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Magneto-optical spectroscopy on semiconductor nano rings and single quantum dots

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2007

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Title: Magneto-optical spectroscopy on semiconductor nano rings and single quantum dots

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Date: June 2007

Report of a graduation project from June 2006 to June 2007, carried out at the Eindhoven University of Technology at the Department of Applied Physics

Supervisors: Ir. N.A.J.M. Kleemans
           Prof. Dr. P. Koenraad
Abstract

Recently, by development of a specific growth procedure InAs/GaAs quantum dots can be transformed into quantum ring structures. Theoretical calculations and recent measurements show that Aharonov-Bohm oscillations occur in ring-structures containing a single electron. In this report we will focus on the study of rings containing an exciton, i.e. an electron and a hole that are present in the ring at the same time. The electron-hole pairs in these rings are generated by photo excitation. Goverov shows that there are different "optical" Aharonov-Bohm effects for both the coupled and uncoupled electron-hole pairs. The Goverov model is rather simple, a more sophisticated model based on realistic parameters developed by Antwerp shows that the "optical" AB effects are much more intricate. They show that for the case of strong Coulomb interaction in realistic rings no Aharonov-Bohm effect related features occur.

Magneto-luminescence measurements show that in a self-assembled quantum ring the coulomb interaction is strong enough to bound the electron-hole pair. No "optical" Aharonov-Bohm effect in these rings up to 30 T is shown in agreement with the theoretical modeling from Antwerp. Also the observations of excited state level splitting, diamagnetic shift and Zeeman splitting are in fair agreement with the theoretical model. For future experiments we want to study single quantum rings using a confocal microscope. A confocal microscope is developed and photoluminescence of a single dot as function of magnetic fields up to 10 T could be studied. The observed diamagnetic shifts is consistent with photoluminescence measurements performed on ensembles of quantum dots.
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Chapter 1

Introduction

Low-dimensional semiconductor structures confine electrons in less than three dimensions. In this thesis self-assembled quantum rings and quantum dots are discussed. These structures are objects with a size in the order of 1–10 nm in height in diameter larger and have been subject of intense research both to uncover physical phenomena [1] and explore their potential in technological applications such as lasers [2], quantum computing [3], single photon sources [4] and many more. In dots the electrons are confined to zero dimensions and have discrete energy levels. Their energy spectra are shell-like, analogous to that of real atoms and therefore quantum dots can be regarded as "artificial atoms".

The study of energy levels in quantum dots or rings is essential for a better understanding of their physical properties. An important tool to investigate the nature and behavior of discrete energy levels are magnetic fields. The energy of the discrete levels is changed by applying a perpendicular magnetic field, resulting in a diamagnetic shift and Zeeman splitting. Since the changes in energy are in the order of meV/T, high magnetic fields are needed to identify and study the energy levels.

One of the most intriguing quantum mechanical phenomena is the existence of the Aharonov-Bohm effect [5]. This effect has no classical analogue at all and is therefore counter intuitive. In short the Aharonov-Bohm effects describes the behavior of a charged particle in the vicinity of an electromagnetic field. Even though this charged particle is in a field free region, there is still a vector potential not equal to zero due to the near-by presence of the electromagnetic field. Since the Hamiltonian in quantum mechanics uses potentials instead of the actual fields, the charged particle is feeling this vector potential and consequently its motion is influenced by it.

The Aharonov-Bohm effect for a electron in a self-assembled quantum ring is recently shown [1]. Furthermore, there has been theoretical effort on a mechanism of phase difference acquired in a magnetic field by a composite and polarizable object with overall zero charge in these quantum rings. Such neutral particles, called excitons, are bound states of an electron and a hole in semiconductors, and are responsible for optical emission of crystals. The quantum phase picked up by such an exciton is influenced by the magnetic field [6]. The exciton emission can be strongly suppressed in certain magnetic-field windows, known as the "optical" Aharonov-Bohm effect [7].

Each dot and ring in an ensemble has slightly different optical properties due to the inhomogeneous nature of dots and rings. Studying single dots or rings will provide details which are masked during ensemble measurements. Confocal microscopy is a well established
single dot and ring spectroscopy technique [8]. The principle of a confocal microscope has two
crucial points, first illuminating a single point of the sample with a focussed beam. Secondly
the use of a blocking pinhole for detection of the signal. This pinhole filters out all the off-axis
and out-of-focus light. A confocal system provides the highest resolution obtainable from
conventional optics, since only light from the focus is imaged the resolution of the system is
limited by the diffraction limit spot size of 260 nm with the use of a solid immersion lens.

This thesis focusses on the magneto-optical behavior of self-assembled quantum rings.
The high magnetic field measurements are performed in the High Field Magnetic Laboratory
in Nijmegen up to 30 T. The ground state and excited states energy levels of the exciton
in a ring as a function of magnetic fields are determined. The measurement are compared
by a model, developed by V. Fomin, V.N. Gladilin and J.T. Devreese of the TFVS group of
the Department of Physics at Antwerp University. Furthermore a single dot/ring confocal
microscope is developed which can be operated in magnetic fields up to 11 T. This confocal
microscope is for single dot or ring characterization and proof of principle is performed with
single dot PL as a function of magnetic field.

The outline of this thesis is as follows: in chapter 2 the theory about the non-optical
and optical Aharonov-Bohm effect is described. Furthermore the behavior of an bound and
unbound exciton is modeled in this chapter. In chapter 3 the different photoluminescence
setups and quantum ring sample are described. Next to that both magnetic and non-magnetic
photoluminescence measurement are discussed. Chapter 4 discusses the development of the
confocal microscope, with every component described elaborately. The proof of principle
of the confocal microscope is performed with test measurements on a quantum dot sample.
Finally, in chapter 5 conclusions and suggestions are presented.
Chapter 2

Theory

2.1 General properties of semiconductors

A wide range of III-V materials has been investigated for their semiconducting properties, mostly based on combinations of the elements Al, Ga, In and As. In figure 2.1 a part of the periodic system is shown, with the most common semiconductor elements. The intrinsic electrical properties of semiconductors are very often modified by doping with group II and IV elements. Depending on the kind of doping, a region of a semiconductor can have more electrons or holes, and is then called N-type or P-type semiconductor, respectively [9]. Different layers of semiconductor compounds are used to form a heterostructure.

The band gap \( E_g \) and the lattice constant \( a_0 \) are two important properties which determine the properties of the different semiconductor materials. In figure 2.2 the lattice constant \( a_0 \) is plotted along the horizontal axis against the band gap energy \( E_g \) and corresponding wavelength \( \lambda \) along the vertical axis for different semiconductor materials. All the values in the graph are for room temperature. The difference in lattice constants for different materials causes strained layers. These layers change the conduction and valence bands and \( E_g \).

An important way to analyse the band structure of semiconductors are photoluminescence (PL) measurements. When a semiconductor is illuminated, an electron absorbs a photon when the energy of the photon is larger then the band gap energy \( E_g \). The electron is excited to the conduction band and leaves a hole behind in the valence band. This absorption process can

\[
\begin{array}{cccccccc}
I & II & III & IV & V & VI & VII & VIII \\
\text{H} & \text{He} & \text{Li} & \text{Be} & \text{B} & \text{C} & \text{N} & \text{O} \text{ F} & \text{Ne} \\
\text{Na} & \text{Mg} & \text{Al} & \text{Si} & \text{P} & \text{S} & \text{Cl} & \text{Ar} \\
\text{K} & \text{Ca} & \text{Ga} & \text{Ge} & \text{As} & \text{Se} & \text{Br} & \text{Kr} \\
\text{Rb} & \text{Sr} & \text{In} & \text{Sn} & \text{Sb} & \text{Te} & \text{I} & \text{Xe}
\end{array}
\]

*Figure 2.1: Group III and V elements are most commonly used in low dimensional semiconductor structures*
Figure 2.2: Bandgap energy and wavelength plotted against the lattice constant for different semiconductor compounds at room temperature [10].
Table 2.1: The band gaps at 0 K and 300 K and lattice constants of InAs and GaAs.

<table>
<thead>
<tr>
<th></th>
<th>Bandgap T = 0 K</th>
<th>Bandgap T = 300 K</th>
<th>Lattice constant T = 298 K</th>
</tr>
</thead>
<tbody>
<tr>
<td>InAs</td>
<td>0.43 eV</td>
<td>0.36 eV</td>
<td>6.068</td>
</tr>
<tr>
<td>GaAs</td>
<td>1.52 eV</td>
<td>1.43 eV</td>
<td>5.653</td>
</tr>
</tbody>
</table>

be used for band gap analysis. PL is the emission of a photon as a result of the recombination of an electron and a hole. The resulting PL spectrum gives crucial information about the structural properties of nanostructures.

In figure 2.2 one can observe two kinds of semiconductors, direct and indirect. When the electrons in the minimum of the conduction band and holes in the maximum of the valence band have the same k-vector, it is called a direct gap. Recombination via a photon is likely process, because the momentum of the hole and electron match. The well-known semiconductor compound GaAs has a direct gap of \( E_g = 1.42 \text{ eV} \), with a corresponding wavelength of \( \lambda = 871 \text{ nm} \). These values are valid a room temperature, for lower temperatures the gap energies are slightly higher, see table 2.1 [11]. Most optoelectronic devices are based on materials with a direct gap. The materials used during this work have a direct gap.

### 2.2 Self-assembled quantum dot & quantum ring formation

Low dimensional semiconductors confine electrons in one or more dimensions. The best known low dimensional semiconductor is the quantum well. There the electrons are confined in one direction and are free to move in the plane perpendicular to this direction. In Quantum Dots (QD’s) and Quantum Rings (QR’s) electrons are confined in all three dimensions [9]. This confinement leads to discrete energy spectra, which are comparable to the spectra we obtain from atoms. Therefore a QD is often referred to as an ‘artificial atom’.

Structures such as QD’s and QR’s can be grown by MBE from the III-V semiconductor materials InAs and GaAs. The bandgaps and lattice constants are given in table 2.1 [11] [12]. With molecular beam epitaxy (MBE) single layers of atoms or molecules can be grown on a substrate. In this way highly homogeneous layers of material can be grown. Parameters affecting the growth include the temperature of the substrate, the flux of the particles and the time the substrate is exposed to the particles. When the lattice constants of the semiconductors differ a few percent, first a layer is grown on the substrate, the wetting layer. Due to strain in the materials, layer by layer growth becomes unfavorable and instead of another layer, islands are formed. This is the Stranski-Krastanov growth mode. The transition to quantum dots occurs after growing 1.5 mono layer (ML) of InAs on GaAs. Droplets of InAs, the quantum dots, are formed on top of a thin layer of the InAs wetting layer. After deposition of more than 2.5 ML dislocations in QD’s are formed, which quenches the optical properties [13].

To form QR’s the dots are capped with a thin layer of GaAs, in the order of 2nm, and subsequentially annealed [14]. The schematic growth process of QR’s is shown in figure 2.2. A detailed description of the growth method of QR’s is given in [15], [16] and [17].

Scanning Probe Microscopy (SPM) is used to determine the structural properties of the nanostructure such as shape, size and composition of dots and rings. Atomic Force Microscopy (AFM) shows that the size of a dot is \( \sim 60 \text{ nm} \) in diameter and \( \sim 10 \text{ nm} \) in height. For a ring
Figure 2.3: The different growing steps of a self-assembled InAs QR on GaAs. (1) The InAs quantum dots are capped with a 2nm thick layer of GaAs (2). After annealing the indium migrates from the center of the dot outwards (3). Then the QR is capped with GaAs giving rise to a second wetting layer (4).

Figure 2.4: AFM image of a self-assembled InAs quantum ring [18].
Figure 2.5: Filled state X-STM image of self-assembled InAs quantum rings cleaved in the [110]-plane (a) and in the [1\bar{1}0] plane (b) showing the asymmetry of the rings in this sample. The inset shows the 3D model of a ring as observed by X-STM [18].

this size is \(~80\,\text{nm}\) in diameter and \(~9\,\text{nm}\) in height. Furthermore QR’s have an asymmetry in diameter and height depending on the growth conditions. In figure 2.4 an AFM image of a QR is shown.

The actual sizes found by Cross-sectional Scanning Tunnelling Microscopy (X-STM) for the dots and rings capped with a thick layer of GaAs, are smaller compared to the sizes found by AFM for surface dots and rings. X-STM shows a diameter for the QR’s of 23 nm and a height of 3.6 nm. QR’s are sometimes called craters or volcanoes, because of their volcano-like shape. The X-STM picture of a typical QR is shown in figure 2.5. The rings are cleaved in two different dimensions showing the asymmetry of the ring as observed by AFM measurements [18] [19]. The difference in size between AFM and X-STM measurements for QD’s and QR’s is well understood and explained in [1].
2.3 Optical transitions of quantum rings in magnetic field

The Aharonov-Bohm (AB) effect is an intriguing quantum mechanical phenomenon, which occurs in ring-like topologies [1]. In this section first a quantum mechanical approach of the AB effect in an ideal ring is discussed, followed by the AB-effect a non-ideal, self-assembled InAs quantum rings. Laser excitation produces an electron-hole pair in the ring. Both the interacting and the non-interacting electron-hole cases are discussed. Finally, the energy spectrum and optical transitions probabilities for an exciton in a non-ideal ring is discussed. These calculations are based on theoretical work of the TFVS group in the Department of Physics at Antwerp University by V. Fomin, V.N. Gladilin and J.T. Devreese.

2.3.1 An electron in an one dimensional ideal ring

Figure 2.6 illustrates Berry’s gedanken experiment of a particle in box, which expels magnetic field. The flux enclosed by the ring is called $\Phi_e$. The Berry phase is accumulated as the particle in the isolating box is slowly rotated around the field lines. The circle trajectory is adiabatic, therefore the system returns to its original state after a full cycle, and accumulates only a topological phase. The phase is given by the total flux $\Phi$ in terms of the quantum flux $\Phi_0$, with $\Phi_0 = \frac{\hbar}{2\pi}$, closed by the path of the box and is equivalent to the AB phase.

The Hamiltonian of a single electron in the presence of a magnetic field $\vec{B}$ is given by:

$$H = \frac{1}{2m_e^*}(\vec{p} - e \vec{A})^2 + V$$  \hspace{1cm} (2.1)

where $V$ contains details of the electronic confinement potential, and $m_e$ is the electron mass. The vector potential $\vec{A}$ is related to $\vec{B}$:

$$\vec{B} = \vec{\nabla} \times \vec{A}$$  \hspace{1cm} (2.2)

The Hamiltonian is connected to the energy $\varepsilon$, via the Schrödinger equation, given by:

$$H\psi = \varepsilon\psi$$  \hspace{1cm} (2.3)

To simplify matters the electronic confinement potential $V$ is set to zero. First the wave function of an electron in an one dimensional ring is calculated in the absence of a magnetic
field, so \( B = 0 \). The wave function only depends on the azimuthal angle \( \phi \) so \( \psi = \psi(\phi) \). Using spherical coordinates the Schrödinger equation leads to:

\[
-\frac{\hbar^2}{2m_e^*} \nabla^2 \psi(\phi) = -\frac{\hbar^2}{2m_e^* R^2} \frac{\partial^2}{\partial \phi^2} \psi(\phi) = \varepsilon \psi(\phi)
\]

(2.4)

Furthermore \( \psi(\phi) = \psi(\phi+2\pi) \), because the wave function is uniquely determined. Then if we take into account that the function is normalized the solution of the Schrödinger equation is given by:

\[
\psi(\phi) = \frac{1}{\sqrt{2\pi}} \exp(\pm i R \frac{\hbar}{\sqrt{2m_e^*}} \phi)
\]

(2.5)

Because of the periodic boundary conditions the energy values \( \varepsilon \) are quantized, such that the discrete energy values \( \varepsilon_m \) are given by:

\[
\varepsilon_m = \frac{m^2 \hbar^2}{2m_e^* R^2}
\]

(2.6)

The corresponding wavefunctions belonging to \( \varepsilon_m \) are \( \psi(\phi) = \frac{1}{\sqrt{2\pi}} \exp(im\phi) \). The energies are labeled with quantum number \( m \) [20], [21], often referred to as the magnetic quantum number.

The magnetic field is included and the wavefunctions and energies will be determined. Using Stoke’s theorem the magnetic flux penetrating the ring is given by:

\[
\oint \vec{A} \cdot d\vec{l} = \int \int \vec{B} \cdot d\vec{S} = \Phi
\]

(2.7)

The wave function is given in the following equation:

\[
\psi(\phi) = \frac{1}{2\pi} \exp(i \Phi \frac{\phi}{\Phi_0} \frac{\Phi_0}{2\pi})
\]

(2.8)

The energy levels belonging to the wave function \( \psi \), are obtained by applying the same procedure as used in the absence of the magnetic field. For a detailed description see [1]. The corresponding energy levels are given as a function of the magnetic quantum number \( m \):

\[
\varepsilon_m = \frac{\hbar^2}{2m_e^* R^2} (m + \frac{\Phi}{\Phi_0})^2, \quad m = 0, \pm 1, \pm 2, ...
\]

(2.9)

The energy levels are functions of the magnetic flux and are given in figure 2.7. For increasing magnetic field, the ground energy state will change from angular momentum quantum number \( m = 0 \) to \( m = -1 \). The states with different \( m \) become the ground state for rational values of the magnetic flux given by \( \Phi = (p + \frac{1}{2})\Phi_0 \), with \( p = 0, \pm 1, \pm 2, ... \). For QR’s containing a single electron the ground state determines the physical properties of the system. Electrons in a self-assembled QR show magnetization behavior, see [1].

### 2.3.2 An electron and a hole in a non-ideal ring

The self-assembled QR’s are non-ideal rings. The ring-structure is modeled\(^1\) with a varying-thickness InGaAs layer embedded in a matrix of GaAs, which is based on the structural

---

\(^1\) V. Fomin, V.N. Gladilin and J.T. Devreese, TFVS, Universiteit Antwerpen, Belgium.
Figure 2.7: Energy of a one dimensional ring as function of magnetic flux penetrating the center of the ring, equation 2.9. At the $\Phi = \left( p + \frac{1}{2} \right) \Phi_0$ values of the magnetic flux the lowest energy state changes from angular momentum quantum number $m$ to $m - 1$.

information obtained from the X-STM measurements performed by P. Offermans [18]. The bottom of the InGaAs layer is assumed to be perfectly flat and parallel to the $x,y$-plane. The analytical equations, with the height of the rim $h$ as function of the radial coordinate $\rho$ and the azimuthal $\phi$, of the ring are:

$$h(\rho, \phi) = h_0 + \frac{(h_M(1 + \xi \cos(2\phi)) - h_0)\gamma_0^2}{R^2} \times \frac{R^2 - (\rho - R)^2}{(\rho - R)^2 + \gamma_0^2}, \rho \leq R \quad (2.10)$$

$$h(\rho, \phi) = h_\infty + \frac{(h_M(1 + \xi \cos(2\phi)) - h_\infty)\gamma_\infty^2}{(\rho - R)^2 + \gamma_\infty^2}, \rho > R \quad (2.11)$$

The radius of the ring is given by $R$, $h_0$ is the height of the ring in the center, $h_M$ the rim height and $h_\infty$ the height of the InGaAs layer far away from the ring which corresponds with the wetting layer. The parameters $\gamma_0$ and $\gamma_\infty$ define the inner and outer slopes of the rim and $\xi$ defines the asymmetry in the rim height. The modeled ring for realistic parameters is shown in figure 2.8.

The indium (In) fraction $x$ of In$_x$Ga$_{1-x}$As is taken from [19] where $x$ is determined by using the surface relaxation and lattice distortion of the cleaved surface measured by X-STM. The shape of the ring and the concentration of In in the ring determine the band parameters such as the effective mass and the band gaps.

In PL experiments we observe the recombination of an electron-hole pair. Therefore we consider the Hamiltonian of an electron and hole in a QR

$$H_{ex} = H_e + H_h \quad (2.12)$$
Figure 2.8: The shape of the self-assembled ring as modeled by equations 2.10 and 2.11, with $R = 11.5$ nm, $h_0 = 1.6$ nm, $h_M = 3.6$ nm, $h_\infty = 0.4$ nm, $\gamma_0 = 3$ nm and $\gamma_\infty = 5$ nm. The anisotropy parameter $\varepsilon$ is set on 0.2.
where $H_e$ is the single particle Hamiltonian of an electron and $H_h$ of a hole. We are interested in the lowest states of an electron and a hole in the QR. Furthermore only heavy holes are considered, therefore we treat the one-band model. The single-particle Hamiltonians in a ring have the following form:

$$H_e = -\frac{\hbar^2}{2} (\nabla - \frac{e}{\hbar} A)^2 \left( \nabla - \frac{e}{\hbar} A \right) + V_e(r_e) + \delta E_e(r_e) - eV_P(r_e), \quad (2.13)$$

$$H_h = -\frac{\hbar^2}{2} (\nabla - \frac{e}{\hbar} A)^2 \left( \nabla - \frac{e}{\hbar} A \right) + V_h(r_h) + \delta E_h(r_h) - eV_P(r_h), \quad (2.14)$$

where $m_e(r_e)$ and $m_h(r_h)$ are the conduction electron mass and heavy hole mass, respectively. $A = \frac{e}{2} \mathbf{B} \rho$ is the vector potential of the uniform magnetic field $B = e_z B$. The bottom of the conduction band is given by $V_e(r_e)$ and the top of the valence band by $V_h(r_h)$. The strain-induced shift of the conduction band

$$\delta E_{\beta} = -a_{\beta}(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) \quad (2.15)$$

depends on the hydrostatic component of the strain tensor $\varepsilon_{jk}$. The shear strains give rise to the piezoelectric potential

$$V_P(r) = -\frac{1}{4\Pi \varepsilon_0 \varepsilon_r} \int \frac{\text{div} \mathbf{P}}{|\mathbf{r} - \mathbf{r}'|} d^3 \mathbf{r}' \quad (2.16)$$

determined by the piezoelectric polarization $P_i = \varepsilon_{ijk} \varepsilon_{jk}$, $\varepsilon_r$ is the relative dielectric constant. From the work of P. Offermans [18], [19] the distributions of the strain-induced shifts of band edges and of the piezoelectric potential are calculated. The single-particle Schrödinger equations, with $\beta$ indicating an electron ($\beta = e$) or a hole ($\beta = h$),

$$H_{\beta} \Psi^{(\beta)}(r) = E \Psi^{(\beta)}(r) \quad (2.17)$$

are solved within the adiabatic approximation, using the Ansatz:

$$\Psi^{(\beta)}(r) = \psi^{\beta}_k(z; \rho, \varphi) \Phi^{\beta}_{kj} (\rho, \varphi) \quad (2.18)$$

where the index $k$ number the subbands due to the size quantization along the x-axis. The detailed description of solving the Hamiltonian is given in Appendix A. In figure 2.9 the adiabatic potential for a single electron in a uniform In$_{0.6}$Ga$_{0.4}$As ring is shown.

Using the theoretical approach described above and the adiabatic ring potential the electron energy as function of magnetic field is determined and shown in figure 2.10 together with the ideal ring case and for a ”disk” shaped ring. One can observe that the AB oscillations survive for the realistic ring compared to the ideal ring. Furthermore, a shift to higher energies for increasing magnetic field is observed, caused by the magnetic confinement. This confinement creates a parabolic potential for the QR’s, this potential increases the energy of the electrons.

For high magnetic fields this potential localizes the electrons deep in the ring, which decreases the amplitude of the AB oscillations at high magnetic fields. The electrons are then confined in the two deep potential wells of the QR, figure 2.9, and show no ”ring”-like behavior. Additionally, there is a reduced potential barrier at the center of the ring caused by the strain, which reduces the effective electron radius. As indicated by the arrows in the
Figure 2.9: The adiabatic potential of a quantum ring with the same parameters as in figure 2.8. The potential has valleys where the rims are higher and hills where the rims are smaller, asymmetry is clearly shown.

Figure 2.10: Energy levels of an ideal ring, disk, and modeled the QR as a function of the magnetic field. The arrows indicate the crossing of the levels where one would expect an AB oscillation.
Figure 2.11: An electron is excited from the valence band to the conduction band in the area outside the barrier material, by a laser. Then the electron becomes trapped in a quantum well \((\Delta \tau \sim \text{ps})\) where they recombine with a hole and emits a photon \((\Delta \tau \sim \text{ns})\).

figure the first AB-oscillation for the ideal ring is shown at 5 T, while the first AB-oscillation for the realistic ring takes place at 15 T. Recent magnetization measurement by N. Kleemans show that the first AB-oscillations occur at 15 T [1]. If we look at the figure for the "disk" shape, no AB-oscillations are shown up to 50T.

We no longer use the angular quantum number \(m\) to label the energies, because in the asymmetric quantum craters this is no longer a good quantum number. The lowest energy level consist of a series of angular momentum quantum number with \(m = 0, -2, -4, \ldots\). The intermixing of states is mainly due to the asymmetry of the system. Although the lowest energy state consists of a series of quantum numbers the \(m = 0\) nature is still strongest present. The second lowest energy level consists of a series of odd angular quantum momentum numbers where the \(m = -1\) dominates. More detailed information can be found in [1].

2.4 Energy spectrum and optical-transition probabilities for an exciton in a QR

When a sample is illuminated by a laser a photon from this excitation source can be absorbed by an electron. This electron is excited from the valence band into the conduction band. The missing electron in the valence band leaves a hole behind, of opposite electric charge, to which it is attracted by the Coulomb force. The bound electron-hole pair is called an exciton and is a neutral particle which is responsible for optical emission. This emission by recombination of an electron-hole pair is called photoluminescence. In figure 2.11 this process is schematic shown for a quantum well. First an electron is excited to the conduction band and will be bound in the quantum well. This process takes in the order of picoseconds. At a time interval in the order of nanoseconds this pair will recombine and emit PL with a wavelength, depending on the energy levels of the electron and hole. Analyzing a PL spectra of a nanostructure sample will give crucial structural information about the sizes and composition of the nanostructures.

We use again Berry’s gedanken experiment to explain the additional phase an electron picks up due to magnetic field. In the exciton case the object consists of two particles see figure 2.12. After one full revolution, the particles obtain a topological phase. These phases will be different for the electron and hole, since they have different trajectories because of their different masses. The phase difference is due to the unequal magnetic fluxes penetrating
Figure 2.12: The origin of the Berry phase for an exciton is illustrated. The electron and hole are placed in separate isolating boxes and rotated along a closed path. The Berry phase, picked up by the electron and the hole, are proportional to the magnetic flux penetrating the electron trajectory, $\Phi_e$. After a full revolution, the electron and hole accumulate different phases because they have a different mass, yielding a relative phase in the pair proportional to the radial dipole moment. As the electron and hole induce opposite currents, the $AB$ effect in this case originates from the magnetic flux through the area between the two trajectories, $\Delta \Phi$. 

Flux lines

\[ \Delta \Phi \]
Figure 2.13: The two limits of an exciton in a ring. In the left image a non-interacting electron and hole pair moving in opposite direction. And the right image shows the interacting electron hole pair, due to coulomb interaction the two move as one.

their paths, causing the "optical" AB-effect.

The "optical" AB-effect is described using an model of an electron and hole rotating in a QR. There are two limits of behavior of an electron and hole pair within a QR. These limits are shown in figure 2.13. The left panel shows the limiting case for no or very small coulomb interaction. The electron and hole are non-interacting and move separately through the ring. At the right panel strong coulomb interaction is considered. The electron-hole pair moves as a neutral particle through the ring [7].

Only electron-hole pairs with zero total momentum can emit photons. This condition arises as the angular momentum of the photon is suppressed by the atomic wave function involved in the optical transition. The optical selection rule $L = 0$ means that the emission intensity of a QR can be dramatically suppressed in magnetic intervals where the exciton ground state acquires a nonzero momentum. These are the so-called "dark" states, $L \neq 0$, no optical transitions possible. States with optical transition possibilities are called the "bright" states, $L = 0$. The suppression of PL as function of magnetic field is a manifestation of the AB effect in the emission spectra of QR’s. The emission intensity as a function of the magnetic field strongly depends on the Coulomb correlation in the exciton.

As shown in figure 2.14 the ground state changes with increasing magnetic field, and may acquire non-zero angular momentum values $L$. For the weakly bound electron-hole pair (left panel) the ground state $(L_e, L_h) = (0,0)$ changes in favor of the states $(0,1), (-1,1), (-1,2), \ldots$, as the field increases. This produces a sequence of electron-hole ground state total angular momentum values, $L_{tot} = L_e + L_h$ of $0,1,0,1, \ldots$, etc. Therefore the weakly bound electron-hole pair switch between "bright" and "dark" states as function of magnetic field. On the other hand, for the strongly bound exciton in the right panel, the ground-state momentum changes increasingly from $L = 0$ to $L = 1, 2, 3, \ldots$, with magnetic field. The strongly bound electron-hole pair will change from "bright" to "dark" at high magnetic fields. The emission spectra of the QR’s are governed by the exciton ground states. For a detailed derivation see the work of Govorov et al [7].

In the next section the energies and PL of the non-interacting and interacting electron-hole pair are determined. In both cases the self-assembled quantum ring modeled previously, figure 2.8, is used in combination with the calculations in section 2.3.2 for an electron and hole in QR.
Figure 2.14: Exciton spectrum, optical emission intensity and magnetization for a weakly (left) and strongly (right) bound electron-hole pair in a ring. Left panel: At low temperature $T$ the exciton emission from a ring is strongly suppressed in well-defined windows of magnetic field. A "bright" ground state has $L = 0$, and it is "dark" otherwise. Right panel: Exciton emission for a strongly bound exciton, the character is different. This leads to the existence of a magnetic field threshold that is well expressed at low temperatures, $T$. 
2.4.1 Energy spectrum of a non-interacting electron-hole pair

Following section 2.3.2 the behavior of an electron and heavy hole in a QR can be calculated with the one band model. Since we have an electron and hole in the QR they are both taken into account. The calculated lowest energy levels of the heavy hole are shown in graph 2.15. The small splitting between the two lowest levels indicate that the hole is almost completely localized. Due to the highly confined state of the hole the energy spectrum is mainly determined by the energy of the electron. The electron is less confined, because the different energy levels for a electron have larger energy difference compared to a hole, due to the difference in effective mass.

In figure 2.16 the energy spectrum calculated from the individual electron and hole spectra. The diagram shows two clear crossings of the ground state at $B \approx 14 \, \text{T}$ and $B \approx 27 \, \text{T}$. These features in the energy diagram of an weakly bound exciton, (a) in figure 2.13 [7], are signatures of the AB effect in PL experiments. At these points the PL switches from "bright" to "dark" states or visa versa.

2.4.2 Exciton states in a strained quantum ring

In this section the aim is to calculate the energy spectrum of a neutral interacting exciton in the same ring, (b) in figure 2.13. The hamiltonian of the interacting exciton in a QR can be written as

$$H_{ex} = H_e + H_h + V_{\text{Coul}}(\mathbf{r}_e, \mathbf{r}_h)$$  \hspace{1cm} (2.19)

This is similar to the Hamiltonian 2.12 with the addition of the coulomb interaction between an electron and a hole with radius-vectors $\mathbf{r}_e$ and $\mathbf{r}_h$, respectively $V_{\text{Coul}}(\mathbf{r}_e, \mathbf{r}_h)$. In order to find exciton eigenstates, we start with constructing the basis functions, which describe...
Figure 2.16: Energy spectrum of a non-interacting electron-hole pair in a self-assembled QR.

A non-interacting $eh$-pair:

$$\Psi_{j_e,j_h}(r_e, r_h) = \Psi_{j_e}^{(e)}(r_e)\Psi_{j_h}^{(h)}(r_h)$$  \hfill (2.20)

Then we diagonalize the Hamiltonian in equation 2.19 with the above basis, looking for the exciton wave functions of the form

$$\Psi_f^{(ex)}(r_e, r_h) = \sum_{j_e,j_h=1}^{j_{max}} A_{j_f,j_e,j_h} \Psi_{j_e,j_h}^{(eh)}(r_e, r_h).$$  \hfill (2.21)

The matrix elements of the Coulomb interaction $V_{Coul}(r_e, r_h)$ are presented as

$$(V_{Coul})_{j_e,j_h} = \int d^2r_e \int d^2r_h [\Psi_{j_e,j_h}^{(eh)}(r_e, r_h)]^\dagger V_{Coul}(r_e, r_h) \Psi_{j_e,j_h}^{(eh)}(r_e, r_h)$$

$$= \sum_{L_{e1},L_{h1},L_{e2},L_{h2}=-L_{max}}^{L_{max}} \int_0^{2\pi} d\varphi_e \int_0^{2\pi} d\varphi_h [\chi_{j_{e1},j_{h1}}^{(e)}(\rho_e,\varphi_e)\chi_{j_{e2},j_{h2}}^{(h)}(\rho_h)]^\dagger$$

$$\times \chi_{j_{e1},j_{h1}}^{(e)}(\rho_e,\varphi_e)\chi_{j_{e1},j_{h1}}^{(h)}(\rho_h)S_{L_{e1}-L_{e2},L_{h1}-L_{h2}}(\rho_e,\rho_h),$$  \hfill (2.22)

where

$$S_{L_{e},L_{h}} = \frac{-\epsilon^2 \rho_e \rho_h}{4\pi \epsilon_0} \int_0^{2\pi} d\varphi_e \int_0^{2\pi} d\varphi_h \int_{-\infty}^{\infty} dz_e \int_{-\infty}^{\infty} \left| \psi_1^{(e)}(z_e; \rho_e, \varphi_e) \right|^2$$

$$\times \left| \psi_1^{(h)}(z_h; \rho_h, \varphi_h) \right|^2 \frac{e^{i\Delta L_{e}+\Delta L_{h}}}{\sqrt{\rho_e^2+\rho_h^2-2\rho_e \rho_h \cos(\varphi_e-\varphi_h)+(z_e-z_h)^2}},$$  \hfill (2.23)

Importantly, the integrals $S_{\Delta L_{e},\Delta L_{h}}(\rho_e, \rho_h)$ do not depend on the magnetic field. Therefore first $S_{\Delta L_{e},\Delta L_{h}}(\rho_e, \rho_h)$ is tabulated and then for each magnetic field $B$, the matrix elements are calculated. The lowest exciton states are found by numerical diagonalization of the Hamiltonian 2.19 in basis 2.20.

In figure 2.17 the calculated energies for the lowest six coulomb states in the quantum ring are shown. As compared to the non-interacting electron hole pair figure 2.16, the Coulomb
interaction leads to a downward shift (∼12 meV) of the energy levels, because the Coulomb interaction decreases the energy difference between the hole and electron. More importantly, when the Coulomb interaction is taken into account the energy-level crossings, which where present for a non-interacting electron-hole pair, disappear. The Coulomb interaction causes the electron hole pair to act as one state, thus no crossing of the electron and hole state. This results in a much smoother dependence of the lowest-state energy on the applied magnetic field as compared to the case of a non-interacting electron-hole pair. Furthermore they enclose less magnetic flux, because their radius is a smaller ring. The calculated diamagnetic shift of the lowest exciton state with increasing the applied magnetic field to 30 T is about 16 meV.

To obtain more higher-lying states, the calculation scheme is modified in what concerns the choice of the basis wave functions of a non-interacting eh-pair, used to diagonalize the exciton Hamiltonian. Now a basis is formed by a fixed number of the lowest eh-states. This basis is more adequate for the higher-lying states. In figure 2.18(a) and (b) the newly calculated lowest energy levels as a function of the applied magnetic field for a non-interacting eh pair and an exciton are plotted. This method differs compared to the energy graphs of the non-interacting electron-hole pair only for the fact that there are more higher-lying states. This basis supplies a more adequate way to determine the complete energy diagram and from this moment on, this method of determining the energy diagram is used.

The number of states is large even in a relative narrow range ∼40 meV above the lowest state. This is shown for both a non-interacting eh-pair and an exciton. However, only a small fraction of these states are characterized by appreciably large transition probabilities for optical excitation or recombination of an exciton. This is illustrated by figure 2.19 (a) and (b), where the spectral distributions of the calculated probabilities $P$ for optical transitions between the exciton vacuum and one-exciton states. For the lowest states of both a non-interacting eh-pair and an exciton, there is a trend to an increase of $P$ with increasing the

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**Figure 2.17**: Energies of the lowest six interacting exciton states in a strained quantum ring as a function of the applied magnetic field.
Figure 2.18: (a) Energies of the lowest exciton states of a non-interacting eh-pair in a isotropic strained quantum ring as a function of magnetic field. (b) Energies of the lowest exciton states in a strained quantum ring as a function of the magnetic field.

applied magnetic field.

In the figures 2.19 the spectral distribution of $P$ and its evolution with $B$ are rather intricate. This is because of the relatively low symmetry of the QR structure, less optical transitions are allowed for a more symmetric ring. As a result, anisotropic QR structure may posses a wide set of allowed optical transitions even in a relatively narrow spectral interval.

2.4.3 An exciton in different isotropic QR’s

In order to clarify the origin of those intricate patterns described in the last section, different isotropic QR’s are considered, figure 2.20. All these seven QR’s have a constant In concentration and strain is not taken into account. Magnetization and AFM measurements on ring-like nanostructures show that the QR’s which we consider are more isotropic than compared to the X-STM sample [1], thus isotropic QR’s are adequate approximations.

In figure 2.20 the different isotropic In$_{0.55}$Ga$_{0.45}$As/GaAs rings used for calculations are shown. From this moment we only consider the luminescence spectra of the rings with an interacting electron-hole pair. The energy diagrams of the different rings can be found in the appendix. In figure 2.21 the spectral probability $P$ for all the seven rings are plotted. Due to the strong selection rules, applicable for the highly symmetric structures shown in figure 2.20, only a small fraction of the energy states are dipole active. Namely, these are the states with zero envelope angular momentum of the eh-pair, $L_e + L_h = 0$. The different QR’s are compared and discussed one by one.

Since the structures under consideration possesses full axial symmetry and the applied magnetic field is parallel to the symmetry axis, the envelope angular momentum of an electron, $L_e$, is good a quantum number ². So, electron states can be labeled as $(n_e, L_e)$ with $L_e = 0, \pm 1, \pm 2, \ldots$ and $n_e = 1, 2, 3, \ldots$, where $n_e$ numbers the different states of the radial motion. The energy levels of the electron correspond then from bottom to top: $(1,0), (1,-1), (1,-2), (1,2), (1,-3), (1,3), (2,0)$.

²Electron spin is not considered
Figure 2.19: Spectral distribution of the transition probabilities $P$ for optical excitation or recombination of a non-interacting eh-pair (a) and an interacting eh-pair (b) as a function of the magnetic field.

Figure 2.20: Shape of the seven QR’s used within the calculations. They are numbered from QR-I to QR-VII and every ring has one different property as compared to QR-I. The gray plot in the QR models is QR-I and the composition is shown in each QR drawing.
Similarly to the case of a conduction electron, the heavy-hole states can be labeled \((n_h, L_h)\). Due to relatively large effective mass of a heavy hole, the number of its energy levels in the energy interval under consideration is large and we will not list all these levels here. The lowest hole states are \((1,0), (1,1), (1,-1), (1,2), (1,-2)\)... For a non-interacting electron-hole pair, the quantum numbers \(n_e, L_e, n_h, L_h\) remain good quantum numbers. Due to selection rules only the states with zero angular momentum, \(L_e + L_h = 0\) remain dipole active. The electron-hole pair states are labeled then from bottom to top: \((1,0;1,0), (1,-1;1,1), (1,1;1,-1), (1,-2;1,2), (1,2;1,-2), (1,-3;1,3), (1,3;1,-3)\) with for all states \(L_e = -L_h\).

The transition probability \(P\) as a function of applied magnetic field for QR-I, figure 2.21(QR-I), shows the same qualitative picture as for the modeled QR, figure 2.19 (b). The two QR spectra differ in energy height for the lowest exciton state, because the shape of QR-I causes the energy to shift downwards. Furthermore an anti-crossing of the lowest two energy levels is clearly shown.

QR-II is compared to QR-I further shifted from the geometry of a doubly connected ring towards the disk-like geometry. The pattern is not very different compared to the one from QR-I. However due to the more "disk-like" shape, the probability density for an electron is somewhat shifted towards the axis as compared to the case of the more "ring-like" shape. This causes the effective radius for the electron angular motion to be smaller and thus the (anti)-crossing shift to higher fields.

The modeled QR-III has a larger radius of the rim than ring our reference ring QR-I. The effective radius of electron states are larger, this causes the transitions fields to be smaller. \(P\)-distributions for an exciton in this ring are mainly determined by the fact that an increase of the rim radius reduces quantization energy for the angular motion of the charge carriers and for their radial motion. Quantitative parameters, which characterize the behavior of the exciton energy spectra and optical-transition probabilities as a function of the applied magnetic field, are very sensitive to the rim radius. Another crucial parameter of QR’s is the thickness of \(\text{In}_{0.55}\text{Ga}_{0.45}\text{As}\) in the central region of the ring. The dependence on the In content and the rim shape is examined. For this the ring structures QR-IV, QR-V and QR-VI, figure 2.20, are developed. QR-IV has a smaller In content, \(\text{In}_{0.45}\text{Ga}_{0.55}\text{As}\) instead of \(\text{In}_{0.55}\text{Ga}_{0.45}\text{As}\), QR-V has a higher and narrower rim and QR-VI has the same width of the rim, but the height is larger. This is all compared to the reference ring QR-I.

Figure 2.21 shows the spectral distribution of the calculated probabilities \(P\) for optical transitions of the different states of an exciton in the different rings, QR-IV, QR-V, QR-VI, are shown. A decrease in In content from 0.55 to 0.45 leads to a significant increase of the exciton energy. Furthermore an increase of the rim height results in a downward shift of exciton energy levels, while a decrease of the rim width increases the exciton energy. Moderate variations of the In content and the rim shape, have a pronounced effect on the exciton energy spectrum.

QR-VII has a larger wetting layer thickness. The small differences in spectral probabilities \(P\) between QR-VII and QR-I are mainly due to the increase of the effective radius of electron states. This due to the fact that there is a larger chance the electron is in the wetting layer, causing the electrons to be less confined in the QR.

The following conclusions can be drawn from the different QR shape calculations. The exciton energy is almost insensitive to moderate variations of the rim radius. While the energy decreases with increasing thickness of InGaAs in the central region of the ring, the rim height and the rim width. Furthermore the exciton energy strongly decreases with increasing In content.
Figure 2.21: Spectral distributions for the seven isotropic rings shown in figure 2.20. Each spectrum is labeled to the shape of the QR. The different energy levels are labeled with \((n_e;L_e;n_h;L_h,)\) from bottom to top: \((1,0;1,0), (1,-1;1,1),(1,1;1,-1),(1,-2;1,2), (1,2;1,-2), (1,-3;1,3), (1,3;1,-3)\).
The diamagnetic shift of the lowest exciton energy levels is almost insensitive to moderate variations of the rim radius. It increases with increasing the thickness of InGaAs in the central region of the ring. Contrary the shift decreases with increasing the rim height and rim width. Furthermore the diamagnetic shift increases with increasing the In content.

Since there is always a distribution of ring sizes it seems worth looking at the effect of the broadening to different states of an exciton. In order to model a non-homogeneous broadening, the distributions are calculated with a Gaussian distribution

\[
P(E) = \sum_i \frac{P_i}{\sqrt{\Pi}} e^{-\left(E-E_i\right)^2/\Gamma^2} \tag{2.24}
\]

with \(\Gamma\) the broadening parameter. \(i\) labels exciton states, \(E_i\) and \(P_i\) are the energy of the \(i\)th state and the corresponding oscillator strength. Preliminary experiments have shown a broadening of \(\sim 15-20\) meV of the PL peaks. Therefore in figure 2.22 the calculated distributions of \(P\) with a broadening of 10, 15, 20 and 25 meV is shown.

As illustrated by these figures, for \(\Gamma \geq 15\) meV the peaks, corresponding to the lowest and first excited electron states, are already non-resolvable in the distributions \(P(E)\) at \(B = 0\). The spectra are fairly simple, but contributions from many states are included.

All the calculations in this section are based on the probabilities of optical transitions to single-exciton states and the exciton-exciton interaction, strain and piezo-electric effect are neglected.
Figure 2.22: Spectral distribution of $P$ for QR-VII, figure 2.20(QR-VII), at: (a) $\Gamma = 10\text{meV}$, (b) $\Gamma = 15\text{meV}$, (c) $\Gamma = 20\text{meV}$ and (d) $\Gamma = 25\text{meV}$.
Chapter 3

QR’s in magnetic field: the determination of the exciton binding

In this chapter the optical measurements on the single layer QR structure will be discussed. First the sample is characterized and the different optical setups are described. Then the optical measurements in magnetic fields on the single QR sample will be discussed. These measurements are separated in two different parts. First the measurements with low excitation energy, where only the ground state of the excitons in a QR is populated. Secondly, the excitation energy is increased and excited states become visible. These measurements of the exciton and excited states at high magnetic fields will be discussed and compared to the theory.

3.1 Characterization of single layer QR sample

In this section the single layer QR sample (G-3861) is characterized by PL and AFM measurements. First a short description of the used PL setup is given followed by the different characterization measurements of the sample.

3.1.1 Characterization PL setup

In figure 3.1 the schematic representation of the characterization PL setup is given. The sample is excited with a Nd:YAG green laser, with an operating wavelength of 532 nm and a power of 25 mW. In front of the laser low pass filter F1 is placed to filter out the higher harmonics of the excitation laser. With the help of lens L1 and a mirror the laser beam is collimated and excites the sample. The spot size on the sample is approximately 4.2 mm$^2$, resulting in an excitation density of approximately 500 mW/cm$^2$.

The PL emitted from the sample is collected and collimated with the help of lens L2. Then the beam is reflected by a gold mirror onto filter F2, where the remaining laser light is filtered out. Finally the PL is focussed on the slit of the monochromator by focus lens L3. This setup uses an InGaAs photodiode array detector with 512 pixels, each with a size of 50x500 µm. The detector has a quantum efficiency larger than 80 % in the near infrared region for wavelength between 0.8 – 1.6 µm.
Figure 3.1: Schematic representation of the PL setup, a laser excites carrier in the sample. The PL due to recombination is measured with the use of a 30 cm monochromator and InGaAs detector.
PL can be collected at temperatures down to 4.2 K, regulated with the use of a continuous flow cryostat, as schematically shown in figure 3.1. The sample is placed in the inner chamber, filled with contact He gas, of the setup. The outer chamber is vacuum to isolate the system. The helium is transported from a helium dewar to the inner chamber where the sample is cooled down. This helium transfers through a heat exchanger which adjusts the flux of the liquid helium. The heat exchanger is also connected to a resistance which can heat the sample, such that temperatures in the range from 4.2 K to 273 K can be reached.

3.1.2 Single layer QR sample G3861

The single layer QR sample is shown in figure 3.2. In this figure a schematic drawing of the single layer ring sample is shown together with an AFM image. The single layer QR’s show a more isotropic shape compared to the X-STM analyzed QR. The QR samples were grown by A.G. Taboada et al by MBE. Two samples were grown, one with a layer of QR’s on top of it to perform AFM measurements and one with a single layer of QR’s in the sample used to perform PL measurements.

From AFM the ring density is determined: $n_{QR} = 8 \times 10^9 \text{ cm}^{-2}$. Furthermore in figure 3.3 the cross sections of a ring in the [110] and [1\bar{1}0] directions are given. The rings have a symmetric round shape and furthermore, the rings have an undulation of height across the

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1. Used by Fomin et al. to model the adiabatic QR potential
2. A.G. Taboada, J.M. Garcia et al. from Instituto de Microelectrónica de Madrid
rim. The typical sizes, obtained by taking height profiles of several rings, are 75 – 95 nm in outer diameter. This inner diameter is 30 – 40 nm and the depth of the depression in the center of the ring is 1 – 1.5 nm. AFM measurements are not suitable to determine the actual ring size underneath the buried material. These AFM pictures give only an impression about the symmetry, density and distribution of the rings. Further X-STM measurements need to be done to be conclusive on the ring sizes.

The PL spectra at 5 K for G3861 is shown in figure 3.4 for two different excitation energies. The peaks have a Gaussian size distribution. The PL profile consists of a mean peak at 1.31 eV and a shoulder that comprises a weaker peak on the low energy side. In figure 3.4 two different excitation densities are shown and the shape of the spectra does not depend on the excitation density. This indicates that the additional peaks are not due to excited states, but most probably causes by a QR’s family with different height. The single layer sample consists of at least two different families of QR’s. The peak of the PL corresponds to a wavelength of $\sim 950$ nm. This wavelength is typically for self-assembled InAs QR’s [22] [23].

### 3.2 Magneto-luminescence measurements

In this section the PL measurements with magnetic field performed in Nijmegen in collaboration with the High Field magnetic Lab (HFML) are shown. First the used setup is treated extensively, after which the PL results for different excitation densities and the magnetic field dependence is discussed. First the PL spectra at different excitation energies are determined, followed by the magnetic field dependence of the exciton in a ring in magnetic fields. All the measurements are performed with QR sample G3861, figure 3.2.
Figure 3.4: PL of single layer QR sample G3861 for two different excitation energies. The mean peak is at 1.31 eV and a peak is appearing on the low energy shoulder. The spectrum is taken at 4.2 K in the characterization PL setup 3.1. The low excitation density spectra is multiplied 10 times. The insert shows a fit with 2 Gaussians, which indicate two different QR heights.
3.2.1 PL spectroscopy in a magnetic field

The setup for magneto-luminescence measurements is given schematically in figure 3.5. The sample is mounted on a sample holder in a cryostat inside a 33 T DC magnet. The excitation laser is a red Dye laser with a wavelength of 620 nm.

First the laser light passes through an filter F1 to filter out the higher order harmonics of the laser. Then to adjust the excitation intensity, a Babinet-Soleil compensator and a polarizer are used. This combination and the fact that the light emitted by the laser is linearly polarized, makes it possible to vary the excitation power over two orders of magnitude. Then with the help of a mirror the laser light is directed downwards onto a microscopic objective. The distance between the mirror and objective is \( \sim 10 \) m. The objective focuses the light on the sample. The sample and objective are placed in a microscope insert. During this project two different combinations of an objective with a microscope insert are used.

For high excitation densities a Newport 10x microscope objective in combination with an insert with \( x, y \) and \( z \) piezo’s as positioners are used. The objective has a working distance of 5.5 mm and a focal distance of 16.5 mm. The objective leads to a small spot size of roughly 1 \( \mu \)m. The setup in combination with this objective is called 3.5-I. This combination is magnetic field dependent and influences the polarization for fields \( \geq 20 \) T, thus no adequate polarization and intensity measurements can be performed.

The second combination is a 10x lens, which has a 100x bigger spot and therefore used for the low excitation measurements. The sample can be positioned by two wedges on the insert, these wedges can move sample about 1 mm in both directions. This combination has no influence on the polarization and is magnetic field independent, we call this combination 3.5-II.

The PL from the sample is reflected by a mirror and both circular polarizations of the luminescence (\( \sigma^+ \) and \( \sigma^- \)) are measured simultaneous. This is done with the use of a Wollaston prism and a polarizer. The PL is focussed by a lens L1 with a focal distance of 10 cm on a 30 cm monochromator with a grating of 300 lines/mm and an slit width of 50 \( \mu \)m. The filter F2 is used to filter out the laser wavelength and the light is detected by an OMAV LN-cooled InGaAs detector with 512x1 pixels.

All measurements in magnetic field are done at liquid helium temperature of 4.2 K. The temperature is determined by a RuO\(_2\) resistor. The magnetic field is generated by a 33 T bitter magnet which consists of four coils. The bore of the magnet is 32 mm and the coils are water cooled. The magnetic field is always perpendicular on the sample.

3.2.2 Magneto-luminescence measurements

In figure 3.6 the typical PL spectra’s of the single layer quantum ring sample are shown. The spectra’s are taken at 4.2 K with \( B = 0 \) and show the different PL spectra at different excitation density. The excitation wavelength is \( \sim 620 \) nm and the PL setup 3.5-I is used. It is shown that due to higher excitation densities two peaks are coming up, these are the first two excited states. The peaks in the spectrum correspond to the transitions indicated in the right panel of figure 3.6.

As shown in figure 3.6 the number of observed peaks depends on the photo-excited carrier density, which can be varied by changing the excitation power. By increasing the excitation power, more charge carriers are created per unit time and more energy levels are filled. The splitting of the ground-state with the two excited states are 40 meV and 74 meV.
Figure 3.5: Schematic setup for the Photoluminescence measurements in high magnetic fields.
Figure 3.6: Left Panel: PL spectra’s at different excitation powers for sample G3861. Measured with setup 3.5-I at 4.2 K at zero magnetic field. The two peaks on the high energy side are coming up at high excitation powers and are the excited states. Right Panel: Dipole allowed transitions at $B = 0$.

Not all optical transitions are allowed, as discussed in section 2.4. For QR’s only recombination of electrons and holes in the same level (such as ground state or excited state) are allowed. This follows from the fact that the orbital momentum of a state must be conserved. The selection rule for spin imply that electrons with spin $S_z = -1/2$ ($S_z = +1/2$) can only recombine with holes with $J_z = +3/2$ ($J_z = -3/2$). These two transitions have a different circular polarization, $\sigma^+$ and $\sigma^-$ respectively, since photon spin carriers $S = \pm 1$ [24]. The difference in polarization is only observed by applying a magnetic field, because at zero field both electron and hole levels are two-fold spin-degenerate.

3.2.3 Low excitation density magneto-luminescence

For this measurements 3.5-II is used. The excitation density is low, therefore only the ground-state of the QR sample is excited. Both polarizations are measured simultaneously for the ground state as a function of applied magnetic field $B$.

The Zeeman effect is the splitting of a energy line into different energy lines in the presence of a magnetic field. The spin Zeeman splitting for electrons in a magnetic field is given by

$$\Delta E = g\mu_B B$$

where the electron Landé $g$ factor determines the magnitude of the Zeeman splitting and $\mu_B = e\hbar/2m_e$ is the Bohr magneton. The Landé factor depends strongly on the size and properties of the investigated QR’s.

In figure 3.7 the PL spectra at different magnetic fields for both polarizations are given. The spectra’s are taken with an collection time of 10 seconds to collect enough PL for a clear signal. The PL spectra are taken with intervals of 0.5 T and only at certain fields the spectra are shown.
Figure 3.7: PL spectra of the ground-state of the QR sample G-3861 shown for both polarization measured at the same time. On the left the $\sigma^-$ and on the right the $\sigma^+$ spectra are shown. The rings are excited at $\lambda \sim 620$ nm with low excitation density. The measure interval is 10 seconds at 4.2 K, slit opening of 250 $\mu$m and a grating of 300 grooves per mm is used. A PL spectra is taken in steps of 0.5 T from 0...30 T, for clarity only six of the 60 measurements are shown.

As seen for both polarizations by increasing magnetic field there is a shift of the energy peak to higher energies, this is the diamagnetic shift and is due to the additional magnetic confinement of the charged carriers in the rings; this magnetic confinement increases the energy levels of the excitons in the ring and the electrons are less confined. The diamagnetic shift is different for the different polarizations, due to Zeeman splitting.

In figure 3.8 the energy peak positions of both polarizations are plotted. The position of the peaks of the PL spectra is determined by the Gaussian size distribution of the QR’s. The inset in figure 3.8 shows the fitting procedure. As mentioned before in this chapter the single layer QR sample consists of at least two different families of rings with different heights, two Gaussians are used to fit the graphs and determine the mean peak position.

From figure 3.8 follows a diamagnetic shift of around 8 meV with a Zeeman splitting between the two polarizations of 3 meV, for 30 Tesla. The diamagnetic shift of QR’s compares well to the value of the diamagnetic shift found for QD’s, for 30 T around 10 – 15 meV [13] [25], although this is of course dependent on the sizes of the QD’s. From the Zeeman splitting the $g$-factor can be determined with equation 3.1. With a zeeman splitting of 3 meV for 30 T the $g$-factor is 1.73. This compares to $g$ factors found for dots [26].

The area of the two Gaussians gives the PL intensity. To check if the PL intensity is reliable, we took measurements at the same magnetic field during sweep up and down, as shown in figure 3.9 for 20 T. The two measurements at the same magnetic field are the same, from this one can conclude that the PL intensity is an accurate parameter.

The intensity measurements are used to determine the relative polarization as a function of the magnetic field. In figure 3.10 the relative polarization $\frac{\sigma^- - \sigma^+}{\sigma^- + \sigma^+}$ is given as a function of the applied magnetic field. The relatief polarization is increasing with increasing magnetic
Figure 3.8: The peak energy positions of the PL spectra of the ground-state of the rings plotted as a function of the magnetic field for both polarizations. The insert shows the fitting procedure: the spectra are fitted with two Gaussians since there are at least two different ring families. The position of the "main" Gaussian is the peak energy in the plot.
field. This because the spins align parallel along with the magnetic field lines, therefore the spins states in the direction of the magnetic field will relatively increase.

3.2.4 High excitation density magneto-luminescence

For the high excitation PL measurements 3.5-II is used. In figure 3.6 three pronounced peaks are shown for high excitation. In this section the effect of an applied magnetic field on the excited states for both polarizations are determined and discussed.

In figure 3.11 the models of Govorov [7] and the Antwerp team are shown for a non-interacting and interacting electron-hole pair in a ring. The model used by Govorov, upper panel, is relatively simply. The model calculated by Antwerp, lower panel, is more sophisticated, because it is based on a realistic ring structure observed by X-STM.

The Govorov model shows crossing of the ground states for both the non-interacting and interacting electron-pair, furthermore it shows no increase in energy for the ground state as function of magnetic field. In the model performed by the Antwerp team there is a shift to higher energies shown as a function of magnetic field and for the interacting electron-hole pair no crossing of the ground state is shown. The ground state for the Antwerp model with interacting electron-hole pair is a smooth increasing curve compared to the non-interacting oscillating curve.

Figures 3.12(a) and 3.12(b) show the experimentally observed PL spectra for different magnetic fields for both circular polarizations $\sigma^-$ and $\sigma^+$ for fields up to 30 T. The ground-state acts similar as for low excitation energy and increases in energy for both polarizations. The first and second excited states split for higher magnetic fields into two peaks.

In the top panel of figure 3.13 the PL spectra of both polarizations are given, with the
Figure 3.10: The relative polarization of the PL spectra for the ground-state of G3861 as a function of the magnetic field. The insert shows the total intensity for the $\sigma^+$ and $\sigma^-$ for the ground states as a function of the magnetic field.

... dashed lines as a guide to the eye. These figures are the similar as in figure 3.12 (a) and (b), but with less spectra shown. The bottom panel shows the energy peak positions of the different peaks as a function of the magnetic field for both polarizations. Quantitatively a clear splitting of the excited states is shown. Furthermore, the second excited state crosses with the first, probably due to the difference in angular momentum $L$ of both states. For one excited state $L$ is opposite to the applied magnetic field, while for the other $L$ is in the same direction. The excited level with $L$ antiparallel to the magnetic field will go to higher energy with increasing magnetic field, while the excited state with the $L$ parallel to the magnetic field will decrease in energy with increasing magnetic field.

A quantitative analysis by fitting a number of Gaussians is not possible for these spectra, since they consist of different QR families and excited states. Therefore the peak positions where determined by eye. The dashed lines are a guide to the eye. By increasing magnetic fields the peak positions are less clear, thus the error in the determined peak position is also increasing. However, this way of fitting for the ground-state is consistent with the data obtained from the low excitation density measurements fitted with the two Gaussians.

The energy diagram for both polarizations shows, as observed before, a Zeeman splitting between the different energy levels. All the three different states show this splitting at 30 T. No accurate values can be determined for the diamagnetic shifts and Zeeman splittings for the excited states, due to the large fitting error.

The measurements are compared to the calculations performed by Govorov and the Antwerp team, both shown in figure 3.11. The interacting electron-hole pair calculations performed by the Antwerp team correspond best to the measurements shown in figure 3.13. The high excitation density PL measurement shows a monotonous diamagnetic shift of the...
Figure 3.11: Exciton spectrums of two models: Govorov and Antwerp for the non-interacting and the interacting eh-pair as a function of the magnetic field. (1) Govorov model for non-interacting eh-pair, (2) Govorov model for interacting electron-hole pair, (3) Antwerp model for non-interacting eh-pair and (4) Antwerp model for interacting eh-pair [7].

Figure 3.12: PL spectra for the two different circular polarizations measured for magnetic fields up to 30 T. The spectra are taken at 4.2 K with an excitation wavelength of ~ 620 nm. The spectra are plotted in steps of 1T.
Figure 3.13: Top panel: PL spectra for the two different circular polarizations for fields up to 30 T. The spectra are taken at 4.2 K with an excitation wavelength 620 nm at 4.2 K. The dashed lines are a guide to the eye. Bottom panel: The peak positions of the ground-state and excited states as a function of magnetic field for both polarizations.
ground state and no crossing of the ground state. This is consistent with the interacting electron-hole pair model by the Antwerp team based on realistic ring parameters. The Govorov model is too simple to describe these self-assembled QR PL measurements.

The interacting electron-hole pair calculations performed by the Antwerp team agree with the measurements and it is clear that the electron-hole pair acts as one neutral particle in our self-assembled QR. Additionally we found no modification of the intensity of the PL lines as predicted by Govorov.

Continuing with the model calculated by the Antwerp team, in figure 3.14 the spectral distribution $P$ for the isotropic QR-VII, figure 2.20(QR-VII), with Gaussian broadening $\Gamma = 20$ meV is given. The graph is in reasonable agreement with the measured spectra, figure 3.12. A remark has to be made regarding the fact that the model is for unpolarized optical transitions.

Figure 3.14: Spectral distribution $P$ of an exciton in QR-VII, figure 2.20(QR-VII), with $\Gamma = 20$ meV.
In the calculations the energy distance between the first and second resolvable peaks of $P(E)$ at $B = 0$ is approximately the same as that between the second and third peak. There is a general qualitative similarity between the spectra of $P(E)$ for $\Gamma = 20$ meV and the measured PL spectra 3.12. Both show one ground-state and two pronounced excited states, furthermore the excited state show similar behavior with increasing field. The measured PL spectra consist of many different excited states, which all contribute to the determined PL spectra.

At the same time, a relatively intricate exciton energy spectrum is rather different from the picture implied by 3.13. The calculations of $P$ cannot be used straightforward comparison with the measured PL spectra. These spectra presumably correspond at high excitation intensities to recombination from few-exciton states and, moreover, are affected by occupation probabilities for these states. Finally, there are indications that the quantum ring structures used throughout the model are slightly different from the one used during the experiments.

Nevertheless, the existing qualitative similarities between the calculated distributions $P(E)$ and measured PL spectra seem indicative. Even for a model without strain, piezoelectric effect, exciton-exciton interaction and realistic In distribution.

A comparison between the peak spacings from the experiments and the theory, one can suppose that the radii of In of the QR structures in the experimental sample are larger than that assumed for QR-I and QR-VII in the modeling. This because the experimental values of the the peak spacing are smaller than those in the model. Furthermore the In content of the rings used in the experiment seems to differ from the content used in the modeling. An increase in In causes a decrease in energy, see QR-IV. Since the energies of the PL spectra in the experiments are higher than in the model, it implies that the In content of the rings measured is lower than the In content determined by X-STM and used in the model [18].
Chapter 4

The confocal microscope: Experimental setup & proof of principle

Each dot and ring in an ensemble has slightly different optical properties due to the inhomogeneous nature of dots and rings. Ensemble measuring masks details that are otherwise available though studying single dots or rings. Self-assembled nano structures have a range of surface density from 0 to 10's of nano structures per $\mu m^2$. The main challenge is therefore to isolate a single dot from the ensemble of millions. Single dot and ring spectroscopy techniques are well established using confocal microscopy [8], the application of nanometer sized aperture masks [27] and lithographic etching of micropillars [28], [29]. During this report a diffraction limited confocal microscope and samples with low dot/ring densities are presented.

For the design of a confocal microscope there are a number of points to consider. The system must be able to operate at low temperatures to reduce the thermal broadening. Long term stability is preferable, since it is often necessary to study one structure for many days. High precision movement is required to bring a single dot accurately into focus. Finally, an important consideration is the adaptability of the system [30].

This chapter describes the confocal microscope elaborately and every component will be discussed in detail. Then the alignment procedure of the system is given and we will proof that our confocal setup works.

4.1 Confocal microscope: principle of operation

Over the years confocal microscopy has become the method for obtaining clear, three dimensional optical images of sample structures. It has been used in many research area’s; from studying biological samples such as cells to fluorescence measurements or examining physical structures like semiconductor quantum dots and nano-optics. The principle of confocal microscopy is shown in figure 4.1 [31].

The confocal imaging system achieves out-of-focus rejection by two strategies, illustrated in figure 4.1. The strategy is to illuminate a single point of the sample with a focussed beam, resulting in a sharp illumination intensity drop above and below the plane of focus. Secondly the use of a blocking pinhole in the conjugate plane to the specimen eliminates the degrading out-of-focus information. In this project a single-mode fiber is used as a blocking pinhole. A
Figure 4.1: Principle of confocal Microscopy
confocal system provides the highest resolution obtainable with conventional optics. As only light from the exact focus of the objective lens is imaged, the system’s spatial resolution is diffraction limited in the case of ideal optical performance. A single dot can be treated as a point source and the diffraction limit for a point source emitter is given by Sparrow’s criterion which describes the full width at half maximum of the central peak of an Airy function\(^1\), \(\Delta x\) as, [30]

\[
\Delta x \approx \frac{0.52 \cdot \lambda}{NA_{obj} \cdot n_{med}}
\]  

\(NA_{obj}\) is the numerical aperture (\(NA\)) of the objective lens, \(n_{med}\) the refractive index of the host material and \(\lambda\) the free space wavelength of the light. Equation 4.1 approximates the spot size assuming a plane wave input and does not account for the gaussian shaped beam profile. This resolution can be further improved by combining confocal microscopy with solid immersion lens microscopy (see section 3.4). With an excitation wavelength \(\lambda = 632.8\) nm and the \(NA_{obj} = 0.65\) gives this a theoretical spotsize of 506 nm.

These points are needed for high resolution optical spectroscopy of semiconductor structures. Investigation of the emitted optical lines of the sample due to changes in the surrounding material, applied voltages, or the deposited optical energy becomes feasible.

4.2 Microscope design

A schematic drawing of the confocal microscope is shown in figure 4.2 and has roughly five sections: the optical head, the cryostat containing the magnet, the excitation unit, the microscope stick with piezo positioners and the detection with a liquid nitrogen cooled CCD camera. The excitation laser and detector are connected to the optical head with the use of single mode fibers. The head sits on the microscopic tube which in turn sits inside the 4 K super conducting 11 T magnet.

4.2.1 The optical head and detection unit

The optical head contains almost all the optics required to direct excitation light to the sample and collect the photoluminescence (PL). A schematic view and a photograph of the optical head is shown in figure 4.3. The optical head consists of three different arms I, II and III shown in figure 4.3. The excitation beam, \(\lambda = 633\) nm, is coupled in using a single mode fiber at arm I and is collimated by a illumination lens (LI) before passing through a beam splitter. The specifications of the lenses and beam splitters used in the head are described in tabel 4.1. The beam splitter reflects 50 % of the laser beam to excite the carriers in the sample. Part of the light reflects back from the sample surface and travels up through the microscope, hitting the topmost beam splitter. 50 % of this light is used by arm II and focussed onto a CCD imaging camera placed directly on the head for alignment, discussed later in this chapter. The other 50 % is collected by the collection fiber at arm III and directed to the detector. The pinhole of the confocal microscope arrangement is provided by the collection fiber with a main field diameter of 5.6 \(\mu m\) [32].

\(^1\)An Airy function is the solution to the differential equation \(y'' - xy = 0\). This is the simplest second-order linear differential equation with a point where the character of the solutions changes from oscillatory to exponential.
Figure 4.2: Schematic view of the confocal microscope system used for single dot and ring studies.
Table 4.1: The optical specifications of the used components in the optical head.

<table>
<thead>
<tr>
<th>Component</th>
<th>Specifications</th>
</tr>
</thead>
</table>
| Illumination lens (L I) | Spectral range: 350 - 1550 nm  
Numerical aperture NA: 0.25  
Focal distance f: 11 mm  
AR-coated range: 600 - 1050 nm |
| CCD lens (L II)     | Spectral range: 350 - 1550 nm  
Focal distance f: 50 mm  
AR-coated range: 400 - 700 nm |
| CCD lens (L III)    | Spectral range: 350 - 1550 nm  
Numerical aperture NA: 0.25  
Focal distance f: 11 mm  
AR-coated range: 1050 - 1600 nm |
| Beam splitter      | Spectral range: 350 - 1550 nm  
Beam splitter cubes: 50:50  
non-polarizing  
AR-coated range: 700 - 1100 nm  
BK7 glass |
| Vacuum window      | Spectral range: 350 - 1150 nm  
Diameter: 25.4 mm  
Thickness: 4.0 mm  
BK7 glass |
| Objective          | Spectral range: 350 - 1500 nm  
Numerical aperture NA: 0.65  
Focal distance f: 2.75 mm  
Working distance: 1.6 mm |
Figure 4.3: on the left a schematic close up of the optical paths inside the microscope head. On the right a photograph of the microscopic head.
The twin beam splitter arrangement also corrects for the beam deviation caused by a single beam splitter. This is also shown in figure 4.3. PL from the sample is offset from the optical axis by the first beam splitter. The second beam splitter counters this offset, redirecting the PL to the optical axis therefore allowing for coupling into the collection fiber in practice. The coupling into the fiber is done with the help of a Collection lens (L III). The clear aperture of this lens is 5.5 mm and the incident light on the collection lens is of 3.6 mm diameter, determined by the objective. The lens is not completely filled and this will give a effective $NA \sim 0.15$. This matches the $NA$ of the collection fiber $NA \sim 0.14$ and gives maximum collection efficiency.

The excitation arm

The excitation beam is provided by a 633 nm laser diode. The laser beam is coupled through a 630 nm single mode fiber onto the microscope head. The excitation arm sits independently from the head. The excitation power can be attenuated by placing absorptive neutral density (ND) filters. To filter the excitation laser out a high-pass filter is placed before the collection fiber. This filter blocks out all the light below 700 nm.

In figure 4.4 the input laser power is plotted as a function of the laser power on a laser detector placed above the vacuum window. This is an indication of the power on the sample, the vacuum window and objective are not included and provide an extra loss. Depending on the spot size of the laser on the sample this gives the excitation power. The theoretical spot size is, according to equation 4.1 $\sim 500$ nm. The corresponding excitation power, with the use of the linear fit from figure 4.4 is then given by:

$$P_{excited}[W/cm^2] = P_{laser}[mW] \cdot 170$$  \hspace{1cm} (4.2)
Figure 4.5: The optical fiber head, coupling the light from the fiber to the detector. The head is designed to optimize the coupling and reduce the losses.

With $P_{\text{excited}}$ the laser power per cm$^2$ at the sample and $P_{\text{laser}}$ the power input of the laser in mW.

The spectrometer

The PL signal is focused on the collection fiber with the collection lens (L III). The fiber is a 850 nm single mode fiber and acts as a pinhole since its core diameter is 5.6 $\mu$m. This single mode fiber has an operating wavelength of 800 - 1050 nm and is connected to the optical head and detector by two FC/PC fiber connectors.

In figure 4.5 the coupling of the fiber into the detector is shown. This is done with an optical fiber connector and the lenses are specially designed to have an optimal coupling between fiber and detector. The spectrometer is a 300 mm monochromator equipped with a 600 grooves per mm and a 150 grooves per mm grating. The camera connected to the end is a back illuminated liquid nitrogen cooled silicon CCD with a 1024 x 256 array from Princeton Instruments. Each pixel measures 26 $\mu$m by 26 $\mu$m. The optimal resolution of the detector is 0.4 nm corresponding to 0.5 meV around $\sim$ 950 nm with a grating of 600 grooves per mm [33]. The camera operates a $T = -90^\circ\text{C}$ and the quantum efficiency of this camera is $\sim$ 90 % at 800-1000 nm [33].

4.2.2 The cryostat

The sample is placed into a cryostat, with a 11 T super conducting magnet. In figure 4.2 a schematic drawing of the cryostat together with the confocal microscope is shown. The length of the microscope stick is such that the sample stage is positioned exactly between the two super conducting magnets.

The cryostat consist of two main area’s separated by a inner vacuum. These area’s are the magnet bath and the insert, both shown in figure 4.2. The magnet bath contains the super conducting magnet. The outside of this bath is isolated with so called super-isolation foil and vacuum to prevent thermal contact with the environment. The height of the helium in the bath is monitored by two resistors placed on different heights. The resistance decreases
when cooled down, the resistance values for respectively room temperature, liquid Nitrogen and liquid Helium: $R_{RT} \sim 30\Omega$, $R_{TN} \sim 7$ and $R_{TH} \sim 2$.

The super conducting magnet is operational when cooled down to 4.2 K. The current through the super conducting magnets is supplied with an external power source and this magnet is made to operate till 11 T. With a super conducting wire the height of the helium level above the magnets is measured. When the magnet bath is filled it is possible to measure two days without refilling. The second area is the insert, in this area the microscope stick is cooled down to 4.2 K. In the insert there is a small helium bath which can be filled [34].

4.2.3 The microscopic stick and x,y,z positioning stage

The microscope stick is used to place the sample in the center of the magnet. The upper part of this microscope stick contains mounts for the optical head. Appropriate electrical cable connectors and a vacuum sealed anti-reflection coated window is present. The stick is inserted in a $\sim 2$ m long stainless steel tube of 50.8 mm diameter. When performing measurements at low temperature the tube is evacuated and then filled with a few mbar of helium exchange gas. This process removes air and moisture from the tube, preventing the formation of air of ice crystals on the sample, positioners and optics when cooled down.

The sample is placed on a x,y,z positioning stage and scanner. Together with the objective lens, mounted on a separate cage plate, the sample and positioners are positioned at the bottom of the $\sim 2$ m long stick. In figure 4.6 the bottom of the stick is shown with the positioning stage and objective lens see table 4.1. The objective is screwed to the top of the titanium tube housing. The lens diameter is 4 mm and the incoming illumination beam is 3.6 mm so the lens is completely illuminated resulting in the optical $NA$. Equation 4.1 shows that the highest resolution is obtained using the highest numerical aperture, therefore full illumination is necessary.

The positioning stage is built from mounting three piezo positioners on top of each other. The piezo’s are moved with the Impact Drive Mechanism [31], this is a method for moving an object under friction by impulsive force. Shown in figure 4.7, the system is considered consisting of two masses $m_1$ and $m_2$ connected by a spring loaded friction mechanism. $m_2$ is connected to a ground with a actuator. During a slow extension of the piezo, $m_1$ is not moving with respect to $m_2$ due to the friction force. After it reaches his final elongation $d$, the piezo is rapidly contracted which induces an inertial force on $m_1$. This force exceeds the static friction, therefore $m_1$ moves along $m_2$ leading to an net displacement $\Delta x$ of the complete system. The piezo’s are operated with an piezo step controller ranging from Voltage steps 0 – 20 V.

To test the exact resolution room temperature a grating with well-known sizes is placed underneath the microscope. The step sizes at room temperature are determined for different voltages. For a step of 20 V the piezo moved $(850 \pm 50)$ nm downwards and $(800 \pm 50)$ nm upwards. Furthermore the step sizes is linear with the applied current 0...20 V. Shown in figure 4.8. When the system is cooled down to 4 K the step size decreases, the resolution of the piezo’s for a 1 V step at 4 K is around $4 - 5$ nm [31].

Apart from positioning the piezo’s are used as a indication of the temperature, since the capacity of the piezo’s changes with temperature. The different capacity values for different temperatures are given in the table 4.2.
Table 4.2: The piezo capacity at different temperatures.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Capacity Value x</th>
<th>Capacity Value y</th>
<th>Capacity Value z</th>
</tr>
</thead>
<tbody>
<tr>
<td>300 K</td>
<td>538 nF</td>
<td>538 nF</td>
<td>530 nF</td>
</tr>
<tr>
<td>77 K</td>
<td>220 nF</td>
<td>219 nF</td>
<td>211 nF</td>
</tr>
<tr>
<td>4 K</td>
<td>119 nF</td>
<td>118 nF</td>
<td>111 nF</td>
</tr>
</tbody>
</table>
Figure 4.7: The inertial drive principle, first the piezo is at its begin position then the sample holder \( m_1 \) after slow extension of the piezo. Rapid contraction of the piezo causes \( m_1 \) to stay on the same position, while the piezo contracts.

Figure 4.8: Step size of the piezo’s as a function of the driving voltage (0 – 20V) for different frequencies at room temperature. (a) Forward motion, (b) backward motion. The filled boxes are measured and the open circles taken from the calibration data [31].
4.3 Solid Immersion Lens technology

A single quantum dot or ring is approximated as a point source, emitting light equally in all directions. The objective lens collects a cone of light emitted from the quantum dot or ring. The solid angle of such a cone is given by

$$\Omega = 2\pi(1 - \cos\theta)$$  \hspace{1cm} (4.3)

where $\theta$ is the angle between the normal and the cone edge. The ratio of $\Omega$ to the solid angle of a sphere, $4\pi$, gives the collection efficiency from the sample as

$$\eta = 0.5(1 - \cos\theta).$$  \hspace{1cm} (4.4)

With the effective NA of the system, $\cos^2\theta = 1 - NA_{eff}^2$, the final collection efficiency is given by:

$$\eta = \frac{1}{2}\left\{1 - \sqrt{1 - \left(\frac{NA_{obj}n_g}{n_s}\right)^2}\right\}$$  \hspace{1cm} (4.5)

With $NA_{obj}$ the numerical aperture of the objective, $n_s$ the refractive index for the host material, 3.5 for GaAs, and $n_g$ the refractive index from the gas above the sample. The large refractive index mismatch leads to an efficiency of 0.87 % for a 0.65 NA objective lens.

The refractive indexes cannot be changed, but the introduction of a numerical aperture increasing lens can increase $n_s$ to reduce the index mismatch. Normally this is done by embedding the system in high refractive index liquid, for example oil. Due to the cryogenic temperatures and surface contacts this option is not possible. Alternatively a Solid Immersion Lens (SIL) can be used [35]. This is a solid hemispherical lens made out of high refractive index material, which is attached to the sample. The hemispherical shape of the lens results in all incident light from the gaussian beam arriving perpendicular to the lens surface. This removes refraction at the SIL/air interface. Furthermore the high refractive index produces a tighter focus of the spot on the sample surface, figure 4.9.

The SIL reproduces two related effects, all linked to the increase in refractive index. The collection efficiency is increased by roughly a factor of $n_{SIL}^2$. The spot size is reduced by a factor of $n_{SIL}$ as shown in equation 4.1.

Attachment of the SIL to the sample is a critical step in the successful use of a SIL. This has to be done with care to avoid damage, dust and contaminates from affecting the SIL or
sample surface. In this project a SIL with $n = 1.83$ is used [36]. To minimize the air gaps and to assist with the positioning of the SIL a custom build SIL application unit was used, shown in figure 4.10.

The SIL is attached using a small amount of two part epoxy glue, applied to the sample with the use of the end of a bare fiber. Subsequent the SIL is placed on the sample with the use of synthetic tweezers. When the SIL is placed at the sample on the glue, the sample and SIL are placed exactly underneath the pressing arm. This arm has a small rubber bung on its end, which is covered with lens tissue to prevent dust on the SIL. Then pressure is gently applied on the SIL and sample with the arm by just releasing it. The rubber bung acts as a buffer to prevent damage on the SIL and helps to provide uniform pressure. After attaching the SIL on top of the sample, they are placed in an oven and baked for 1 hour at 70º. This process dries and makes it stronger in order to make sure it stays intact during cooling down to 4.2 K.

The spot size on the sample is important for single dot measurements. With a small spot size on the sample less dots are excited and will emit PL. With the use of equation 4.1 the theoretical spot is determined: 504 nm.

Experimentally the spot size is determined with an interference setup schematically shown in figure 4.11. A laser is coupled into a collection fiber and is focussed with the use of the same objective as for the confocal microscope on the sample. This light is reflected and the reflection is coupled back into the same fiber. The reflected light from this fiber is collected by a detector with the same wavelength as the laser. To acquire an image of the sample surface,
Figure 4.11: A schematic drawing of the interference setup used to determine the spot size of the confocal microscope. A laser is coupled into a fiber and focussed 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 [37].

In figure 4.12(a) a cross section of such a scan is fitted. Then the derivative is taken and fitted assuming a Gaussian beam profile. The Full Width Half Max (FWHM) can be directly compared to the theoretical diffraction limit of 506 nm. In figure 4.12(b) the derivative is taken and the FWHM is determined: (498 ± 30 nm).

Figure 4.13 shows two images of the grating one image without SIL (a) and one with SIL (b). This is an example of the magnification (factor 3.25), produced by imaging through a n=1.83 SIL onto a 2µm grating. The images are non-linear, this is due to the piezo’s which have different step sizes when operated near there maximum range.

The improved spatial resolution obtained by using a SIL can be measured performing an identical resolution measurement described in the section about the spot size without a SIL. With a SIL of $n_{SIL} = 1.83$, equation 4.1 predicts an improvement in resolution from 504 to 275 nm. Figure 4.14 shows the same measurement as figure 4.12.

The horizontal axis of figure 4.14(a) is corrected with the magnification 3.25 determined before. Then a FWHM of (260 ± 20 nm) is measured. This consistent the values predicted by the calculations. This is also shown with the following equation of the spot size without SIL divided with the spot size with SIL.

\[
\frac{498}{260} = 1.92 \quad (4.6)
\]
This is comparable to the expected 1.83, the $n$ of the SIL.

4.4 System alignment

The importance of accurate and precise system alignment should not be overlooked, since the signal intensity is very low and every photon counts. Each optical element in the system, except the lens for the imaging camera, has accurate $x, y$ and $z$ positioning. The entire microscope head sits on a tilt stage allowing for translation in the $x$ and $y$ direction of the entire head relative to the microscope stick. These minor corrections are required to keep a straight optical path through the microscope body.

Alignment of the system for PL measurements involves roughly four steps. First the excitation laser and collection arms of the optical head are aligned separately by collimating the laser beams coming from these arms. Then both spots are positioned on the same position at a distance of $\sim 5 \text{ m}$. The diameter of the excitation laser need to be $\sim 3.6 \text{ mm}$ to fully illuminate the objective. Accurate collimation at the collection arm has been shown to be essential in demonstrating diffraction limited performance.

The second step is to place the optical head on top of the microscope stick. First the laser is placed in the middle of the objective using the positioners of the tilt stage. Then the positioners of the sample holder are used to place the sample in focus of the laser and in the center of the SIL. The center of the SIL roughly determined by eye and then precise alignment is done by the positioners to determine the exact center. When the spot is in focus on the sample or SIL a refraction pattern is shown on the camera display.

After the head is aligned and the spot is positioned on the sample the microscope stick is put into a vacuum tube and flushed five times with He contact gas. After flushing the tube is filled with $\sim 20 \text{ mbar}$ of He contact gas. To reach low temperatures the tube is place into the...
Figure 4.13: A 2 μm “chess” grating imaged by scanning the surface, (a) without the SIL and (b) with SIL. This shows the magnification properties of the SIL.
cryostat and cooled down to 4.2 K. The temperature is monitored by looking at the capacity of the piezo’s.

When the piezo’s reach liquid He temperature the third alignment step will take place, since the cooling down and possible vibrations during handling changed the position of the head. This has to be corrected with the positioners of the tilt stage until the focus is found again. A strong ND filter is placed in front of the laser and a small laser spot is shown on the camera, when the focus is on the center of the SIL. With the use of the tilt stage and the excitation arm $x,y,z$ positioners this spot has to be made concentric. To check if this spot is exactly the focus one has to move the $z$-piezo up and down, during this handling the spot has to stay on the same position and remains concentric.

Finally, the PL has to be focussed on the collection fiber. This is done with the $x,y,z$ positioners of the collection arm. First the laser peak at 633 nm on the detector has to be maximized by positioning the collection arm. Then the spectrometer is moved to the desired wavelength of interest and there the PL signal is maximized by only using the collection arm. Since the laser is focused on the top of the sample or SIL, the sample needs to be moved with the $z$-piezo to focus the light on the sample layer where the dots or rings are. By changing the $z$-piezo the PL signal is optimized. When the PL is maximized with the collection arm a high pass filter ($\lambda \geq 700$ nm) has to be placed before the collection fiber.

It is a difficulty getting optimal alignment of the system. When no PL is shown on the first sight, then one should remove the ND filter and excite hard. Then the wetting layer and the material of the substrate, mostly GaAs, will come up. This PL signal is then used to optimize and align the system. Then the ND filters are placed back in front of the laser and one can start measuring.
4.5 Proof of principle of the confocal microscope on single QD’s

In this section the single dot sample R80 is described and characterized. This sample is used to proof the principle of our setup. Secondly, in this section the results are given of the test measurements of the confocal microscope. The results are shown for measurements at 77 K and 4.2 K.

4.5.1 Single layer QD sample R80

To test the confocal microscope, single dot PL is investigated. For this purpose single layer self-assembled QD’s are needed with a low density of dots. To accomplish this a single layer of InGaAs quantum dots is grown under specific growth conditions on top of a GaAs substrate and then covered with GaAs and covered with a AFM top layer. The sample is grown by solid source MBE at the Eindhoven University of Technology\textsuperscript{2}. An schematic drawing of this sample together with an AFM picture of the surface of the sample is shown in figure 4.15.

From AFM the dot density is determined: $n_{QD} = 4 \times 10^8 \text{ cm}^{-2}$. Furthermore in figure 4.16 the cross sections of a dot in the [110] and [1¯10] directions are given. The QD’s are of similar sizes and have a isotropic shape. The typical sizes of our QD’s, obtained by taking height profiles of several dots, is 60 – 70 nm in diameter and 7 – 10 nm in height.

The macro PL spectra at 4.2 K of the low density dots sample R80 are shown in figure 4.17 for two different excitation powers. These measurements are performed with the PL characterization setup 3.1. The PL-profile consists of peak at 1.27 eV and at the high energy shoulder there are some weaker peaks. These peaks are independent on excitation density. This means that the additional peaks are not excited states. The peaks can most probably

\textsuperscript{2}Grown by R.G. Hamhuis and R. Nötzel
be described to QD’s with different heights, as we already observed for the rings.

4.5.2 Single dot measurements at 77 K

The 77 K measurements are performed outside the magnet, this microscope stick is put into a cryostat filled with liquid Nitrogen. In figure 4.18 single dot PL spectra are shown for different excitation densities. The high intensity lines are PL signals from individual dots. It is not possible to determine whether one sees the ground state or an excited state. This is because we excite to many dots.

The single dot lines are on top of a broad PL peak and the lines itself are broadened. This is due to the thermal broadening of the PL at 77 K. Another reason the single dot lines are quite broad is due to the resolution of the spectrometer, \( \sim 1 \) meV.

To prove that the lines on top of the broad PL peak are single dot PL lines, two measurements on the same position on the sample at very low excitation density and long measurement interval are performed. These measurements are shown in figure 4.19 and the measuring interval is 600 s. The spectra are placed above each other for clarity reasons. The spectra reproduce nicely, indicating that the confocal microscope is stable.

Furthermore, a final test of measuring at different positions of the sample is performed. The single dot lines should appear and disappear when going to different position of the sample. In figure 4.20 four different PL spectra of single dot PL lines are plotted. All the four spectra are on a different position of the sample by using the x and y piezo’s to move the sample a couple of hundred nanometers. The PL lines we obtain are lines from single dots, because with the movement of the sample single dots go in and out of the laser spot and therefore emit more or less PL. That is the cause of peaks appearing and disappearing.

In figure 4.20 one can clearly see that lines are disappearing by looking at the line indicated with the arrow at \( E = 1.328 \) eV. At position 2 the PL line is very intense, while it is completely disappeared at position 3. At the two other positions the peak is also shown, but less intense as compared to position 2.

It is possible to follow the single dot lines and see them appear and disappear. The resolution of the monochromator together with the thermal broadening is limiting the resolution.
Figure 4.17: PL of single layer QD sample R80 for two different excitation energies. The mean peak at 1.267 eV consists of several peaks indicated with the arrows. The exposure time is 0.1 s and the slit width is 250 μm. The spectrum is taken at 4.2 K using a grating of 600 grooves/mm. The low excitation density graph is taken with 25% of the original excitation density.
The dots are excited with $\lambda = 632.8 \text{nm}$ and measured with a 600 grooves/mm at time intervals of $t = 60 \text{s}$ at $T = 77 \text{K}$ with a slit width of 250 $\mu \text{m}$.

In the next section measurements are done at $T = 4.2 \text{K}$ to exclude the effect of thermal broadening.

4.5.3 Single dot measurements at 4.2 K

After the proof of principle of the confocal microscope at 77 K, measurements are performed at 4.2 K with the microscope placed in the magnet. In figure 4.21 single dot PL lines of different dots at 4.2 K of the single layer quantum dot sample R80 are shown. Compared to the 77 K measurements the 4.2 K measurements show more sharp individual lines. Less thermal broadening is shown, therefore the background shown in the 77 K measurements disappears.

With the use of the $x$ and $y$ step piezo’s the sample is moved in the order of hundreds of nanometers. Again it is tested if the single dot PL line appear and disappear at different positions. In figure 4.22 the result of four different positions measurements are shown. The laser spot is placed on a position with a low density of dots, so few lines are visible, furthermore a low excitation density is used. As an insert in the graphs a position plot is shown, this is the exact position of the measurement compared to the begin position shown in the top left graph (position 1). Two different individual lines are marked in all the four graphs. One line with a square ($\lambda = 1.326 \text{eV}$) and the other one with a circle ($\lambda = 1.272 \text{eV}$). When the squares are followed the lines show the same behavior as observed before at the 77 K dot measurements. The peaks appear and disappear nicely. The line indicated with the square is clearly shown in positions 3 and 4, while is is completely disappeared in 1 and 2.
Figure 4.19: PL spectra of R80 taken on the same position during 600 seconds and extremely low excitation density. The spectra reproduce nicely, measured at 77 K with a grating of 600 grooves/mm and a split width of 150 µm. The intensities of both measurements are exactly the same, but for clarity one spectra is placed above the other. The high intensity lines are due to cosmic rays.
Figure 4.20: PL spectra of single dot PL lines on four different positions at 77 K. One can see the appearance and disappearance of the line indicated with an arrow at $E = 1.328$ eV. This shows that we excite and measure PL from single dots.
The lines indicated with a circle are more intense in position 1 than in the rest of the positions, but the peak is shown in all four positions. The difference between position 2 and 3 is 700 nm and with a spot size of $\sim 260$ nm one would expected it to disappear, since that particular dot should be out of the laser spot. However, the relative high excitation density, due to the small spot size, causes surrounding dots also to emit by diffusion. Therefore not only the dots in the laser spot are excited, but also in the surrounding area. This problem is hard to solve by reducing the excitation density, since the PL signal becomes extremely low and measuring intervals too large.

The setup is proven at 4 K to monitor single dot PL and it is possible to follow single dot lines. As a final test the single dot PL lines behavior with a magnetic field is determined. In figure 4.23 a series of single dot PL spectra are as function of magnetic field. The resolution of the monochromator prevents the lines to have a Gaussian of Lorentzian distribution. This bad resolution makes impossible to see any splitting of energy levels at high magnetic field, as shown in chapter 3. In order to see the shift of the peak of the single dot line a dashed line as a guide to the eye is fitted. This is the diamagnetic shift, discussed before.

The intensity of the PL is decreasing with higher magnetic fields. While increasing the field the piezo positions of the microscopic stick change a bit. For every Tesla increase in magnetic field the $z$-piezo is needs to be corrected with 1 or 2 piezo steps of 20 V. Better result can be obtained by proper corrections.

In figure 4.24 the peak position of the single dot lines in figure 4.23 are shown. The peak position is determined by using a Lorentz function, Gaussian function and by ”eye” to fit the graphs. The three different fitting procedures show the same trend and values.
Figure 4.22: PL spectra of single dot PL lines on four different positions at 4.2 K. One can see the appearance and disappearance of the lines as shown before in figure 4.20. Two different single dot lines are indicated with a square (λ = 1.326 eV) and a circle (λ = 1.272 eV).
Figure 4.23: PL spectra of single dot PL lines for magnetic fields up to 10 T for unpolarized light. The spectra are taken at 4.2 K with an excitation wavelength of 633 nm. The dashed line is a guide to the eye for the peak position of the single dot PL line.
The diamagnetic shift is clearly shown and in the order of 1 meV. This is consistent with measurements in literature [13] and with the single layer of ring measurements described in this chapter.

In this section it is shown that with the confocal microscope single dot PL measurements can be performed. For both 77 K and 4 K single PL lines are shown. When moving the sample with the positioners, single dot PL lines appear and disappear. The system is stable to perform long interval measurements and a diamagnetic shift of \( \sim 1 \) meV is shown for fields up to 10 T. The resolution of the system is a problem, one will not be able to show splitting of the individual lines of excited states due to the resolution of 1 meV, FWHM of peak in figure 4.23.

In figure 4.25 a single QD spectrum is shown for different gate voltages \( V_g \). The neutral exciton, singly and doubly charged excited states of a single QD are indicated by respectively \( X^0, X^+ \) and \( X^{++} \). As one can see the resolution of the PL lines is better than 1 meV. The \( 2X^0 \) peak corresponds with the presence of two excitons in the QD, bi-exciton state. The splitting of the lines, shifts and the appearing of the charged excited states are within the area of 1 meV. This indicates that our confocal microscope setup with the 30 cm monochromator is not suitable for accurate single PL measurements. This problem is solved by the installation of a triple monochromator. The resolution of this monochromator is 20 \( \mu \)eV, so in combination with the confocal microscope the resolution will become better and single dot/ring PL as shown in figure 4.25 can be performed. No test measurements are performed with this monochromator, because the triple monochromator was only installed in the last weeks of my research project.
Figure 4.25: Single dot PL spectra for two separate dots as a function of hole charging from -1.9V (lower trace) to +0.3V (upper trace). The $X^0$, $X^+$ and $X^{2+}$ emission lines are observed corresponding to the neutral exciton and singly and doubly positively charged excitons, respectively. Bi-exciton emission ($2X^0$) is also observed. The interesting area of this figure is located within 1 meV indicating that a monochromator with the resolution of 1 meV is not suitable to perform accurate single dot PL measurements [38].
Chapter 5

Conclusions and suggestions

5.1 Conclusions

We studied the behavior of an electron-hole pair, called an exciton, in a self-assembled quantum ring by polarized photoluminescence in high magnetic fields up to 30 T. The exciton ground state in a quantum ring shows a diamagnetic shift of 8 meV and a Zeeman splitting of 3 meV at 30 T. The degree polarization in the direction of the magnetic field increases. The \( g \)-factor of the quantum rings in the sample used for the magneto-luminescence measurements is 1.73, this is in the same order as the \( g \)-factor determined for dots \[26\]. For high excitation density measurements on the single layer quantum ring sample, excited states of the exciton in a ring appear. The excited states show energy level splitting and Zeeman splitting as function of magnetic field.

More interestingly, Goverov shows that there are "optical" Aharonov-Bohm effects for both a coupled and uncoupled electron-hole pair in a ring. The Goverov model is rather simple, while a more sophisticated model is developed by the Antwerp team based on realistic ring parameters. The Antwerp model shows that in the case of strong Coulomb interacting electron-hole pair in a ring, there are no variations in photoluminescence intensity contrary to what was predicted by Goverov. Secondly, no crossing or deviation from a monotonous diamagnetic shift of the ground state photoluminescence lines is determined. The model predicts the absence of "optical" Aharonov-Bohm effects for an exciton in a self-assembled quantum ring up to magnetic fields of 30 T.

The model explains quantitatively the experimentally observed photoluminescence of the exciton ground- and excited states as a function of magnetic field in our self-assembled quantum rings. The behavior of the electron-hole pair in a ring is consistent with the model developed by the Antwerp team. The coulomb interaction in self-assembled quantum rings is strong enough to bound the electron-hole pair and let it act as a neutral exciton.

In the future we will study the photoluminescence of single quantum rings structures and therefore a confocal microscope is developed and tested on a single layer quantum dot sample. At 77 K and at 4.2 K single dot photoluminescence lines are measured and one is able to study the photoluminescence lines of the single dots. Single dot measurements are performed up to 10 T and a diamagnetic shift of \( \sim 1 \) meV at 10 T is observed and this value corresponds with other observations in the literature \[13\]. The resolution of the monochromator (1 meV) is the limiting factor at the moment to see splitting of energy levels, for instance due to Zeeman effects or the formation of charged excitons and bi-excitons. This will be resolved with the
installation of a triple monochromator with a resolution of $20 \, \mu\text{eV}$.

5.2 Suggestions

In order to determine the photoluminescence intensity and accurate degree of polarization, the high excitation density measurements of the exciton need to be repeated in combination with an insert without magnetic field and polarization dependence. Then one would be able to comment on the increase or decrease of spins states and system preferability. To get more electronic information on the rings, capacitance-voltage experiments should be performed on the electrons in a quantum ring. One can tune how many electrons there are in a ring and determine the behavior of the ring containing a different amount of electrons at different magnetic fields. This will give an conclusive answer to the behavior of one electron in a ring.

To further explain the high magnetic field exciton measurements, the model has to take polarization into account, furthermore exciton-exciton interaction, strain and In content are not taken into account at this model. These parameters will improve the understanding of the high excitation density photoluminescence measurements. X-STM measurements on our more isotropic rings will lead to a better parameterization of the ring we actually measured on.

To exclude the exciton-exciton interaction, Photoluminescence excitation (PLE) measurements have to be performed. Here one excites with a wavelength range of for example 800 – 1000 nm. Therefore the ground state and excited states will appear separately and only one exciton is in the ring. Time-resolved photoluminescence measurements need to be performed to see better if dark-bright oscillations occur in our ring geometry.

The next step in the confocal microscope project is to measure single ring photoluminescence. The problem of the confocal microscope is the resolution of the monochromator. Therefore two crucial improvements for the confocal microscope: a high resolution monochromator, and the installation of polarizers to measure left and right circular polarized light. This will be extremely interesting, since than one can follow single dot or ring photoluminescence line with high resolution for both polarizations. Splitting of energy levels for a single ring can be monitored by sharp photoluminescence lines. Already a triple monochromator is installed with a resolution of $\sim 20 \, \mu\text{eV}$.
Chapter 6

Acknowledgements

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Appendix A

If one solves the single-particle Schrödinger equations, with \( \beta \) indicating an electron \((\beta = h)\) or electron \((\beta = e)\),

\[
H_{\beta} \Psi^{(\beta)}(\mathbf{r}) = E \Psi^{(\beta)}(\mathbf{r}) \tag{A.1}
\]

using the Ansatz:

\[
\Psi^{(\beta)}(\mathbf{r}) = \psi^{(\beta)}_{k}(z; \rho, \varphi) \Phi^{(\beta)}_{kj}(\rho, \varphi) \tag{A.2}
\]

where the index \( k \) number the subbands due to the size quantization along the \( x \)-axis. The motion is separated into a fast motion along the \( z \)-direction a slow motion along the \( x, y \)-plane. The confinement energy in the \( z \)-direction as higher compared to the \( x, y \)-direction due to the smaller height compared to the lateral dimensions.

This separation gives the following Schrödinger equation for the "fast" motion:

\[
\left[ -\frac{\hbar^2}{2} \frac{\partial^2}{\partial z^2} + \frac{1}{m_{\beta}(\rho, \varphi, z)} \frac{\partial}{\partial z} \pm V_{\beta}(\rho, \varphi, z) \pm \delta E_{\beta}(\rho, \varphi, z) \right] \psi^{(\beta)}_{k}(z; \rho, \varphi) = \xi^{(\beta)}_{k}(\rho, \varphi) \psi^{(\beta)}_{k}(z; \rho, \varphi) \tag{A.3}
\]

The upper/lower sign stands for conduction electrons/ heavy holes, respectively. The equation is numerically solved for each node of a two-dimensional grid in the \((\rho, \varphi)\)-plane. As a result, we obtain the adiabatic potentials \( \xi^{(\beta)}_{k}(\rho, \varphi) \), tabulated on the aforementioned grid.

The Schrödinger equations for the "slow" motion,

\[
\left[ -\frac{\hbar^2}{2} \nabla^2 + \frac{e}{\hbar} \mathbf{A} \right] \frac{1}{m^{(\beta)}_{k}(\rho, \varphi)} + \xi^{(\beta)}_{k}(\rho, \varphi) \Phi_{kj}^{(\beta)}(\rho, \varphi) = E_{kj}^{(\beta)} \Phi_{kj}^{(\beta)}(\rho, \varphi) \tag{A.4}
\]

with again the upper and lower sign for, respectively electrons and holes. The adiabatic potential \( \xi^{(\beta)}_{k}(\rho, \varphi) \) and the effective masses \( m^{(\beta)}_{k}(\rho, \varphi) \) determine the eigenstates of the in-plane motion, which are labeled by the index \( j \). The eigenstates are determined by a numerical diagonalization of the Hamiltonian. To simplify a basis of eigenfunctions of a Hamiltonian is chosen such that:

\[
\xi^{(\beta)}_{k}(\rho) = v^{(\beta)}_{k} + \omega^{(\beta)}_{k} \rho^2. \tag{A.5}
\]

Since this potential is isotropic, the corresponding in-plane eigenfunctions can be written down as

\[
\Phi_{knL}^{(\beta)}(\rho, \varphi) = \chi_{knL}^{(\beta)}(\rho) e^{iL\varphi} \tag{A.6}
\]
where \( n \) labels the radial solutions.

Inserting the adiabatic potential A.5 instead of \( \chi_{i\, k}^{(\beta)} \) into equation A.4 This in combination with a constant \( m_i^{(\beta)} \) and wave functions in the form of equation A.6, gives the one-dimensional Schrödinger equation

\[
(\chi_{k\, nL})'' + \frac{1}{\rho} (\chi_{k\, nL})' + \left[ \frac{2m_k^{(\beta)}}{\hbar^2} (E_{k\, nL} - v_k^{(\beta)}) + \frac{L^2}{\rho^2} - \frac{L^2}{\rho^2} - (\alpha_k^{(\beta)} \rho)^2 \right] \chi_{k\, nL} = 0 \tag{A.7}
\]

where \( l = \sqrt{\hbar/(eB)} \) is the magnetic length,

\[
\alpha_k^{(\beta)} = \sqrt{\frac{1}{4l^4} + \frac{2m_k^{(\beta)} w_k^{(\beta)}}{\hbar^2}}. \tag{A.8}
\]

The corresponding eigenfunctions are:

\[
\chi_{k\, nL}^{(\beta)}(\rho) = C_{k\, nL} \rho^{|L|} \exp\left(-\frac{\alpha_k^{(\beta)} \rho^2}{2}\right) L_n^{(|L|)}(\alpha_k^{(\beta)} \rho^2), n = 0, 1, 2, \ldots \tag{A.9}
\]

where \( L_n^{(m)}(x) \) are generalized Laguerre polynomials and \( C_{k\, nL} \) are normalized constants. The eigenenergies are

\[
E_{k\, nL}^{(\beta)} = v_k^{(\beta)} + \frac{\hbar^2}{m_k^{(\beta)}} \left( (n + \frac{1}{2} - \frac{L^2}{2}) - \frac{1}{l^2} + |L| \alpha_k^{(\beta)} \right) \tag{A.10}
\]

For each value of the applied field, the electron and hole eigenstates in the quantum ring are found by numerical diagonalization of the adiabatic Hamiltonian. As a result of diagonalization, we obtain the energies, \( E_{ij}^{(\beta)} \), and the wavefunctions,

\[
\Psi_{ij}^{(\beta)}(r) = \psi_1^{(\beta)}(z; \rho, \varphi) \sum_{L=-L_{\max}}^{L_{\max}} \sum_{n=0}^{n_{\max}} a_{jnL} \chi_{1nL}^{(\beta)}(\rho) e^{iL\varphi} = \psi_1^{(\beta)}(z; \rho, \varphi) \sum_{L=-L_{\max}}^{L_{\max}} \chi_{1L}^{(\beta)}(\rho) e^{iL\varphi}, \tag{A.11}
\]

of the lowest electron and hole states in the quantum ring as a function of the applied magnetic field.