10±1 ML), and (b) 4 BL (8±1 ML) height. The bright contrast corresponds to InAs, whereas the dark contrast corresponds to GaAs. The distribution of the different heights of the dots is given in (c). The inset in (a) shows the typical disk shape of our dots. The X-STM measurements were performed by M. Bozkurt.
as quantum dots with discrete height differences of 1 ML and the dots were modeled accordingly. In the same way we attribute the different peaks to a multimodal height distribution of our dots. Quantum dots emitting around P9 at the low energy side of the spectrum have the largest height, whereas dots emitting at the high energy side have the smallest height. The width of the peaks is due to the dot to dot variation of the diameter and composition. The structure present in peak P3 is most likely related to water vapour absorption. A redistribution of carriers over the dots having different heights occurs for increasing temperatures. At elevated temperatures the excitons in the QDs with smaller height thermally escape and to the higher dots, where they recombine.

(C) Cross-sectional Scanning Tunneling Microscopy

To characterize the dot size, shape and composition we performed Cross-sectional Scanning Tunneling Microscopy (X-STM), as was recently done on similar dots [167]. The measurements have been performed in constant current mode. Three different quantum dots are shown in Figs. 6.2(a) and (b). The images were obtained at a voltage of $-3$ V. At these voltages the contrast is mainly caused by topographic effects due to strain induced surface relaxation [168]. The bright contrast corresponds to InAs with the largest lattice constant and the dark contrast is identified as the GaAs interlayer with the smallest lattice constant. From these measurements we determine the height of the dots with bilayer (BL) precision. Note that in X-STM individual ML cannot be distinguished. Fig. 6.2(a) shows two different dots having a height of 3 and 5 BL and Fig. 6.2(b) shows a dot with a height of 4 BL, which correspond to a height of $(6\pm 1)$ ML, $(10\pm 1)$ ML and $(8\pm 1)$ ML respectively. For more than 50 dots the height was measured and the resulting distribution is shown in Fig. 6.2(c). The quantum dots best resemble circular discs, as depicted in the inset of Fig. 6.2(a), and therefore we assume that the height of the dot is independent of where the dot is cleaved. The height distribution shows that we have quantum dots with heights varying between 5 and 15 ML, which matches quite well with the 9 peaks we observe in the macro PL. A height of 5 ML would then correspond to dots belonging to macro PL peak P1. Moreover, most dots have a height between 7-9 ML corresponding to the part of the PL spectrum which is most intense (P3-P5).

The X-STM images also show that the lateral sizes of the quantum dots are less well defined. The largest diameter found by X-STM is 30 nm and corresponds to the value found by AFM [168]. The GaAs interlayer is not located between the InAs dot and the InP substrate, but the InAs dots are rather embedded in the GaAs layer. Although the GaAs layer suppresses the As/P exchange reaction, the actual role of this layer in the growth of these dots is still a matter of further investigation. There appears to be no strong intermixing of Ga and P inside the quantum dot and therefore we conclude that our dots consist of almost pure InAs. For all the studied dots comparable compositions are found. Furthermore, Figs. 6.2(a), (b) show that the dot formation preferentially takes place at the step edges introduced by the miscut of the substrate.

6.2.3 Magnetoluminescence of individual quantum dots

In order to study the PL of individual quantum dots we use an aluminium mask on top of the sample, with openings varying between 500 nm and 1400 nm.
Figure 6.3: PL of three individual quantum dots showing from left to right a positive exciton $g$-factor, a quenched $g$-factor and a negative $g$-factor. The spectra are shown for magnetic fields of 0 T, 5 T and 10 T in the Faraday configuration.

Most measurements have been performed on openings of 1 $\mu$m. The excitation is provided by a 635 nm wavelength cw laserdiode. We studied quantum dots emitting between 1200 nm and 1600 nm using the confocal microscopy setup described in chapter 2. The PL was analyzed in the Faraday configuration in magnetic fields up to 10 T aligned parallel with the growth direction [68, 146]. The polarization is analyzed using an achromatic quarter wave plate and a linear polarizer. The luminescence was dispersed by a 75 cm monochromator and detected by an InGaAs array. The linewidth varies from dot to dot, and is of the order of 100 $\mu$eV, limited by the quantum dot linewidth itself. In order to exclude biexciton luminescence we performed power dependent measurements and excluded all lines with a superlinear dependence on the excitation density [87].

(A) Correlation between emission energy, exciton $g$-factor and diamagnetic shift

The emission energy $E(B)$ of an exciton in a quantum dot in a magnetic field $B$ is in good approximation given by (see chapter 3):

$$E(B) = E_0 \pm \frac{1}{2} g_{ex} \mu_B B + \alpha_d B^2,$$

(6.1)

where $E_0$ is the emission energy at $B = 0$ T, $\mu_B = +5.79 \times 10^{-5}$ eV/T is the Bohr-magneton, and $\alpha_d$ is the diamagnetic coefficient. The second term of Eq. 6.1 is the Zeeman term which gives rise to a spin induced splitting of the exciton PL in a magnetic field, whereas $\alpha_d$ is linked to the exciton radius.
Figure 6.4: The exciton $g$-factor as function of the emission energy $E_0$ for 164 quantum dots. A sign change of $g_{ex}$ is observed for dots emitting at low energies. The quantum dots having a small height have a more negative $g$-factor as compared to dots having a large height. Moreover, dots having both a small height and a small diamagnetic coefficient $\alpha_d$ (blue stars), i.e., small lateral size, have the largest negative $g$-factor. The colors represent different intervals of $\alpha_d$, and correspond to the colors as shown in the histogram in Fig. 6.5.

The magnetoluminescence spectra of three individual quantum dots emitting at different energies are shown in Fig. 6.3 for magnetic fields of $B = 0$ T, 5 T and 10 T. We observe a clear sign change of the polarization of the Zeeman splitted lines for the low energy quantum dot as compared to the high energy dot. Moreover, for the quantum dot emitting around 850 meV we observe no Zeeman splitting at all for magnetic fields up to 10 T. All three dots exhibit a diamagnetic shift towards higher energies for increasing magnetic field. In order to analyze the data we define $g_{ex}$ as:

$$g_{ex} = \frac{E(\sigma^+) - E(\sigma^-)}{\mu_B B}. \quad (6.2)$$

Figure 6.3 shows, from left to right, a dot with $g_{ex} > 0$, $g_{ex} \simeq 0$ and $g_{ex} < 0$. In order to verify the sign of $g_{ex}$, we also measured control samples with known $g_{ex}$ in a given direction of the magnetic field and known angle between the axes of the quarter lambda plate and the linear polarizer. To reveal the relation between $g_{ex}$ and the emission energy we investigated the exciton $g$-factor of in
Figure 6.5: The diamagnetic coefficient as function of the emission energy. There is only a weak correlation between the diamagnetic coefficient and the emission energy. The inset shows the histogram of the different values of $\alpha_d$. Blue corresponds to small values of $\alpha_d$, white to the average values of $\alpha_d$, and red to the large values of $\alpha_d$.

The dependence of $g_{ex}$ on $E_0$ is shown in Fig. 6.4. A strong correlation between $E_0$ and $g_{ex}$ is observed. At large emission energy the exciton $g$-factor changes its sign and becomes increasingly negative. The exciton $g$-factor changes from +0.5 to −2 for dots emitting at 775 meV to 1050 meV. Since the emission energy of the dot is mainly determined by the height of the dot, as is inferred from the macro PL, the dots having a smaller height have a more negative $g_{ex}$.

From the magnetic field dependence of the exciton lines we also extract the diamagnetic coefficient $\alpha_d$, which is to good approximation proportional to the spatial extension of the exciton wave function, and is therefore a measure for the lateral size of the dot [169] (see also chapter 3). To verify that the emission energy is mainly determined by the height of the dot, we plot $\alpha_d$ against the emission energy in Fig. 6.5. There is only a weak correlation between emission energy and the diameter of the dots. We therefore conclude that the change from positive to negative values of $g_{ex}$ is governed by the quantum dot height. The weak correlation between $E_0$ and $\alpha_d$ indicates that dots of smaller (larger) height have on average a smaller (larger) lateral size. Fig. 6.4 shows that quantum dots emitting at the same energy have a large variation of the diamagnetic shift. In
Figure 6.6: The exciton $g$-factor as function of the diamagnetic coefficient for different emission energies $E_0$. There is a strong correlation between $\alpha_d$ and $g_{ex}$. The filled red symbols correspond to the emission range indicated in the separate graphs. The emission intervals correspond to the discrete peaks P1-P6 in the macro PL spectrum and thus to dots of different height. The lowest dots which have a smallest lateral size have the most negative exciton $g$-factor.
order to analyze the importance of the lateral size of the dot on $g_{ex}$, we specify in Fig. 6.4 three different ranges of $\alpha_d$. These ranges are determined from the distribution of $\alpha_d$ as shown in the inset of Fig. 6.5, and correspond to quantum dots with small (blue), average (white) and large (red) $\alpha_d$. We find that dots emitting at the same energy, but having a smaller lateral size, have a more negative $g_{ex}$. Thus reducing the size of the dots, i.e. either height or diameter, will result in more negative values of $g_{ex}$.

The relation between $\alpha_d$ and $g_{ex}$ is plotted in Fig. 6.6. We find a strong correlation between $\alpha_d$ and $g_{ex}$. In general there is an increase of the exciton g-factor for increasing $\alpha_d$. The filled red symbols in the different panels correspond to different emission wavelengths, which correspond to the energy ranges around the peaks in the macro PL. The filled symbols in the lower right panel correspond to emission energies around peak P1 of the macro PL data and correspond to the dots lowest in height. Quantum dots of the same height, but of smaller diameter, have a more negative $g_{ex}$. Figure 6.6 thus shows that quantum dots having the smallest diameter and height, i.e. the overall smallest size, have the most negative $g_{ex}$. Increasing the size of the dot results in a sign change of $g_{ex}$, where the dots with the overall largest size (filled symbols in the upper left panel) have the most positive $g_{ex}$.

Up to now we assumed that the change in the emission energy of the dots did not arise from the change in composition of the dots. We can exclude that composition plays a large role as X-STM did not show significant compositional variations over the different dots. Moreover, the influence of the composition on InAs/GaAs dots has been addressed in several papers, which conclude that there is only a small effect on $g_{ex}$ [89].

(B) Anisotropy splitting

Analysis of the single dot spectra showed anisotropy splittings ($\Delta E_{as}$) for 24 quantum dots with a magnitude up to 250 µeV, whereas the other dots did not have a resolvable anisotropy splitting. The measured values of $\Delta E_{as}$ are comparable with those found for InAs/GaAs quantum dots [68]. As an example a contour plot of the magnetoluminescence of a quantum dot with $\Delta E_{as} = 160$ µeV is shown in Fig. 6.7(a). Recently, there has been discussion about the origin of this splitting [80], but it is generally believed to arise from the asymmetry of the footprint of the dot [88, 77, 170]. To demonstrate the dependence of $\Delta E_{as}$ on the quantum dot size, we plot $\Delta E_{as}$ as a function of the emission energy in Fig. 6.7(b). In this analysis we only treat the subset of quantum dots that exhibit an anisotropy splitting resolved in our experiments. As shown in Fig. 6.7(b), dots having a smaller height, i.e. larger $E_0$, have in general a larger anisotropy splitting. We believe this is due to the fact that for quantum dots of lower height the exciton wave functions are more squeezed in the lateral directions. Therefore they are more sensitive to the asymmetry of the footprint of the dot, resulting in larger values of $\Delta E_{as}$. Nevertheless, higher dots are still sensitive to the confinement potential asymmetries when they have a large lateral size. This is depicted in Fig. 6.7(b) by making a distinction between dots which have a small and large $\alpha_d$. The anisotropy splitting for the higher dots is only observed for dots having a large diamagnetic coefficient ($\alpha_d > 7$ µeV/T²). In general we find that both small and large lateral sizes give rise to an anisotropy splitting for quantum dots of lower height. It should be noticed that the anisotropy splitting
Figure 6.7: (a) Contour plot of the magnetoluminescence of a dot showing an anisotropy splitting of $\Delta E_{as} = 160 \, \mu eV$ at $B = 0 \, T$. The blue (red) color corresponds to low (high) PL intensity. The peak positions used in the fitting procedure are indicated with the circles and are fitted by the lines using Eq. 6.1. For this particular dot $g_{ex} = (-1.00 \pm 0.09)$ and $\alpha_d = (7.1 \pm 0.2) \, \mu eV/ T^2$. (b) The anisotropy splitting $\Delta E_{as}$ of in total 24 quantum dots as function of their emission energy $E_0$. The filled (empty) circles indicate dots having a small (large) diamagnetic coefficient.
does not occur for the negatively and positively charged exciton, which supports our assumption that we are considering the neutral exciton.

6.3 Theoretical results

In this section an understanding is provided on the correlation between $g_{ex}$ and the emission energy based on the concept of orbital momentum quenching [66, 171]. This concept is best illustrated by discussing spherical symmetric InAs nanocrystals, placed between 10 eV high barriers. These high barriers prevent the leakage of the wave function outside the nanocrystals. Moreover, the InAs nanocrystals are unstrained. Therefore the complicating factors of shape, composition and strain are canceled in these calculations and only the quantum dot confinement will have its influence.

The Roth formula can be used to predict the behavior of the electron $g$-factor $g_e$ in bulk material [172, 173], and is given by:

$$g_e = g_0 - \frac{2E_p}{3E_g} \left( \frac{\Delta}{E_g + \Delta} \right),$$

(6.3)

where $E_p$ is the interband coupling between the conduction and valence band, known as the Kane energy [174, 175], which is similar for the different III-V semiconductor compounds; $E_p \sim 21$ eV. The dependence of $E_p$ on the band gap energy $E_g$ and the spin-orbit coupling $\Delta$ can be neglected. In general, the value of the electron and hole $g$-factor are mainly determined by the interband couplings, which are determined by $E_p, E_g,$ and $\Delta$. Equation 6.3 actually reflects that for small interband couplings, i.e. large band gaps or small spin-orbit couplings, the electron $g$-factor is the free electron $g$-factor $g_0$. The Roth formula is used to represent $g_e$ of a quasi InAs-like bulk material with a varying bandgap. The result for $g_e$ as function of the emission energy ($E_g$) is shown in Fig. 6.8 for the Roth formula (red dotted line) and for calculated values of $g_e$ using eight band $k \cdot p$ theory [66]. In these calculation the radius of the nanocrystals is varied from 20 to 2 nm to span the energy range between 420 – 980 meV. The value of $g_e$ varies between the bulk InAs value of -14.6 and the free electron value of +2 for small nanocrystals. The trend of $g_e$ predicted by the Roth formula is due to the decoupling of the conduction and valence band by increasing the bandgap. In the limit of $E_g \to \infty$, both the Roth formula and the calculated values of $g_e$ result in the free electron $g$-factor. However, at intermediate emission energies, the calculated $g_e$ approaches the free electron value faster compared to the predictions of the Roth formula. This is solely due to the quantum confinement, which is the only difference between the calculations and the formula, and this can be explained in the context of the orbital momentum quenching. The interband coupling results in an effective orbital momentum of the electron states in the conduction band due to the intermixing with the states in the valence band [66, 69]. This orbital momentum is calculated for the quantum confinement case, shown by the green line in Fig. 6.8. The quantum dot confinement causes the quenching of the orbital momentum. This can be understood as follows. The ground state, either in bulk or in the nanocrystal, is always a mixture of all eight bands due to the interband couplings. The confinement in a nanocrystal will tend to modify this mixture such that the ground state becomes more and more dominated by the conduction band states. Therefore the nature of the
confined states change by confinement. Confinement will quench the orbital momentum gained by the admixture of the valence band states into the ground state. The modification by the confinement is proportional to $1/\delta E$, where $\delta E$ is the energy separation between the ground state and first excited state [66]. A large (small) nanocrystal has a small (large) $\delta E$ and therefore a large (small) admixture of the different interband states resulting in a large (small) orbital momentun.

An eight band $\mathbf{k} \cdot \mathbf{p}$ approximation is applied to calculate the electron and hole $g$-factors for the InAs/InP quantum dots as function of the emission energy in order to have both a qualitative and quantitative comparison between the experimental data and the calculated values of $g_{ex}$. The model includes strain and assumes a homogeneous InAs disk (as shown in the inset of Fig. 6.2(a)) surrounded by a matrix of InP. The details of the calculations can be found elsewhere [69, 158], and here only the results on the electron, hole and exciton $g$-factors are discussed.

The electron $g$-factor as function of the emission energy $E_0$ is shown in Fig. 6.9 for different radii. The emission energy is altered by changing the height of the quantum dots from 4.5 nm to 1.5 nm, corresponding to the lowest and highest emission energy, respectively. This height variation is in agreement with the different heights measured by X-STM. The electron $g$-factor is overall

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Figure 6.8: The electron $g$-factor $g_e$ of strain-free InAs nanocrystals as function of the emission energy obtained by the Roth formula (dotted red line). The value starts from the bulk InAs value and quenches to the free electron value at high emission energies. For the calculated values of $g_e$ (red solid line) the free electron value is obtained for much lower emission energies. The quenching of the orbital momentum is shown by the green line. The figure is adapted from Ref. [66].

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The calculation have been performed in collaboration with C. E. Pryor and M. E. Flatté, Department of Physics and Astronomy and Optical Science and Technology Center, University of Iowa, Iowa, USA.
Figure 6.9: The calculated electron $g$-factor $g_e$ of as function of the emission energy for InAs/InP quantum dots with various radii. The emission energy is determined by the height of the quantum dots and varies from 1.5 nm to 4.5 nm, corresponding to the highest and lowest emission energy, respectively. The trend is comparable to the InAs nanocrystals and can be understood in terms of orbital momentum quenching.

varying between $-4$ and $+0.5$ in an energy range of 700 – 1250 meV and is not dependent on the radius of the dots. In contrast, the height of the quantum dot is dominating the tendency of $g_e$ on the emission energy. This can be understood in terms of the orbital momentum quenching: a larger quantum dot confinement energy causes a larger orbital momentum quenching. As the confinement energy is mainly dominated by the height, the electron $g$-factor is relatively unsensitive to the radius.

As shown in Fig. 6.10, the hole $g$-factor $g_h$ has a zero (Fig. 6.10(a)) and second order (Fig. 6.10(b)) component on the magnetic field; i.e. the hole $g$-factor is given by $g_h = g_{h0} + g_{h2} B^2$. The zero-order hole $g$-factor $g_{h0}$ has a strongly dependence on the radius\(^4\) and determines the value of $g_h$. The non-linear behavior of the Zeeman splitting, due to the second order hole $g$-factor $g_{h2}$, has been observed in experiments on bulk and quantum wells \cite{176, 177} and is a result of the admixture between heavy hole and light hole states. At zero magnetic field the heavy hole and light hole bands are split by strain and confinement. The splitting between the heavy hole and light hole state is dependent on the magnetic field and therefore the nature of the confined hole states depends on the magnetic field, causing $g_h$ to be non-linear, as shown in Fig. 6.10(c). The overall magnitude of the $g_{h0}$'s of InAs/InP is smaller as compared to InAs/GaAs quantum dots (compare values with Ref. [171]). The reduced strain in InAs/InP quantum dots relative to InAs/GaAs quantum dots reduces the splitting of the

\(^4\)The calculation of the hole $g$-factor is complicated and it is therefore difficult to give a "hand-waving" argument for this strong dependency.
Figure 6.10: (a) The calculated hole $g$-factor $g_h$ has a zero order component $g_h^0$, which is linear in magnetic field and (b) a second order component $g_h^2$, which is quadratic in magnetic field. The second order component is due to the heavy hole-light hole admixture in magnetic field. The inset shows the calculated Zeeman splitting for the holes for a dot of 11 nm radius for different heights in nm. There is a clear deviation from the linear dependence on the magnetic field.
Figure 6.11: The calculated exciton $g$-factor $g_{ex} = g_0^h - g_e$ as function of the emission energy for various sets of radii. The emission energy is again varied by changing the height of the dots. The offset between the different sets of radii is dominated by $g_h$, although the main trend is governed by the contribution of $g_e$. The experimental data points (black squares) and the calculated values of $g_{ex}$ display the same correlation with the emission energy: $g_{ex}$ decreases with increasing emission energy.

heavy hole and light hole band edges in the dot, and thus there is more light hole character in the hole ground state of an InAs/InP quantum dot than in that of an InAs/GaAs quantum dot. As the light hole $g$-factor is less negative than the heavy hole $g$-factor, this effect leads to less negative $g_n$’s in InAs/InP dots than in InAs/GaAs dots.

In order to calculate the exciton $g$-factor, we only take into account the linear contribution of the $g_0^h$ as in the experiments the non-linearity has not been observed for magnetic fields up to 10 T. The exciton $g$-factor $g_{ex}$ is plotted as function of the emission energy in Fig. 6.11. Whereas the difference between the subsequent sets of radii originates from $g_0^h$, the trend is governed by the $g_e$. The black points in Fig. 6.11 correspond to the experimentally obtained values of $g_{ex}$ and are well-described both qualitatively and quantitatively for dots with a radius of 13 – 15 nm, in agreement with the values found by X-STM.

6.4 Summary

Macro PL and X-STM measurements showed that the studied InAs/InP dots have a multimodal height distribution. Single quantum dot luminescence, carried out on a large number of dots, showed a strong correlation between exciton $g$-factor, diamagnetic coefficient and emission energy. The strong dependence
of $g_{ex}$ on the emission energy results in a sign change of the exciton $g$-factor. The trend in $g_{ex}$ is mainly governed by the height variation. We demonstrated that the value of $g_{ex}$ is correlated with the diamagnetic coefficient and conclude that dots with a large diameter have a smaller $g_{ex}$. In general, dots having a smaller overall size will have a more negative $g_{ex}$ as compared to quantum dots of larger overall size. We even showed that for several quantum dots the exciton $g$-factor is quenched. This opens the possibility to evenly tune the sign of $g_{ex}$ by using for instance electric fields. We also observed anisotropy splittings for InAs/InP quantum dots, and demonstrated that low dots can give rise to a larger anisotropic splitting. We conclude that quantum dots with large height and small lateral size are the most suitable candidates to be used as an entangled photon source, since this application relies on dots having small anisotropy splittings. [26]

In order to provide a qualitative and quantitative understanding on the trend of $g_{ex}$ with the emission energy an eight band $k \cdot p$ model is applied to calculate the electron and hole $g$-factors as function of the emission energy. The behavior of $g_{e}$ is well-understood within the context of orbital momentum quenching and is due to the variation of the height of the dots, whereas the hole $g$-factor $g_{h}$ is dominated by the diameter of the dots. The calculations showed that the trend of $g_{ex}$ with emission energy is due to $g_{e}$, whereas the value of $g_{ex}$ is determined by $g_{h}$. A quantitative agreement between the calculations and experiments is obtained for dots with a 11 $-$ 13 nm radius [158]. Our experimental and theoretical study gives a detailed insight into the exciton $g$-factor in quantum dots and opens a possibility of engineering and controlling the $g$-factor in individual quantum dots.

6.5 Outlook

As is shown in Fig. 6.2, the GaAs interlayer is not underneath the QDs but rather surrounding the InAs quantum dots. New X-STM measurements on samples containing different GaAs interlayer thicknesses can give more insight in the growth dynamics of these dots. Moreover, the GaAs can also influence the value of the exciton $g$-factor. Therefore the model needs to be extended in order to investigate the effect of the interlayer on the $g$-factor and diamagnetic shift.

The first measurements on charge tunable InAs/InGaAsP quantum dots have been performed.\(^{5}\) Up to now the charging of quantum dots compatible with the telecommunication wavelengths have only been reported for InAs/InP quantum dots positioned on pyramidal nanotemplates [178]. A typical spectrum as function of the gate voltage $V_g$ is shown in Fig. 6.5 for an excitation power of 0.20 mW, showing clearly the different charging plateaus of one or possibly two dots. The challenge now becomes to identify the different charging plateaus. For this, new measurements are needed which also determine the polarization of the lines. This will help to distinguish between the neutral exciton and the singly charged exciton by analyzing the fine structure splitting. Moreover, the behavior of the different exciton complexes in magnetic field will result in an extended description of the energy level structure in these quantum dots.

\(^{5}\)In the case of InAs/InP quantum dots it is not possible to grow a blocking barrier (a semiconductor compound with a larger bandgap) on top of the structure.
Figure 6.12: A contour plot of the PL as function of the gate voltage $V_g$ for an excitation power of 0.20 mW. The color scale relates to the detector counts per 60 s. The PL was dispersed by the single stage of 750 nm with the 750 grooves/mm grating and detected by the InGaAs array detector.

In principle this system also allows to investigate many-body effect for these quantum dots, as was done for the InAs/GaAs quantum dots (see chapter 4), by positioning the quantum dots more closely to the back contact.
CHAPTER 6.
Chapter 7

Electronic behavior in self-assembled InAs/GaAs quantum rings in magnetic fields

7.1 Abstract

In quantum mechanics, particular attention is paid to phenomena occurring due to the phase coherence of charge carriers in doubly-connected (ring) topologies. Electrons confined to a submicron ring manifest a topologically determined quantum-interference phenomenon, known as the Aharonov-Bohm (AB) effect [179, 180], as a result of the oscillatory behavior of their energy levels as a function of an applied magnetic field. This behavior is usually associated with the occurrence of oscillatory persistent currents in the ring [181, 182, 183]. Experimental evidence for AB oscillations has been detected in the mesoscopic regime in metallic [184, 185, 186] and semiconducting [187, 188] rings, containing many electrons. We address the occurrence of the AB effect in defect-free self-assembled semiconductor nanostructures [76, 99, 105, 189, 190]. The ability to fill nanostructures with only a few (1-2) electrons offers the unique possibility to detect magnetic field induced oscillations in the persistent current carried by single electron states. In this chapter the behavior of electrons in self-assembled InAs/GaAs quantum rings is investigated by measuring the magnetization of an ensemble of quantum rings in which each ring is populated with 1 – 2 electrons.¹

We report the first direct measurement by means of ultra-sensitive magnetization experiments of the oscillatory persistent current carried by a single electron in self-assembled InAs/GaAs “volcano-like” nanostructures. Remarkably, this single electron current occurs even in the absence of an opening [50] in our nanostructures, which is required for the AB effect in the standard treatment.

The magnetic field at which the first oscillation in the magnetic moment arises is much higher than expected from the diameter of the quantum rings as determined by AFM [190]. However, the experiments are in good agreement with a model based on the structural parameters as determined with X-STM measurements [191].

The origin of the AB effect and its signatures in magnetization are discussed in the next section. The growth of self-assembled InAs/GaAs quantum rings, the structural analysis by AFM and X-STM, and the sample design are discussed in the section 7.3. The following section discusses the torque magnetometer setup. Section 7.5 reports the experimental results on the AB effect in these rings. The model based on the X-STM analysis on these nanostructures is described in section 7.6 and is in qualitative agreement with the experiments. The chapter is concluded by a brief summary.

7.2 The Aharonov-Bohm effect

Electrons confined to a submicron ring manifest a topologically determined quantum-interference phenomenon, known as the Aharonov-Bohm (AB) effect, as a result of the oscillatory behavior of their energy levels as a function of an applied magnetic field. The phenomenon was first predicted by Y. Aharonov and D. Bohm in 1959 [179] and was observed for the first time in a superconducting ring by Tonomura et al. [192]. In order to understand the AB effect we first discuss the case of a magnetic field only concentrated in the center of a ring. We treat the ring as an one-dimensional wire, bent to a circle of radius $R$. This situation is shown in Fig. 7.1.

Although the electrons confined to the ring are not affected by the magnetic field itself, they are affected by the vector potential of the magnetic field. This is a consequence of the Hamiltonian in quantum mechanics, which contains vector potentials instead of the actual fields. The single electron Hamiltonian in the presence of a magnetic field is given by Eq. 3.2. We will now follow the same approach\(^2\) as in Ref. [181]. To simplify matters we set the electronic confinement potential to zero. In absence of a magnetic field, $\mathbf{B}=0$, we calculate the wave function $\psi$ of an electron in an one-dimensional ring. This wave function only

\(^2\)Ref. [183] gives a clear alternative representation of the AB effect in rings.
depends on the azimuthal angle $\phi$, so $\psi = \psi(\phi)$. Using spherical coordinates the Schrödinger equation leads to:

$$\frac{-\hbar^2}{2m^*_e} \nabla^2 \psi(\phi) = -\frac{\hbar^2}{2m^*_e} \frac{1}{R^2} \frac{\partial^2}{\partial \phi^2} \psi(\phi) = E \psi(\phi)$$  \hspace{1cm} (7.1)

The wave function is single valued and therefore the periodic boundary condition $\psi(\phi) = \psi(\phi + 2\pi)$ applies. The energy values $E$ are quantized because of this periodic boundary condition. This leads to the discrete energy values:

$$E_{m_z} = \frac{m_z^2 \hbar^2}{2m^*_e R^2}, \quad m_z = 0, \pm 1, \pm 2, \pm 3, \ldots$$  \hspace{1cm} (7.2)

with the corresponding wave functions $\psi(\phi) = \frac{1}{\sqrt{2\pi}} \exp\left(\frac{i m_z \phi}{\hbar}\right)$. The energies and wave functions are labeled with quantum number $m_z$, analogues to the projection of the angular momentum quantum number along the $z$-axis \([11]\), sometimes denoted as magnetic quantum number.\(^4\)

Now the wave functions and corresponding energies in the presence of a magnetic field are calculated. The magnetic field shown in Fig. 7.1 is assumed to be homogeneous and time independent and directed perpendicular to the plane of the ring. At the rim of the ring no magnetic field is present, since it is only penetrating through the opening of the ring. This leads to $B = 0$ at the rim of the ring, resulting\(^5\) in $A = \nabla \chi$. The magnetic flux $\Phi$ penetrating the ring is, using Stokes’s theorem, related to $\chi$ by:

$$\Delta \chi = \oint A \cdot d\mathbf{l} = \int \int B \cdot d\mathbf{S} = \Phi$$  \hspace{1cm} (7.3)

A new wave function $\psi'$ is defined:

$$\psi' = \psi \exp\left(\frac{i e}{\hbar} \chi\right)$$  \hspace{1cm} (7.4)

The advantage of this representation is the elimination of the vector potential $\mathbf{A}$ in the Schrödinger equation \([193]\). Using definition 7.4 together with the periodic boundary condition leads to a relative change in the phase of the wave function given by:

$$\frac{\psi'(\phi)}{\psi'(\phi + 2\pi)} = \frac{\psi(\phi) \exp\left(\frac{i e}{\hbar} \chi(\phi)\right)}{\psi(\phi + 2\pi) \exp\left(\frac{i e}{\hbar} \chi(\phi + 2\pi)\right)}$$  \hspace{1cm} (7.5)

The original wave function $\psi$ is single valued and applying Eq. 7.3 the relation between $\psi'(\phi)$ and $\psi'(\phi + 2\pi)$ is given by:

$$\psi'(\phi) = \psi'(\phi + 2\pi) \exp\left(2\pi \frac{\Phi}{\hbar \Phi_0}\right),$$  \hspace{1cm} (7.6)

\(^3\) Spherical coordinates: $\nabla^2 = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial}{\partial r}\right) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial}{\partial \theta}\right) + \frac{1}{r^2 \sin^2 \theta} \frac{\partial^2}{\partial \phi^2}$. For the one-dimensional ring the wave function only depends on the azimuthal angle $\phi$ and the polar angle $\theta = \pi$ and $r$ is equal to the radius of the ring $R$. Therefore $\nabla^2$ simplifies to $\nabla^2 = \frac{1}{R^2} \frac{\partial^2}{\partial \phi^2}$.

\(^4\) The solution of the azimuthal part of the Schrödinger equation in spherical coordinates leads to $\exp\left( i m_z \phi \right)$, with $m_z$ the magnetic quantum number. This solution is identical to the solution of the Schrödinger equation of an one-dimensional ring.

\(^5\) See chapter 3, and from vector calculus: $\nabla \times \nabla \chi = 0$
where the elementary flux quantum \( \Phi_0 = \frac{h}{2e} \) is introduced. The consequence of the gauge transformation is thus a change in the phase of the wave function \( \psi' \) when the azimuthal angle \( \phi \) changes by \( 2\pi \). This additional phase picked up by the electron gives rise to a persistent current. Moreover from Eq. 7.6 a periodic dependence of \( \psi' \) on \( \Phi \) is obtained with period \( \Phi_0 \). In general \( \psi'(\phi) \) is given by:

\[
\psi'(\phi) = \frac{1}{\sqrt{2\pi}} \exp(i m_z \phi) \exp(i2\pi \Phi \Phi_0 / 2\pi)
\] (7.7)

The corresponding energy levels are:

\[
\mathcal{E}_{m_z} = \frac{\hbar^2}{2m^*_e R^2} (m_z + \Phi / \Phi_0)^2, \quad m_z = 0, \pm 1, \pm 2, \ldots
\] (7.8)

The energy levels as function of the magnetic flux penetrating the center of the ring are given in Fig. 7.2. In the case that the magnetic field is oriented along the positive \( z \)-axis, as in Fig. 7.2, the value of \( m_z \) becomes more negative for increasing \( B \). The transitions in \( m_z \) occur for rational values of the magnetic flux given by \( \Phi = (p + \frac{1}{2})\Phi_0 \), with \( p = 0, \pm 1, \pm 2, \ldots \).

The transitions in angular momentum quantum number are intimately related to jumps in the magnetization. The relation between the energy and the magnetization \( M \) of a many particle system is given by one of the thermodynamical Maxwell relations:

\[
M = - \frac{\partial F}{\partial B} |_{N,T},
\] (7.9)

where \( F \) is the total free energy, \( N \) the total number of electrons and \( T \) the temperature. In the case of an electron in a one-dimensional ring and at zero temperature the free energy is equal to the energy given in Eq. 7.8. In Fig. 7.3 the magnetic moment of a single electron in a one-dimensional ring \( \mu \) is plotted.

Figure 7.2: Energy of an one-dimensional ring as function of magnetic flux penetrating the center of the ring, \( \mathcal{E}_{m_z} = \frac{\hbar^2}{2m^*_e R^2} (m_z + \Phi / \Phi_0)^2 \). At the values of the magnetic flux \( \Phi = (p + \frac{1}{2})\Phi_0 \) the lowest energy state changes from angular momentum quantum number \( m_z \) to \( m_z - 1 \).


The magnetic moment $\mu$ against the magnetic flux penetrating the center of the ring. The magnetic moment abruptly changes sign at the values of the magnetic flux $\Phi = (p + \frac{1}{2})\Phi_0$ and is zero at $\Phi = p\Phi_0$, where $p$ is an integer.

The magnetic moment is zero for integer values of $\Phi_0$.

The different steps in the formation process of a self-assembled InAs QR on GaAs. The InAs quantum dot on GaAs (1) is capped with a 2 nm layer of GaAs (2). Annealing the structure leads to a migration of indium from the center of the dot outwards (3). Subsequently to the annealing step the QR is capped with GaAs giving rise to a second wetting layer (4).

against the magnetic flux. The linear relation between magnetic field and magnetic moment is expected; the energy depends quadratically on the magnetic flux and the magnetic moment is the derivative of the energy. The jumps in the magnetic moment appear for rational values of the magnetic quantum flux $\Phi_0$. The magnetic moment is zero for integer values of $\Phi_0$.

### 7.3 Growth and characterization

The studied quantum rings are created out of MBE grown quantum dots (see chapter 2) by some additional growth steps. The quantum dots are capped with a thin layer of GaAs (typically 2 nm) and are subsequential annealed. The growth of self-assembled InAs/GaAs QRs was first performed by García et al. [189]. The proces is schematically shown in Fig. 7.4. The annealing process

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6The quantum ring samples have been grown by A. G. Taboada, D. Granados, and J. M. García, CSIC, Instituto de Micro electrónica de Madrid, Spain.
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Figure 7.5: (a) An AFM image of QRs. The scanned area is $2.17 \times 2.17 \mu m^2$ and the z-range is 3.2 nm. The rings are elongated along the [110] direction. The inset shows an enlargement of an area with a length and width of $0.35 \times 0.32 \mu m^2$. (b), (c) X-STM images of different cross-sections of two stacked “nano-volcanoes”. The images reveal the presence of indium (bright spots) in the center of the nanostructures, and a clear difference in the height of the rim between the [110] and [1\bar{1}0] directions. The inset shows the three-dimensional model of the rings observed by X-STM. The X-STM measurements were performed by P. Offermans [50].
causes a migration of indium away from the center of the dot giving rise to a second wetting layer. The remaining parts of the quantum dot now form a quantum ring. For a detailed description of the growth method of QRs see Refs. [194, 195, 196].

Figure 7.5(a) shows an AFM image of QRs. The AFM shows QRs that are elongated in the [110] direction due to the preferable migration of indium in this direction [76]. Furthermore there is a variation of the height of the rim within each ring. The QR density of this sample is determined on $n_{QR} = 9 \times 10^9$ cm$^{-2}$. The outer size of the QRs is about 100 nm by 70 nm and an average height of 1 nm is obtained. The QRs have thus a larger diameter and smaller height as compared to the dimensions of QDs.\footnote{QDs have typically a diameter of 70 nm and a height of 5 nm}

It has been shown by Offermans et al. that the actual sizes found by X-STM for the rings capped with a thick layer of GaAs, are considerably different compared to the sizes found by AFM for uncapped rings [50], see also chapter 2. Most importantly, the shape of these nanostructures differs considerably from that of ideal rings in two respects: (i) the presence of indium in their center resulting in the absence of a hole in the nanostructures and (ii) a distinct anisotropy of the nanostructure, i.e. the height of the rim is larger in the [110] than in the [110] direction [76, 197]. From the X-STM measurements, shown in Fig. 7.5(b) and (c), we find a typical diameter of 23 nm and a height of 4 nm for the rings. These exact sizes are strongly dependent on the exact growth conditions and the sizes differ from nanostructure to nanostructure (a typical size distribution of $\sim 5\%$ is obtained from the homogenous broadening of the PL in chapter 8). The inset of Fig. 7.5(c) shows the three-dimensional model of the InAs quantum rings based on the X-STM measurements.

7.4 The torque magnetometer

The magnetization measurements are performed at the High Field Magnet Laboratory.\footnote{The measurements have been performed in collaboration with I. M. A. Bominaar-Silkens, U. Zeitler, P. C. M. Christianen, and J. C. Maan.} The HFML is committed to research in the highest available magnetic fields: up to 33 T of continuous magnetic field and 60 T of pulsed magnetic field.\footnote{web address: http://www.ru.nl/hfml/}

A torque magnetometer is used to measure the magnetization. The principle
Figure 7.6: Layer structure of the InAs/GaAs QR sample for the magnetization experiment, consisting of 29 layers of these self-assembled nanostructures. The QRs are located between two 24 nm GaAs layers. The repeated sequence contains a modulation doped layer, which provides electrons to the QR layers. Additional doped layers (blue/dark grey) are inserted to accommodate for the depletion towards the capping layer and the undoped substrate.

is based on a torque induced by the magnetization in a magnetic field. This torque is translated into a rotation, which is detected. The torque $\Gamma$ experienced by the magnetization $\mathbf{M}$ in a magnetic field $\mathbf{B}$ is given by 7.10:

$$\Gamma = \mathbf{M} \times \mathbf{B} + \mathbf{r} \times (\mathbf{M} \cdot \nabla)\mathbf{B},$$

(7.10)

with $\mathbf{r}$ the distance from the rotation axis to a place on the sample. When the sample is mounted on the rotation axis and placed in a homogeneous magnetic field, the second term in equation 7.10 vanishes. A torque will only be detected if the sample is magnetized anisotropically at an angle with respect to the magnetic field. The larger the angle of the sample normal $\mathbf{n}$ with respect to the magnetic field $\mathbf{B}$, the higher the torque. However, implementing the sample under an angle results in an in-plane magnetic field, whereas in most experiments a magnetic field perpendicular to the sample is preferred. For the used setup, an optimized situation is obtained for an angle of 10° of the sample normal with respect to the magnetic field. Figure 7.7 gives a schematic representation of the setup.

The sample is mounted on a 20 mm-long phosphorbronze wire, which is stretched between two epoxy posts. The torque is translated into rotation using Eq. 7.11:

$$\Gamma = \left( \frac{\pi R^4 G}{2l} \right),$$

(7.11)

10 An homogeneous magnetic field leads to $\nabla \mathbf{B} = 0$. In addition $\mathbf{r} = 0$ if the sample is mounted on the rotation axis.
where $R$ is the radius of the wire, $G$ is the torsional spring constant of the material and $l$ is the length of the wire. The radius of the wire is approximately 12.5 $\mu$m, which leads to a rotation of $1.4 \times 10^{-4}$ degrees/pNm. The torque wire is glued on a removable part of the magnetometer to allow easy access for sample mounting. As shown in Fig. 7.7 the rotation of the sample is measured using a laser. A laser diode with a wavelength of 790 nm and variable intensity is coupled to a fiber which enters the magnetometer. Here the light leaves the fiber and is focused on a quadrant detector using a mobile lens. The system can be aligned using two adjustable mirrors. The first mirror reflects the laser onto the sample and the second mirror reflects the beam coming from the backside of the sample onto the quadrant detector. In order to make the backside of the sample reflective, an aluminium coated glass plate is mounted underneath the sample. The quadrant detector consists of four fibers. Each of these fibers is connected to four identical silicon diodes, which are used to determine the intensity distribution of the laser among the fibers. From this intensity distribution a normalized coordinate of the laser spot is deduced, which is independent of fluctuations in the total laser intensity. The quadrant detector can resolve rotations as small as $3 \times 10^{-6}$ degrees.

A small coil is mounted underneath the sample serving as feedback coil. This feedback coil is connected with a current source. A current flowing through the feedback coil induces a magnetization given by 7.12:

$$|M| = NIA,$$

(7.12)

where $N$ is the number of windings, $I$ the current through the coil and $A$ the area of the coil. This feedback current is regulated such that the normalized coordinate is kept constant. Operation in this feedback mode has the important
advantage that by keeping the normalized coordinate constant, it immediately
gives the magnitude of the magnetization from the dimensions and number of
windings of the coil. We used a coil of diameter 6 mm made from 18 µm thick
Cu wire.

Unfortunately the movement of the sample is not only caused by the mag-
etization. Generally, it is necessary to use an active feedback to damp the
unwanted additional motion of the magnetometer due to the mechanical noise.
Mechanical coupling to the cryostat typically induces two types of resonances.
A slow movement of approximately 7 Hz belonging to the rotational eigenfre-
quency of the system and a fast movement of approximately 117 Hz belonging
to the vertical vibration eigenfrequency of the system. The vibrational motions
usually damp in seconds and are not always present whereas the rotational
motions are never completely absent and take minutes to dampen. An active
feedback is therefore aimed at damping the vibrations. A feedback current is
generated by a proportional-integral-differential (PID) controller. The input of
this PID controller is the normalized position X, consisting of a DC component
we wish to measure and two AC components with the frequencies of the two
different eigenfrequencies of the system. Nevertheless, the mechanical noise is
limiting the sensitivity of this setup. The sensitivity of the magnetometer is
\(2.8 \times 10^{-12} \text{ J/T, i.e. } 3 \times 10^{11} \mu_B \) at \( B = 14 \text{T} \). More details of the setup are
given in Ref. [200].

7.5 The magnetization measurements

The magnetization measurements are performed at \( T = 1.2 \text{K} \) and \( T = 4.2 \text{K} \) in
magnetic fields up to 15 T. The total magnetization of the sample is due to about
\(1.5 \times 10^{11} \) nanostructures with a total number of electrons \( N \sim 2.2 \times 10^{11} \). These
numbers are based on the AFM measurements, the sample size of \(7 \times 8 \text{ mm}^2\), the
Si flux during the growth, the thickness and the number of the doping layers.
Figure 7.8(a) shows the raw experimental data as a function of the magnetic
field \( B \) for \( T = 4.2 \text{K} \) and \( T = 1.2 \text{K} \). Over the entire magnetic field range we
observed a relatively large background signal, which is due to the substrate and
to dia- and paramagnetic materials close to the sample [201, 202, 203]. On top
of this background, one clearly observes an oscillation of the magnetization at
a magnetic field of about 14 T. To enhance the visibility of this oscillation, a
linear background is subtracted from the signal. Furthermore we normalized
the signal to the total number of electrons \( N \) in the sample, resulting in the
magnetic moment per electron \( \mu = M/N \) (Fig. 7.8(b)). The mechanical noise
is about 8% of the experimentally observed oscillation magnitude.

We performed several tests to ensure that the oscillatory signal does origi-
nate from the magnetic moments of the ensemble of nanostructures. First, we
polished away the epilayers of the sample, leaving behind only the substrate, and
repeated the magnetization experiment. This measurement yielded a smooth
background magnetization curve without an oscillation around 14 T. Secondly,
we performed Shubnikov-de Haas experiments by measuring the magnetoresis-
tance of the sample up to magnetic fields of 21 T. We found a high value of
the resistivity without any oscillatory behavior, which proves that there are no
free carriers in the epilayers. Finally, we verified that the oscillatory signal is
not related to de Haas-van Alphen oscillations [157, 193]. By estimating the
Figure 7.8: (a) Experimental curve of the magnetization $M$ at $T = 4.2\, \text{K}$ and 1.2 K. An Aharonov-Bohm oscillation is observed around 14 T, superimposed on a smooth background. (b) Oscillation in the magnetic moment per electron ($\mu = M/N$), obtained at 1.2 K and at 4.2 K, after subtracting the linear background from the signal. The signal is divided by the total number of electrons $N$, and averaged over several measurements. (c) Calculated magnetic moment of a single electron in a nanostructure at different temperatures. The calculations are based on the structural parameters of the “nano-volcanoes”, as obtained by X-STM measurements, and accurately reproduce the Aharonov-Bohm oscillation at $B = 14\, \text{T}$ (see section 7.6).
electrons that would be necessary to reproduce the measured oscillation period, we found an electron density, which is two orders of magnitude larger than is expected from the doping levels. Therefore we conclude that the observed oscillations in the magnetization are caused by the electrons confined in the self-assembled nanostructures.

A first step to interpret this result is to compare our quantum ring to an ideal In$_{0.55}$Ga$_{0.45}$As ring with a similar diameter of 11.5 nm [50]. The electron energy $E^{(e)}$ spectrum as a function of the magnetic field, for such a ring, is shown in Fig. 7.9. The arrow indicates a changeover from the state with angular momentum $m_z = 0$ to the state with $m_z = -1$ as the ground state. The changeover of the ground state of an ideal ring of 11.5 nm induces a jump in the magnetization around 5 T. As we observe a distinct oscillation near 14 T, the simple model of an ideal ring is clearly not sufficient to describe our “volcano-like” nanostructures. Therefore we determine the energy spectrum as a function of the magnetic field of a more realistic model based on the X-STM characterization of the nanostructure [191].

7.6 The model

Based on the structural information of the QRs extracted from the X-STM measurements [50], the ring structure is modeled by a varying-thickness In$_x$Ga$_{1-x}$As layer embedded in a matrix of GaAs [191]. The bottom of the In$_x$Ga$_{1-x}$As layer is assumed to be perfectly flat and parallel to the $xy$-plane. The height of the In$_x$Ga$_{1-x}$As layer as a function of the radial coordinate $\rho$ and of the angular
coordinate $\varphi$ is modeled by:

$$h(\rho, \varphi) = h_0 + \frac{[\tilde{h}_M(\varphi) - h_0] \left\{ 1 - [\rho/R(\varphi) - 1]^2 \right\}}{\left\{ [\rho - R(\varphi)]/\tilde{\gamma}_0(\varphi) \right\}^2 + 1} \quad \rho \leq R(\varphi)$$  \hspace{1cm} (7.13)$$

and

$$h(\rho, \varphi) = h_\infty + \frac{\tilde{h}_M(\varphi) - h_\infty}{\left\{ [\rho - R(\varphi)]/\tilde{\gamma}_\infty(\varphi) \right\}^2 + 1} \quad \rho > \tilde{R}(\varphi)$$  \hspace{1cm} (7.14)$$

with

$$\tilde{h}_M(\varphi) = h_M \left( 1 + \xi_h \cos 2\varphi \right)$$  \hspace{1cm} (7.15)$$

$$\tilde{\gamma}_0(\varphi) = \gamma_0 \left( 1 + \xi_\gamma \cos 2\varphi \right)$$  \hspace{1cm} (7.16)$$

$$\tilde{\gamma}_\infty(\varphi) = \gamma_\infty \left( 1 + \xi_\gamma \cos 2\varphi \right)$$  \hspace{1cm} (7.17)$$

$$\tilde{R}(\varphi) = R \left( 1 + \xi_R \cos 2\varphi \right)$$  \hspace{1cm} (7.18)$$

Here, $h_0$ corresponds to the thickness at the center of the crater, $h_M$ to the rim height and $h_\infty$ to the thickness of the In$_x$Ga$_{1-x}$As layer far away from the ring-like structure. The parameters $\gamma_0$ and $\gamma_\infty$ define the inner and outer slopes of the rim, respectively. The parameters $\xi_h$, $\xi_\gamma$, and $\xi_R$ describe the ring-shape anisotropy. A typical shape of a QR is shown in Fig. 7.10 for $R = 11.5$ nm, $h_0 = 1.6$ nm, $h_M = 3.6$ nm, $h_\infty = 0.4$ nm, $\gamma_0 = 3$ nm, $\gamma_\infty = 5$ nm, $\xi_h = 0.2$, $\xi_\gamma = 0$, $\xi_R = 0$. In this particular case ($\xi_\gamma \xi_R = 0$), Eqs. 7.13 and 7.14 lead to the model considered in Refs. [50, 204].

The fraction $x$ of In$_x$Ga$_{1-x}$As is determined using the surface relaxation and lattice distortion of the cleaved surface observed in X-STM. This method is described intensively in Ref. [10] and the result of the determined indium gradient in the QRs can be found in Ref. [191]. For now we assume a homogeneous indium concentration of 55%. The shape of the ring and the concentration of indium in the ring determines the band parameters such as the effective mass and the band gaps. These parameters are used to calculate the potential of the ring. We use a model based on the Hamiltonian of an electron in a strained self-assembled nanostructure [205, 206] and include piezo-electric effects [207]. The strain tensor and the distribution of indium for the “volcano-like” geometry of the self-assembled nano-structures are calculated following a three-dimensional finite-element method of elasticity theory [204]. The Schrödinger equation is solved using the adiabatic approximation [208], separating electron degrees of freedom into “fast” (the motion along the growth axis) and “slow” (the in-plane motion) components. The calculated in-plane adiabatic potential $E_{11}^{(c)}$ for an uniform In$_{0.55}$Ga$_{0.45}$As is shown in Fig. 7.11. Note that for a non-uniform indium distribution the adiabatic potential is more complicated. The potential has a finite central maximum at $x = y = 0$, which makes the structure singly-connected, as distinct from a doubly-connected ideal ring. Furthermore, the potential possesses two pronounced minima at $x = \pm 11.5$ nm and $y = 0$ separated by potential barriers at $x = 0$ and $y = \pm 11.5$ nm, reflecting the anisotropy of the nanostructure.
CHAPTER 7.

Figure 7.10: Height of a QR as a function of the radial and the azimuthal coordinates as modeled by the Eqs. 7.13 and 7.14.

Figure 7.11: The adiabatic potential $E_1^{(c)}$ of a quantum ring with the same parameters as in Fig. 7.10 for an uniform indium concentration of 55%. The potential has valleys where the rims are higher and hills where the rims are smaller. The potential reflects the asymmetry of the ring. In the $xy$-plane the top view of the ring of Fig. 7.10 is shown.
With the anisotropic adiabatic potential $E^{(c)}_1$ as depicted in Fig. 7.11, the Schrödinger equation for the “slow” degrees of freedom determines the eigenstates of the in-plane motion. Finally, the electron energy eigenvalues in the nanostructure were obtained by diagonalizing the adiabatic Hamiltonian for the “slow” degrees of freedom in the basis of the in-plane wave functions with 20 radial and 25 azimuthal functions and using Eq. 7.9 the magnetization is calculated.

More details on the calculations can be found in Ref. [191]. The purpose of the following part of this section is to show how the parameters determine the magnetization behavior of the quantum rings and how the actual profile of the confining potential is essential to interpret the phase-coherent electronic properties of these nanostructures.

Figure 7.12 shows the magnetic moment calculated for quantum rings with different radii. The rings in our experiments are expected to have a diameter of approximately 11.5 nm. According to Fig. 7.12 this would correspond to a jump in the magnetization for fields lower than 10 T. For rings with large radii, the jump in the magnetization occurs at lower magnetic field compared to rings with small radii; obviously a ring with a larger inner area needs a smaller magnetic field to enclose one flux quantum. The decrease in amplitude is due to the asymmetry of the ring. This asymmetry of the ring leads to a localization of the electrons in the parts where the rim is highest (see Fig. 7.11). For larger rings the localization in the potential minima is stronger as the part of the ring with a higher potential gets more extended. Effectively this results in a decrease of the persistent current in larger rings as compared to smaller rings as it is more difficult for electrons to tunnel through the broader potential barriers in larger rings.

The effect of the ring-height anisotropy, described by the parameter $\xi_h$, and the ring-slope anisotropy, described by the parameter $\xi_\gamma$, on the oscillations of the calculated electron magnetic moment $\mu$ as a function of the magnetic field $B$ has been studied in Ref. [204]. Well-pronounced oscillations of $\mu(B)$
can be expected even for QRs with a strong shape anisotropy, provided that the width of the rim changes as a function of $\varphi$ in anti-phase with the rim height. Remarkably, this condition is satisfied for realistic QRs as characterized by X-STM, where the parameters $\xi_h$ and $\xi_\gamma$ have opposite sign, so that the height and the width of the rim vary with $\varphi$ in such a way that the effects of these variations on the cross-sectional area of the rim (in the $\rho z$-plane) partially compensate each other (see also Ref. [191]). However, the asymmetry of the ring results in the intermixing of states with different $m_z$. Therefore the energy levels are no longer indicated with the use of the angular momentum quantum number $m_z$, since in the asymmetric quantum craters this is no longer a quantum number. The lowest energy level consists of a series of angular momentum quantum numbers with $m_z = 0, -2, -4, \ldots$. Although the lowest state consists of a series of even angular momentum quantum numbers, the $m_z = 0$ nature is still strongest present. The second highest energy level consists of a series of odd angular momentum quantum numbers where the $m_z = -1$ behavior dominates. Moreover, the intermixing between odd and even states becomes possible when the $C_2$-symmetry of the quantum ring is broken. In our model we can still apply a rotation over $\pi$ resulting in the same quantum ring shape. In reality this will not be the case and the ground state will consist of a mixture of all angular momentum quantum numbers with a dominating $m_z = 0$ behavior of the ground state at low magnetic fields. However, as a result of the intermixing of the quantum levels, there are no longer any crossings of the energy levels and instead so called anti-crossings occur. Instead of an instantaneous transition of a state with even angular momentum quantum number to a state with odd angular momentum momenta, there will be a more gradual change of a state which is dominated by $m_z = 0$ to a state which is dominated by $m_z = -1$. Effectively this changes the jumps in the magnetization into smooth transitions. For now we will consider rings with $C_2$-symmetry.

In Fig. 7.13(a) (Fig. 7.13(b)) the energy levels without (with) the effect of strain are shown for an asymmetric quantum ring. The strain results from a lattice mismatch of the InGaAs QR material with the GaAs host matrix. The energy levels including strain have increased in energy as compared to the unstrained case. Since the potential for an electron in the strained QR is relatively shallow, there exist fewer discrete energy levels below the continuum of states in the GaAs barrier as compared to the unstrained case. Due to a reduced potential barrier at the center of a strained ring, the effective electron radius decreases when taking into account strain. Correspondingly, the transition magnetic fields, where the ground- and first excited electron energy levels interchange, are higher in a strained ring than in an unstrained one. The diamagnetic shift causes the energy levels to increase for increasing magnetic field.

The increase of the energy of the electron levels as a result of the strain, leads to a reduction in the magnitude of the variations of the adiabatic potential as a function of the azimuthal angle. As a result, at low magnetic fields, the mixing of electron states with different magnetic quantum numbers, which occurs due to the shape anisotropy of a QR, is weakened in a strained ring as compared to the case when strain is absent. The consequences of the weakening of state mixing are clearly seen in Fig. 7.13. At $B < 10$ T the energy spacing between the lowest electron state (which arises from the state with $m_z = 0$ in a circularly symmetric ring) and the first excited state (which arises from the state with $m_z = -1$ in a
Figure 7.13: Electron energy spectra in a QR at $T = 0$ K, calculated without (a) and with (b) effects due to strain. Energies are counted from the bottom of the conduction band in unstrained InAs. A dashed line, indicating the region of the continuum as obtained from our numerical simulation, is a guide to the eye.
circularly symmetric ring) is enhanced due to strain. Also the zero-field splitting between the first and second excited states (which correspond respectively to \( m_z = -1 \) and \( m_z = 1 \) in a circularly symmetric ring) is significantly reduced when taking into account the strain-induced effects.

The energy levels allow us to calculate the magnetization behavior of the rings using Eq. 7.9. The effect of strain on the magnetic moment \( \mu \) of an electron is given in Fig. 7.14. The magnetic moment both with and without taking into account strain are shown. Most importantly, our model accurately explains the position of the observed AB oscillation around 14 T, rather than at 5 T expected for an ideal ring of the same radius. The difference in the position of the AB oscillations is due to the influence of strain in the self-assembled “volcano-like” nanostructures as well as to the singly-connectedness of these nanostructures. At low fields the electronic ground state has a dominant component with angular momentum zero and is relatively sensitive to the confining potential. In an ideal ring of radius \( R \) the ground state wave function is concentrated at \( \rho = R \) and vanishes at the center of the ring (\( \rho = 0 \)), where the potential is infinitely high. For a singly-connected structure the adiabatic potential has a finite height and thus a non-zero wave function at \( \rho = 0 \). In comparison to an ideal ring, the electron density is therefore shifted towards the center, leading to a smaller effective radius and a higher magnetic field for the first AB oscillation. We compare our “volcano-like” nanostructure also with a disc-shaped quantum dot. Although both nanostructures are singly-connected, the behavior of the disc-shaped quantum dot is fundamentally different: the nature of the ground state does not change (compare Figs. 7.13(b) and 7.15) and consequently there are no AB oscillations due to the absence of a central maximum of the potential.\(^{12}\)

\(^{12}\)The potential of the quantum disc differs from the parabolic confinement potential, which is generally observed for quantum dots. Nevertheless, the behavior of the ground state energy does not show any crossings in both cases. We do note that due to the different confinement potential the second excited state is not triply, but only doubly degenerate, in contrast to
Figure 7.15: The electron energy spectrum $E^{(e)}$ as a function of $B$ for a disc-shaped quantum dot at $T = 0$ K. There are no crossings of the lower lying energy levels.

Therefore, the actual profile of the confining potential is essential to interpret the phase-coherent electronic properties of these nanostructures.

In Fig. 7.8(c) the calculated results are plotted for three different temperatures. Without including size variations of the nanostructures, the calculated amplitude of the AB oscillations increases with decreasing temperature [191]. However, the negligible temperature effect on the electron magnetic moment in our model (see Fig. 7.8(c)) is due to the nanostructure ensemble averaging of 5% in the radius. This explains the negligible temperature effect in the measurements (see Fig. 7.8(b)). We notice that the experimental value for the oscillation magnitude of the magnetic moment per electron, $\Delta \mu \sim 17 \mu_B$, is higher than the $4 \mu_B$, which we calculated based on the X-STM analysis. Calculations showed that the magnitude is sensitive to the exact structural properties, such as the indium concentration and the shape. A magnitude of $17 \mu_B$ at the observed transition magnetic field position is readily achieved for slightly modified nanostructures. It is realistic to assume that the nanostructures in the sample used for the magnetization measurements are somewhat different from those in the reference sample used for the X-STM characterization. Furthermore, the number of electrons confined in the quantum rings and the size and shape of the quantum rings determine the magnetization as a function of the magnetic field [209].

Figure 7.8(c) also shows the calculated magnetic moment for higher magnetic fields that are not yet accessible by magnetization experiments. Comparison of the higher order AB oscillations in realistic nanostructures with those in ideal quantum rings discloses two major differences. First, the ratio of magnetic fields, at which the two first jumps in the magnetic moment occur, is equal to 1:3 for ideal rings (Fig. 7.9) and is approximately 1:2 for the self-assembled “volcano-like” nanostructures. As stated above, a reduced effective radius of the electron state with $m_z = 0$ in the nanostructure leads to a pronounced shift of what has been observed by others (see chapter 8).
the first AB oscillation towards higher fields. This effect is less prominent for the states with higher angular momentum $m_z$ that determine the ground state for higher fields. As a result the shift of the AB oscillations towards higher fields is relatively smaller for states with higher $m_z$. The second difference between the calculated results for the “volcano-like” nanostructures and for the ideal rings consists in that for realistic nanostructures the higher order AB oscillations are strongly damped. This is a consequence of the presence of the magnetic field in the rim of the nanostructures, which enhances the electron localization close to the minima of the adiabatic potential.

7.7 Summary

In conclusion, we demonstrate, using advanced growth capabilities, experimental characterization and theoretical modeling, the existence of an oscillatory persistent current in self-assembled nanostructures containing only a single electron. We measured the first Aharonov-Bohm oscillation at a field of 14 T, in perfect agreement with our model based on the structural properties determined by X-STM measurements. Even though the nanostructures under investigation are singly connected and anisotropic, they show the AB behavior that is generally considered to be restricted to ideal (doubly connected) topologies. These results demonstrate the possibility to design and fabricate non-magnetic semiconductors with magnetic properties, which can be controlled by tuning the size and shape of self-assembled nanostructures.
Chapter 8

Excitonic behavior in self-assembled InAs/ GaAs quantum rings in high magnetic fields

8.1 Abstract

We investigate the exciton energy level structure of a large ensemble of self-assembled InAs/GaAs quantum rings by photoluminescence spectroscopy in magnetic fields up to 30 T for different excitation densities. The confinement of an electron and a hole in these quantum rings along with the Coulomb interaction suppress the excitonic Aharonov-Bohm effect. Nevertheless, the ring geometry is reflected in the non-equidistant exciton energy levels and the fact that they split up in only two levels in magnetic field. The model introduced in chapter 7, based on realistic parameters of the self-assembled quantum rings, allows us to interpret the essential features of the observed PL spectra in terms of the calculated optical transition probabilities.

To overcome the inhomogeneous broadening observed in the PL of an ensemble of rings, we also analyzed the PL of a large number of single quantum rings in magnetic field up to 10 T (measured with the confocal microscope in Eindhoven) and up to 30 T (measured at the HFML in Nijmegen). In contrast to what has been observed for InAs/GaAs quantum dots emitting at a similar wavelength (see chapter 3), the exciton $g$-factor and the diamagnetic coefficient $\alpha_d$ are uncorrelated to the emission energy. The Zeeman splitting is linear in magnetic fields up to 30 T, indicating a negligible magnetic field induced heavy hole-light hole mixing. Interestingly, a quadruplet splitting is observed for several rings, which is different from the one observed for quantum dots. This

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quadruplet splitting is analyzed in detail and possible origins are discussed.

8.2 Magneto-optical properties of an ensemble of quantum rings

8.2.1 Introduction

The excitonic energy structure of self-assembled quantum dots (QDs) is well studied [12, 210]. By magnetoluminescence experiments it has been demonstrated that the electronic energy levels in a QD can be described by the Fock-Darwin model for a two-dimensional harmonic oscillator in a magnetic field [62, 63, 72, 97, 211, 212, 213] (see also chapter 3). Changing the QDs to ring-like structures modifies the energy spectrum, and gives rise to the Aharonov-Bohm (AB) effect, which reflects the oscillatory behavior of charge carriers in a ring-like geometry in the presence of a magnetic field [179] (see chapter 7).

The optical emission of self-assembled InAs/GaAs quantum rings (QRs) [76, 189] has been studied experimentally without a magnetic field [105], and in magnetic fields not higher than 9 T [214]. In general, excitons are neutral excitations, thus we do not expect any sensitivity to the magnetic flux. However, since the exciton is a polarizable composite particle, the area between the different trajectories of the electron and the hole determines the phase picked up by the exciton [215]. Therefore the possible prominence of the AB effect for excitons strongly depends on their polarization. In order to explain this in more detail we use Berry’s gedanken experiment [216] in a similar way as was done in Ref. [215]. An electron and a hole are present in an ideal ring as shown in Fig. 8.1(a). Both charge particles are placed in an isolating box and are rotated along a closed trajectory. Analogous to the case of an electron in a ring discussed in chapter 7, both the electron and the hole obtain a topological phase. These phases will be different for the electron and hole, since they have different trajectories because of their different effective masses. The phase difference due to the different trajectories gives rise to the “optical” AB-effect.

In the case of a weakly interacting electron-hole pair both the electron and hole change their angular momentum $\ell$ at different magnetic fields. Figure 8.1(b) shows the energy diagram for a weakly interacting electron-hole pair. The “exciton” ground state is $(m^e_\ell, m^h_\ell) = (0,0)$, where $m^e_\ell$ ($m^h_\ell$) is the magnetic quantum number of the electron (hole) (see chapter 7). For increasing magnetic field the hole changes its magnetic quantum number $m^h_\ell$ first as it traverses the largest circumference. As the magnetic field increases the state therefore changes in favor of the state $(0,1)$, followed by $(-1,1)$, $(-1,2)$, etc., resulting in an oscillatory ground state energy. Moreover, according to the optical selection rules [74] the recombination of a hole with $m^h_\ell = 1$ and an electron with $m^e_\ell = 0$ is forbidden. As a result the optical emission from a weakly interacting electron-hole pair turns from bright to dark and vice versa as function of the magnetic field (depicted as the bright and dark areas in Fig. 8.1(b)). Depending on the polarization of the exciton (i.e. the difference in trajectory of the electron and hole) the dark and bright windows are changed.

It may be clear that the above sketched picture is highly simplified as in our quantum rings the exciton binding is considerable and the polarization small. Moreover, realistic quantum rings are asymmetric causing intermixing of the
different energy levels, as was discussed in chapter 7, and thereby preventing possible bright to dark transitions. Nevertheless calculations of the PL spectra of type I GaAs/AlGaAs QRs and several type II structures showed that a weak reminiscent feature of the AB effect in the PL spectrum might be observed [217, 218, 219]. Experimentally the optical AB effect has been shown in different ring-like structures [45, 99, 220]. Recently, the exciton energy spectra for various models of the InAs/GaAs self-assembled QRs were calculated as a function of the applied magnetic field and it was shown that the spectra are very sensitive to the details of the QR shape [221].

In this section we will consider the excitonic properties of self-assembled InAs/GaAs QRs in magnetic fields up to 30 T. Using different excitation densities we probe the magneto-PL of the ground and excited states. The essential features in the magneto-PL spectra are reproduced in calculations based on a realistic QR model [191, 209, 221]. We will demonstrate that QRs, in contrast to QDs, have non-equidistant energy levels and exhibit a magnetic field induced splitting of the higher excitonic energy levels into two levels. Furthermore, we will show that the confinement of an electron and a hole along with the Coulomb interaction suppress the excitonic AB effect in these QRs.

8.2.2 Setup

For the PL studies, a sample containing a single layer of QRs [76, 189] is mounted in a liquid-helium bath cryostat at \( T = 4.2 \) K. The excitation is provided by a Dye laser operating at 2 eV. The excitation power is varied with a Babinet-
Soleil compensator in combination with a linear polarizer. A Wollaston prism allows for simultaneous detection of both circular polarizations. The PL signal is dispersed by a single grating spectrometer, and the detection is performed by a liquid-nitrogen-cooled CCD camera. Static magnetic fields up to 30 T were applied parallel to the growth direction and the PL is detected in the Faraday configuration.

8.2.3 Measurements

The dependence of the QR emission energy on the excitation density is shown in Fig. 8.2(a). The ground state emission energy of the QRs is centered around 1.308 eV, typical for these nanostructures [76]. The ground state emission has an inhomogeneous broadening with a FWHM of 20 meV. With increasing excitation density two additional peaks can be resolved. These peaks have an energy of 39 meV and 63 meV above the ground state energy. The wetting layer (WL) emission is centered around 1.438 eV (not shown), which is 67 meV above the highest observed confined-state energy of the QRs.

We determine the energy of the ground state PL by fitting the spectra at low excitation densities by a Gaussian. The observed ground state emission energy of an exciton in a QR for relatively small $B$ is approximately given by $E(B) = E_0 \pm \frac{1}{2} g_{ex} \mu_B B + \alpha_d B^2$ [222]. Here $E_0$ is the emission energy at $B = 0$ T, $g_{ex}$ the exciton $g$-factor, $\mu_B = +5.79 \times 10^{-5}$ eV/T the Bohr-magneton, and $\alpha_d$ the diamagnetic coefficient. The second term is the Zeeman term which gives rise to a spin induced splitting of the exciton PL in a magnetic field. We defined $g_{ex} = \frac{E(\sigma^+) - E(\sigma^-)}{\mu_B B_0}$, and find $g_{ex} = -1.7$, in correspondence with previously reported values obtained on individual QRs, comparable to values for QDs [99, 146] and in agreement with the values found for individual rings (see next section). In Fig. 8.2(b) the diamagnetic shift $\Delta E_{\text{dia}}$ is shown, defined by $\Delta E_{\text{dia}} = \frac{E(\sigma^-) + E(\sigma^+)}{2} - E_0$. The diamagnetic shift has a smooth dependence on the magnetic field. From the quadratic fit (red solid line) we find $\alpha_d = 10 \mu eV/T^2$, in agreement with previous reported values for QRs [214] and QDs [146].

To investigate the influence of the ring-like geometry on the excitonic behavior in the excited states of the QRs, we measured the magneto-PL of these structures for higher excitation intensities. Figure 8.2(c) shows the higher excitation data in $\sigma^-$ polarization as function of $B$ in intervals of 5 T. The dashed lines are a guide to the eye and follow the peak positions. We have carefully assigned the PL peak positions as function of $B$ (see Fig. 8.3) by comparing the PL spectra at different $B$. As implied by Fig. 8.2(c), both resolvable excited states split up in two separate peaks. Each of the PL peaks of the QRs Zeeman splits further with a smaller energy separation into two peaks of opposite circular polarization.

8.2.4 The model

To understand the energy structure of the excitons we use a model based on the structural properties of these QRs obtained by X-STM measurements, which was introduced in chapter 7 [50, 191, 209]. The model is used to calculate the single-exciton optical transition probability spectrum. Slightly different param-
Figure 8.2: (a) PL as a function of excitation density, for which the lowest (highest) excitation density is $10^2 \text{Wcm}^{-2}$ ($10^5 \text{Wcm}^{-2}$). Two excited states can be distinguished for higher excitation density located 38 meV and 63 meV above the ground state emission energy. (b) The diamagnetic shift $\Delta E_{\text{dia}}$ of the ground state. The quadratic fit (red line) is used to determine the diamagnetic coefficient $\alpha_d$. (c) Excited states as a function of $B$ in $\sigma^-$ polarization for an excitation density of $10^5 \text{Wcm}^{-2}$. The dashed lines are guides to the eye in order to follow the evolution of the peak positions in $B$. The arrow indicates the emission energy at which for QDs a third peak is present.
Figure 8.3: The energy diagram showing the peak position in $B$ in both $\sigma^-$ (empty circles) and $\sigma^+$ (filled circles) polarization. The QRs exhibit splittings into two states of the different excited states, in contrast to QDs where a third peak (indicated by the dashed line) is observed.

eters are used to model the QRs as compared to chapter 7, in order to have a good qualitative agreement between the calculations and the experimental data [221]. It is realistic to assume that the sample analyzed in this chapter consists of slightly different rings as compared to the sample analyzed in the magnetization measurements. This is a result of the slightly different growth conditions. The actual shape and asymmetry of the rings is very sensitive on the exact growth conditions. The results are shown in Figs. 8.4(a) and 8.4(b), for a non-interacting and an interacting electron-hole pair, respectively. For a non-interacting electron-hole pair we calculate that around $B = 15$ T there is a crossover in the ground state energy, in agreement with magnetization experiments [223]. The inclusion of the Coulomb interaction results into a smooth behavior of the ground state energy as function of $B$, as shown in Fig. 8.4(b). At a magnetic field of 15 T the calculated spectrum shows that the first excited state has a reduced optical spectral probability. This is due to the redistribution of the oscillator strength between the first excited state and the ground state in favor of the latter. In the case of the interacting electron-hole pair the ground state energy is lowered by the energy of the electron-hole Coulomb attraction, which is 13 meV.

8.2.5 Discussion and conclusion

In the following we compare the experimental results with the theoretical calculations. The calculated ground state emission energy is 1.34 eV, in reasonable agreement with the measured ground state PL energy (1.31 eV). In general we find that the calculated energies are $\sim 30$ meV higher than the experimental values. The PL of the continuum states in the WL is calculated to be at 1.43 eV, which corresponds well to the measured value of 1.44 eV. Moreover, the calculations at $B = 30$ T show a 10 meV shift of the ground state to higher energy, where the experimental value is 8 meV. We note that the Zeeman effect is not
Figure 8.4: Calculated optical transition probabilities for a realistic QR in the case of (a) a non-interacting electron-hole pair and (b) an interacting electron-hole pair. The grey scale is logarithmic where black (white) corresponds to the highest (lowest) transition probability. The arrows correspond to the first excitonic AB resonance in the ground state.

Figure 8.5: Calculated broadened optical transition probabilities $P$ as a function of the emission energy $E$ for $B=0$ to $30$ T in $5$ T steps.
taken into account in the calculations. Both experimentally and theoretically we find a smooth dependence of the ground state emission energy on $B$, which is a consequence of the Coulomb interaction. In contrast, in the absence of Coulomb interaction a kink in the energy is expected (see Fig. 8.4(a)).

To interpret the higher lying energy states, we will focus only on the states in the model having a large spectral transition probability (cf. Fig. 8.4(b)), and compare them with the experimentally observed PL peaks. The first excited state is expected at 20 meV above the ground state emission energy. However, in our experimental data we cannot resolve this peak due to the inhomogeneous broadening. The second excited state in our model is at 58 meV above the ground state emission energy and corresponds to the second peak in our experiment, whereas the calculated energy level at 1.42 eV, 82 meV above the ground state emission, corresponds to the third peak we observe. In order to better compare the calculated spectra to the experimental spectra we introduce a Gaussian broadening $\Gamma$, which simulates the inhomogeneous broadening of the ensemble. For $\Gamma = 10\text{ meV}$, we find the best comparison of the calculated spectra with the experimental data. Figure 8.5 shows the calculated PL spectra for $B$ up to 30 T in steps of 5 T. The calculated and measured spectra (cf. Fig. 8.2(c)) show a qualitative resemblance, although the absolute values of the energy splittings are different. Importantly, the introduced broadening indeed shows that the first excited state is not resolvable in the magneto-PL. We do note that based on our model we assign the measured PL peaks to different excitonic states in the QRs as compared to the identification based on PLE measurements on single QRs [224]. However, within the theoretical model, which was successfully applied to explain the magnetization behavior of QRs on similar samples [223, 191], we found that for all realistic ring parameters the PL of the first excited state is concealed by the ground state luminescence.

The excitonic behavior characteristic for ring-like structures manifests itself in the magneto-PL under high excitation conditions. We observe the splitting of the excited states into two states as well as non-equidistant energy level splittings. In contrast to our measurements, experiments on QDs resulted in a magnetic induced splitting of the $d$-state into three states and equidistant energy levels [97, 211, 213]. This $d$-state corresponds to the second peak in Fig. 8.2(c), which for QRs has predominantly an $\ell = 2$ character, where $\ell$ is the orbital angular momentum quantum number. The dashed line in Fig. 8.3 indicates the position of the third energy level as observed for QDs. However, we observe a minimum in PL intensity at this emission energy (see arrow in Fig. 8.2(c)), which shows the absence of this third peak.

In the calculations the strongest effect on the oscillator strength is expected for the first excited state with predominantly an $\ell = 1$ character. This state is not resolved in our measurements due to the inhomogeneous broadening. The oscillator strength of the ground state of the single exciton of our modeled QR does not significantly change with $B$, as was confirmed in the experiments. In contrast to the AB effect of single electrons in these rings, we neither observe nor expect an excitonic AB effect based on our model. The absence of prominent oscillations in the ground state energy of the calculated exciton spectra as compared to the case of a non-interacting electron-hole pair is a consequence of the Coulomb interaction.

The details of the calculated spectra are very sensitive to the size, shape and composition of the QR. Nevertheless, based on the X-STM measurements we
do find a relatively good quantitative agreement between the calculated optical transition probabilities and the measured PL spectra [221]. The model does not include many-exciton complexes and charged excitonic states, which influence the optical transition probability spectra. We estimate this will only be a small effect as the exciton binding energy is an order of magnitude larger compared to the exciton-exciton interaction and additional charging energies. In order to calculate the actual PL spectra from the optical transition probability spectra one needs to take into account a non-equilibrium distribution function for excitons in a strong laser field and use a response theory [225]. Despite the preliminary character of our model, we are able to find a qualitative agreement between the measurements and the calculations and thereby we can explain the essential features in our measurements.

8.3 Excitonic behavior in individual quantum rings

In order to study the excitonic behavior in self-assembled InAs/GaAs quantum rings in more detail, we analyzed a large number of single QRs. In this section the description of the experimental setups and the sample are discussed. The correlation between the exciton $g$-factor $g_{ex}$, the diamagnetic coefficient $\alpha_d$ and the emission energy $E_0$ are analyzed for a large number of quantum rings and $g_{ex}$ and $\alpha_d$ have been studied in magnetic fields up to 30 T. Finally we investigate and discuss the anomalous quadruplet splitting observed for several quantum rings.

8.3.1 Experimental setup, sample and characterization

The presented experiments up to 10 T are performed using the confocal microscope setup as described in chapter 2. The PL was studied at $T = 4.2 \text{K}$ and the excitation was provided by a 635 nm laser diode. The PL is dispersed in the triple additive configuration and detected with the Si CCD detector. The grating combination was 1100-900-900 grooves/mm resulting in a resolution of 48 $\mu$eV at 950 nm.

For the PL studies up to magnetic fields of 30 T, the sample is mounted in a liquid-helium bath cryostat at $T = 4.2 \text{K}$. The excitation is provided by a HeNe laser operating at 633 nm ($\sim 1.96 \text{eV}$). The excitation power is varied with a Babinet-Soleil compensator in combination with a linear polarizer. A 40$\times$ objective (with a numerical aperture of 0.65 and a working distance 5.3 mm) is used to focus the excitation spot to $\sim 1 \mu \text{m}^2$ as well as to collect the PL. A lens with a focal length of 300 mm is used to image the collection spot on the slit of the monochromator. The magnification of the objective and the imaging lens together is $\sim 60 \times$, resulting in a collection spot of $\sim 60 \mu \text{m}$. The PL signal is dispersed by a single grating spectrometer with a focal length of 64 cm and a 1200 grooves/mm grating. The detection is performed by a Si CCD camera of 1340 $\times$ 100 pixels with a pixel size of 20 $\mu \text{m}^2$. The resolution of the system is

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2These measurements have been performed in close collaboration with J. H. Blokland, P. C. M. Christianen and J. C. Maan, HFML, IMM, Radboud University Nijmegen, The Netherlands.
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Figure 8.6: (a) A $2 \times 2 \mu m^2$ AFM image of the sample with a low density of QRs. From these measurements a quantum ring density of $3 \times 10^9 \text{cm}^{-2}$ is determined. The arrows indicate QRs with a larger diameter as compared to the majority of the QRs. The AFM image was measured by A. G. Taboada. (b) The macro-PL spectrum of the sample with a low QR density. The PL peak has an asymmetric shape and contains a low energy tail, which is attributed to the rings of larger diameter.

$\sim 82 \mu eV$. For both experiments the static magnetic fields are applied parallel to the growth direction and the PL is detected in the Faraday configuration.

The measured sample consisted of a single layer of QRs from the outer part of the wafer. As a result of the gradient of nanostructures over the wafer, this sample contained a reasonably low QR density of $3 \times 10^9 \text{cm}^{-2}$ as compared to the sample of the same wafer used in the ensemble PL measurements ($8 \times 10^9 \text{cm}^{-2}$). The QR density is determined from the AFM image, shown in Fig. 8.6(a). A SIL is mounted on top of the sample and was used in the experiments up to 10 T. The macro-PL spectrum is shown in Fig. 8.6(b). The PL peak has an asymmetric shape and contains a low energy tail (indicated by the arrow). Remarkably, this is an opposite asymmetry as compared to the PL of InAs/GaAs quantum dots reported in chapter 3. No noticeable multimodal height distribution is present and therefore we attribute the low energy tail to QRs of somewhat larger radius. This interpretation is supported by the AFM images, which indeed also show rings with a considerable larger radius (indicated with the arrows in Fig. 8.6(a)).

8.3.2 The exciton $g$-factor and diamagnetic coefficient of quantum rings

The exciton $g$-factor $g_{ex}$ and diamagnetic coefficient $\alpha_d$ are determined for a large number of QRs. For the InAs/GaAs QRs the same sign of the $g$-factor is obtained as for the InAs/GaAs QDs, i.e. the Zeeman splitted line with $\sigma^-$-polarization is at higher energy as compared to the one with $\sigma^+$-polarization. Figure 8.7(a) and (b) show $g_{ex}$ and $\alpha_d$ as function the emission energy at $B =$
Figure 8.7: (a) The exciton g-factor $g_{ex}$ as function of the emission energy $E_0$. The red dashed line is linear regression indicating a negligible trend, in contrast to what has been observed for InAs/GaAs and InAs/InP QDs [38, 36]. The red filled circles correspond to the largest $\alpha_d$ and the blue stars correspond to the smallest values of $\alpha_d$. (b) The diamagnetic coefficient $\alpha_d$ as function of $E_0$. The color scale relates to the three different intervals of $\alpha_d$ used in (a). (c) $g_{ex}$ as function of $\alpha_d$ for two different energy intervals.
Figure 8.8: Three different quantum rings are measured up to $B = 30\,\text{T}$. The blue (white) color corresponds to a low (high) PL intensity.

0\,\text{T} ($E_0$) for 70 quantum rings. No clear trend between the emission energy and $g_{\text{ex}}$ and $\alpha_d$ is observed. The values of $g_{\text{ex}}$ and $\alpha_d$ obtained for the QRs are comparable to the ones found for the QDs [38] and reported for QRs in literature [146, 226]. A linear regression through the data results in a negligible dependence in contrast to the results of the quantum dots. The absence of a clear trend is likely to be caused by the absence of a multi-modal height distribution of QRs (see Fig. 8.6(b)), as the height is of major influence on $g_{\text{ex}}$ [36, 38]. Figure 8.7(c) shows the correlation between $g_{\text{ex}}$ and $\alpha_d$ for three energy intervals. In contrast to what has been observed for the QDs, the QRs with the lowest emission energy do not have the most positive value of $g_{\text{ex}}$. It rather appears that QRs with larger $\alpha_d$ have a more positive $g_{\text{ex}}$; the quantum rings having emission at higher energy have on average the same height but a larger diameter. This weak correlation between the diameter and $g_{\text{ex}}$ is in agreement to what has been observed for the InAs/InP and InAs/GaAs quantum dots [36, 38]. It is also reflected in Fig. 8.7(a) where on average the data points with the largest $\alpha_d$ (red points) are above the ones with smallest $\alpha_d$ (blue points), although there are considerable deviations from this main trend.

Several individual QRs have been analyzed in magnetic fields up to 30\,\text{T}. Figure 8.8 shows a contour plot of three different QRs. The exciton peaks of quantum ring 2 are fitted up to 30\,\text{T} and the Zeeman splitting and the diamagnetic shift are shown in Fig. 8.9(a) and (b). The Zeeman splitting of this and other rings is perfectly linear up to the highest available magnetic fields. This is in contrast to what has been observed for InAs/GaAs quantum dots [227, 228], where for higher magnetic fields the Zeeman splitting deviated from a linear dependence. This deviation is attributed to an enhancement of the heavy hole-light hole mixing in magnetic field (see also chapter 6), as was observed for
Figure 8.9: (a) The Zeeman splitting as function of $B$ is linear up to the highest available magnetic field. From the fit (dashed line) we determine $g_{ex} = -2.66$. (b) The diamagnetic shift $\Delta E_{dia}$ as function of $B^2$. The linear fit for $B \leq 11$ T describes the diamagnetic behavior in relatively small magnetic fields for $\alpha_d = 8.23 \mu eV$. For $B > 16.5$ T there is a strong deviation from the quadratic dependence on the magnetic field.

quantum wells [176, 177]. Heavy hole-light hole mixing is mainly determined by the confinement potential and the strain. In general, a decrease in the height of the nanostructure results into less intermixing of the heavy hole and light hole band as the splitting between the heavy hole and light hole bands increases. On the other hand, an increase of the strain results into a larger splitting between the heavy-hole and light-hole band but also to the intermixing of the states. As a result of the annealing step used during the growth of the quantum rings, the height of the studied quantum rings is relatively small as compared to quantum dots of Ref. [227, 228]. During this growth step a large part of the original quantum dot is dissolved and diffused away. It is likely that the resulting additional confinement causes a negligible heavy hole-light hole intermixing and will thus result in a linear Zeeman splitting.

Figure 8.9(b) shows the diamagnetic shift, which is as large as 6 meV at 30 T. In order to determine $\alpha_d$, the diamagnetic shift is fitted up to 11 T. The quadratic dependence on magnetic field with $\alpha_d = 8.23 \mu eV/T^2$ describes the
diamagnetic shift accurately for magnetic fields up to 16.5 T. For higher fields there occurs a strong deviation of the diamagnetic shift. This is in reasonable agreement to what has been observed for the ensemble of rings (see inset Fig. 8.2(a)). For these high magnetic fields the magnetic confinement is no longer a perturbation compared to the lateral confinement of the quantum ring as $\hbar \omega_c \simeq \hbar \omega_{x,y}$, resulting in a linear contribution of the diamagnetic shift to the lateral confinement energy. The magnetic length at $B = 16.5$ T is $l_B \sim 6.5$ nm, which is comparable to the width of the rim of the QR of about $\sim 6$ nm (see Fig. 7.5). We note that due to the absence of a hole in the center of the QRs the actual lateral confinement is somewhat larger. It the actual lateral confinement is comparable for InAs/GaAs QDs with a relatively large diameter [228, 212] comparable values of the magnetic field are reported at which the linear behavior of the diamagnetic shift starts to dominate.

### 8.3.3 The anomalous quadruplet splitting

While analyzing the magnetoluminescence of individual quantum rings with the triple monochromator in triple additive mode, 4 out of 7 quantum rings exhibited an anomalous quadruplet splitting. In Fig. 8.10 the quadruplet splitting observed for three different rings is shown in three contour plots. For relatively small $B$ the exciton line splits into a conventional Zeeman doublet with opposite circular polarization. At a critical field $B_c$ each of the two circular polarized lines split further in two lines of the same polarization, resulting in a quadruplet structure. For the 4 rings investigated up to 10 T the emission energy $E_0$, the critical magnetic field $B_c$, and the energy splitting between the lines of equal polarization at 10 T ($E_{\sigma-}$ and $E_{\sigma+}$) are given in table 8.1. Although the statistics are poor, there is no relation between the $E_0$ and $B_c$ and for all 4 rings we find $E_{\sigma-} < E_{\sigma+}$.

Table 8.1: The emission energy $E_0$ at $B = 0$ T, the critical magnetic field $B_c$ at which the quadruplet structure appears, and the energy splitting $E_{\sigma-}$ and $E_{\sigma+}$ between the lines of equal circular polarization at 10 T for four quantum rings.

<table>
<thead>
<tr>
<th>$E_0$ (eV)</th>
<th>$B_c$ (T)</th>
<th>$E_{\sigma-}$ ($\mu$eV)</th>
<th>$E_{\sigma+}$ ($\mu$eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.2824</td>
<td>5</td>
<td>400</td>
<td>431</td>
</tr>
<tr>
<td>1.2922</td>
<td>8.2</td>
<td>142</td>
<td>154</td>
</tr>
<tr>
<td>1.2955</td>
<td>4.5</td>
<td>138</td>
<td>157</td>
</tr>
<tr>
<td>1.2980</td>
<td>6.6</td>
<td>158</td>
<td>174</td>
</tr>
</tbody>
</table>

In order to determine the character of the exciton in the quantum rings exhibiting the quadruplet structure, power dependency measurements have been performed. For the different rings a similar behavior is observed and here only the quantum ring of Fig. 8.10(c) is analyzed in detail. The PL spectra at $B = 0$ T are shown in Fig. 8.11(a) for different excitation powers. Even for the lowest excitation powers ($\sim 100$ nW) the exciton line is present. Therefore we exclude the possibility that the exciton line is a result of the recombination of an electron...

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3For the QR shown in Fig. 8.10(b) there are two PL lines in the background which split and show a negative diamagnetic shift. Further discussion on these lines can be found in Chapter 5.
Figure 8.10: The contour plots of three different rings showing the quadruplet splitting. The blue (red/white) color corresponds a low (high) PL intensity. At a critical magnetic field $B_c$, each of the conventional Zeeman lines split further in two lines of the same circular polarization. The inset in (c) shows the quadruplet splitting in more detail.
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Figure 8.11: (a) The PL spectra for different excitation powers of the quantum ring shown in Fig. 8.10(c) at $B = 0$ T. (b) From the PL intensity as function of excitation power a slope of 0.96 is found.

Figure 8.12: (a) The PL spectra for different excitation powers of the quantum ring shown in Fig. 8.10(c) at $B = 10$ T. For low excitation powers the lowest energy lines of each circular polarization disappear. For higher excitation powers the low energy lines become even more intense than the high energy lines. The dashed lines show that the peak positions are independent of the excitation power. (b) The PL intensity as function of the power shows a superlinear dependence for the low energy lines of the two doublets.
Figure 8.13: The integrated PL intensity of the $\sigma^-$ doublet as function of magnetic field. The arrow indicates where a resolvable splitting occurs (see inset Fig. 8.10(c)).

and hole from the $\ell = 1$ shell. For increasing power there is an increase of the background due to the charging of the WL, and a possible biexciton feature at a lower energy of $\sim 1$ meV appears. Analyzing the PL intensity of the exciton line for different powers results in the dependency shown in Fig. 8.11(b). The slope is 0.96, typical for the powerdependence of the exciton, whereas the biexciton has usually a slope in the interval between 1 and 2 [87]. Therefore the exciton in the QR exhibiting the quadruplet splitting is either the neutral exciton $X^0$, or the singly positively ($X^+$) or negatively charged exciton ($X^-$). We can exclude higher charged exciton complexes, as for these complexes electron-electron, hole-hole and electron-hole exchange energies are expected to give rise to a set of lines [105, 120]. For the neutral exciton a fine structure splitting is expected, as is discussed in chapter 3. No feasible splitting is resolved by analyzing the linear and circular polarization of the PL at $B = 0$ T with. However, for the InAs/GaAs quantum dots analyzed in chapter 3 the FSS is also not resolved. It could well be that for these relatively shallow nanostructures the FSS is very small and therefore not resolvable within the experimental resolution. As a final remark, the sample under investigation is undoped. Nevertheless, there is always a residual $p$-doped background present in MBE grown samples and there have been a large number of reports studying charged exciton complexes in undoped samples.

Figure 8.12(a) shows the PL spectra for the same QR at $B = 10$ T. Surprisingly, only for higher excitation densities the low energy lines appear. For 100 nW of excitation power only the conventional Zeeman doublet is observed, whereas for excitation powers larger than 10000 nW the split off lines are more intense as compared to the conventional Zeeman doublet. The energy positions of the different PL peaks are not dependent on the excitation power. The PL intensity of the 4 lines of the quadruplet as function of the power is shown in Fig. 8.12(b). Whereas the slope of the high energy lines ($\sigma^-$ (high) and $\sigma^+$

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4As is already discussed in the previous section and chapter 7, the first excited state of the rings is not a pure $\ell = 1$ state.
Figure 8.14: (a) The energy of the PL lines of the QR shown in Fig. 8.10(c) as determined by a Lorentzian fitting procedure. Clearly, the conventional Zeeman doublet (filled symbols) behaves normally, whereas the low energy peaks of each polarization split off at a critical magnetic field. (b) The Zeeman splitting between the filled and unfilled symbols result in the same $g_{ex}$. (c) The diamagnetic shift $\Delta E_{dia}$ of the conventional Zeeman doublet (filled symbols) exhibits a purely quadratic dependence on the magnetic field, whereas the diamagnetic shift of the split off lines (unfilled symbols) around $B \approx B_c$ cannot be described by a quadratic dependence.
Figure 8.15: (a) The contour plot of a QR analyzed up to 30 T. (b) The PL spectra for the $\sigma^+$ polarization. A quadruplet splitting is resolved at $B \sim 16$ T. The splitting is indicated with the arrows for $B = 18$ T. (c) The energy of the PL peaks as determined by a Lorentzian fit as function of magnetic field.
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(high)) is smaller than 1, a superlinear dependence is found for the low energy lines of each polarization. As the low energy lines only appear for higher excitation densities, it is probable that the low energy lines are a result of a multi-exciton complex which is only stable at a critical magnetic field. This is also reflected by analyzing the PL intensity of the lines before and after the splitting. Figure 8.13 shows the integrated intensity of the $\sigma^-$-peaks as function of the magnetic field. Around the critical magnetic field $B_c$ there is a strong increase in the total integrated intensity, instead of a redistribution of the oscillator strength over both states.

The PL lines of the QR are fitted with Lorentzian curves and the result is shown in Fig. 8.14(a). From the fit it is clear that the conventional Zeeman doublet (filled circles and squares) continues to behave normally and the low energy lines split off at $B_c$. The Zeeman energies between the normal doublet and the split off branches are given in Fig. 8.14(b). Remarkably, the measured value of $g_{ex}$ is identical. This behavior is reproduced for all four rings. As in general there is a large spread of $g_{ex}$ for different rings (see Fig. 8.7(a)), an identical value of $g_{ex}$ supports that the quadruplet states is arising from (an) exciton state(s) in the same ring. The exact behavior of the diamagnetic shift of the split off branch varies from ring to ring, whereas the diamagnetic shift of the normally behaving lines are well described with a quadratic dependence.

The quadruplet splitting is also analyzed in the experiments up to 30 T. The contour plot of the such a ring is shown in Fig. 8.15(a). The splitting is hardly visible in this representation, and therefore the PL peaks between 16 and 20 T are shown for the $\sigma^+$-polarization in Fig. 8.15(b). Fitting the peaks with Lorentzian curves result in the energy position as function of magnetic field as shown in Fig. 8.15(c). The energy splittings $E_{\sigma^-}$ and $E_{\sigma^+}$ are $\sim 90\mu$eV. Importantly, the splitting does not disappear for increasing magnetic field.

8.3.4 Discussion

(A) Excluded explanations

For self-assembled InAs/GaAs QDs a quadruplet splitting is reported in chapter 3, which is attributed to a reduced symmetry in the quantum dot system, resulting in a mixing of the bright and dark states. However, the dark states have in general a different exciton $g$-factor as compared to the bright states, in contrast to what we observe for the quantum rings. Moreover, as function of the excitation power the relative intensity between the dark and bright states is not altered in the experiments on the quantum dots, whereas for the quantum rings the relative intensity of the quadruplet lines is determined by the excitation power. We therefore exclude that the quadruplet splitting observed for the rings is of similar origin. Furthermore, we also exclude that magnetic field induced heavy hole-light hole mixing is causing the quadruplet splitting, as in the experiments up to 30 T the exciton $g$-factor is perfectly linear.

(B) Possible explanations

The transitions could be related to singlet to triplet transitions when a singly charged exciton, either $X^-$ or $X^+$, is related to the quadruplet splitting. For small magnetic fields the singlet state of the exciton is the ground state. For
the singlet state the spins of the two holes (electrons) of the $X^+$ ($X^-$) are antiparallel, whereas for the triplet state the spins of the holes (electrons) are parallel. Due to the Pauli exclusion principle two holes (electrons) with a parallel spin must reside in a different energy level. The energy difference between the singlet and triplet splitting is given by the exchange energies between the charges and the energy level splitting between the ground state and the first excited state. For increasing magnetic field the energy difference between the first excited state and the ground state decreases (depicted in Fig. 8.16). When this energy difference is comparable to the exchange energy, the triplet state becomes energetically favorable. The ground state and first excited states for holes are split by only several meV whereas for electrons this splitting is $\sim 15$ meV. Therefore, it is more likely for holes to have a singlet to triplet transition at relatively small magnetic fields. Although this picture explains the magnetic field induced transition as well as the polarization of the lines, we do not yet understand the dynamics. For one, for magnetic fields smaller than $B_c$ the triplet should already be visible at higher energies due to the thermal distribution over the singlet and triplet configuration. Also, in the proposed picture it is not clear why the triplet ground state is only visible at higher
Another possibility is that the asymmetric ring potential as shown in Fig. 7.13 (see chapter 7) results in a magnetic field induced localization in each of the two potential pockets. It is most probable that the two potential pockets are not identical for a realistic quantum ring. The picture now arises that in a magnetic field the exciton becomes more and more localized in either one of the two potential pockets. As these pockets are not identical this will give rise to an energy splitting between the two localized states. This will thus result in a quadruplet splitting. In this picture the evolution of the energy levels in magnetic field and the power dependence are not understood up to now.

Finally, we note that for InAs/GaAs WL states a similar quadruplet splitting has been observed [229]. However, no clear explanation of this behavior has been given. As the similarity between these natural quantum dots and our quantum rings is the relatively small height of these nanostructures, this might be related to the origin of the quadruplet splitting. Note that due to the non ring-like geometry of the natural dots, it is more likely that the singlet to triplet transition is causing the quadruplet splitting instead of the asymmetric ring-like potential.

8.4 Summary

We have analyzed the emission energy of a large ensemble of self-assembled InAs/GaAs QRs in high magnetic fields. Our model shows that the confinement of an electron and a hole along with the Coulomb interaction suppress the excitonic AB effect in these nanostructures. Nevertheless, the ring character of our nanostructures results in non-equidistant energy level splittings in the exciton diagram and into a magnetic field induced splitting of each excited state into two states. This is different to what has been observed in measurements on quantum dots; the quantum dots are well-described within the harmonic oscillator model. The optical transition probabilities are calculated within our model, based on the characterization of a realistic QR. Comparing these calculations with our experimental data we find a qualitative agreement, which allows us to identify the different PL peaks and helps to explain the excitonic behavior in magnetic field.

In order to study the excitonic behavior in the QRs in more detail, a large number of individual rings have been studied. Due to the absence of a multimodal height distribution there is no observable trend between the emission energy, exciton $g$-factor and the diamagnetic coefficient. By investigating the Zeeman splitting up to 30 T we show that for QRs have is a negligible magnetic field induced heavy hole-light hole mixing. The diamagnetic shift is well described by a quadratic dependence up to 16.5 T, comparable to what has been reported on quantum dots. By analyzing exciton lines with high resolution an anomalous quadruplet splitting is revealed, which is much different as the one reported for the InAs/GaAs quantum dots. Although the origin of this splitting is still unclear, it is most likely related to the singlet to triplet transition of singly charged excitons.
8.5 Outlook

The excitonic AB effect is likely to be more pronounced for charged exciton complexes [99]. Therefore a charge tunable quantum ring sample is needed to determine unambiguously the charged state of the studied exciton [105]. This would allow to study the ”optical” AB effect for both the positively and negatively charged excitons and opens the opportunity to investigate the role of the Coulomb interactions in the quantum ring. Moreover, the identification of the charged state of the exciton helps to unravel the origin of the quadruplet splitting. To understand the excitation dependence of the quadruplet splitting it is useful to study the relaxation processes in the quantum ring. This can be done by time-resolved photoluminescence, PLE and correlation measurements.
Chapter 9

The magnetoluminescence of type II self-assembled InP/GaAs quantum dots

9.1 Abstract

In this chapter the experimental results on the magnetoluminescence of both an ensemble and individual type II self-assembled InP/GaAs quantum dots are discussed. For these type II quantum dots the band offset of the InP quantum dot material relative to the GaAs host material is such that the hole is located outside the dot, whereas the electron is confined in the quantum dot. Nevertheless, the Coulomb interaction between the electron and hole gives rise to an indirect exciton. The radiative exciton recombination time of these indirect excitons is relatively long (up to $\sim \mu$s) \[230\] as compared to the recombination time of direct exciton in type I quantum dots ($\sim$ns), which is for instance attractive for optical memories \[231, 232\]. Interestingly, it has been shown that these indirect excitons can give rise to the "optical" Aharonov-Bohm (AB) effect \[45, 220\]. Instead of the topologically determined ring structure (see chapter 7, 8), the type II alignment can give rise to a ring-like potential for the hole, as shown in Fig. 9.1.

We performed magnetoluminescence measurements on a large ensemble of InP/GaAs quantum dots. In Ref. \[45\] clear AB oscillations were observed in the ensemble magnetoluminescence energy of these type II InP/GaAs quantum dots. Surprisingly, we do not observe any oscillations in the ensemble magnetoluminescence energy measured on the same sample. As the reported oscillations are relatively small ($\sim 500 \mu$eV) compared to the full width at half maximum ($\sim 35$ meV) of the ensemble PL, we performed the ultimate experiment by measuring the magneto-PL of individual InP/GaAs quantum dots on the same sample. For the single ring experiments the line width of $\sim 120 \mu$eV is much smaller than the observed oscillation. In these decisive experiments we do not observe the AB oscillatory behavior within the experimental resolution of $40 \mu$eV. This result shows the necessity to study subtle magnetoluminescence properties of nanostructures on an individual dot level, rather than on a large
CHAPTER 9.

Figure 9.1: (a) A simplified picture of the conduction and valence band profiles for the InP dots in GaAs. (b) The top view of the quantum dot plane. The holes are confined to a ring around the quantum dot due to the Coulomb interaction with the electron confined in the quantum dot. Figure adapted from Ref. [45].

9.2 Sample

The sample containing the InP/GaAs quantum dots\(^1\) is the same sample as measured in Ref. [45]. The sample is grown by MOCVD on semi-insulating GaAs [001] substrates at 550 °C. The structures consisted of a 300 nm undoped GaAs buffer layer grown at 600 °C followed by the QD layer. The dots are capped by a 50 nm undoped GaAs layer. The QD sizes are determined from cross-sectional TEM measurements and are (32 ± 6) nm and (4 ± 2) nm in diameter and height, respectively. More details on the growth and characterization are reported elsewhere [233, 234]. The quantum dot density is \(\sim 3 \times 10^{10} \text{ cm}^{-2}\). Due to the high quantum dot density the single dot luminescence experiments are challenging. In order to measure single dots the sample is prepared with a mask (see chapter 2).

9.3 Ensemble magnetoluminescence

The ensemble PL is both measured with the macro-PL setup described in chapter 2 and with the confocal microscope setup in magnetic fields. The ensemble PL for \(B = 0 \text{T}\) is shown in Fig. 9.2(a) for different excitation powers. For increasing excitation density a blue shift is observed. This blue shift has been reported previously in Refs. [235, 230, 236, 237, 238]. The origin of this blue shift is debated, but usually it is attributed due to the charging energy in the system with separate electron-hole confinement, and it serves as a fingerprint of the II-type heterojunction. However, some authors attribute the blue shift due to the presence of excited states for higher excitation densities [235]. Figure 9.2(b) shows the PL as function of temperature. Whereas for the type I InAs/GaAs quantum dots of chapter 3 and the InAs/InP quantum dots of chapter 6 an increase of the PL intensity is observed for temperatures up to several tens of

\(^{1}\)This sample has been grown by E. Ribeiro, W. Carvalho, Jr., and G. Medeiros-Ribeiro, Laboratório Nacional de Luz Síncrotron, Campinas, Brazil.

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Figure 9.2: (a) The macro-PL spectra at $T = 6$ K for three different excitation powers. A blue shift is observed for increasing excitation powers. (b) The temperature dependence of the ensemble luminescence. The PL intensity monotonously decreases with increasing temperature.

Kelvin as compared to 5 K, we observe a monotonous decrease of the PL intensity for the type II dots. The charge separation in type II dots strongly reduces the electron-hole interaction and therefore the Coulomb interaction is less compared to conventional type I dots. Increase of the temperature results effectively to a further reduction of the electron-hole interaction, resulting in a reduced PL intensity.

The magnetoluminescence of the ensemble of type II dots is shown in Fig. 9.3. The experiments have been performed for excitation powers of $\sim 10^3$ W/cm$^{-2}$ and for a below bandgap excitation wavelength of 825 nm at a temperature of 2 K. The observed structure on top of ensemble luminescence is due to individual quantum dots. Nevertheless the PL peak can be nicely fitted by a Gaussian (see inset Fig. 9.3(b)). The PL spectra as function of the magnetic field are presented in Fig. 9.3(a) for both circular polarizations at $B = 0$ and 9 T. The PL peak with $\sigma^-$-polarization is at higher energy as compared to the $\sigma^+$-polarization resulting in a negative exciton $g$-factor. The energy peak positions as function of magnetic field are shown in Fig. 9.3(b). The diamagnetic shift $\Delta E_{dia}$ is shown in Fig. 9.3(c). For larger magnetic fields the diamagnetic shift deviates from the quadratic dependence. This is more clearly shown by plotting $\Delta E_{dia}$ as function of $B^2$ (see Fig. 9.3(d)). The origin of the deviation of the quadratic behavior for magnetic fields larger than 5.4 T is unknown and can be related to complicated redistribution processes in a magnetic field. Analyzing the results we obtain a large diamagnetic coefficient of $\alpha_d = 32 \mu eV/T^2$ up to 5.4 T and $g_{ex} = -0.99$. In order to resolve any oscillatory behavior, the quadratic diamagnetic shift is subtracted from the data and the residu is shown in Fig. 9.3(e). The inset shows the residual energy up to 5.4 T. Whereas Ref. [45]

2The experiments have also been repeated for lower powers and an excitation wavelength of 635 nm. The results are consistent with the results shown in Fig. 9.3.

3These measurements have been performed with the confocal microscope setup. The collection has been performed by a multimode fiber with a diameter of 200 $\mu$m.
Figure 9.3: (a) The spectra for $\sigma^-$ (red dashed line) and $\sigma^+$ (solid line) polarization at $B = 0$ and 9 T. (b) The energy position of both circular polarizations as function of $B$. The energy positions are determined by fitting the spectra with a Gaussian (see inset). (c) The diamagnetic shift $\Delta E_{\text{dia}}$ as function of $B$ and (d) $B^2$. For magnetic fields up to 5.4 T the diamagnetic shift can be well described by a quadratic dependence. The fit (red line) gives $\alpha_d = 32 \text{µeV/T}^2$. (e) The residual energy given by $\Delta E_{\text{dia}} - \alpha_d B^2$. The inset shows the absence of an oscillatory behavior within the experimental resolution. (f) The emission energy as function of $B$ as reported by Ribeiro et al. [45] shows oscillations with a magnitude of $\sim 500 \text{µeV}$ for the same sample.
reported a clear oscillation of 500 $\mu$eV (see Fig. 9.3(f)), we surprisingly do not reproduce these results. In fact, no oscillations are resolved within the experimental resolution of 40 $\mu$eV. In order to resolve the discrepancy between our experiments and the experiments presented in Ref. [45] single quantum dots are analyzed.

### 9.4 Single dot magnetoluminescence

#### 9.4.1 Experimental results

The magnetoluminescence of several individual InP/GaAs quantum dots is analyzed using the confocal microscope setup. The PL is dispersed with the 750 grooves/mm grating and detected by the Si CCD detector. The FWHM of the peaks is typically $\sim 150$ $\mu$eV. The excitation wavelength is 825 nm and $T = 2$ K. Due to the high QD density, the individual lines are superimposed on a background due to the emission of different quantum dots. A typical single dot spectrum obtained on this sample is shown in Fig. 9.4(a). The PL line indicated with the arrow is analyzed in magnetic field. The spectra for $B = 0$ T, 2 T and 5 T are shown in Fig. 9.4(b) for both circular polarizations. The single line at $B = 0$ T Zeeman splits into two lines of opposite circular polarization, where $\sigma^-$ is the highest energy line. Figure 9.4(c) shows the peak positions as function of the magnetic field. The peak positions are determined by fitting the peaks with a Gaussian. The $\sigma^-$ ($\sigma^+$) branch is fitted by $E(B) = E_0 \pm \frac{1}{2} g_{ex} \mu_B B + \alpha_d B^2$ (see chapter 3). From these fits we obtain for this quantum dot $g_{ex} = -1.34 \pm 0.09$ and $\alpha_d = 7.3 \pm 0.8 \mu$eV/T$^2$. The difference between the fits and the experimental determined values of the peak position is shown in the inset of Fig. 9.4(c). Importantly, there are no observable oscillations up to magnetic fields of 6 T in agreement with the measurements on the ensemble of quantum dots.

<table>
<thead>
<tr>
<th>$E_0$ (eV)</th>
<th>$g_{ex}$</th>
<th>$\alpha_d$ (\mu eV/T$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.32470</td>
<td>$-1.00 \pm 0.03$</td>
<td>6.2 \pm 0.1</td>
</tr>
<tr>
<td>1.33403</td>
<td>$-1.87 \pm 0.02$</td>
<td>5.6 \pm 0.2</td>
</tr>
<tr>
<td>1.34925</td>
<td>$-1.34 \pm 0.09$</td>
<td>7.3 \pm 0.8</td>
</tr>
</tbody>
</table>

Two other dots have also been analyzed and showed a similar behavior. $E_0$, $g_{ex}$ and $\alpha_d$ for all three QDs are listed in table 9.1. The values of $g_{ex}$ are in good agreement with the ones obtained from the macro-PL. Surprisingly, we observe a large difference between the single dot diamagnetic coefficient ($\sim 6 \mu$eV/T$^2$) and the diamagnetic coefficient of the ensemble luminescence ($\sim 32 \mu$eV/T$^2$). The analyzed single dots are on average on the higher energy side, whereas in the macro-PL all dots contribute to the diamagnetic shift. Indeed a large spread
Figure 9.4: (a) A typical spectrum obtained on one of the apertures of the InP/GaAs quantum dot sample. Due to the high QD density the number of lines in the spectrum are high and contribute to the background. The single dot line indicated with the arrow is analyzed in magnetic field. (b) The single dot luminescence spectra for both circular polarizations at $B = 0$ T, 2 T and 5 T. The line Zeeman splits and shows a diamagnetic shift. (c) The energy of the PL for both circular polarizations determined by fitting the single dot spectra by a Gaussian. From the energy positions we extract for this dot $\alpha_d = 7.3 \pm 0.8 \, \mu eV / T^2$ and $g_{ex} = -1.34 \pm 0.09$. The inset shows the residual energy for both polarization using the determined values of $\alpha_d$ and $g_{ex}$. 


in $\alpha_d$ over the energy range has been reported on similar quantum dots grown by chemical vapor epitaxy (CBE) [239]. Nevertheless, the results on $\alpha_d$ for the quantum rings (chapter 8) showed a good agreement between the ensemble and single dot luminescence. Therefore other processes, such as a redistribution of charge carriers from the relatively shallow dots to the more confined dots induced by the magnetic field can not be excluded.

9.4.2 Discussion

The absence of any oscillatory behavior, both in the macro-PL and single dot PL, convincingly shows that within the resolution of 40 $\mu$eV no AB related features are present for these type II dots. We note that compared to ensemble measurements, single dot measurements are the ultimate test in order to detect subtle magnetoluminescence properties, such as the optical AB effect. In contrast to the PL of individual nanostructures, the ensemble PL energy of quantum dots is also governed by a possible magnetic field induced redistribution over the different quantum dots. Also, as shown in Fig. 9.2, the exact peak position is sensitive to the excitation power, which can vary due to the misalignment in a magnetic field. These effects do not influence the PL energy of individual quantum dots [235], but might have been of influence on the observed oscillation in the ensemble PL as reported in Ref. [45]. It is therefore necessary to study subtle magnetoluminescence properties of nanostructures on an individual dot level, rather than on a large ensemble of dots. Furthermore, we note that it is not clear for these dots whether the hole outside the quantum dot is located in the plane of the dot (as depicted in Fig. 9.1) or on top of the quantum dot. The latter configuration will even not allow for the AB effect. Finally, we remark that single dot measurements on type II InP/GaAs quantum dots grown by chemical beam epitaxy and emitting at higher energy also did not show any resolvable AB related phenomena [240, 239].

9.5 Summary

An ensemble and individual type II InP/GaAs quantum dots have been investigated in the presence of a magnetic field. For increasing excitation power the ensemble PL showed a blue shift indicating the type II confinement. The magnetoluminescence of the ensemble of dots did not resolve any oscillatory behavior related to the AB effect in contrast to experiments reported on the same sample [45]. Decisive magnetoluminescence measurements on individual dots, for which the PL energy is determined more accurately as compared to the ensemble PL energy, did not reveal any AB related phenomena within the experimental resolution of 40 $\mu$eV. Our results show the necessity to study subtle magnetoluminescence properties of nanostructures on an individual dot level, rather than on a large ensemble of dots.
Bibliography


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Abstract

Magneto-optical properties of self-assembled III-V semiconductor nanostructures

In this thesis the optical properties of self-assembled III-V semiconductor nanostructures are investigated by analyzing the photoluminescence (PL) with and without an external magnetic field. The studied PL is a result of the recombination of electrons and holes confined in the nanostructures. In this thesis we directly correlate the size, shape, composition and topology of the nanostructures to their magnetic and optical properties and study the influence of an electrical contact in the close proximity of these nanostructures.

One of the most interesting nanostructures are quantum dots (QDs), which are able to confine charge carriers in all three dimensions. Both the exciton g-factor $g_{ex}$ and the exciton diamagnetic coefficient $\alpha_d$ determine the behavior of excitons confined in a QD in a magnetic field and provide further insight in the energy level structure of these nanostructures. The exciton Zeeman splitting is proportional to $g_{ex}$ and is a result of the different spin configurations of the electron and hole, which constitute the exciton. The diamagnetic shift is a result of the additional confinement provided by the magnetic field $B$, and is proportional $\alpha_d B^2$, where $\alpha_d$ scales with the lateral extend of the quantum dot. PL experiments on a large ensemble of InAs/GaAs QDs, as well as on individual InAs/GaAs QDs are performed. For these dots $\alpha_d$ and $g_{ex}$ have been determined. Importantly, there is a trend between $g_{ex}$ and the emission energy $E_0$: for larger emission energy a more negative value of the exciton g-factor is observed. From the power and temperature dependence of the ensemble PL it is shown that the sample consists of QDs with different height, where the highest dots correspond to the smallest emission energies. Moreover, QDs with larger $\alpha_d$ have a more positive g-factor. From this it is inferred that quantum dots with an overall larger size have a less negative value of $g_{ex}$.

By implementing the QDs in a Schottky device in close proximity to an electrical contact it is possible to tune the charged state of the exciton in the dot. We show that charge-tunable InAs/GaAs quantum dots also allow to study many-body interactions between a localized QD state and a Fermi sea of electrons. As a result of these many-body interactions new optical transitions are observed in the PL spectra. The same charge-tunable quantum dots are also investigated in a magnetic field. Instead of the positive diamagnetic shift observed for the majority of the quantum dots, two different types of negative diamagnetic shift are reported. The shallow character of our quantum dots causes a negative quadratic diamagnetic shift for the highly negatively charged exciton

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complexes in line with predictions for shallow quantum dots. The second type of negative diamagnetic shift is observed even for the neutral exciton and is strongly linearly dependent on the magnetic field.

We also investigated quantum dots grown from different semiconductor materials. InAs/InP quantum dots are studied using AFM, X-STM, macro-PL and micro-PL. Macro PL and X-STM measurements show that the studied InAs/InP dots have a multimodal height distribution. Single quantum dot luminescence, carried out on a large number of dots, shows a strong correlation between exciton $g$-factor, diamagnetic coefficient and emission energy. In fact, the strong dependence of $g_{ex}$ on the emission energy results in a sign change of the exciton $g$-factor. In correspondence with what we found on the InAs/GaAs QDs, we find that dots having a smaller overall size will have a more negative $g_{ex}$ as compared to quantum dots of larger overall size. We also show that for several quantum dots the exciton $g$-factor is quenched. A theoretical model is used to calculate the effect of the quantum dot size on the $g$-factor. The model is both qualitatively and quantitatively in good agreement with the experimental obtained results.

Nanostructures of a different topology are self-assembled InAs/GaAs quantum rings (QRs). Using magnetization measurements on a large ensemble of quantum rings we demonstrate the presence of the Aharonov-Bohm (AB) effect in these nanostructures. Moreover, a model based on the X-STM measurements on these nanostructures reproduces the magnetic field position of this oscillation. The optical properties of these QRs are investigated for magnetic fields up to 30 T for a large ensemble of quantum rings and for individual rings. Although the excitonic AB effect is suppressed in these nanostructures, the ring character of our nanostructures gives rise to non-equidistant energy level splittings and into a magnetic field induced splitting of each excited state into two states in the ensemble PL. This is different to what has been observed in measurements on quantum dots. The calculations based on the same model show a qualitative agreement with the experimental data, and allow us to identify the different PL peaks. Furthermore, analyzing exciton lines with high resolution revealed an anomalous quadruplet splitting, which is different as the one reported for InAs/GaAs quantum dots. Although the origin of this splitting is still unclear, it is most likely related to the singlet to triplet transition of singly charged excitons.

Finally the magneto-luminescence experiments on type II self-assembled InP/GaAs quantum dots are discussed. For these dots the hole is located outside the quantum dot, whereas the electron is confined inside the quantum dot creating a type II exciton. The magneto-luminescence of the ensemble of dots did not resolve any oscillatory behavior related to the AB effect in contrast to experiments reported on the same sample. Decisive magneto-luminescence measurements on individual dots did also not reveal any AB related phenomena within the experimental resolution of 40 $\mu$eV. These results show the necessity to study subtle magneto-luminescence properties of nanostructures on an individual dot level, rather than on a large ensemble of dots.
Samenvatting

Magneto-optical properties of self-assembled III-V semiconductor nanostructures

In dit proefschrift worden de optische eigenschappen van zelf-georganiseerde III-V halfgeleider nanostructuren onderzocht aan de hand van de fotoluminescentie in de aan- en afwezigheid van een extern magneetveld. Fotoluminescentie is een gevolg van de recombinatie van een elektron-gat paar dat is ingevangen in de nanostructuren. In dit proefschrift bestuderen we de correlatie tussen de grootte, vorm, compositie en topologie van verscheidene nanostructuren en de resulterende magnetische en optische eigenschappen. Verder laten we zien wat de invloed is van een elektrische contact in de nabijheid van de nanostructuren.

Eén van de meest interessante nanostructuren zijn kwantum punten waarin ladingsdragers in alle drie de dimensies opgesloten kunnen worden. De exciton $g$-factor $g_{ex}$ en de diamagnetische coefficient $\alpha_d$ bepalen het gedrag van excitonen in kwantum punten in een magneetveld en geven ons een belangrijk additioneel inzicht in de energieniveau structuur van deze nanostructuren. Zo is de exciton Zeeman splitsing proportioneel met $g_{ex}$ en wordt deze splitsing veroorzaakt door de verschillende spin configuraties van het elektron en het gat, die samen het exciton vormen. De diamagnetische verschuiving schaalt met $\alpha_d$ en is een resultaat van de additionele opsluiting van de excitonen in het magneetveld, waarbij $\alpha_d$ gerelateerd is aan de kwantum punt diameter.

Fotoluminescentie experimenten op zelf-georganiseerde InAs/GaAs kwantum punten zijn uitgevoerd op meer dan 80 individuele kwantum punten en op een groot ensemble van kwantum punten. De enkele kwantum punten zijn bestudeerd in een magneetveld en voor al deze kwantum punten zijn $g_{ex}$ en $\alpha_d$ bepaald. Het meest opvallende resultaat is dat we een trend zien tussen de emissie energie $E_0$ en $g_{ex}$: voor een hogere emissie energie vinden we gemiddeld een negatievere waarde van $g_{ex}$. Door het analyseren van de excitatie dichtheid en temperatuur afhankelijkheid van de fotoluminescentie van het ensemble kwantum punten tonen we aan dat de hoogste kwantum punten corresponderen met de laagste emissie energieën. Daarbij hebben kwantum punten met een grotere waarde van $\alpha_d$ (wat correspondeert tot een grotere diameter) een meer positieve exciton $g$-factor. Hieruit concluderen we dat kwantum punten met een groter volume een minder negatieve waarde voor $g_{ex}$ hebben.

Wanneer we de kwantum punten in bouwen in een Schottky structuur geeft dit ons de mogelijkheid om de ladingstoestand van het exciton te bepalen. Wij laten zien dat ladinginstelbare InAs/GaAs kwantum punten ook de mogelijkheid geven om veel-deeltjes interacties te onderzoeken tussen een gelokaliseerde
SAMENVATTING

toestand in een kwantum punt en een Fermi zee van elektronen in een elektrisch
contact. Het resultaat van de veel-deeltjes interacties is terug te vinden in de
fotoluminescentie spectra, waar nieuwe optische transities gedetecteerd worden.
Wanneer we dezelfde kwantum punten in een magneetveld analyseren vinden we
dat meerdere van de bestudeerde kwantum punten een negatieve diamagnetische
verschuiving laten zien, in plaats van een positieve diamagnetische verschuiving
wat normaal wordt waargenomen voor deze kwantum punten. Voor verscheidene
kwantum punten observeren we dat de hogere negatief geladen excitonen
een negative diamagnetische verschuiving ondergaan. Dit gedrag kunnen we
relateren aan de relatief ondiepe opsluitingspotentiaal van deze kwantum pun-
ten. Verrassend genoeg vinden we voor verschillende quantum punten zelfs een
sterke negatieve diamagnetische verschuiving voor het neutrale exciton.

Kwantum punten bestaande uit verschillende halffeder materialen zijn
ook onderzocht. Zo zijn InAs/InP kwantum punten bestudeerd met behulp van
AFM, X-STM en fotoluminescentie. Vanuit de X-STM en ensemble fotolumines-
centie metingen laten we zien dat de kwantum punten bestaan uit verschillende
hoogtes. Verder tonen we aan dat er een sterke correlatie bestaat tussen $E_0$,
gex en $\alpha_d$. Net als voor de InAs/GaAs kwantum punten vinden we dat een
grooter volume een meer positieve waarde van $g_{ex}$ oplevert. We kunnen nu zelfs
aantonen dat het teken van de exciton $g$-factor te veranderen is door de grote
van de kwantum punten aan te passen en dat we $g_{ex}$ ook gelijk aan nul kun-
nen maken. Een geïntroduceerd model gebaseerd op de X-STM metingen komt
zowel kwantitatief als kwalitatief overeen met de experimenten.

Nanostructuren met een andere topologie zijn InAs/GaAs kwantum ringen.
Met behulp van magnetisatie metingen op een groot aantal ringen tegelijk tonen
we het bestaan aan van het Aharonov-Bohm (AB) effect in deze ringen. Een
model gebaseerd op de structurele eigenschappen reproduceert het magneetveld
waarop de eerste AB oscillatie plaatsvindt. Ook de optische eigenschappen van
een ensemble en enkele kwantum ringen zijn onderzocht in magneetvelden tot
30 T. Het ring karakter van onze nanostructuren komt tot uiting in de niet-
equidistante energieniveau structuur en de opsplitsing van de verschillende en-
ergieniveaus in tweeën in de ensemble magneto-luminescentie metingen. Dit is
in tegenstelling tot wat is gevonden voor kwantum punten. Bovendien geeft
ons model dat ook gebruikt is voor de magnetisatie metingen een kwalitatieve
overeenkomst met het experiment en stelt ons in staat om de metingen te in-
preteren. Opmerkelijk is dat we voor deze nanostructuren een kwadruplet
splitting observeren wanneer we de fotoluminescentie van enkele kwantum rin-
gen detecteren. Deze splitsing is van andere origine als degene geobserveerd
voor kwantum punten en is waarschijnlijk gerelateerd aan de singlet naar triplet
transitie van enkel geladen excitonen.

Tenslotte is ook de fotoluminescentie van type II InP/GaAs kwantum pun-
ten gemeten. Voor deze kwantum punten zijn de gaten buiten de kwantum punt
gelokaliseerd en de elektronen in de kwantum punt, waardoor een ring-
achtige opsluiting potentiaal kan ontstaan die aanleiding geeft tot het AB ef-
fect. In tegenstelling tot eerder gerapporteerde metingen vinden wij geen AB
effect wanneer we een ensemble van InP/GaAs kwantum punten meten. In het
beslissende experiment op individuele type II kwantum punten wordt ook geen
AB effect waargenomen. Dit resultaat toont aan dat het noodzakelijk is om
subtiele effecten in de magneto-fotoluminescentie te bestuderen op het niveau
van enkele nanostructuren.
List of publications

Papers

2009


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2008


2007


Oral presentations international conferences

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2007


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Poster presentations international conferences

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2007

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Curriculum vitae

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