Optical Detection of the Hyperfine Interaction

in a Positively Charged

Self-Assembled Quantum Dot

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Abstract

In this dissertation, optical studies of the interaction between either a single hole spin or a single electron spin confined to a self-assembled InGaAs quantum dot (QD) with the nuclear spin ensemble of the host material are presented. QDs are semiconductor structures that allow for a charge carrier confinement in all spatial directions; this leads to a quantization of the electronic states. Dipole allowed transitions between the electronic states enable an optical spectrum similar to the one of atoms. In fact, a QD consists of $10^5$ atoms with nonzero nuclear spin. A single hole spin or electron spin confined to the QD couples to this ensemble of nuclear spins via hyperfine interaction. Optical orientation of the electron spin polarization can be transferred to nuclear spins via hyperfine interaction, leading to dynamic nuclear spin polarization (DNSP). On the other hand the nuclear spin polarization acts back on the electron spin, causing an energy splitting of the electronic spin levels. This characteristic can be exploited to optically measure DNSP by performing spectroscopy.

In the first part of this thesis, we investigate the evolution of nuclear spin polarization in a transverse magnetic field via a single QD electron spin. While in a magnetic field along the growth direction a pronounced DNSP is expected, in a transverse magnetic field the nuclear spin polarization should vanish because of the Larmor precession of the electron. Experimentally, nuclear spin polarization compensates transverse magnetic fields $B_x$ up to $\sim 1.7T$. Therefore, the orientation of the nuclear spin polarization has to be mainly perpendicular to the optically defined electron spin polarization. To further investigate this anomalous behavior, we develop a pump probe technique that allows to fully characterize the nuclear spin polarization as function of $B_x$ by resonant spectroscopy. The aim is to find out whether the evolution of nuclear spin polarization in transverse magnetic fields can be mapped to quantum optics models. We find that the nuclear spin polarization in transverse magnetic fields strongly depends on the sample structure and is very likely an inherent property of self-assembled InGaAs QDs due to their strain.

In the second part of this thesis, coherence properties of single hole spins confined in a QD are studied with time resolved measurements. By optically charging the QD in an n-doped field-effect structure we can benefit from the superior electronic properties of such devices. We find a coherence time $T_2^*$ of 240ns which is an or-
der of magnitude larger than what was previously measured in p-doped samples. Additionally, we observe that the hole spin coherence measurements are strongly influenced by nuclear spins.
Zusammenfassung

In dieser Arbeit werden optische Studien der Interaktion zwischen einem einzelnen Lochspin oder einem einzelnen Elektronenspin, der in einem selbstorganisierten InGaAs Quantenpunkt (QP) eingeschlossen ist, mit dem Kernspinensemble des umgebenden Materials vorgestellt. Quantenpunkte sind Halbleiterstrukturen, die eine dreidimensionale räumliche Eingrenzung von Ladungsträgern erlauben, was eine Quantisierung der Elektronenzustände ermöglicht. Erlaubte Dipolübergänge zwischen den Elektronenzuständen führen zu einem ähnlichen optischen Spektrum wie in einem Atom. Im Gegensatz zu einem Atom besteht ein Quantenpunkt jedoch aus $10^5$ Atomen, die alle einen endlichen Kernspin besitzen. Ein einzelner Lochspin oder Elektronenspin im Quantenpunkt koppelt zu diesem Kernspinensemble über die Hyperfeinwechselwirkung. Deshalb kann die optisch verursachte Polarisation des Elektronenspins auf die Kernspins übertragen werden, was zu einer dynamischen Kernspinpolarisation (DKSP) führt. Andererseits bewirkt die Hyperfeinwechselwirkung eine Aufspaltung der Energiezustände des Elektronenspins, wenn das Elektron in Kontakt mit den spinpolarisierten Kernen steht. Diese Eigenschaft wird verwendet, um den Grad der DKSP mit Hilfe von spektroskopischen Methoden optisch zu bestimmen.

Im ersten Teil dieser Arbeit wird die Entwicklung von Kernspinpolarisation in einem transversalen Magnetfeld mit Hilfe von einem einzelnen Elektronenspin im Quantenpunkt untersucht. Während in einem Magnetfeld entlang der Wachstumsrichtung des QPs eine starke DKSP erwartet wird, sollte die Kernspinpolarisation in einem transversalen Magnetfeld aufgrund der Larmorpräzession des Elektrons verschwinden. Experimentell kann eine Kompensation des externen Magnetfeldes $B_x$ durch die Kernspinpolarisation bis zu $\sim 1.7T$ festgestellt werden. Dazu muss die Kernspinpolarisation mehrheitlich senkrecht zur optisch definierten Polarisation der Elektronenspins orientiert sein. Um diesen ungewöhnlichen Polarisationsmechanismus der Kernspins zu studieren, wird eine Pump-Probe Technik entwickelt, die eine vollständige Charakterisierung von der Kernspinpolarisation als Funktion von $B_x$ mit Hilfe von resonanter Spektroskopie erlaubt. Das Ziel dabei ist herauszufinden, ob die Entwicklung der Kernspinpolarisation in einem transversalen Magnetfeld mit quantenoptischen Modellen beschrieben werden kann. Wir stellen fest, dass die Kernspinpolarisation in transversalen Magnetfeldern sehr stark von der Probenstruktur
abhängt und sehr wahrscheinlich eine inhärente Eigenschaft von selbstorganisierten InGaAs QP aufgrund ihrer Gitterverspannung ist.

Contents

Title a

Abstract d

Zusammenfassung e

Contents g

List of symbols and abbreviations i

1. Introduction 1
   1.1. Motivation and scope of this thesis ................................. 2

2. Self-assembled Quantum Dots and Nuclear Spins 5
   2.1. Charge tunable self-assembled Quantum Dots ......................... 5
       2.1.1. Growth of self-assembled quantum dots ........................... 6
       2.1.2. Quantum dot level scheme and optical selection rules ........ 7
       2.1.3. Charge control ................................................. 9
       2.1.4. Quantum confined Stark effect ................................. 11
   2.2. Nuclear Spins in Quantum Dots ................................... 13
       2.2.1. Electron and Hole Spin System ................................. 13
       2.2.2. Nuclear Spin System .......................................... 14
       2.2.3. Isotropic and Anisotropic Hyperfine Interaction ............. 16
       2.2.4. Electron Spin Decoherence .................................... 18

3. Experimental Methods 21
   3.1. Photoluminescence ............................................... 21
   3.2. Resonant Rayleigh Scattering .................................... 22
   3.3. Resonance Fluorescence .......................................... 24
   3.4. Pump Probe Measuring Methods .................................... 24
       3.4.1. Optical detection of Nuclear Spin Polarization .............. 25
       3.4.2. Pump Probe Method with Resonant Rayleigh Scattering ...... 26
       3.4.3. Pump probe Method with Resonance Fluorescence ............ 34
# List of symbols and abbreviations

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$g_{el,h,ex}$</td>
<td>$g$-factor (for electron, hole and exciton, respectively)</td>
</tr>
<tr>
<td>$j$</td>
<td>Total angular momentum (spin and orbital)</td>
</tr>
<tr>
<td>$l$</td>
<td>Orbital angular momentum</td>
</tr>
<tr>
<td>$A_i$</td>
<td>Hyperfine coupling constant of nucleus $i$</td>
</tr>
<tr>
<td>$B_{ed}$</td>
<td>Knight field</td>
</tr>
<tr>
<td>$B_{ext}$</td>
<td>External magnetic field</td>
</tr>
<tr>
<td>$B_{loc}$</td>
<td>Local dipolar nuclear field</td>
</tr>
<tr>
<td>$B_{nuc}$</td>
<td>Overhauser field</td>
</tr>
<tr>
<td>$E_F$</td>
<td>Fermi energy</td>
</tr>
<tr>
<td>$\hat{I}^i$, $I^i$</td>
<td>Nuclear spin operator and quantum number</td>
</tr>
<tr>
<td>$N$</td>
<td>Number of quantum dot nuclei</td>
</tr>
<tr>
<td>$Q$</td>
<td>Nuclear quadrupolar moment</td>
</tr>
<tr>
<td>$\hat{S}, S$</td>
<td>Electron spin operator and projection on $z-$axis</td>
</tr>
<tr>
<td>$T_1$</td>
<td>Spin relaxation time</td>
</tr>
<tr>
<td>$T_2, (T_2^*)$</td>
<td>Spin dephasing time (ensemble averaged)</td>
</tr>
<tr>
<td>$V_g$</td>
<td>Applied gate voltage</td>
</tr>
<tr>
<td>$\gamma_i$</td>
<td>Gyromagnetic ratio of nucleus $i$</td>
</tr>
<tr>
<td>$\gamma_0$</td>
<td>Unit cell volume</td>
</tr>
<tr>
<td>$\rho_c^+ (\rho_c^-)$</td>
<td>Circular polarization of PL light (under $\sigma^+$- ($\sigma^-$-) polarized excitation</td>
</tr>
<tr>
<td>$\sigma^+ (\sigma^-)$</td>
<td>Circular light polarization of positive (negative) helicity</td>
</tr>
<tr>
<td>$\sigma_x$</td>
<td>Linear light polarization along $x$</td>
</tr>
<tr>
<td>$\omega_Q$</td>
<td>Quadrupolar coupling strength</td>
</tr>
<tr>
<td>$\Psi$</td>
<td>Electron wave function</td>
</tr>
<tr>
<td>$\Omega_{el}$</td>
<td>Electron Larmor frequency</td>
</tr>
<tr>
<td>$\Delta E_{OS}$</td>
<td>Overhauser shift</td>
</tr>
<tr>
<td>$\Delta E_{el,e,ex}^Z$</td>
<td>Zeeman splitting (for electron, hole and exciton, respectively)</td>
</tr>
<tr>
<td>$\uparrow, \downarrow (\uparrow\downarrow, \downarrow\uparrow)$</td>
<td>Electron (hole) spin up, electron (hole) spin down</td>
</tr>
<tr>
<td>Symbol</td>
<td>Abbreviation</td>
</tr>
<tr>
<td>--------</td>
<td>-------------------</td>
</tr>
<tr>
<td>$1 \text{eV} = 1.6 \cdot 10^{-19} \text{J}$</td>
<td>1 electronvolt</td>
</tr>
<tr>
<td>$e = 1.6 \cdot 10^{-19} \text{A} \cdot \text{s}$</td>
<td>Electron charge</td>
</tr>
<tr>
<td>$g_0 = 2$</td>
<td>Free electron $g$-factor</td>
</tr>
<tr>
<td>$\hbar = 6.582 \cdot 10^{-10} \mu \text{eV} \cdot \text{s}$</td>
<td>Reduced PLANCK’s constant</td>
</tr>
<tr>
<td>$\mu_B = 58 \mu \text{eV} / \text{T}$</td>
<td>BOHR magneton</td>
</tr>
<tr>
<td>$k = 86 \mu \text{eV} / \text{K}$</td>
<td>BOLTZMANN constant</td>
</tr>
<tr>
<td>$\mu_0 = 4\pi \cdot 10^{-7} \text{N/A}^2$</td>
<td>Permeability of free space</td>
</tr>
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</table>

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
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</thead>
<tbody>
<tr>
<td>As</td>
<td>Arsenic</td>
</tr>
<tr>
<td>CB</td>
<td>Conduction band</td>
</tr>
<tr>
<td>CCD</td>
<td>Charge coupled device (camera)</td>
</tr>
<tr>
<td>DNSP</td>
<td>Dynamical nuclear spin polarization</td>
</tr>
<tr>
<td>CPT</td>
<td>Coherent population trapping</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full width at half maximum</td>
</tr>
<tr>
<td>Ga</td>
<td>Gallium</td>
</tr>
<tr>
<td>HH</td>
<td>Heavy hole</td>
</tr>
<tr>
<td>In</td>
<td>Indium</td>
</tr>
<tr>
<td>LH</td>
<td>Light hole</td>
</tr>
<tr>
<td>MBE</td>
<td>Molecular beam expitaxy</td>
</tr>
<tr>
<td>PL</td>
<td>Photoluminescence</td>
</tr>
<tr>
<td>PLE</td>
<td>Photoluminescence excitation</td>
</tr>
<tr>
<td>QD</td>
<td>Quantum dot</td>
</tr>
<tr>
<td>QI</td>
<td>Quadrupolar interaction</td>
</tr>
<tr>
<td>RF</td>
<td>Resonance fluorescence</td>
</tr>
<tr>
<td>SIL</td>
<td>Solid immersion lens</td>
</tr>
<tr>
<td>SNR</td>
<td>Signal to noise ratio</td>
</tr>
<tr>
<td>Ti</td>
<td>Titanium</td>
</tr>
<tr>
<td>VB</td>
<td>Valence band</td>
</tr>
<tr>
<td>$X^n$</td>
<td>Exciton of charge n</td>
</tr>
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</table>
1. Introduction

We all use modern electronic devices in daily life for work, communication and entertainment. Year by year, the computing power of such devices grows. One of the main reasons for this development is the miniaturization of the electronic components. Hence an increasing amount of functionality can be realized on the same area of the computer chip. As a consequence a computer processor consists of millions of transistors, where the smallest have a gate length of 10nm. The ability to fabricate devices at such small length scales has also inspired many research areas. The growth and study of objects where at least one of the spatial dimensions extends to only a few nanometers is called Nanotechnology. If objects are made smaller and smaller, the classical description fails at low temperature and quantum mechanical effects play an important role.

The study of charge carriers that are confined in low dimensions led to the discovery of interesting electrical and optical properties such as the Aharanov-Bohm effect [1], Coulomb blockade [2] and the quantum Hall effect [3]. In this thesis we focus on semiconductor structures with a confinement in all spatial directions, quantum dots (QD). On one hand, a QD shows a discrete energy spectrum and optical properties like an atom, on the other hand it is an object consisting of $10^4 - 10^5$ atoms. The confinement allows for a study of the interaction of a single electron or hole with its environment. One example is the interaction with a nearby electron or hole reservoir that results in interesting many-body physics [4–6]. Furthermore, the electron spin in a QD couples to the nuclear spins of the atoms the QD consists of via hyperfine interactions [5, 7, 8].

Single electrons confined in a QD are considered as promising candidates for the use as spin qubits in quantum information processing [9, 10]. Compared to atoms, the on-chip realization is promising for the scalability of the system. Additionally, QDs exhibit long spin lifetimes and coherence times due to the weak interaction with the environment [11]. This is absolutely crucial for quantum information storage. Last but not least, the coherent optical control of electron spins in self-assembled QDs allows for a fast manipulation of the quantum state on picosecond timescales [12, 13]. Therefore a prospering research field was established which addresses the problem of initialization, manipulation and readout of the
However, a single electron spin in a QD is not completely isolated from its environment. On the one hand, coupling to the environment is needed to manipulate and read out its state and on the other hand this coupling constitutes a source of decoherence. A particular interaction is the strong coupling of the electron spin to nuclear spins. Since the nuclear spins are randomly oriented in a QD the electron spin experiences a fluctuating effective magnetic field $B_{\text{eff}}$. As a consequence, nuclear spins are the dominant source of decoherence for electron spins [17]. It is well known that a long coherence time is important for quantum information processing, therefore the strong coupling to nuclear spins might be a drawback. One possible way to prolong the coherence time is preparing nuclear spins in a state which exhibits less fluctuations [18]. Additionally, by controlling nuclear spins it should be possible to influence the electron spin decoherence. For the realization of such a scenario, the interaction between electron and nuclear spins has to be studied in detail.

The hole on the other hand, which is a missing electron form the valence band, exhibits very little interaction with the nuclear spins. Because of the $p$-symmetry of the hole wave function the effective overlap of holes and the nuclei is strongly reduced compared to electrons. Hence, the hyperfine interaction to nuclear spins is weak, leading to longer coherence times [19].

1.1. Motivation and scope of this thesis

This thesis focuses on the optical study of nuclear spin interactions either with a single electron or a hole that is confined in a self-assembled InGaAs QD and is structured as follows.

Chapter 2 is an introduction to self-assembled InGaAs QDs and nuclear spins in QDs. In a first part we describe the growth of self-assembled QDs and the resulting discrete energy levels with their optical selection rules. Furthermore the charge control is discussed, which is realized by embedding the QDs in a diode structure. For the study of nuclear spins it is important to know the interactions in the nuclear spin system as well as the coupling to a single electron or hole spin. We discuss the corresponding interactions in the second part of chapter 2. Finally, the influence of the electron - nuclear spin coupling on coherence properties of the electron spin are lined out.

To gain insights into the interaction between nuclear spins and the spin of a single charge carrier confined in a QD, the spin properties of the single charge carrier are studied optically. A variety of different spectroscopy methods are introduced in chapter 3. While non-resonant excitation methods are important for a first charac-
terization of the QD, resonant spectroscopy techniques, namely resonant Rayleigh scattering and resonant fluorescence, allow for a direct access to optical transitions with high spectral resolution. Polarization of nuclear spins leads via the hyperfine interaction to a shift of the electron energy levels, the so-called Overhauser shift. The second part of chapter 3 covers the development of a pump probe method that allows for a resonant measurement of the Overhauser shift. Especially the increased spectral resolution compared to non-resonant measurement methods is promising to get new insights in the interactions between the electron spin and the nuclear spin reservoir.

Chapter 4 focuses on the evolution of nuclear spin polarization in a transverse magnetic field $B_x$. We will show that the nuclear spin polarization is very likely to cause a cancellation of $B_x$ for the electron spin up to $B_{\text{crit}} = 1.7\text{T}$. This is rather unexpected, since nuclear spins are optically polarized along the z-axis resulting in an effective magnetic field $B_{z,\text{eff}} \sim 500\text{mT}$ that is much smaller than the measured $B_{\text{crit}} = 1.7\text{T}$. There are several indications that the generated nuclear spin polarization is rotated to the x-axis [20]. To further investigate the nuclear spin evolution during this process, we measured the Overhauser shift resonantly for two different sample structures.

In chapter 5, time resolved measurements on the coherence properties of a single hole spin confined in a QD are presented. In contrast to electrons, holes interact weakly with nuclear spins, which leads to longer coherence times. To additionally prolong the coherence time, we created the hole optically in an n-doped sample, which results in less charge noise compared to p-doped samples. We find a coherence time of $T_2^* = 240\text{ns}$. Spin echo measurements reveal a $T_2$ of $1\mu\text{s}$. 
2. Self-assembled Quantum Dots and Nuclear Spins

In the first part of this chapter the basic principles of self-assembled InGaAs QDs are explained. We start with a section about the growth of self-assembled QDs. Subsequently, the consequences of such a quantum confinement potential in three spatial directions and the optical selection rules are discussed. Furthermore the diode-structure used for controlling the number of excess charges in the QD is presented. In the second part of this chapter, the theoretical background of the nuclear spin system and a single electron or hole spin confined in a QD are introduced. While the coupling of the electron-nuclear spin system is dominated by the contact hyperfine interaction, the hole couples through the anisotropic dipolar hyperfine interaction to the nuclear spin ensemble. Finally, we will discuss the influence of nuclear spins on the electron spin decoherence.

2.1. Charge tunable self-assembled Quantum Dots

A quantum dot (QD) is an artificial structure that exhibits a strong confinement in all three spatial dimensions. In other words, it is a zero dimensional object confining single charge carriers. There are different ways to realize such a confinement. One possibility is to deplete a two-dimensional electron gas with electric gates or by oxidation, which results in a spatial confinement for electrons. Such a QD is usually coupled via tunnel barriers to electronic reservoirs which allow for a deterministic charging with single electrons. To investigate the physical properties, current and voltage probes are attached to these reservoirs. Therefore electronic properties can be measured [21].

A second possibility is to generate confinement by a semiconductor surface which is realized in semiconductor nanocrystals [22]. These QDs are grown by crystallization in a colloidal solution, and are therefore called colloidal QDs [23]. Nanocrystals are optically active, the transition energy depends on the size of the crystal, which is a growth parameter. On the one hand, the performance of colloidal quantum dots
for quantum optics experiments is strongly affected by spectral diffusion leading to a broadening of the optical linewidth of several 100µeV [24]. Additionally, colloidal QDs are suffering from blinking and bleaching. On the other hand, the chemical stability allows to use them as fluorescence markers in biology [23].

In self assembled semiconductor QDs [25] the charge confinement is given by a combination of semiconductor materials with different band gaps and a type I interface (see section 2.1.1). Compared to colloidal QDs, the optical line widths are much narrower and near life-time limited (∼1.6µeV) [26]. Additionally, a single self-assembled QD behaves like a quantum emitter which manifests in perfect antibunching of the emitted photons [27]. Hence a denotation as artificial atom is very common. However, a self-assembled QD is still a mesoscopic object which consists of 10^4 – 10^5 atoms. Many interesting physical effects arise due to the interaction of the confined electron with the host material of the QD and the resulting strain from the growth process.

### 2.1.1. Growth of self-assembled quantum dots

The InGaAs QDs studied in this work were grown by molecular beam epitaxy (MBE). This method allows depositing single crystals with a rate of less than 3000nm per hour and therefore a very precise control over the material composition is achieved. MBE takes place in high vacuum and the deposited particles are heated in separate effusion cells. The gaseous particle beams condense subsequently on the wafer. For growth of InGaAs QDs, a GaAs wafer is heated to 600°C under ultra-high vacuum. First, a buffer layer of several 100nm of GaAs is grown to clean the surface of the wafer. In a second step, the evaporated gallium (Ga) particle beam is replaced with indium (In). There is a large lattice mismatch between GaAs and InAs of 7%, which leads to an abrupt change of the growth characteristics at a critical layer thickness of InAs. After an initially two dimensional growth of InAs (Fig. 2.1 (a)) the mechanical strain is released at 1.7 monolayers by the formation of three dimensional InAs islands (Fig. 2.1 (b)). At this critical point it is energetically favorable to minimize strain energy by increasing surface energy. The growth process is stopped after 2.7 monolayers of InAs. A continuation would again result in a two dimensional layer of InAs with crystal defects which relax the strain. The formed InAs QDs are randomly distributed and have a diameter of 20-30nm and a height of 10nm. This growth process, which results in the spontaneous formation of InAs QDs, is called Stranski-Krastanow method [28].

Typical transition wavelengths for these QDs are at 1100nm, which is a rather inconvenient spectral range for efficient silicon based photodetectors. Therefore the QDs are partially covered with GaAs (Fig. 2.1 (c)) and the temperature is increased. In this annealing process the intermixing between Ga and InAs is increased, result-
2.1. Charge tunable self-assembled Quantum Dots

Figure 2.1.: Growth of self-assembled QDs. (a) Two dimensional growth of InAs. (b) Strain driven formation of InAs islands after 1.7 monolayers. (c) Partial capping layer of GaAs is grown. (d) Annealing leads to a reduced height of the QDs. (e) Protective overgrowth with GaAs.

ing in a smaller QD height (Fig. 2.1 (d)). The stronger confinement in the growth direction leads to a shift of the transition wavelength to the blue i.e. 950nm, which is a much better spectral range for silicon based photodetectors. This method to shift the wavelength is known as the partially covered island (PCI) technique [29]. As a last step the InAs QD are covered with a GaAs layer to avoid surface effects (Fig. 2.1 (e)).

Even after the formation of InAs droplets, there is a thin two dimensional layer of the same material remaining, which is called the wetting layer. This quantum well structure can be optically accessed and is energetically shifted compared to the QD spectra. The emission of the wetting layer is expected at 840nm-860nm.

Self-assembled QDs exhibit a distribution in size, shape, In and Ga content. This leads to a distribution of their transition energies. Usually, an ensemble of QD shows an inhomogeneous broadening in the order of 10meV [30], compared to the natural line width of $\sim 1\mu\text{eV}$ of a single QD [31]. To resolve the line width isolation of a single QD is desirable. Additionally, the growth process allows to control the QD density as follows. To assure a homogeneous result of the growth process, the wafer is rotated. During the formation of the QDs the rotation of the wafer is stopped. Therefore a gradient of the QD density develops according to the distance from the indium source. By choosing the corresponding piece of the wafer, a low density region can be selected to perform studies on single QDs.

2.1.2. Quantum dot level scheme and optical selection rules

The surrounding of the InAs QD with GaAs that exhibits a larger band gap leads to a quantum confinement potential in all three dimensions. In Fig. 2.2 the potential landscape perpendicular to the growth direction of the QD is shown, which can be approximated by a 2D harmonic potential. Several discrete energy states that can
be occupied by electrons in the conduction band (CB) and holes in the valence band (VB) are formed. In analogy to atomic states, the discrete energy states of the confining potential are labeled s, p, d,... Furthermore, the quantum confinement leads to a separation of the hole band into a heavy hole (HH) band and a light hole (LH) band [32] that is additionally enhanced by the strain in self-assembled QDs. The LH band is shifted by more than 10 meV to lower energies such that only the HH band is considered for further studies on QDs.

The Bloch wave function of the lowest energy states in the conduction band exhibit an s-type character, which means that the total angular momentum of an electron occupying the CB is provided by its spin $S_z = \pm \frac{1}{2}$. On the other hand the highest energy states of the valence band have a p-type wave function, leading to an angular momentum of 1. Therefore the total angular momentum projection of the HH spin is $J_z = \pm \frac{3}{2}$. Further consequences of the different wave function symmetries of electrons and holes will be discussed in detail in section 2.2.

An electron can be lifted from the valence band to the conduction band in the QD i.e. by absorbing a photon. The electron in the conduction band is bound to the hole in the valence band by the Coulomb attraction, resulting in a quasi-particle called exciton. Subsequently a recombination of this electron hole pair can take place, by emission of a photon. Until now we only considered neutral excitons ($X^0$). By confining extra charge carriers into the QD other exciton types can be formed. A negatively charged exciton ($X^-$) is generated if an excess electron is inside the QD. Typically the emission wavelength for $X^-$ is red shifted by $\sim 6$ meV as compared to $X^0$. Similarly a positively charged exciton ($X^+$) can be created with an additional

\begin{figure}[h]
\centering
\begin{tabular}{cc}
(a) Lateral confinement: & (b) Harmonic approximation: \\
\includegraphics[width=0.4\textwidth]{fig2a.png} & \includegraphics[width=0.4\textwidth]{fig2b.png}
\end{tabular}
\caption{Confinement in an InGaAs QD. (a) Confinement potential of an InGaAs QD perpendicular to the growth plane. Discrete energy states are labeled with s,p,d,... similar to atomic states. (b) An approximation of the confinement potential is realized with a 2D harmonic oscillator where the energy states are separated by $\hbar\omega_e$ for electrons in the conduction band (CB) and by $\hbar\omega_h$ for holes in the valence band (VB). Figure taken from [33].}
\end{figure}
hole inside the QD. In this case the electron hole recombination energy depends strongly on the QD and can be a few meV larger or smaller than that of the neutral exciton. For all following experiments we focused on a positively charged QD. Based on the twofold spin degeneracy of the electron and the hole inside the QD four different excitons can be generated for $X^+$. Their total angular momentum projection is given by $M_z = S_z + J_z$ leading to $M_z = \pm 1$ or $M_z = \pm 2$. Because of the conservation of angular momentum, the emitted photon upon exciton recombination carries the spin of the electron hole pair. Therefore only the exciton with $M_z = \pm 1$ can decay radiatively, resulting in a $\sigma^+$, respectively a $\sigma^-$ polarized photon. These excitons are called bright. The so-called dark excitons with $M_z = \pm 2$ cannot recombine optically. For a deterministic charging with a single electron or hole, the QD is exposed to an electric field and coupled to a nearby electron reservoir. This can be realized by embedding the QD layer into a field effect structure [34, 35], which is explained in detail in the following section.

### 2.1.3. Charge control

The samples studied in this thesis consist of a quantum dot layer which is embedded in a field effect structure as shown in Fig. 2.3. Similar to QD fabrication, this electronic structure is grown by MBE. In order to apply an electric field to the QD, the QD layer is sandwiched between a negatively doped GaAs back contact and a metallic top gate. A layer of intrinsic GaAs acts as a tunnel barrier between the back contact and the QD layer. Additionally a blocking barrier, consisting of AlGaAs on top of the capping layer, is needed to prevent current flow in the sample. Finally the electronic structure ends with a layer of intrinsic GaAs that prevents the aluminum in the blocking barrier from oxidation. After the growth process a semitransparent metallic top gate, consisting of 2nm titanium and 8nm gold, is deposited on top of the sample.

The electrostatic energy of an electron confined in a QD is determined not only by the applied voltage $V_g$ between the back contact and the top gate but also by an offset voltage $V_g^0$, arising from the Schottky interface at the top gate. Since the Fermi energy $E_F$ is pinned to the conduction band (CB) at the back contact, the applied gate voltage changes the energy of the QD states depending on the lever arm $\eta = d/b$, where $d$ is the distance of the top gate from the back contact and $b$ is the thickness of the tunneling barrier.

By choosing the gate voltage such that the electronic state of the QD is shifted below $E_F$ an electron tunnels into the QD from the back contact, which acts as an electron reservoir [35]. Such a deterministic charging of the QD with single electrons is only possible due to the Coulomb repulsion which prevents a second
Figure 2.3.: Field effect structure. Conduction band (CB) and valence band (VB) edge of the field effect structure. The n-doped back gate fixes the Fermi energy $E_F$ to the CB. With an externally applied gate voltage ($V_g$) between the top gate and the back contact, the slope of the band edges can be controlled. Figure taken from [33].

electron from tunneling into the QD. By applying a more positive $V_g$ the Coulomb repulsion can be overcome, resulting in a QD charged with two electrons in a singlet state. Similarly, the p-shell states of the QD can be successively filled.

Until now, we have only considered charging of the QD with electrons. There are two possibilities to achieve this for holes. Either the back contact is p-doped and acts as a hole reservoir or the holes are optically generated in an n-doped sample. For the experiments in this thesis we used the latter. A detailed comparison of the two methods is given in chapter 5.

In order to charge the QD optically with a single hole, an electron is excited to the conduction band leaving a single hole in the valence band. Subsequently, both charge carriers relax into the QD. If the applied gate voltage is chosen such that the electronic state lies above the Fermi energy, the electron tunnels out of the QD (Fig. 2.4(b)). The criteria for the hole to stay in the QD not given by $E_F$ but rather by the position of the two dimensional continuum of hole states at the interface to the superlattice [4, 36]. If the lowest subband energy of the hole continuum is shifted below the QD hole state, the hole cannot tunnel out of the QD. Consequently the hole is trapped inside the QD and a positively charged exciton $X^+$ can be created by adding an optically generated electron hole pair (Fig. 2.4(c)) [37]. This state is only stable due to the additional binding energy of the second hole that results in an electronic state below the Fermi energy (Fig. 2.4(d)).

In order to charge the QD with two excess holes, a more negative gate voltage $V_g$ has to be applied. In this case, also the second electron tunnels out of the QD because it overcomes the attractive Coulomb energy. Therefore two holes are trapped inside the QD, which leads to a $X^{2+}$ state upon optical excitation. The
Figure 2.4: Charging n-doped samples with a single hole. (a) After the optical excitation of the electron to the CB, the electron hole pair relaxes into the QD. (b) Since the electronic state of the QD lies above the Fermi energy, the electron tunnels out of the QD. On the other hand the hole remains trapped because of the QD hole state laying below the two-dimensional continuum of states. (c) A second optical excitation leads to the formation of a positively charged exciton. (d) This $X^+$ state is stable due to the Coulomb energy of the second hole which results in an electronic state laying below the Fermi energy. Figure taken from [33].

number of observed charge states depends strongly on the confinement potential. While for InAs QD studied by Ediger et al. [38] states from $X^{6+}$ to $X^{7−}$ were observed, in Fig. 2.5 charged excitons from $X^+$ to $X^{−3}$ are shown.

2.1.4. Quantum confined Stark effect

Besides the discrete jumps in the photoluminescence (PL) measurements as a function of the gate voltage due to the different charging states, a slight shift of the energy plateaus with the gate voltage is observable in Fig. 2.5. This is due to the quantum confined Stark effect which arises on the one hand from the permanent dipole moment $d_{\text{exc}}$ of the exciton and on the other hand from the induced dipole moment. To quantify the latter we introduce the polarizability $\alpha$. As a consequence
Figure 2.5.: Charging diagram measured in PL. Colour plot of the PL intensity as a function of gate voltage and emitted wavelength, red means high and blue low intensity. The measurement was taken on a 25nm tunneling barrier sample. The jumps in energy between the different charging states are observable. The differently charged excitons are labeled with $X^+$ for a positively charged exciton, $X^0$ for the neutral exciton, $X^-$ for the negatively charged exciton and so on.

the exciton energy changes as a function of the effective electric field. This can be described with the following formula [39]

$$E = E_0 - d_{\text{exc}} F - \alpha F;$$  \hspace{1cm} (2.1)

where $E_0$ is the energy of the unperturbed exciton and $F$ is an external electric field. Especially from the experimental point of view the quantum confined Stark shift is very beneficial. It allows to scan across the exciton resonance by varying $V_g$ without changing the laser wavelength.
2.2. Nuclear Spins in Quantum Dots

As previously mentioned, a self-assembled QD consists of $10^4 - 10^5$ atoms and a single confined charge carrier is not completely decoupled from its environment. In particular, the confined electron spin in a QD couples to the nuclei of the atoms that the dot consists of. This leads to several interesting physical phenomena, for example locking of a QD transition to a resonant pump laser [40, 41] or bistabilities of the nuclear spin system that exhibit memory effects (non-Markovian behavior) [42, 43]. Furthermore, the random fluctuations of the nuclear spin system are the main source of decoherence for a single electron spin confined in a QD [17]. The prospect of prolonging the coherence time to the extent that a single spin manipulation is possible, was one of the main motivations that led to increased interest in nuclear spin physics in QDs [18]. Here, we will give an overview of the electron/hole spin system, the nuclear spin system and the coupling between the two. Last but not least we consider the influence of these couplings on the coherence properties of a single electron spin in a QD.

In an externally applied magnetic field, the total Hamiltonian for a single electron/hole interacting with an ensemble of nuclear spins is given by

$$H_{\text{tot}} = H_{\text{el}z} + H_{\text{nuc}z} + H_{\text{ihf}} + H_{\text{ahf}} + H_{\text{dip}} + H_Q,$$

(2.2)

where $H_{\text{el}z}$ and $H_{\text{nuc}z}$ are the electron and nuclear Zeeman Hamiltonians, respectively. While for the electron the isotropic hyperfine interaction $H_{\text{ihf}}$ is the dominant coupling to the nuclear spin system, the anisotropic hyperfine interaction $H_{\text{ahf}}$ describes the most important interaction between the hole spin and the nuclear spins. $H_{\text{dip}}$ denotes the nuclear dipole-dipole interaction while $H_Q$ are the nuclear quadrupolar interactions. In the following section we will discuss the different terms in detail.

2.2.1. Electron and Hole Spin System

In the framework of this thesis we studied mainly positively charged QDs. While in chapter 4 the electron spin in the positively charged exciton is investigated, the experiments in chapter 5 focus on the hole spin in the ground state. Therefore we will consider the interaction of an electron spin and a hole spin to an external magnetic field in this section.

For a QD electron spin, the application of a magnetic field $\vec{B}$ leads to a Zeeman splitting:

$$H_{\text{el}}^z = g_\text{el} \mu_B \vec{S}_\text{el} \cdot \vec{B},$$

(2.3)
where \( g_{el} \) is the effective electron g-factor varying from dot to dot between 0.4 to 0.6, \( \mu_B \) denotes the Bohr magneton and \( \vec{S}_{el} \) the electron spin operator. In analogy the Zeeman interaction for the hole is given by

\[
H^h_z = -g_h \mu_B \vec{S}_h \cdot \vec{B},
\]

(2.4)

with the effective hole g-factor \( g_h \) and the hole spin operator \( \vec{S}_h \). In contrast to \( g_{el} \), the effective hole g-factor depends strongly on the magnetic field direction. While for magnetic fields along the growth direction \( g_h \) is around 1-1.5, for transverse magnetic fields \( g_h \) is between 0 and 0.4 [44, 45].

### 2.2.2. Nuclear Spin System

The material composition of the considered QDs is mainly given by InAs and has some additional Ga contributions due to diffusion processes [46]. The nuclear spin system is therefore given by the naturally occurring isotopes of the three atomic species: \(^{115}\text{In} \ (95.3\%), \ ^{113}\text{In} \ (4.7\%), \ ^{75}\text{As}, \ ^{69}\text{Ga} \ (60.1\%), \ ^{71}\text{Ga} \ (39.9\%)\). The natural abundance is denoted in brackets. While Ga and As have a total nuclear spin of \(3/2\), In has a spin of \(9/2\). In a static magnetic field, each nucleus \( i \) experiences the following Zeeman interaction:

\[
H^\text{nuc}_z = -\sum_i \gamma_i \vec{I}_i \cdot \vec{B},
\]

(2.5)

with \( \gamma_i \) the gyromagnetic ratio and \( \vec{I}_i \) the spin operator of the \( i \)th nucleus.

#### Nuclear Dipolar Coupling

The nuclear dipole-dipole interaction is an important coupling within the nuclear spin system. A nuclear magnetic moment \( \gamma_i I^i \) will produce a dipolar magnetic field which acts on a nuclear spin \( j \). This interaction is strongly dependent on the relative position \( r_{ij} \) between two nuclear spins \( i \) and \( j \) [47].

\[
H^\text{dip}_{ij} = \frac{\mu_0 \gamma_i \gamma_j}{4\pi r_{ij}^3} \left( I^i \cdot I^j - 3 \frac{(I^i r_{ij})(I^j r_{ij})}{r_{ij}^2} \right)
\]

(2.6)

The fluctuating local magnetic field created at the nucleus \( j \) by the other nuclei can be estimated from the bulk value in GaAs, \( B_{\text{loc}} \approx 1.5 \cdot 10^{-4} \text{T} \) [48].

The nuclear dipolar coupling (2.6) can be decomposed into "secular" parts which commute with \( H^\text{nuc}_z \) (energy conserving) and into "non-secular" parts.
The "secular" parts have the following form

\[ H_{\text{sec}}^{\text{dip}} \sim I_z^2 I_z^2 - \frac{1}{4} \left( I_z^2 I_z^2 + I_z^2 I_z^2 \right). \]  

(2.7)

Clearly, \( H_{\text{sec}}^{\text{dip}} \) conserves the nuclear spin. The second term in (2.7) describes flip-flops between nuclear spins of the same species at different sites, which leads to nuclear spin diffusion.

"Non-secular" terms are only important for \( B_{\text{ext}} \leq B_{\text{loc}} \). They do not conserve the total spin of the nuclei, it is transferred to the crystal lattice. Therefore a depolarization within \( t_{\text{nuc}} \approx 10^{-100} \mu s \) takes place [21], if quadrupolar interactions are absent (see following section).

### Nuclear Quadrupolar Interactions

After looking into magnetic interactions between nuclear spins we will now focus on the influence of an electric field on nuclei. The electrostatic energy of a localized charge distribution \( \rho(\vec{x}) \) placed in an external potential \( \phi(\vec{x}) \) can be written with a multipole expansion [49]:

\[
W = q\phi(0) - pE(0) - \frac{1}{6} \sum_i \sum_j Q_{ij} \frac{\delta E_j}{\delta x_i}(0) + \ldots
\]  

(2.8)

\[
p = \int x' \rho(x') d^3 x' \quad \text{dipole moment}
\]

\[
Q_{ij} = \int (3x_i'x_j' - r^2 \delta_{ij}) \rho(x') d^3 x' \quad \text{quadrupole moment}
\]

Due to symmetry reasons, the nuclei do not have an electric dipole moment and are therefore insensitive to homogeneous fields [47]. Nuclear spins with \( I > 1/2 \) possess a finite electric quadrupole moment which couples to an electric field gradient. Generally the quadrupole moment describes the ellipticity of a charge distribution in a nuclei and is strongly dependent on the material.

If the nuclear environment has cubic symmetry (as in bulk GaAs), there is no electric field gradient and therefore no quadrupolar coupling [47, 50]. Crystal strain in self-assembled QDs gives rise to nonzero electric field gradients at the position of the nuclei. Assuming the electric field gradients have cylindrical symmetry and are oriented along \( z' \), the quadrupolar interactions can be described by

\[
H_Q = \frac{\hbar \omega_Q}{2} \left( I_{z'}^2 - \frac{I(I+1)}{3} \right)
\]  

(2.9)

where \( \omega_Q \) is the quadrupolar coupling strength and \( I_{z'} \) the angular momentum projection on the \( z' \)-axis [47]. Without any external field, the \( (2I+1) \) spin lev-
els are paired into \( m_{z'} = \pm 1/2, \pm 3/2, ... \pm I \) and separated from each other by \( 1, 2, ...,(I - 1/2) \hbar \omega_Q \).

By setting \( g_N \mu_B B_Q = \hbar \omega_Q \), where \( g_N \mu_B = \sum \gamma_i \), an effective quadrupolar magnetic field can be calculated. In self-assembled InAs QDs \( B_Q \) is estimated and measured to be several 100mT [51–53]. This nonlinear energy splitting between different spin levels leads to a strong suppression of nuclear spin diffusion and therefore to long nuclear polarization storage times [52].

### 2.2.3. Isotropic and Anisotropic Hyperfine Interaction

#### Isotropic Hyperfine Interaction

The dominant interaction between a single QD electron spin and an ensemble of nuclear spins is the isotropic hyperfine interaction [17, 54]. The Hamiltonian is given by

\[
H_{\text{hf}} = \nu_0 \sum_k A_k |\psi(\vec{R}_k)|^2 \vec{S} \cdot \vec{I}_k, \tag{2.10}
\]

where \( \nu_0 \) denominates the atomic volume of InAs, \( \vec{R}_k \) denotes the position of the \( k \)th nucleus and \( \psi(\vec{R}_k) \) is the electron envelope wave function. The hyperfine coupling constant is \( A_k = \frac{16\pi}{3} \mu_0 g_0 \mu_B \hbar \gamma_k |u(\vec{R}_k)|^2 \) with \( u(\vec{R}_k) \) the Bloch amplitude at the position of the nuclei and \( g_0 \) the free electron g-factor. There are only non-vanishing terms of \( A_k \) if the Bloch function is non zero at the position \( \vec{R}_k \) of the nuclei. This focuses the attention on the wave function in a QD. In GaAs the top of the valence band has a p-like symmetry and the bottom of the conduction band has an s-like symmetry. Contrary to p-like symmetry, the probability of an s-like wave function symmetry does not disappear at the site of the nuclei. Therefore the isotropic hyperfine interaction has only non-vanishing contributions for electrons in the conduction band. The strength of the total hyperfine constant of the main components indium, gallium and arsenide are \( A_{\text{In}} \approx 56 \mu eV, A_{\text{Ga}} \approx 42 \mu eV \) and \( A_{\text{As}} \approx 46 \mu eV \) [55].

The Hamiltonian of the isotropic hyperfine interaction (2.10) can be divided into a static and a dynamical part [56]:

\[
\vec{S} \cdot \vec{I} = \frac{I_+^k S_+}{\text{static part}} + \frac{1}{2} \left( I_+^k S_- + I_-^k S_+ \right) \text{ dynamical part}. \tag{2.11}
\]

\[
\begin{align*}
S_+ &= S_x + iS_y \quad \text{raising operator} \\
S_- &= S_x - iS_y \quad \text{lowering operator} \tag{2.12}
\end{align*}
\]

The static part of the effective Hamiltonian affects energies of the electron and nuclear spins leading to an "effective magnetic field", which is only experienced by
2.2. Nuclear Spins in Quantum Dots

the electron due to spin polarized nuclei (Overhauser field) or by the nuclei due to a spin polarized electron (Knight field).

- **Overhauser Field**

\[
H_{\text{hf}}^{\text{static}} = \left( \sum_k A^k \right) S_z \quad \Rightarrow \quad B_{\text{nuc}} = \frac{1}{g_e \mu_B} \sum_k A^k I^k \quad (2.13)
\]

A fully polarized nuclear spin system in GaAs can lead to an effective magnetic field of several Tesla [57], which is experienced by the electron.

- **Knight Field**

\[
H_{\text{hf}}^{\text{static}} = \sum_k \left( A^k S^k \right) I^k \quad \Rightarrow \quad B_{\text{el}} = -\frac{1}{g_N \mu_N} A^k S^k \quad (2.14)
\]

The maximal effective magnetic field due to a polarized electron spin is \( B_{\text{el,max}} = 13\text{mT} \) for Ga and \( B_{\text{el,max}} = 22\text{mT} \) for As [57].

The dynamical part of the effective Hamiltonian allows transferring angular momentum between the two spin systems. If the external applied magnetic field \( B_{\text{ext}} \) is larger than the local magnetic field \( B_{\text{loc}} \), direct electron nuclear flip-flop processes will vanish due to the large energy mismatch of the electronic and nuclear Zeeman splitting (electron: \( g_e \mu_B = 10^{-2}/\text{T meV} \), nuclei: \( g_N \mu_N = 10^{-5}/\text{T meV} \)). In this case higher order nuclear spin flips can still take place via an excited or virtual state [58].

**Anisotropic Hyperfine Interaction**

In addition to the contact hyperfine interaction discussed in the previous section, the electron or hole can couple to a dipole-like hyperfine interaction to the nuclear spins. The Hamiltonian of this anisotropic hyperfine interaction is given by

\[
H_{\text{ahf}} = \frac{\mu_0}{4\pi} g_0 \mu_B \sum_k \gamma_k \left( \frac{3(\vec{n}_k \cdot \vec{S})(\vec{n}_k \cdot \vec{I}_k) - \vec{S} \cdot \vec{I}_k}{R^2_k} \right), \quad (2.15)
\]

where \( \vec{n}_k = \vec{R}_k / R_k \) is the normalized position vector of the nuclear spin. In the case of an s-type wave function the anisotropic hyperfine interaction is much smaller than the isotropic hyperfine interaction. Conversely, for p-type wave functions the anisotropic dominates over the isotropic hyperfine interaction. Theoretical predictions, based on the anisotropic hyperfine interaction, estimated the interaction strength of a QD hole spin with nuclear spins to be one order of magnitude less than for a QD electron spin [19], which was experimentally confirmed [44].
2.2.4. Electron Spin Decoherence

In addition to an external applied magnetic field $\vec{B}_{\text{ext}}$, the QD electron spin experiences a nuclear magnetic field $\vec{B}_{\text{nuc}}$. Therefore spin precession about the vector of the total magnetic field is expected ($\vec{B}_{\text{tot}} = \vec{B}_{\text{ext}} + \vec{B}_{\text{nuc}}$). Especially the longitudinal component $B_{\text{nuc}}^z$, i.e., the component oriented parallel or opposed to $\vec{B}_{\text{ext}}$, changes the precession frequency independent of the strength of the external field. If $\vec{B}_{\text{nuc}}$ would be fixed and precisely known, the resulting phase shift could be exactly calculated. This is not the case in a QD; the nuclear field, generated by $N \sim 10^5$ nuclear spins, has a random and unknown value and orientation. Hence, the electron spin picks up a random phase and decoherence occurs. For an unprepared nuclear spin state, a Gaussian distribution of the nuclear field values $B_{\text{nuc}}^z$ with a standard deviation of $\Delta B_{\text{nuc}} \approx A \sqrt{\frac{N g_s \mu_B}{2 N_{\text{el}}}} \approx 20 - 40 \text{mT}$. [59] The dephasing of the electron spin is of the form $\exp\left[-\frac{t^2}{T_2^*}\right]$ with

$$T_2^* = \frac{\hbar \sqrt{2}}{g \mu_B \sqrt{\langle (B_{\text{nuc}}^z)^2 \rangle}}$$

(2.16)

This leads to a typical dephasing time of $T_2^* \sim 1\text{ns}$ [21]. Experimentally, the $T_2^*$ time can be determined with a Ramsey experiment, which works as follows. Assuming the external magnetic field is applied along the z-axis, the electron spin is first rotated into the x-y plane. After a spin precession for a certain time interval, the electron spin is rotated again by an angle of $\pi$ and measured along the z-axis. Because of the different phases picked up during the precession, the measured signal is oscillating as a function of the chosen time interval. The dephasing can be measured by the decay of the oscillation amplitude (see chapter 5).

For determining $T_2^*$ an average over several experiments with different $B_{\text{nuc}}$ is taken. Due to the nuclear field fluctuations a short coherence time can be observed. The situation completely changes, if only one experiment with a single spin is considered. Obviously the time scale $T_2$ on which the phase of the electron spin is lost would be longer than the previously considered value $T_2^*$. Experimentally, the random spin evolution during a certain time interval $\tau$ can be reversed by applying a 180° rotation pulse (spin-echo pulse) and letting the spin evolve for a second time interval of the same duration $\tau$ [60]. It is an important requirement that the time constant of the nuclear field fluctuations is long compared to the spin echo sequence. Therefore the effect of the random nuclear field can only be partially eliminated, resulting in a spin echo $T_2$ time. For simplification the spin echo $T_2$ time will be referred to as $T_2$ time in this thesis. Recent spin echo measurements on QDs determined a value of $T_2 \sim 3\mu\text{s}$ [61]. Since this decoherence is not irreversible, with more advanced pulse techniques even longer coherence times exceeding $200\mu\text{s}$
can be measured [10].
After looking into the dephasing mechanisms of the QD electron spin, we now focus on electron spin flips caused by the nuclear field. For $B_{ext} \ll B_{nuc}^{z,y}$ electron-nuclear spin flip-flops are allowed and lead to electron spin relaxation times of $T_1 \sim 10\text{ns}$. As $B_{ext}$ increases, direct electron nuclear flip-flops become suppressed and $T_1$ rapidly increases. At high magnetic fields $T_1$ decreases again due to interaction with acoustic phonons [9].
3. Experimental Methods

In this chapter an overview of the different spectroscopy techniques used to investigate InGaAs QDs is given. While Photoluminescence (PL) is mainly applied for a first characterization of the QDs, resonant spectroscopy techniques namely resonant Rayleigh scattering and resonance fluorescence allow for a direct access to the optical transitions in the QD. Furthermore, the development of a pump probe method that allows for a resonant measurement of the Overhauser shift is described.

3.1. Photoluminescence

Photoluminescence (PL) can be applied to optically characterize charge states of single QDs [30, 35, 62]. After exciting the QD sample with a non-resonant laser above the band gap of GaAs, electron hole pairs are formed which diffuse near the vicinity of the QD trap potential. Subsequently the exciton is trapped by the QD potential and a relaxation to the QD ground state takes place by phonon scattering on ps timescale [63]. Finally, the exciton recombines spontaneously after its lifetime $\tau \sim 1\text{ns}$ [64] and a photon is emitted.

Non-resonant excitation allows to excite different charge states of the QD which can be spectrally resolved as a function of the gate voltage, leading to a charging diagram as shown in Fig. 2.5. While PL is advantageous to achieve an overview of the exciton energies of a particular QD, a drawback of non-resonant excitation is the charging of defects in the QD environment. This can lead to shifted or broadened transition energies compared to resonant measurements [65]. Additionally, the spectral resolution is constricted and a lifetime limited line width of $h/\tau \sim 1\mu\text{eV}$ cannot be resolved with a standard spectrometer. This problem can be overcome with an additional Fabry-Pérot cavity filter which suffers on the other hand from a low photon throughput [66]. The polarization conservation of a PL experiment is strongly dependent on the laser excitation energy. A non-resonant excitation to the p-shell states of the QD (referred to as photoluminescence excitation (PLE)) can lead to a maximal polarization preservation of $\sim 80\%$. This plays an important role in polarization of nuclear spins which will be explained in detail in section 3.4.1.
3.2. Resonant Rayleigh Scattering

Besides the limited spectral resolution in non-resonant experiments an additional reason to perform resonant spectroscopy is the conservation of the coherence during the optical excitation process. To perform resonant spectroscopy a narrow band laser is scanned across the excitonic resonance. Subsequently, the intensity of the transmitted or reflected light is detected with a photodiode. The measured intensity is given by

\[ I_{\text{det}} \propto \langle | E_L(x_{\text{det}}) + E_{\text{QD}}(x_{\text{det}}) |^2 \rangle \]

\[ = \langle | E_L(x_{\text{det}}) |^2 \rangle + 2\langle | E_L(x_{\text{det}})E_{\text{QD}}(x_{\text{det}}) | \rangle + \langle | E_{\text{QD}}(x_{\text{det}}) |^2 \rangle, \]

where \( E_L(x_{\text{det}}) \) is the electric field of the laser and \( E_{\text{QD}}(x_{\text{det}}) \) is the electric field of the QD at the detector. The time averaging during the measurement is represented with brackets. Due to the stabilized laser intensity, the first term results in a constant offset to the signal. By considering the fact that \( | E_{\text{QD}} | \ll | E_L | \) the third term in (3.1) can be neglected. Therefore the measured intensity is given by

\[ I_{\text{det}} \propto \langle | E_L(x_{\text{det}})E_{\text{QD}}(x_{\text{det}}) | \rangle. \]

(3.2)

The response of the QD to the incident laser field is given by \( E_{\text{QD}} = \chi(\delta)E_L(0) \), where \( \chi(\delta) \) is the susceptibility as a function of the laser detuning \( \delta \) and \( E_L(0) \) describes the laser field at the position of the QD. For a complex susceptibility \( \chi(\delta) = \chi'(\delta) + i\chi''(\delta) \) the first term relates to the dispersion while the second term stands for the absorption. By simplifying the QD to a two level system, the susceptibility can be expressed as follows

\[ \chi'(\delta) \propto \frac{\gamma\delta}{\delta^2 + \gamma^2}, \quad \chi''(\delta) \propto \frac{\gamma^2}{\delta^2 + \gamma^2}, \]

(3.3)

with the radiative damping rate \( \gamma = \Gamma/2 \) and the laser detuning \( \delta = \omega_0 - \omega_L \) between the laser energy \( \hbar\omega_L \) and the excitonic transition energy \( \hbar\omega_0 \). Additionally, one has to take into account the phase shift of \( \pi \) while the laser beam is propagating through its focus, the Gouy phase [67, 68]. For this reason the laser field is phase shifted by \( \pi/2 \) compared to the electric field of the scattered light by the QD. This leads to a detected intensity given by

\[ I_{\text{det}} \propto \text{Re}\langle E_L^2(0)e^{i\pi/2}(\chi' + i\chi'') \rangle. \]

(3.4)

Due to the fact that only the absorptive part of the QD response is in phase with the laser field at the detector, its interference with the incoming field is measured.
Experimentally, the measured quantity in a differential transmission experiment is the transmission contrast \( \frac{dT}{T} \), which is given by

\[
\frac{dT}{T} \propto \frac{\sigma_0}{A} \gamma^2 \delta^2 + \gamma^2, \tag{3.5}
\]

where \( A \) corresponds to the spot size of the laser and \( \sigma_0 \) is the scattering cross section. Alternatively, the reflection signal \( \frac{dR}{R} \) can be investigated since the QD is emitting light in all spatial directions. A detailed derivation of equation (3.5) is given by Karrai et al. [69]. For simplicity differential transmission and reflection are referred as \( dT \) and \( dR \), respectively. The measured \( dT \) signal as a function of the laser detuning leads to a Lorentzian signal as shown in Fig. 3.1. The slight asymmetry in the Lorentzian can be explained with an additional small component from the dispersion. Such a situation occurs if the emitted light from the QD experiences an additional phase shift due to scattering effects at the sample surface or the gate.

Resonant Rayleigh scattering revealed the two-level nature of a QD that manifests in power broadening and a saturation of the signal [70]. With an increase of the laser power, the incoherent part of \( E_{QD} \) increases and therefore the measurement contrast decreases resulting in a saturation of the signal.

A particularity of the resonant measuring method differential transmission

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure3.1.png}
\caption{\textbf{dT/T measurement for X$^-$}. Typical dT/T signal versus detuning for X$^-$ below saturation. The optical line width is 1.7\,\mu eV.}
\end{figure}

\( (dT) \) is the challenge to resolve a change in the reflection of \( \sim 0.1\% \). Usually, an increase of the signal to noise ratio (SNR) is achieved by using Lock-In technique. To perform a Lock-In measurement, the resonance energy of the exciton needs to be modulated. This is realized with the quantum confined Stark effect, which allows
to control the exciton transition energy via the gate voltage (see chapter 2.1.4). To resolve the true line shape of the resonance, a square wave modulation of $V_{pp} = 100\text{mV}$, which is an order of magnitude larger than the line width of the resonance, is applied to the gate. Therefore two replica of the resonance at $V_- = V_r - 1/2V_{pp}$ and $V_+ = V_r + 1/2V_{pp}$ can be observed in the measured spectrum. In a Lock-In detection only the first Fourier component of the modulation frequency is integrated and all other frequencies are filtered out. Therefore a higher SNR can be achieved [31, 71].

### 3.3. Resonance Fluorescence

An alternative approach to perform resonant spectroscopy is to measure directly the response of the QD to the incident laser field, which corresponds to $\langle |E_{QD}(x_{\text{det}})|^2 \rangle$ in equation 3.1. The main difficulty is to achieve a sufficient suppression of the laser such that the resonance fluorescence photons (RF) from the QD are not overwhelmed by the laser background. The approach by Muller et al. [72] relies on a spatial filtering of the resonant laser. Alternatively, a polarization suppression of the laser by a factor of $10^6$ allows for measuring resonance fluorescence [73, 74]. Experimentally, this is realized by operating the microscope in a dark-field configuration [75]. A linear polarizer that is perpendicular to the incoming linearly polarized laser field is placed in front of the collection fiber. Subsequently the RF photons are sent to an avalanche photodiode (APD). In contrast to a dT measurement where the polarization of the signal is given by the laser, only the photons that have a polarization component orthogonal to the excitation laser are collected in RF measurements. Typically $10^3 - 10^6$ counts/sec. are detected from a single QD.

### 3.4. Pump Probe Measuring Methods

To explore the electron nuclear spin system with increasing transverse magnetic field in chapter 4, it would be ideal to directly observe the dynamics of the electron and nuclear spins. However, only the electron spin is directly accessible with current measurement techniques. In contrast, nuclear spin polarization can only indirectly be measured via the electron spin. This is possible due to the hyperfine interaction that generates an energy shift (Overhauser shift $\Delta E_{os}$) on the electron spin levels which is proportional to the nuclear spin polarization. In this section we describe a method to efficiently polarize nuclear spins and optically measure the magnitude of nuclear spin polarization. Furthermore, two different pump probe measuring methods are developed to access $\Delta E_{os}$ with resonant spectroscopy.
3.4.1. Optical detection of Nuclear Spin Polarization

There exist several methods to polarize nuclear spins in an InGaAs QD [66, 76, 77]. Most of them relay on the scalar form $\mathbf{S} \cdot \mathbf{I}$ of the hyperfine interaction that conserves the total spin and the energy. By the relaxation of an electron spin via this interaction, the spin angular momentum is transferred to a nuclear spin in the so-called flip-flop process. Therefore, a large electron spin polarization obviously helps to create significant nuclear spin polarization. Due to the selection rules in a QD, which correlate the electron spin states with the light polarization, a large electron spin polarization can be achieved by optical means. Since a sizable nuclear spin polarization at $B_x = 0$ is required for the investigation of nuclear spin dynamics in a transverse magnetic field $B_x$, we polarize nuclear spins by exciting the QD into one of its excited p-shell states [66].

To guarantee a sizable nuclear spin polarization a single electron spin has to be in the ground or excited state of the exciton, which is only fulfilled in a charged QD. Additionally, we would like to monitor the electron spin polarization via the polarization of the fluorescence, therefore the electron spin has to be in the excited state. Out of these reasons we will focus our studies on the $X^+$ state. A schematic drawing of the $X^+$ energy diagram is shown in Fig. 3.2 (b). The QD is excited into the p-shell with a $\sigma^+$ polarized laser. The subsequent relaxation to the s-shell takes place within picosecond timescales. The relaxation occurs due to a combination of phonon-mediated relaxation and co-tunneling through the electron

![Diagram](image.png)

Figure 3.2.: Optical detection of nuclear spin polarization in $X^+$. (a) Nuclear spin polarization is achieved by resonantly pumping a single QD to one of its excited states in the p-shell with a circularly polarized laser (e.g. $\sigma^+$, green arrow). Subsequently the emitted co- and cross-polarized photons are detected. (b) PLE spectra of $X^+$ in zero magnetic field measured under co-polarized ($\sigma^+/\sigma^+$) and cross-polarized ($\sigma^+/-\sigma^-$) excitation/detection configuration. The energy splitting $\Delta E_{os}$ is induced by the Overhauser field.

The figure shows the PLE spectra of $X^+$ with different excitation/detection configurations, where $\sigma^+$ and $\sigma^-$ represent the polarization directions of the laser and detected photons, respectively. The energy splitting $\Delta E_{os}$ is highlighted with a red arrow. The spectra are measured in zero magnetic field.
reservoir. Finally the $X^+$ state recombines to $|\downarrow\rangle (|\uparrow\rangle)$ under the emission of a $\sigma^+$ ($\sigma^-$) polarized photon. These photons can be analyzed with a spectrometer. The polarization configuration of the p-shell laser and the emitted photons is denoted $(\sigma^\alpha/\sigma^\beta)$ where $\sigma^\alpha$ is the excitation polarization and $\sigma^\beta$ represents the detection polarization. Due to nuclear spin polarization an energy shift between $|\downarrow\uparrow\rangle$ and $|\uparrow\downarrow\rangle$ occurs. This so-called Overhauser shift $\Delta E_{OS}$ is proportional to the magnitude of the nuclear spin polarization in z-direction and can be measured by separately analyzing the co- and cross- polarized emitted photons. Such a polarization sensitive detection method allows for a high spectral resolution up to $4 \mu eV$. For all measurements the p-shell state was excited to saturation, which results in a high polarization preservation. The polarization preservation can be quantified with the degree of circular polarization which is defined as $\rho^+ = (I^+ - I^-)/(I^+ + I^-)$, where $I^\alpha$ is the PLE intensity in the $(\sigma^+, \sigma^x)$ polarization configuration. Experimentally, a polarization preservation of up to $\rho^+ \sim 80\%$ can be measured. The emitted PLE photons as a function of the energy detuning are shown for co- and cross- polarized detection with respect to the excitation polarization in Fig. 3.2 (a). Clearly a high degree of polarization preservation is exhibited. The energy shift of $\Delta E_{OS} = 18 \mu eV$ between the two polarization configurations corresponds to an effective magnetic field experienced by the electron of $B_{eff} = \hbar \omega_{OS}/g_e \mu_B \sim 500 mT$. In summary, resonant excitation into the p-shell allows to polarize nuclear spins. In order to measure the nuclear spin polarization the energy splitting of co- and cross-polarized detection is extracted. Additionally, in $X^+$ the electron spin polarization can be measured determining the polarization of the emitted photons. To achieve a higher spectral resolution for measuring nuclear spin polarization compared to a spectrometer and to have a coherent measuring method to directly investigate the optical transitions, resonant spectroscopy techniques in combination with p-shell excitation were investigated. In this case the limiting factors for the resolution are given by the optical line width that is determined by the homogeneous broadening and the inhomogeneous broadening that arises from charge fluctuations and distortions of the optical line shape.

### 3.4.2. Pump Probe Method with Resonant Rayleigh Scattering

One possibility to access the Overhauser shift resonantly is to combine the p-shell pumping with resonant Rayleigh scattering, namely dR, which is described in section 3.2. We performed a dR measurement on $X^+$ for different p-shell laser powers. The results of this measurements are shown in Fig. 3.3 (black). Obviously, there is an ideal PLE laser power of $12 \mu W$ to achieve the largest reflection contrast. The signal
strength in resonant absorption measurements, like dR, is strongly dependent on the population of the ground state of the optical transition \([15, 78]\). Only with a sizable population of the ground state, a dR signal can be measured. Since the ground state of the \(X^+\) consists of a single hole inside the QD, the PLE laser is populating the QD with holes which is explained in detail in chapter 2. For weaker PLE laser power than 12\(\mu W\) the reflection contrast is smaller due to reduced population of the \(X^+\) ground state. A larger PLE laser power than 12\(\mu W\) results in a population of excited states of \(X^+\). Therefore the single hole population inside the QD is reduced, which leads to a smaller reflection contrast.

To investigate nuclear spin polarization in a transverse magnetic field (see chapter 4) we are interested in a large Overhauser shift \(\Delta E_{os}\) in addition to a sizable reflection contrast. A comparison of the reflection contrast (black) and \(\Delta E_{os}\) (blue) as a function of the PLE laser power is shown in Fig. 3.3. Sizable nuclear spin polarization of \(\Delta E_{os} = 19\mu eV\) can only be achieved for a large PLE laser power of several 100\(\mu W\), where the reflection contrast is small.

One possibility is to choose the p-shell laser power such that a sizable reflection contrast is reached at the cost of a reduced nuclear spin polarization. Another approach is to combine the large Overhauser shift with a sizable reflection contrast in a pulsed pump probe technique. While nuclear spins are polarized in the pump sequence, a reduced p-shell laser power enables a sizable reflection contrast for the resonant measurements in the probe sequence. To polarize nuclear spins, a p-shell laser power of several hundred \(\mu W\) is needed. In order to perturb the system as little as possible, the dR measurements take place with an additional weak p-shell laser that is needed to charge the QD with a single hole. The probe signal depends dramatically on the time constants of the experiment. If the measurement time

![Figure 3.3: Comparison between dR and \(\Delta E_{os}\). Reflection contrast (black) and \(\Delta E_{os}\) (blue) as a function of PLE laser power.](image)
Figure 3.4.: Nuclear spin dynamics measured with p-shell emission. (a) Setup for pump probe measurement. The p-shell excitation laser is pulsed with an AOM. The pump laser is blocked with a mechanical shutter in front of the spectrometer. (b) Pulse sequence for the pump probe experiment. A long pump pulse $T_{\text{pump}}$ polarizes nuclear spins, after a waiting time $T_{\text{wait}}$ a short p-shell laser ($T_{\text{probe}}$) is needed to probe the sample. (c) $\Delta E_{\text{os}}$ is measured as a function of $T_{\text{pump}}$. An exponential fit (red) leads to a buildup time of nuclear spin polarization $\tau_{\text{buildup}}=2\text{ms}$. (d) $\Delta E_{\text{os}}$ is measured as a function of $T_{\text{wait}}$. An exponential fit (red) leads to a decay time of nuclear spin polarization $\tau_{\text{decay}}=66\text{ms}$.

$(T_{\text{probe}})$ is larger than the decay time of nuclear spins $(\tau_{\text{decay}})$, $\Delta E_{\text{os}}$ is vanishes and only one resonance is measured with a linearly polarized probe laser. In the case of $T_{\text{probe}}<\tau_{\text{decay}}$ the Overhauser splitting can be determined with the energy splitting between the two circularly polarized resonances. Hence this technique allows access to the nuclear spin decay time by varying $T_{\text{probe}}$.

In all following experiments we rely on the detection of the maximal Overhauser shift. Therefore the time constants for build up and decay of nuclear spin polarization in $X^+$ have to be determined. We realized this by a pump probe experiment based on PLE as follows. In order to achieve well defined pump and probe pulses, the cw laser amplitude is modulated with an AOM. Furthermore a mechanical shutter is used to block the pump pulses while allowing the probe pulses to reach
the spectrometer. A schematic picture of the pulse sequences is shown in Fig. 3.4. For measuring the buildup (decay) of nuclear spin polarization, $T_{\text{pump}}$ ($T_{\text{wait}}$) are varied while keeping all other parameters fixed. To improve the signal to noise ratio, the pump-probe sequences are repeated several times during the measurement process, while the signal is accumulated on the spectrometer CCD.

In Fig. 3.4 (c) and (d) the resulting buildup and decay curves for nuclear spin polarization in $X^+$ are shown. Fitting the data with an exponential curve gives a buildup time of $\tau_{\text{buildup}} = 2\text{ms}$ and a decay time of $\tau_{\text{decay}} = 66\text{ms}$.

While the buildup time is determined by the method which is used to polarize nuclear spins, the decay time is a characteristic of the QD state. Similar to previous experiments performed by Maletinsky et al. [8] a buildup time of several ms is measured. Since for $X^+$ there is a single hole in the ground state, the fast electron mediated nuclear spin decay should be strongly suppressed [8, 79]. Therefore a long nuclear spin decay time for $X^+$ is expected and was measured to be on the order of seconds [8]. We find a much shorter $\tau_{\text{decay}}$ of 66ms, which is most likely due to less well defined charge state compared to the experiments performed by Maletinsky et al. [8].

Not only for the nuclear spin polarization but also for the signal to noise ratio (SNR) of the probe signal the timescales of a pump probe measurement are important. The ratio between $T_{\text{probe}}$ and $T_{\text{pump}}$ determines the scaling factor for the integration time which is needed to arrive at the same SNR as for a cw experiment. Hence the measured $\tau_{\text{decay}} = 66\text{ms}$ in combination with a short buildup time $\tau_{\text{buildup}} = 2\text{ms}$ should result in a reasonable signal to noise ratio.

Usually, differential reflection dR is measured with a Lock-In technique (see section 3.2). Therefore a closer look at the time constants of the Lock-In amplifier is needed for performing a resonant pump probe measurement in combination with dR. A commercially available Lock-In amplifier has a time constant $T_c$ that can be chosen between several hundred $\mu$s up to seconds. Obviously a larger $T_c$ results in a better signal to noise ratio. Considering a change of the input signal many $T_c$ are needed to reach the output signal. This is due to the response time of the low-pass filter. The timing and synchronization of the pump probe pulses is controlled with a data acquisition (DAQ) device based on a clock period of 2$\mu$s. Because of its finite memory the modulation sequences are restricted to $\sim 2\text{s}$. During the waiting time between the modulation sequences the feedback loop of the EOM intensity modulator is performed and results in a new pulse sequence which has to be written on the DAQ device. The combination of the waiting time between the modulation sequences with the response time of the Lock-In amplifier ($\tau_{\text{Lock-In}}$) leads to a substantial amount of time where no signal can be acquired (Fig. 3.5). To circumvent this problem we tested an alternative approach consisting of a
Figure 3.5.: Measurement scheme for pump probe with dR. (a) Measurement of the pump laser power $\Omega_{\text{pump}}$ versus time. The gap between the modulation sequences arises from writing the sequence on the DAQ device memory. (b) Corresponding dR signal from Lock-In amplifier versus time. The strong pump laser between the modulation sequences leads to an increase of the dR signal. (c) Digital signal to be multiplied with Lock-in signal. To avoid memory effects the modulation of the dR signal is shifted by $\tau_{\text{Lock-In}}$.

software based Lock-In amplifier, which is implemented directly on the DAQ device. With this method a synchronization of the Lock-In amplifier with the pump probe pulses is possible such that the response time of the Lock-In amplifier can be reduced. To compare the commercially available Lock-In amplifier from Signal Recovery (SR) with the software based Lock-In amplifier, a cw experiment was performed. The result is shown in Fig. 3.6. For the software based Lock-In amplifier a decrease of the signal to noise ratio by a factor of ten is exhibited, which is caused by the finite bandwidth of the used digital to analog converter on the DAQ device. Even though a faster response time can be reached with a Lock-In amplifier implemented on the DAQ device compared to the SR Lock-In amplifier, the combination with the strongly reduced signal to noise ratio will not result in a better performance for the pump probe experiment. Therefore the pump probe experiments were realized with a commercially available Lock-In amplifier, taking
3.4. Pump Probe Measuring Methods

![Image of dR measurement with software based Lock-In amplifier](image)

**Figure 3.6:** dR measurement with software based Lock-In amplifier. dR measurements as a function of gate voltage performed with a SR Lock-In amplifier (black) and a software based Lock-In amplifier (DAQ device) (red).

into account the slower response time compared to a Lock-In amplifier which is based on the DAQ device.

For a first attempt at pump probe measurement with dR only the $\sigma^+$ polarized p-shell laser is pulsed while the resonant laser and a hole generating weak PL laser is continuously on. During the entire measuring process, the gate is modulated for the Lock-In detection (see Fig. (b)). In this experiment a fiber based EOM intensity modulator served as a fast switch of the p-shell laser intensity. Besides the faster rise time compared to an AOM, the fiber based intensity modulator has a larger throughput. The intensity modulation is achieved with two EOMs that are forming a Mach-Zehnder interferometer. By applying a voltage to the EOM the phase of the light in one interferometer arm can be shifted with respect to the other. If the light after recombination is out of phase a destructive interference is observed, resulting in a reduced intensity. Unfortunately the voltage needed to apply to the EOM for achieving a maximal suppression is drifting due to temperature fluctuations. Either the environmental temperature or the heating of the device with a strong pump laser can lead to temperature changes inside the intensity modulator. These changes are rather slow therefore the voltage needed for a maximal suppression can be stabilized with a control loop feedback mechanism (PID).

Similar to the co- and cross- circularly polarized probing performed in non-resonant measurements, we compare $\sigma^+$ and $\sigma^-$ resonant absorption. In Fig. 3.7 (a) dR signal for both polarizations is plotted as a function of the laser detuning. From top to bottom the measuring time between the pump pulses which is indicated with $T_{\text{wait}}$ is decreased. Clearly, there is an observable energy shift up to $5\mu$eV between $\sigma^+/\sigma^+$ and $\sigma^+/\sigma^-$. Additionally, the asymmetry increases as the ratio between $T_{\text{pump}}$ and $T_{\text{wait}}$ increases which can be explained as follows. The consequence
Figure 3.7.: Pump probe measurement with dR. (a) Pump Probe measurements of the Overhauser shift for different polarizations of the resonant laser. DR signal versus detuning for a resonant $\sigma^+$ polarized laser is shown in black, while for a resonant $\sigma^-$ polarized laser it is shown in red. The waiting time $T_{\text{wait}}$ and therefore the measurement time is decreasing from top to bottom. (b) Measurement scheme. The pump laser is pulsed with an EOM, while the resonant laser and a weak PL laser (to generate the hole for $X^+$) is continuously on. Gate voltage is modulated during the measurement process and even on resonance during $T_{\text{pump}}$. (c) Energy level diagram for $X^+$. The $\sigma^+$ pump laser polarizes nuclear spins by exciting to the p-shell (shown in green). Resonant measurements can be performed with $\sigma^+$ (red arrow) or $\sigma^-$ (blue arrow). A large population of $|\downarrow\uparrow\downarrow\rangle$ by the pump laser leads to gain (indicated with dotted orange arrow).

of a strong p-shell laser is a large population of the $|\downarrow\uparrow\downarrow\rangle$ state which leads to stimulated emission (gain) in a dR measurement (Fig. 3.7 (c)). Therefore a negative dR signal can be observed for a $\sigma^+$ polarized resonant laser. Especially the dependence on the ratio between $T_{\text{pump}}$ and $T_{\text{wait}}$ can be understood by the fact that the large population in $|\downarrow\uparrow\downarrow\rangle$ is increasingly dominating for shorter $T_{\text{wait}}$, leading to the observed stimulated emission. In order to extract the energy splitting $\Delta E_{\text{os}}$ between $\sigma^+$ and $\sigma^-$ polarized resonant excitation, the gain process is avoided by modulating the gate voltage away from the $X^+$ resonance during the strong pump pulse.
3.4. Pump Probe Measuring Methods

![Graph](image)

**Figure 3.8.:** Pump probe measurement with dR for different $T_{\text{wait}}$. dR signal versus detuning for a linearly polarized resonant laser is shown. The waiting time and therefore the measurement time is decreasing from top to bottom. (a) With a waiting time of 3.5 ms, nuclear spin polarization is already decayed to $\Delta E_{\text{OS}}=3.5 \mu\text{eV}$. (b), (c) A further decrease of the waiting time leads to $\Delta E_{\text{OS}}=7 \mu\text{eV}$.

The high spectral resolution of dR compared to PLE allows to measure the Overhauser shift directly in a linear basis. In Fig. 3.8 the dR signal as a function of the laser detuning is shown for decreasing measurement time $T_{\text{wait}}$ between the pump pulses from top to bottom. With the realization of a gate modulation away from the $X^+$ resonance during the strong pump laser, two separated resonances with a Lorentzian dR signal can be measured for a linearly polarized probe laser. The maximal nuclear spin polarization is reached for $T_{\text{wait}}=1.4\text{ms}$. Because of the constant pump time $T_{\text{pump}}$ the ratio between the measuring time and $T_{\text{pump}}$ is decreasing from top to bottom and thus the signal to noise ratio (SNR).

Such a short nuclear spin depolarization time is surprising, especially since previous measurements exhibited a $\tau_{\text{decay}}$ of 66 ms. One reason for a shortened decay time could be the influence of the weak PL laser or the probe laser which are continuously...
on. For a better understanding a detailed study including a pulsed weak PL laser and probe laser would be needed.

3.4.3. Pump probe Method with Resonance Fluorescence

Although a resonant measurement of the Overhauser shift can be realized with a pump probe method based on dR, the need for a Lock-In technique leads to a substantial amount of time when no signal can be acquired as discussed in the previous section. Alternatively resonance fluorescence (RF), that is explained in detail in section 3.3, can be used as a resonant measuring method. This has the advantage that no Lock-In technique is needed and therefore a higher SNR in a pump probe experiment should be achieved compared to dR. Switching from resonant Rayleigh scattering to RF does not alter the experiment substantially, only changing the polarization of the excitation laser is a much more time consuming task for RF than for dR because of the alignment of the polarizer in order to achieve a high polarization suppression.

In Fig. 3.9 (a) a scheme for the pump probe measurements with RF is shown. Similar to the previous pump probe technique, the pump pulses are generated with an EOM while a weak hole generating PL laser and the resonant laser are continuously on. Especially during $T_{\text{pump}}$ a suppression of the background is difficult because the emitted PLE photons cannot be distinguished from RF photons. Therefore only the photons between two pump pulses, during $T_{\text{wait}}$ are detected. The resulting RF count rate for a $\sigma^-$ polarized pump laser and a linearly polarized resonant laser as a function of the laser detuning is shown in Fig. 3.9 (b). Compared to the measurement shown in Fig. 3.8 a reasonable signal to noise ratio is achieved with less integration time.

To conclude, a resonant pump probe technique to measure nuclear spin polarization is realized with dR and RF. A comparison of these methods shows an easier polarization control for dR which is contrasted by the improved signal to noise ratio of RF measurements.
Figure 3.9.: Pump probe measurements with RF. (a) Scheme for a pump probe experiment measuring the RF. The pump laser is pulsed with an EOM, while the probe laser and the weak hole generating PL laser are continuously on. The RF photons are only counted during the time period $T_{\text{wait}}$, when the strong pump laser is off. (b) RF pump probe measurement as a function of the laser detuning. The pump laser is $\sigma^-$ polarized, while the resonant laser is linearly polarized.
4. Nuclear Spin Polarization in Transverse Magnetic Fields

We investigated nuclear spin polarization in individual self-assembled InAs/GaAs quantum dots for different sample structures in transverse magnetic fields. Hanle depolarization curves are measured to explore nuclear spin dynamics, leading to an anomalous Hanle curve with a broadening up to 1T. Even though nuclear spin polarization is comparable for different sample structures at zero magnetic field, the broadening of the anomalous Hanle curve is substantially different. The critical field, at which the photo-created electron spin polarization breaks down, depends strongly on the electron spin lifetime. For further investigation of direction and strength of the Overhauser field causing the broadening of the Hanle curve, a resonant measurement technique is applied.

4.1. Introduction

A phase transition is the transformation of a physical system from one state to another, such as vaporization and condensation. These transitions manifest in unconventional and unexpected physical behavior and do not only occur at finite temperature. Even at zero temperature or $h\omega > k_B T$ quantum fluctuations (at energy $h\omega$) are expected to lead to so-called quantum phase transitions. In order to have experimental access to a quantum system, a coupling to the environment is needed which leads to dissipation. Dissipative phase transitions can occur if an environmental parameter is varied such as a magnetic field or an external coherent source (e. g. a laser) [80–82]. Investigations on dissipative phase transitions (DPT) are expected to lead to a deeper understanding of the unexpected physical behavior of systems at low temperatures.

A model exhibiting first- and second- order DPT is the central spin system which is externally driven and decays through interaction with a Markovian environment [83]. In its simplest form, a central spin system consists of a set of spin-1/2 particles, uniformly coupled to a single spin-1/2. A possible experimental
implementation of such a system is a negatively charged InGaAs QD, where nuclear spins represent the set of spin-1/2 particles and the electron is the single spin-1/2 particle. Accordingly, the nuclear spin ensemble is expected to undergo phase transitions. The behavior of the nuclear spin ensemble can be monitored via the interaction between nuclear and electron spins. If a negatively charged InGaAs QD is driven by a laser, it experiences spontaneous decay and therefore satisfies the requirements of the theoretical model studied by Kessler et al. [83]. On the other hand the model does not consider inhomogeneous hyperfine coupling, the electron Zeeman splitting and the nuclear quadrupolar interactions [51, 52] in InGaAs QDs, which raises the question to what extent an experimental observation of dissipative phase transitions is possible in this system. Still, in recent experiments on InGaAs QDs indications for a first order phase transition in the nuclear spin ensemble were observed, including optical bistability as a function of a transverse magnetic field $B_x$ [20]. Therefore we want to further investigate the electron - nuclear spin system under the influence of $B_x$ with an increased spectral resolution to identify discontinuities in the polarization of the nuclear spin ensemble. An abrupt change in electron and nuclear spin polarization would signal a dissipative phase transitions and lead to new insights into the electron nuclear spin system.

4.2. Hanle experiment

In a Hanle measurement, the dynamics of an electron spin in a transverse magnetic field $B_x$ is determined by optical means. In the course of such an experiment an exciton is created with a circularly polarized excitation laser. A schematic drawing of the measurement geometry is shown in Fig. 4.1(a). Because of the selection rules in a QD, the electron spin polarization $S_0$ is optically defined along the z-axis parallel to the Poynting vector of the excitation laser. In the case a finite magnetic field $B_x$ is applied, the optically generated electron spin undergoes Larmor precession around the x-axis at frequency $\Omega_{el} = g_d \mu_B B / \hbar$ (see Fig. 4.1(b)). Due to the fact that the projection of the electron spin onto the z-axis defines the degree of circular polarization of the emitted photons, a depolarization of the PL light can be observed as a function of $B_x$. This dependence allows to determine both, the electron spin lifetime $\tau_s$ and the recombination time $\tau$ of the exciton [48].

The electron spin precession together with spin pumping, spin relaxation and recombination can be described with the following equation of motion for the spin vector $\vec{S}$:

$$\frac{d\vec{S}}{dt} = \vec{\Omega}_{el} \times \vec{S} - \frac{\vec{S}}{\tau_s} - \frac{\vec{S} - \vec{S}_0}{\tau}.$$  \hspace{1cm} (4.1)
4.3. Anomalous Hanle Effect

Figure 4.1.: Scheme illustrating Hanle measurements. (a) An external magnetic field $B_x$ is applied transverse to the Poynting vector of the i.e. $\sigma^+$ polarized laser (orange arrow). Electron spin dynamics are investigated with a polarization analysis of the emitted light (black arrow). (b) The electron spin polarization is optically defined along the z-axis. For finite $B_x$ the electron spin (red arrow) precesses around the x-axis leading to a decay of the polarization along the z-axis.

The first term represents the spin precession in the magnetic field, the second term describes the spin relaxation and the third term denotes the generation of the spin by optical excitation ($S_0/\tau$) and its decay ($-S/\tau$). In steady state ($\frac{dS}{dt} = 0$) and in the presence of a magnetic field transverse to $S_0$ ($\tilde{\Omega}_{el} \perp \vec{S}_0$) one obtains

$$S_z(B) = \frac{S_z(0)}{1 + \left(\frac{\Omega_{el}}{\tau^*}\right)^2}, \quad (4.2)$$

with the effective time $\tau^* = (\tau^{-1} + \tau_s^{-1})^{-1}$. Hence, a Lorentzian with a half width of

$$B_{1/2} = (\Omega_{el}\tau^*)^{-1} = \frac{\hbar}{(g_{el}\mu_B\tau^*)} \quad (4.3)$$

results for the spin projection $S_z$ and therefore the degree of circular polarization of the emitted light as a function of $B_x$.

4.3. Anomalous Hanle Effect

Running a Hanle experiment is one of the few techniques to monitor the behavior of nuclear spin polarization in transverse magnetic fields. Theoretically, the behavior of nuclear spins should closely resemble the one of electron spins. In a transverse magnetic field $B_x \geq B_{\text{Knight}}$, nuclear spins should exhibit a Larmor precession, which would then lead to a fast decay of the optically generated nuclear spin polarization. In that case, the Hanle curve is expected to exhibit a Lorentzian line shape, as
described by Eq. 4.3, with a FWHM of 30mT for a $g_e = 0.5$ and an effective time dominated by the trion radiation lifetime $\tau^* \sim \tau = 0.7\text{ns}$.

A typical Hanle depolarization curve, measured according to section 3.4.1, is shown in Fig. 4.2 (a). The most striking anomaly to the theoretical predictions is the 40 times broadening of the FWHM. This unexpected broadening is termed anomalous Hanle effect. In addition, the Hanle curve exhibits a large hysteresis of 0.5T. The degree of circular polarization is continuously decreasing for magnetic fields up to 0.5T, where it reaches a minimum value of 50%. For larger magnetic fields the degree of circular polarization increases again until a sudden drop down from 70% to 0% is observed at a critical magnetic field $B_{\text{crit}} = 1.1\text{T}$. Similar results have been presented in the detailed studies by Krebs et al. [20].

Together with the hysteresis, the broadening of the Hanle curve is a strong indication for nuclear spin effects. By modulating $\sigma^+ / \sigma^-$ polarization at kHz frequency, an optical polarization of nuclear spins can be prevented [8]. This effect was exploited in an additional measurement. The resulting Hanle curve exhibited a narrowing. Therefore we conclude that the nuclear spin polarization is the cause for the anomalous broadening of the Hanle curve in InGaAs QD.

In addition to the PL circular depolarization data, the Overhauser shift can be extracted from the same experiment (Fig. 4.2 (b)). The results show a decrease of nuclear spin polarization along the z-axis with $B_x$. At the same time a high degree of PL circular polarization is measured (Fig. 4.2 (a)). This leads to the conclusion that a nuclear spin polarization along the x-axis is very likely to compensate $B_x$ and prevents the electron spin from performing Larmor precession. The underlying mechanism that could cause a rotation of the nuclear spin polarization from the z-axis to the x-axis as a function of $B_x$ is not fully understood. Such a situation could result from a tilted effective magnetic field, that is stabilized along the z-axis by a $B_{\text{nuc}}^z$ due to nuclear quadrupolar interactions (QI) [20]. This is further supported by the modest broadening of the Hanle curves measured on strain-free droplet QDs, which do not exhibit QI [84]. Should there exist a tilted effective magnetic field, a finite nuclear spin polarization along the z-axis must be observed up to $B_{\text{crit}}$. The data shown in Fig. 4.2 (b) do not allow to draw a clear conclusion in this respect. To verify the existence of such a nuclear polarization component an experimental technique with increased resolution needs to be applied.
4.4. Hanle measurements for samples with different electron spin lifetimes

In the previous section, the Hanle effect was studied for a sample with 25nm tunnel barrier (sample B) and exhibits a spectacular Hanle curve broadening because of nuclear spin polarization. For a further investigation of the connection between nuclear spin polarization and $B_{\text{crit}}$ we measured samples with different electron spin lifetimes. This is of particular interest since the decay of nuclear spins can be caused by a randomization of the electron spin [5]. Therefore the increased electron spin lifetime of a 35nm tunnel barrier sample (sample A) compared to a 25nm tunnel barrier sample (sample B) should result in a better preservation of nuclear spin polarization which could lead to an increased magnitude of nuclear spin polarization and to even larger values for $B_{\text{crit}}$.

In this section, we present a series of Hanle measurements for different values of nuclear spin polarization at $B_x = 0$ on the same QD to verify to what extent this
Figure 4.3.: Hanle measurements for different nuclear spin polarizations. (a) Hanle depolarization curves for a QD on sample B with a 25nm tunneling barrier. Arrows indicate field sweep direction. (b) Corresponding energy splitting $\Delta E$ between co- and cross- polarized detection. (c) Extracted $B_{\text{crit}}$ for different $\Delta E_{\text{os}}$ at $B_x = 0$T.

A typical Hanle depolarization curve for sample B is shown in Fig. 4.3(a). Similar to the measurements in the previous section, an increased broadening up to $B_{\text{crit}} = 0.75$T and a hysteresis can be observed. The corresponding nuclear spin polarization in z-direction is extracted and plotted as a function of $B_x$ (Fig. 4.3(b)). At $B_x = 0$T a large Overhauser splitting of $\Delta E_{\text{os}} = 20\mu$eV is obtained that decreases continuously as a function of $B_x$ until $B_x = B_{\text{crit}}$. At this point an abrupt drop down to zero is observed. To further investigate the broadening of the Hanle curve for different initial values of $\Delta E_{\text{os}}$ at $B_x = 0$T, we varied the laser power for the p-shell pumping. The results clearly show an increase of $B_{\text{crit}}$ as a function of $\Delta E_{\text{os}}$ at $B_x = 0$T (Fig. 4.3(c)).
To compare the two sample structures A and B with different electron spin lifetimes, we first investigated the magnitude of the nuclear spin polarization at $B_x = 0$. Testing several p-shell resonances for different QDs did not reveal any significant difference for the maximal $\Delta E_{os}$. This result can be explained in two possible ways. Either the dominant depolarization mechanism is not smaller for the longer electron spin lifetime or the p-shell pumping is less effective for the longer electron spin lifetime. Since p-shell pumping relies on a well defined $X^+$ state to polarize nuclear spins, we contrasted the fluorescence spectrum from sample A with the one from sample B (Fig. 4.4). For sample B, a very clean spectrum is measured, whereas sample A can obviously be excited to a multitude of states. This is due to the fact that a longer electron spin lifetime leads to less well defined charge states [62, 85]. Therefore a smaller nuclear spin polarization rate for sample A is expected. Hence a longer electron spin lifetime does not result in a larger $\Delta E_{os}$ at $B_x = 0$.

According to previous experiments the broadening of the Hanle curve scales with

![Figure 4.4: Emission at the s-shell. P-shell excitation at saturation for sample A with a 35nm tunneling barrier (a) and sample B with a 25nm tunneling barrier (b).](image)

$\Delta E_{os}$ at $B_x = 0$ for the same QD on the same sample (Fig. 4.3 (c)). To investigate
Figure 4.5.: Hanle measurements for different sample structures. (a-b) Hanle depolarization curves for a QD on sample B with a 25nm tunneling barrier and a QD on sample A with a 35nm tunneling barrier. Arrows indicate field sweep direction. (c-d) Corresponding energy splitting $\Delta E$ between co- and cross-polarized detection.

the influence of the electron spin lifetime on $B_{\text{crit}}$ we performed Hanle measurements with similar values for $\Delta E_{\text{os}}$ at $B_x = 0$T in sample A and B. In Fig. 4.5 (a-b) typical Hanle depolarization curves for the two different sample structures A and B are shown. The most striking difference is the broadening of Hanle curve which is 0.4T for sample A and 1.5T for sample B. A second difference is the hysteresis, which reaches up to 0.7T in sample B and vanishes completely for sample A. The third difference concerns the line shape. All Hanle curves in sample A show a smooth line shape while for sample B abrupt jumps appear at $B_x = B_{\text{crit}}$. This leads to the conclusion that the nuclear spin polarization at zero magnetic field is not the only parameter that influences the broadening of the anomalous Hanle curve. In contrast to what was observed in GaAs droplet dots [84] the magnitude of the nuclear spin polarization increases during the Hanle measurements and the mechanism which rotates nuclear spin polarization from the z-axis to the x-axis is unknown. This makes direct conclusions for the impact of nuclear spin polarization along the
4.5. Resonant measurements on a 35nm tunnel barrier sample

z-axis on $B_{\text{crit}}$ very difficult. Further studies are necessary to fully understand the increasing nuclear spin polarization along the x-axis in a Hanle measurement. In addition to the Hanle curve we investigate nuclear spin polarization in z-direction with the energy splitting $\Delta E_{\text{os}}$ between co- and cross- polarized detection of the emitted light (Fig. 4.5 (c)-(d)). For both samples $\Delta E_{\text{os}}$ continuously decreases from 12$\mu$eV at $B_x = 0$ to around zero at 0.4T. While for sample A 0.4T corresponds to $B_{\text{crit}}$, sample B exhibits a broadening of the Hanle curve up to 1.1T. For sample B the measured $\Delta E_{\text{os}} = 2\mu$eV in the region between 0.4T and 1.1T is at the resolution limit for a non-resonant measurement method (see section 3.4.1). Therefore it is very difficult to claim a persistence of a small nuclear magnetic field along the z-axis up to $B_{\text{crit}}$. In order to achieve a better understanding of the nuclear spin polarization dynamics in a transverse magnetic field, an increased spectral resolution is needed.

A valuable alternative to the analysis of the QD fluorescence with a spectrometer is the resonant excitation of the QD, which was performed with resonance fluorescence (RF). A resonant measurement offers an at least four times higher spectral resolution which is limited due to the optical line width (see chapter 2). In contrast to non-resonant measurements, RF is an absorption experiment which allows for a direct access of the optical transitions in the s-shell. With the determination of the polarization and the energy difference of these transitions, information about magnitude and direction of nuclear spin polarization is gained.

4.5. Resonant measurements on a 35nm tunnel barrier sample

To prove that the outcome of the experiment can be reproduced by resonant spectroscopy, we performed a pump probe measurement with RF on sample A with a 35nm tunnel barrier. In former measurements a narrow Hanle curve with no abrupt change in the polarization conservation was obtained. This means that for the electron spin there is no complete compensation of the external field $B_x$ caused by the nuclear spin polarization, which can suddenly break down. Therefore we would also expect a gradual change from the Overhauser splitting to the Zeeman splitting for increasing $B_x$.

To study the dynamics of $\Delta E_{\text{os}}$ resonantly, a $\sigma^+$ polarized pump laser is used and the $X^+$ state was resonantly probed with a weak diagonally polarized laser. At zero magnetic field, two resonances are observed, which are energetically separated by the Overhauser shift $\Delta E=10\mu$eV (Fig. 4.6). With increasing magnetic field $B_x$ a decrease of $\Delta E_{\text{os}}$ is observed. As a result the two resonances merge at $B_x = 0.1T$. 
For $B_x = 0.2T$ an evolution into two resolvable resonances is measured because of the Zeeman splitting. Finally, four resonances can be observed for a large magnetic field of $B_x = 1T$. This is due to the fact that in Voigt geometry (transverse magnetic field) all four transitions of $X^+$ are allowed and linearly polarized [78, 86, 87]. Similar to non-resonant measurements on the same sample structure, a gradual decrease of the Overhauser shift can be observed. From the smooth Hanle curve we expect no abrupt change in the nuclear spin polarization. This is confirmed in resonant measurements. Hence the resonant pump probe technique allows for investigations of nuclear spin dynamics in transverse magnetic fields with an enhanced measurement resolution compared to non-resonant measurements.

4.6. Resonant measurements on a 25nm tunnel barrier sample

Hanle measurements on sample B with a 25nm tunnel barrier showed an anomalous broadening of the Hanle curve. First, we would like to reproduce these features which are the abrupt jumps from circular to linear polarization of the emitted photons, hysteresis and a large value for $B_{\text{crit}}$ on the order of 1.5T. In a second step we aim at further investigating the mechanism which rotates the nuclear spin.
4.6. Resonant measurements on a 25nm tunnel barrier sample

polarization from the z- to the x-axis.

4.6.1. Gate properties

In order to ensure good charge screening, we rely on the diode structure and in particular a well grounded back contact. Without the latter, the optically generated charges get trapped and can result in a reduced frequency bandwidth and thus in a large $RC$ time constant. It turned out that the back gate of the sample to be studied was difficult to contact. This is most certainly due to the degradation of the sample material. Therefore special attention has to be paid to the contact quality.

To test the frequency bandwidth of the gate, we applied a square wave modulation with a large amplitude such that the two Stark shifted resonances can be resolved with a spectrometer (Fig. 4.7). By increasing the modulation frequency to 700Hz a merging to one broad resonance is observed. This is a consequence of the reduced frequency bandwidth and confirms an incomplete grounding of the back contact. Therefore the charge screening in the sample structure is restricted.

Not only for a fast gate modulation a good charge screening is needed. Particularly, a strong p-shell pump laser generates a lot of charge carriers in the sample. Without a well grounded back contact, these charges are trapped and can lead to fluctuations and a broadening of the resonances in the subsequent measuring process. In a pump probe experiment performed on sample B, very broad optical line widths were measured independent of the polarization of the pump laser. As a

![Figure 4.7: Gate modulation at different frequencies.](image)

**Figure 4.7:** Gate modulation at different frequencies. PL counts versus wavelength for different gate modulation frequencies. At 2Hz there are two Stark shifted, well separated resonances observable. For a modulation frequency of 200Hz already a merging of the two resonances starts, which evolves at 700Hz to one broad resonance. This is due to the incomplete grounding of the back contact.
consequence the Overhauser splitting could not be resolved. Even though the sample exhibits a reduced frequency bandwidth, a continuous wave (cw) RF measurement is still possible because it does not rely on fast charge screening. A drawback of a cw experiment compared to a pump probe experiment is the increase of the background counts which arises from the incoherent fluorescence. Whereas in PLE measurements incoherent fluorescence was spectrally resolved, it is an additional constant signal for cw RF experiments since the two fluorescence signals cannot be distinguished. Therefore the signal to noise ratio of a cw experiment is substantially reduced compared to a pump probe experiment.

4.6.2. Resonance fluorescence background

Performing a RF measurement as in section 4.5 but for a sample with a smaller tunnel barrier leads to a substantially different outcome. Color coded RF counts for a decreasing magnetic field $B_x$ versus scanned gate voltage are plotted in Fig. 4.8 (a). A sudden change of the background together with an increase of the number of resonances can be observed at $B_{\text{crit}}=1.2T$. As discussed earlier, the main contribution of the RF background arises from the PLE emission. Therefore the averaged RF background and the emitted $\sigma^+$ photons in a PLE experiment versus $B_x$ are shown in Fig. 4.8 (b). In both cases, an abrupt decrease at $B_{\text{crit}}$ can be observed. As discussed above, the PLE photons exhibit a strong polarization preservation i.e. $\sigma^+$ for $B_x < B_{\text{crit}}$. For larger magnetic fields this polarization preservation breaks down, resulting in a linearly polarized emission (see section 4.4). As a consequence, the PLE $\sigma^+$ count rate exhibits a sudden decrease of 75% at $B_{\text{crit}}$. The same step would be observed for a $\sigma^+$ polarized RF background. However, the measured decrease in the RF background is only 5%, corresponding to a 7% circularly polarized RF basis. This is below the measurement uncertainty of the polarization measurement equipment. Therefore the sudden decrease of the RF background has a physical origin and is an indication for the breakdown of the nuclear spin polarization. Additionally, an increase of the RF background from zero to 0.3T can be seen in Fig. 4.8 (b). This is also observed for the PLE $\sigma^+$ emission. Although, the reason for this increase is unclear, we can rule out any change in the position of the QD, since the resonant signal does not change its magnitude. Moreover the increase of the RF background is not a general feature and was only observed for this particular QD. This is not particularly surprising since the line shape of the anomalous Hanle curve differs from QD to QD.
4.6. Resonant measurements on a 25nm tunnel barrier sample

Figure 4.8.: CW RF measurements sample B. (a) RF measurements for decreasing in-plane magnetic field $B_x$ versus gate voltage. White lines are a guide to the eye indicating the Zeeman splitting. Performed on a 25nm tunneling barrier sample (B). (b) Averaged background from RF measurements (blue) as a function of $B_x$ in comparison with the emitted $\sigma^+$ photons in a PLE experiment (black). An abrupt jump at $B_{\text{crit}}$ is observable in both cases.

4.6.3. Extraction of the resonance positions

Not only the abrupt change in the RF background at $B_{\text{crit}}$ is an indication for the breakdown of nuclear spin polarization but also the number of resonances before and after $B_{\text{crit}} = 1.2$T. In contrast to PLE measurements, the increased spectral resolution of the RF measurements allows to measure four resonances above $B_{\text{crit}}$ for the first time. Line cuts below and above $B_{\text{crit}}$ of the measurements shown in Fig. 4.8 (a) are plotted in Fig. 4.9 (a). Below $B_{\text{crit}}$ we observe a pair of split resonances separated by the hole Zeeman splitting ($|g_h|\mu_B B_x$). This observation is fully consistent with the cancellation of $B_x$ with an in-plane Overhauser field. Above $B_{\text{crit}}$ the nuclear spin polarization breaks down, resulting in an additional electron Zeeman splitting ($|g_e|\mu_B B_x$) [20]. The expected fourth resonance lies just...
nuclear spin polarization mainly along the x-axis for $B_x \sim B_{\text{crit}}$. To investigate the evolution of the nuclear spin polarization direction between these two points in

4.6.4. Transition between Overhauser shift and hole Zeeman splitting

Nuclear spins are polarized along the z-axis at $B_x = 0$T and Fig. 4.8 implies a nuclear spin polarization mainly along the x-axis for $B_x \sim B_{\text{crit}}$. To investigate the evolution of the nuclear spin polarization direction between these two points in

Figure 4.9.: Extracted positions of the resonances (a) Plot of two line cuts from the measurements shown in Fig. 4.8(a). Below $B_{\text{crit}}$, at $B = 1.05$T only two resonances are observed (black), that are split by the hole Zeeman energy $g_h \mu_B B_x$. Above $B_{\text{crit}}$, at $B = 1.35$T the nuclear spin polarization breaks down, resulting in the expected electron and hole Zeeman splitting (red). The background is subtracted. (b) Extracted energy shift $\Delta E$ between different energy transitions of the measurements shown in Fig. 4.8(a). Arrows indicate the color coded field sweep direction. Black lines show expected Zeeman splitting; electron and hole g-factors were determined in an independent measurement.

not within the measurement range.

The energy differences between the various resonances extracted from Fig. 4.8 (a) are plotted as a function of the magnetic field $B_x$ in Fig. 4.8 (b). The data for an increasing magnetic field is taken from an additional measurement. The electron and hole Zeeman splitting is indicated with black lines, corresponding g-factors were determined in independent measurements. The data shows a hysteresis, as it is expected from Hanle experiments (Fig. 4.5). At $B_{\text{crit}}$ the resonances jump to the energy positions that are calculated from the electron and hole g-factors.
more detail, a resonant RF measurement was performed in an increasing magnetic field below $B_{\text{crit}}$ and with a smaller step size of $B_x = 0.025$T. The result is shown in Fig. 4.10 (a) after background subtraction. Three different regions can be distinguished. At $B_x = 0$T the Overhauser splitting is visible. An intermediate regime exists at $B_x = 0.45$T before the hole splitting dominates at $B_x = 1$T. Respective line cuts that represent the three different regimes are shown in Fig. 4.10(a).

In the first regime, at $B_x = 0$T the two resonances are not equally strong. This height difference arises due to depopulation of the ground state caused by the pump laser. To illustrate the influence of the pump laser on the RF signal we performed a simulation of the underlying four level system. The calculations are based on the master equation solved in steady state; incoherent transitions are included with a Lindblad form [88]. An energy level diagram that indicates also the considered decay rates is shown in Fig. 4.11 (a). For simplification we did not consider a dynamic nuclear spin polarization (DNSP) that scales with the pump

Figure 4.10.: Transition between Overhauser shift and Zeeman splitting. (a) Resonant measurements for increasing in-plane magnetic fields below $B_{\text{crit}}$. Resonant excitation laser is vertically polarized with respect to the QD axes. (b) Line cuts for selected magnetic fields $B_x$. 
laser power. As a result the two excited states $|\downarrow\uparrow\downarrow>$ and $|\uparrow\downarrow\uparrow>$ exhibit a constant splitting $\Delta E_{os}$ as a function the laser power. The influence of the pump laser on the system is simulated with a rate $\Gamma_{pump}$ from $|\downarrow>$ to $|\downarrow\uparrow\downarrow>$. Additionally the spontaneous emission rate $\Gamma$ from both excited states was set to $(2\pi\tau)^{-1} = 250\text{MHz}$, where $\tau$ is the exciton lifetime. A coupling between the two excited states arises from co-tunneling and is simulated with $\gamma_1 = 0.001\Gamma$. To consider incomplete hole spin pumping, the two ground states are coupled with $\gamma_2 = 0.001\Gamma$. The resonant linearly polarized laser has a Rabi frequency of $\Omega_L = 0.1\Gamma$, which is below saturation and drives both transitions, $|\downarrow>\rightarrow|\downarrow\uparrow\downarrow>$ and $|\uparrow>\rightarrow|\uparrow\downarrow\uparrow>$. The detuning is given as $\delta$ in Fig. 4.11 (a). Because the emitted RF photons from the transitions $|\downarrow\uparrow\downarrow>\rightarrow|\downarrow>$ and $|\uparrow\downarrow\uparrow>\rightarrow|\uparrow>$ are measured, we simulate the RF spectra with the population of the excited states ($\rho_{33} + \rho_{44}$) multiplied with the spontaneous decay rate $\Gamma$. The simulated RF spectra for different rates $\Gamma_{pump}$ are shown in Fig. 4.11 (b). Without considering a pump laser the two peaks, which are split by $\Delta E_{os}$, are equally strong. Once the pump laser is considered, an asymmetry can be observed. In particular, the emitted RF photons from the excited state $|\uparrow\downarrow\uparrow>$ are more pronounced compared to $|\downarrow\uparrow\downarrow>$. This is due to the pump rate $\Gamma_{pump}$, which leads via cotunneling $\gamma_1$ to an increased population of the state $|\uparrow\downarrow\uparrow>$. Experimental data in Fig. 4.10 (b) show, that the asymmetry is partially cancelled at $B_x = 0.1\text{T}$. The reason is most likely a mixing of the ground states with a finite transverse magnetic field which corresponds to an increased rate $\gamma_2$.

Concerning the intermediate regime at $B_x = 0.45\text{T}$, four resonances would be expected due to the superposition of the Overhauser shift and the hole Zeeman splitting. It is clear that the strength of these resonances depends on the optical
4.6. Resonant measurements on a 25nm tunnel barrier sample

Figure 4.12.: Polarization dependence at $B_x=0.45\text{T}$. (a) Energy diagram of $X^+$ in a transverse magnetic field $B_x < B_{\text{crit}}$ for the special case $B_{n,x} = -B_x$. (b) Resonant Hanle measurement at $B_x = 0.45\text{T}$ for circular and linear resonant excitation.

selection rules. To gain a better understanding of the polarization of the optical transitions we once more consider the Hanle measurements. The main property of anomalous Hanle curves is the PL circular polarization conservation up to $B_{\text{crit}}$. This implies an electron spin polarization along the laser excitation axis ($z$-axis), resulting in circularly polarized transitions. This situation is visualized in Fig. 4.12 (b). It shows an energy level diagram for $X^+$, where the excited states are polarized along the $z$-axis and separated in energy by $\Delta E_{\text{os}}$. In contrast to the electrons, the holes are only influenced by the externally applied in plane field $B_x$. This leads to the eigenstates $|\downarrow\rangle - |\uparrow\rangle$ and $|\downarrow\rangle + |\uparrow\rangle$ which are separated by the hole Zeeman splitting. The polarization of the four possible transitions is given by the electron spin polarization of the excited state. Therefore, the transitions $|\downarrow\downarrow\uparrow\rangle \rightarrow (|\downarrow\rangle - |\uparrow\rangle)$ and $|\downarrow\uparrow\downarrow\rangle \rightarrow (|\uparrow\rangle + |\downarrow\rangle)$ should be $\sigma^+$ polarized while $|\uparrow\downarrow\uparrow\rangle \rightarrow (|\downarrow\rangle - |\uparrow\rangle)$ and $|\uparrow\uparrow\downarrow\rangle \rightarrow (|\uparrow\rangle + |\downarrow\rangle)$ should be $\sigma^-$ polarized. As a consequence a resonant measurement with a linearly polarized laser results in four resonances which are separated by the hole Zeeman splitting and $\Delta E_{\text{os}}$. These two energy shifts are most likely very similar in the measurement shown in Fig. 4.10 (b) at $B_x = 0.45\text{T}$. As a result, one large resonance arises from the two diagonal transitions and is flanked by two smaller ones.

To further confirm the polarization of the involved optical transitions, we performed a resonant measurement with circularly polarized light at $B_x = 0.45\text{T}$. RF spectra for a linear, $\sigma^+$ and $\sigma^-$ polarized laser excitation are shown in Fig. 4.12 (a) after subtraction of the background. While three resonances are observed for the linear RF spectra, only two resonances are measured for both circularly polarized excitations. The two resonances are separated by the hole Zeeman splitting. Due
to the strong variations in the background, we measured only a short detuning range of the laser. With additional measurements it was verified that there exist no additional resonances for $\sigma^+$ and $\sigma^-$ excitation. Moreover, the resonance positions between the different polarizations of the resonant laser cannot be compared. During the change of the RF polarization basis it is very likely to trap charges in the sample which lead to an energy shift. The signal to noise ratio differs for various polarizations which can be explained as follows. For a resonant $\sigma^-$ excitation, $\sigma^+$ polarized RF photons are detected and a large background is measured due to the mainly $\sigma^+$ polarized PLE emission. Therefore the signal to noise ratio for the $\sigma^-$ RF spectrum is worse than for the $\sigma^+$ RF spectrum in Fig. 4.12 (a).

One hast to keep in mind that for RF only the photons that have a polarization component orthogonal to the resonant laser polarization can be detected. According to the transitions plotted in the level diagram of Fig. 4.12 (b) no signal should be measured for $\sigma^+$ or $\sigma^-$ excitation. This situation changes if there is an interaction between the two excited states $|\downarrow\uparrow\downarrow>$ and $|\uparrow\downarrow\uparrow>$, such that an excitation with $\sigma^+$ and $\sigma^-$ can be followed by a decay of $\sigma^-$ and $\sigma^+$ polarized light, respectively. Such an interaction could arise due to a net field along the x-direction. Additionally, PLE experiments show a polarization conservation of only 60\% for $B_x = 0.45$T. Therefore, an electron spin flip has to take place either in the p- or in the s-shell of the QD, which further supports the hypothesis of an interaction between the two excited states $|\downarrow\uparrow\downarrow>$ and $|\uparrow\downarrow\uparrow>$. 
4.7. Discussion

By resonantly measuring nuclear spin polarization with RF in transverse magnetic fields we achieved on the one hand a higher spectral resolution and on the other hand a direct measurement of the $X^+$ transitions. Particularly, the polarization of the $X^+$ transitions as well as the population of the ground states can be determined. Due to the fact that only two resonances are obtained for circular RF spectroscopy at $B_x = 0.45\,\text{T}$ and because they exhibit a splitting of exactly $|g_h|\mu_B B_x$, we conclude that the electron in the excited state defines the polarization of the optical transitions. Hence the transitions $|\downarrow\uparrow\uparrow\rangle\rightarrow (|\uparrow\downarrow\downarrow\rangle - |\uparrow\uparrow\uparrow\rangle)$ and $|\downarrow\uparrow\downarrow\rangle\rightarrow (|\uparrow\uparrow\downarrow\rangle + |\downarrow\downarrow\downarrow\rangle)$ are $\sigma^+$ polarized while $|\uparrow\downarrow\uparrow\rangle\rightarrow (|\downarrow\downarrow\downarrow\rangle - |\uparrow\uparrow\downarrow\rangle)$ and $|\uparrow\uparrow\uparrow\rangle\rightarrow (|\uparrow\uparrow\downarrow\rangle + |\downarrow\downarrow\downarrow\rangle)$ are $\sigma^-$ polarized. This conclusion is also supported by the linear RF spectrum which shows that all four transitions contribute to the signal. The observation of circularly polarized optical transitions are fully consistent with a cancellation of the external field by $B_{\text{eff}}^x$, that in turn hinders the electron spin rotation.

To allow for a better understanding of the evolution of the nuclear spin polarization direction in a transverse magnetic field, $B_{\text{nuc}}^z$ and $B_{\text{nuc}}^x$ were extracted from the measurements shown in Fig. 4.8 (a) and plotted vs $B_x$ (Fig. 4.13). This extraction is only possible under the assumption that $B_{\text{nuc}}^z$ cancels completely the external magnetic field $B_x$ for circularly polarized transitions in $X^+$. Starting from $B_{\text{nuc}}^z=0.67\,\text{T}$ at $B_x = 0\,\text{T}$, the nuclear field is gradually decreasing to 0 at $B_x = 0.5\,\text{T}$. This is far below $B_x = B_{\text{crit}}$. On the contrary, the magnitude of the nuclear field along the x-axis is increasing. A complete cancellation of the applied magnetic field $B_x$ can be observed up to $B_{\text{crit}} = 1.7\,\text{T}$, where the nuclear spin polarization breaks down and where the Zeeman splitting that is expected for an experiment in Voigt geometry is restored. The magnitude of the nuclear field at $B_{\text{crit}}$ along the x-axis is roughly 2.5 times larger than the nuclear field along the z-axis for $B_x = 0\,\text{T}$. This means that the nuclear field does not only rotate into the xy-plane but also increase with increasing in-plane magnetic field.

Sallen et al. [84] explained the origin of the transverse nuclear spin polarization in Hanle experiments on droplet QDs with the Knight field. The magnetic field experienced by the nuclear spins is given by a superposition of the external transverse magnetic field and the Knight field. The total magnetic field is therefore slightly tilted towards the z-axis. Thus, the nuclear spin polarization has a contribution along the x- and the z-axis. This model was successfully applied to describe the line shape of Hanle curves in droplet QD [84]. For the case of droplet QDs the broadening of the Hanle curve is in the order of a few 10mT, which is exactly the range of the Knight field.

These broadening effects occur for one order of magnitude smaller values of $B_x$ than the anomalous Hanle effect repeated in this work for InGaAs QDs. Compared
Nuclear Spin Polarization in Transverse Magnetic Fields

Figure 4.13.: Nuclear field changes in $B_x$. Extracted nuclear field magnitude and direction versus transverse magnetic field $B_x$ from Fig. 4.10.

to droplet GaAs QDs, self assembled InGaAs QDs are heavily strained which leads with the associated nuclear quadrupolar interactions (QI) to a sizable $B_{nuc}^z$ [40, 89]. This strain is an inherent property of the self-assembled growth process which relies on the lattice mismatch between GaAs and InAs. As a result the QD strain gradient coincides predominantly with the QD growth direction (z-axis) [90]. Hence the QD experiences an electric field gradient along the z-axis. Due to the non spherical charge distribution of nuclear spins, they couple to the electric field gradient via quadrupolar moments (see chapter 3). The corresponding level separation leads with the relation $g_N\mu_N B_Q = \hbar \omega_Q$ to an effective magnetic field $B_Q$ of several 100mT [51, 91]. Particularly, in a small transverse magnetic field, the nuclear spins do not feel the presence of $B_z$ to first order. Therefore $B_{nuc}$ is always oriented along the z-axis. Such a static magnetic field along the growth direction can lead to a nuclear spin polarization along the x-axis as follows. The total magnetic field experienced by the electron is given by $B_{tot} = B_{nuc} + B_x$. Quadrupolar interactions stabilize the nuclear spin polarization along the z-axis, resulting in a tilted $B_{tot}$. Therefore the electron spin precesses around this tilted magnetic field. Via hyperfine interaction the electron spin polarization is transferred to nuclear spins, resulting in a nuclear spin polarization in the xz-plane. In addition, the polarization along the x-axis is further stabilized by $B_x$. Furthermore, the resulting $B_{nuc}^x$ compensates $B_x$ and reduces the total electron spin splitting which leads to a high dynamic nuclear spin polarization (DNSP) rate.

However, this model cannot be confirmed with the data shown in Fig. 4.13 because $B_{nuc}^z$ is below the measurement resolution of 0.1T for $B_x > 1T$. Probably the model is too simple to fully describe the influence of QI on the Hanle effect. The assumption of a strain gradient along the z-direction is only valid for a spatial average over
the QD. Most nuclei do not experience a strain gradient along the z-direction, which leads to a different behavior of the nuclear spin system in a transverse magnetic field. Not only the direction but also the strength of strain can vary within the QD. An exact description of various inhomogeneities might lead to a better understanding of the experimental data.

On the other hand, the abrupt jump of nuclear spin polarization along the x-axis together with electron spin polarization to zero in combination with a hysteresis could indicate an alternative view on the anomalous Hanle effect, namely the occurrence of a spontaneous polarization and phase transition. However, these results are quantitatively different from the theoretically predicted quantum phase transitions [80, 83], since an abrupt decrease of the nuclear spin polarization along the z-axis at $B_{\text{crit}}$ has not been observed. Even though there are a lot of similarities, the experimental system deviates in several points from the theoretical model. Especially, quadrupolar interactions and the inhomogeneous hyperfine interaction are strongly influencing nuclear spin dynamics and might lead to a different behavior than what is theoretically predicted. To rule out the influence of the QI, a study of strain free QDs should be considered that exhibit the same degree of electron spin polarization as in InGaAs QD which is around 80%. For GaAs droplet QDs only 40% electron spin polarization was achieved. This value might be improved with a quasi-resonant pumping [84]. It would be interesting to investigate Hanle measurements on GaAs droplet QDs with the same degree electron spin polarization as in InGaAs QD, to see if the nuclear spin evolution is closer to the theoretical predictions of phase transitions.
5. Coherence of Optically Generated Hole Spins

To realize a quantum bit a single hole spin in a semiconductor quantum dot has potential advantages over an electron spin. One major advantage of holes is that they have a strongly reduced hyperfine interaction with nuclear spins, which leads to less dephasing. We measured the coherence time $T_2^*$ and $T_2$ of an optically generated hole inside an InAs/GaAs QD embedded in an n-doped diode structure in time domain.

5.1. Introduction

Due to their relatively long lifetimes, semiconductor qubits are promising candidates for quantum information processing. Further interesting properties are their potential for integration on a chip [14, 92, 93] and ultrafast coherent optical control [12, 61, 94, 95]. In particular, electron spins confined in self-assembled InAs/GaAs QDs have been extensively studied as semiconductor qubits over the past years. However, such electron spin qubits strongly couple to the nuclear spin reservoir via hyperfine interaction (see chapter 4). The variations in the effective magnetic field that result from the fluctuating nuclear spins, induce electron spin precessions at various frequencies. As a consequence, reduced coherence times of $T_2^* = 1 - 2\text{ns}$ have been measured [61]. One method to work around the adverse effect of the slowly fluctuating effective magnetic field is the spin echo experiment. In such an experiment the phases of the spins are refocused. As a result, an increased dephasing time of $T_2 = 3\mu\text{s}$ can be achieved [61].

A completely different approach to realize a longer coherence time is to work with a hole instead of an electron. In contrast to an electron, the coupling to nuclear spins is at least one order of magnitude weaker for a hole [19, 44, 96–98]. Therefore, longer coherence times are generally observed. Two different methods were used in recent experiments to charge the QD with a single hole. While hole δ-doped samples are stochastically charged, embedding the QD layer into a
p-doped diode structure allows for a deterministic charging of the QD with holes. Both sample structures were studied by De Greve et al. [99], and they observed similar values for of $T_2^* = 2.3\text{ns}$ for both samples. Several measurements were performed on QDs embedded in a p-doped structure, exhibiting hole coherence times of $T_2^* = 20-100\text{ns}$ [11, 100, 101]. Is the latter method combined with a spin echo measurement, a coherence time of $T_2 = 1.1\mu\text{s}$ can be obtained [99].

One common disadvantage of p-doped samples is the broad $X^+$ line width of $\sim 7\mu\text{eV}$ as compared to the 2-3$\mu\text{eV}$ of n-doped structures. At the ultimate limit of lifetime broadening, the coherence time is equal to the radiative lifetime $\tau = 1\text{ns}$. This would correspond to an exciton line width of $\Gamma = \hbar/\tau \sim 1\mu\text{eV}$ for the investigated QDs. For high quality n-doped samples, usually optical line widths of twice this value are measured [15, 31, 65]. While this broadening can either be explained by enhanced charge fluctuations in the vicinity of the QD or nuclear spin fluctuations [26, 65], the larger value for the $X^+$ line width in p-doped samples is very likely due to charge fluctuations. With the lower mobility of p- compared to n-doped samples, the charge screening is less distinct leading to more charge fluctuations. According to De Greve et al. [99] this charge noise contributes to inhomogeneous dephasing which limits the coherence time $T_2^*$. Therefore prolonged coherence times should result for a method that combines the advantage of holes together with the reduced charge noise in n-doped samples. This is the focus of the presented work in this chapter.

The question still remains how to most accurately measure the coherence times. One possibility to determine $T_2^*$ is coherent population trapping (CPT) [102, 103]. The width and depth of the dip at the two photon resonance of a Λ system determines the coherence of its ground states. CPT was used for determining coherence properties of holes in QD with p-doped back contacts [11] and singlet triplet states in coupled QDs [104]. The main advantage of CPT is that it is based on spectroscopy and that there is no accurate control over optical pulses needed. Disadvantages are the limited accuracy of determining the depth of the CPT dip which is given by the signal to noise ratio [11] and that the method is restricted to determining $T_2^*$ times. Additionally, CPT on electron spin states is superimposed by dragging [40] which makes a distinction between nuclear spin effects and electron spin coherence properties very difficult.

Latest experiments are based on a more versatile tool to measure $T_2^*$ and $T_2$ in the time domain. Press et al. [12] were the first to achieve a complete coherent control over an initialized electron spin state in a quantum dot with picosecond optical pulses. Subsequently, de Greve et al. [99] demonstrated a successful implementation of the same measuring method for a single hole qubit on a p- doped sample.
In this chapter we present results from coherence time measurements that are performed for the first time on optically generated holes in n-doped samples. In analogy to Press et al. [12] we measure $T_2^*$ and $T_2$ in the time domain.

5.2. Spectroscopy on $X^+$

The sample structure consists of a single layer of self-assembled InGaAs QDs, which is embedded into an n-doped diode structure. This allows for a deterministic charging of the QD with single electrons. To perform optical spectroscopy on a positively charged QD, an additional laser is used to generate a positive charge. Further details on generating the positively charged hole by optical means are given in Chapter 2 of this thesis.

A typical resonance fluorescence (RF) spectrum of $X^+$ as a function of the energy detuning is shown in Fig. 5.1. From the Lorentzian fit we can extract an optical line width of $2.3\mu$eV. Such a narrow optical line width is characteristic for an InGaAs QD embedded into an n-doped structure, similar values have been measured for the $X^-$ transition in the same QD. In comparison, p-doped samples exhibit with $7\mu$eV [101] much broader optical line width on $X^+$. This broadening of the optical line width is most likely given by enhanced charge fluctuations in p-doped samples compared to n-doped structures, as lined out in the previous section. This charge noise contributes to inhomogeneous dephasing which limits the coherence time $T_2^*$. Therefore prolonged coherence times should result for optically generated holes in n-doped samples.
5.3. Initialization and Readout

Reliable initialization and readout mechanisms are a prerequisite for a coherent manipulation of a single hole spin. In the first part of this section the initialization of the hole qubit into the spin up state is discussed. For this purpose, hole spin pumping is used [15, 78]. The initialization mechanism is schematically shown in Fig. 5.2 (b). First, we apply a finite transverse magnetic field (Voigt geometry). This leads to a Zeeman splitting of the hole spin states. In Voigt geometry, the diagonal transitions $|\downarrow \uparrow \downarrow \rangle \rightarrow |\uparrow \rangle$ and $|\uparrow \downarrow \uparrow \rangle \rightarrow |\uparrow \rangle$ are allowed. After exciting the $|\downarrow \rangle \rightarrow |\downarrow \uparrow \downarrow \rangle$ transition with a laser, the decay takes place via the diagonal transition to $|\uparrow \rangle$ or back to the ground state $|\downarrow \rangle$ with equal probabilities. Since the pumping rate to $|\uparrow \rangle$ is larger than the decay rate of the hole spin, an initialization into the hole spin up state $|\uparrow \rangle$ can be achieved leading to a reduced absorption of the laser and here in a reduced RF signal. The sample used in this work exhibits incomplete spin pumping. This is why there is still 30% of the signal left. Experimental evidence that the diagonal transitions are allowed is given with a single laser RF measurement. Corresponding data is shown in Fig. 5.2 (a). Four transitions are visible in the color coded plot of RF count rate as a function of gate voltage and laser wavelength.

In contrast to $X^-$, spin pumping for holes is possible without any external magnetic field. This is due to the limited hyperfine interaction of holes with nuclear spins resulting in a larger pump rate compared to the hole spin relaxation time [78]. Due to the fact that diagonal transitions are allowed in Voigt geometry, hole

Figure 5.2: $X^+$ in Voigt geometry. (a) RF measurement of $X^+$ versus gate voltage and laser wavelength in an in-plane field of 4T (Voigt geometry). At the gate voltage indicated with A, Ramsey measurements are performed. The results are presented in section 5.7. (b) Energy level diagram for hole spin pumping. After resonantly exciting $|\downarrow \rangle \rightarrow |\downarrow \uparrow \downarrow \rangle$, the allowed diagonal transition in Voigt geometry, $|\downarrow \uparrow \downarrow \rangle \rightarrow |\uparrow \rangle$, leads to hole spin pumping.
spin pumping is even more efficient for finite magnetic fields and shows the same magnetic field dependence as for electron spin pumping [15, 105]. An advantage of this simple initialization process in Voigt geometry is the fact that it can be used as well for the readout. If the laser is resonantly applied to the $|↓\rangle \rightarrow |↓↑↓\rangle$ transition, the RF signal strength is proportional to the occupation of the ground state $|↓\rangle$.

The coherent oscillation between the two hole spin states $|↓\rangle$ and $|↑\rangle$ is measured as a projection onto the z-axis. The respective oscillation of the occupation in the ground state $|↓\rangle$ is therefore directly measured via the RF signal strength.

5.4. Rabi Oscillations

![Figure 5.3](image)

**Figure 5.3.: Scheme for Rabi oscillations.** (a) Level diagram in x basis for finite magnetic field $B_x$. A slightly detuned (210MHz) circularly polarized rotation pulse is driving all four transitions, which are linearly polarized in Voigt geometry. (b) Level diagram in z basis for finite magnetic field $B_x$. $B_x$ results into a Larmor precession of the hole spins around the x-axis (red arrow).

After initializing the hole spin state by optical pumping, a spin rotation (Rabi oscillation) is performed with a 4ps long, $\sigma^+$ polarized laser pulse. To avoid a population of the excited state, the laser is additionally detuned by $\Delta = 210$GHz. The effect of the laser pulse can be easily understood with the help of the schematic drawing in Fig. 5.3. In the x-basis the hole spin in the ground state is either parallel or antiparallel to the applied magnetic field $B_x$. All optical transitions are linearly polarized due to the optical selection rules in Voigt geometry. Because we use a circularly polarized laser pulse, all four transitions are driven (Fig. 5.3 (a)). Now we consider the same situation in the z-basis (Fig. 5.3 (b)), where the hole spin orientation is along the growth direction. In this case the two allowed optical transitions are circularly polarized, which means that $\Omega_{\text{rot}}$ drives only one
optical transition. Additionally, the Zeeman splitting due to $B_z$ results in coherent mixing of the ground and excited states. After the initialization of the hole spin in $|\uparrow\rangle_z = e^{i\phi/2} |\uparrow\rangle_z + e^{-i\phi/2} |\downarrow\rangle_z$, the laser pulse induces an ac-Stark shift of $|\uparrow\rangle_z$ relative to $|\downarrow\rangle_z$. Thereby, a relative phase shift $\phi$ occurs, that is proportional to the laser pulse duration and the laser power $P_{\text{rot}}$. In other words, a laser pulse is equivalent to an effective magnetic field along the z-axis which induces coherent oscillations between $|\uparrow\rangle$ and $|\downarrow\rangle$. (If not indicated with an index, $|\uparrow\rangle$ and $|\downarrow\rangle$ are in the x-basis.)

Due to the fact that every hole spin state can be represented by a superposition of the states $|\downarrow\rangle$ and $|\uparrow\rangle$ and their relative phase, spin manipulations are best visualized on a unit sphere called Bloch sphere. A schematic drawing of the Rabi oscillations on the Bloch sphere is shown in Fig. 5.4 (a). After the initialization in the $|\uparrow\rangle$ state, a rotation of the hole spin around the z-axis by an arbitrary angle can be achieved if the rotation pulse power $P_{\text{rot}}$ is chosen correspondingly.

To determine the laser pulse power corresponding to a $\pi$ pulse, the emitted RF photons are measured according to the readout scheme as described above. Fig. 5.4 (b) shows the oscillating RF count rate as a function of the normalized laser power $P_{\text{rot}}$. As a consequence of the applied readout scheme, the maximal number of photons indicates the maximal occupation of the $|\downarrow\rangle$ state. Because the experiment is initialized in the $|\uparrow\rangle$ state, the laser power at the maximal count rate corresponds to a $\pi$ rotation on the Bloch sphere. As a result of the previously mentioned imperfect hole spin pumping, the visibility of the Rabi oscillation is

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**Figure 5.4.: Rabi oscillations.** (a) Visualization of Rabi oscillations on the Bloch sphere. The dark red arrow indicates the initialization in the $|\uparrow\rangle$ state. With the laser power, the rotation angle between $|\uparrow\rangle$ and $|\downarrow\rangle$ can be controlled (light red arrows). (b) Rabi oscillations between the hole spin states are observed in the oscillating RF signal as the rotation pulse power is increased.
reduced. This explains that the minimal observed photon number is 250cts/0.1s above the dark count rate of the APD. A reduced visibility, however, has no influence on the accuracy of the determination of $P_\pi$. The accurate control over the rotation angle between $|\uparrow\rangle$ and $|\downarrow\rangle$ allows to conduct Ramsey experiments and hence the determination of the coherence time $T_2^*$. 

5.5. Ramsey measurements

Figure 5.5.: Ramsey scheme. (a) After initialization in the $|\uparrow\rangle$ state, a $\pi/2$ pulse is applied. (b) The in plane field leads to a rotation of the hole spin around the vertical axis at the Larmor frequency. (c) The rotation is stopped after a delay time $\tau$ with a second $\pi/2$ pulse. Depending on the phase evolution, the $\pi/2$ pulse brings the spin back to its initial state $|\uparrow\rangle$ or to the opposite spin state $|\downarrow\rangle$.

With the Rabi experiments in the previous section, we demonstrated that a rotation of the hole spin by an arbitrary angle around the z-axis is feasible. To have a full control over the Bloch sphere, a rotation around a second axis is needed. With the in plane magnetic field causing a Larmor precession of the hole spin, a rotation around the x-axis is established. This effect is exploited for Ramsey interferometry. In a first step after initialization, a $\pi/2$ pulse is applied, that brings the hole spin to the equator of the Bloch sphere. During the subsequent delay time $\tau$ the spin rotates around the x-axis at the Larmor frequency. The phase evolution determines the probability of bringing the spin back to either the initial state or to the opposite spin state with a second $\pi/2$ pulse applied after the time $\tau$ (Fig. 5.5). Finally, the measurement of the spin population as a function of the delay time $\tau$ leads to a sinusoidal oscillation of the RF counts at the Larmor frequency (Ramsey fringes). The effective magnetic field causing the Larmor precession varies slightly in time and so does the precession frequency. Therefore, averaging that is needed to obtain a reasonable signal strength leads to an exponential decay of the Ramsey fringes with a time constant $T_2^*$.

To experimentally achieve two $\pi/2$ pulses separated by a delay time $\tau$, the laser
beam is split into two optical paths A and B (see Fig. 5.6). Practically, there are two different possibilities to control the delay time $\tau$. The first one comprises a constant prolongation of the optical path A with an optical fiber. The second one delays the optical path A continuously by means of a translation stage. Discrete steps of the delay time represent measurements performed with inserted fibers of different lengths. Continuous change of $\tau$ over an interval of 2.5ns is realized with a translation stage.

At a gate voltage of -1.113V corresponding to point A in Fig. 5.2 (a) a decay of the oscillation amplitude can be observed at $\tau = 20$ns, which is followed by a revival of the signal at $\tau = 35$ns. Ramsey fringes for $V_g = -1.13$V before and after the signal has recovered are shown in Fig. 5.7 (b). In both cases a sawtooth shape of the fringes can be observed. For a further investigation of the strong amplitude modulation of the Ramsey fringes, measurements have been performed at $B_x = 4 T$ and for delay times up to $\tau = 400$ns. To extract the amplitude of the Ramsey oscillations, a Fourier transformation is carried out. The resulting fringe amplitudes at the Larmor frequency are plotted as a function of the delay times in Fig. 5.8. Data with similar amplitude modulations for the Ramsey fringes have been observed by Carter et al. [101]. They attribute this modulation to nuclear spin interactions and propose a theoretical model that predicts an envelope modulation for the Ramsey fringes of twice the $^{115}$In precession frequency of 75MHz. They argue that even though there are several other isotopes present leading to nuclear spin effects, $^{115}$In is dominant due to the large nuclear spin of 9/2 and its high abundance. We applied this model to fit the data in Fig. 5.8 with an exponentially decaying cosine function. An amplitude modulation frequency of 74.6MHz and a decay time of $T_2^* = 240$ns are extracted.

**Figure 5.6.:** Delay time control.
Figure 5.7.: Ramsey fringes. (a) Ramsey fringes measured for two different transitions in $X^+$, shown in Fig. 5.2 (a). A decay and revival of the oscillation amplitude can be observed. (b) Magnification of the of a 1.5ns delay window highlighted with a red box in (a), to display the Ramsey fringe shape. After the revival of the oscillation amplitude, at $\tau = 36$ns, oscillations can still be observed with almost unchanged visibility.

The modulation frequency matches within the measurement uncertainty twice the expected $^{115}$In precession frequency of 75MHz. Not only the amplitude modulation but also the sawtooth pattern of the Ramsey fringes can be explained with nuclear spin polarization as follows [101]. Because a different polarization is achieved for each delay time, the Overhauser shift changes the optical transition relative to the laser. Therefore, the initialization and measurement process is disturbed, resulting in the observed sawtooth pattern of the Ramsey fringes. Similar asymmetric patterns were observed in Ramsey experiments with electron spins. [106, 107].
5.6. Spin Echo measurements

As it was discussed earlier, $T_2^*$ can be limited due to inhomogeneous dephasing that arises from slow fluctuations in the nuclear spin polarization as well as from charge fluctuations. To circumvent such slow sources of decoherence, spin echo measurements are carried out.

The advantage of the spin echo measurement over Ramsey experiments is explained by means of the schematic illustrations in Fig. 5.9. After initialization of the hole state, a $\pi/2$ pulse is applied. The effective magnetic field varies for consecutive measurements. Some spins precess slower around the vertical axis because of the smaller field strength (green arrow) and some spins precess faster due to a larger field strength (blue arrow). By applying a $\pi$ pulse after a time delay $T$, the slower spins are now ahead of the faster precessing spins. Complete refocusing takes place after a time delay $T$. Depending on the phase evolution, the $\pi/2$ pulse brings the spin back to its initial state or to the opposite spin state.
5.6. Spin Echo measurements

Figure 5.10.: Spin echo measurements. Spin-echo signal as the time offset $\tau$ is varied, for a time delay of $2T = 1467\text{ns}$, $630\text{ns}$, $240\text{ns}$ from top to bottom. Magnetic field $B_x=4$ T.

spin into $|\uparrow\rangle$, a $\pi/2$ pulse is applied. As lined out before, the effective magnetic field and therefore the precession frequency around the x-axis are slightly different for consecutive measurements. This leads to different precession frequencies in the horizontal plane of the Bloch sphere, as indicated with colored arrows in Fig. 5.9 (b). To rephase the system, an additional $\pi$ pulse is applied after a delay time $T$. Similar to the Ramsey measurements, a second $\pi/2$ pulse is then applied and the spin population is measured as a function of the time offset $\tau$ [60, 61]. This measure to rephase the spins in different realizations of the experiment is effective, as long as the fluctuations of $B_{\text{eff}}$ are slow compared to $T$.

The spin-echo signal for three different time delays $T$ is shown as a function of the time offset $\tau$ in Fig. 5.10. Between a time delay of $T = 204\text{ns}$ and $T = 630\text{ns}$, only very little decay in the amplitude is observed. For the longest time delay at $T = 1467\text{ns}$ only 10% of the signal amplitude remains. Even though there are not enough data points for fitting an exponential decay to the signal amplitudes, the
5.7. Discussion

We demonstrated experimentally that ultrafast coherent control techniques work as well for optically generated hole spins as for electron spins in n-doped samples. From Ramsey measurements we extracted a $T^*_2$ time of $240\text{ns}$ for optically generated hole spins. This is a 1-2 order of magnitude larger value than what was measured with the same method for hole spins in p-doped samples ($T^*_2 = 2.3 - 19\text{ns}$) [99, 101]. As discussed in the introduction the $T^*_2$ time is the longer, the less pronounced the effects of inhomogeneous dephasing are. In other words, n-doped samples exhibit either less charge noise or weaker fluctuations in the nuclear spin polarization. A comparison of the optical line widths in $X^+$ reveals for n-doped samples 2-3$\mu\text{eV}$ which is a much smaller value than the measured 7$\mu\text{eV}$ for p-doped structures. This large difference can only be caused by charge fluctuations. For that reason smaller charge induced spectral diffusion does most likely lead to a longer $T^*_2$ coherence time in n-doped samples compared to p-doped structures. To summarize our results, the method of optically generating holes in QDs brings clear advantages over conventional techniques.

By contrast, the measured spin echo time $T_2$ for optically generated holes ($T_2 = 1\mu\text{s}$) and p-doped samples ($T_2 = 1.1\mu\text{s}$) are comparable. Most interestingly also the $T_2$ times for electrons are with $3\mu\text{s}$ in the same range. Based on dipole dipole interactions of nuclear spins Witzel et al. [108] and Yao et al. [109] predict $T_2 \sim 1 - 10\mu\text{s}$ for a single electron in a QD. In self-assembled QDs this nuclear spin diffusion is negligible because of the quadrupolar interactions. Recent experiments revealed that electron mediated nuclear spin diffusion and co-tunneling rather limit the $T_2$ time [79]. The measured values for $T_2$ are rather surprising, since the weaker interaction of hole spins with nuclear spins compared to electrons should result in a longer $T_2$ value. Hence, the limiting factor for the spin echo time of holes cannot be nuclear spin effects. For p-doped samples, it is very likely that charge induced spectral diffusion, which already limits $T^*_2$, sets an upper boundary for $T_2$ as well. Regarding charge noise, optically generated holes in n-doped samples are less affected which should result, compared to p-doped structures, in a longer spin echo time. Because such a behavior is not observed, it is very probable that the hole generating process with a weak off-resonant laser is limiting the coherence properties. To test this hypothesis, hole spin lifetime measurements should be carried out as a next step.

As expected from the weaker hyperfine interaction of holes with nuclear spins compared to electrons in InGaAs QDs [19, 44, 96–98], the measured decoherence
time $T_2^* = 240\text{ns}$ is longer than for electrons ($T_2^* = 1 - 2\text{ns}$) [61]. The hole spin decay is dependent on the orientation of the external magnetic field. This arises from the simple Ising form $\sum_k A_h^k I_z^k s_z$ that couples the hole to the nuclear spin system, where $A_h^k$ is the coupling of the $k$th nucleus and $I_z^k$ and $s_z$ denote the $z$ component of the $k$th nuclear spin and the hole spin, respectively. In the case of a transverse magnetic field $B_x$ the nuclear spin fluctuations are perpendicular to the applied field $B_x$, resulting in a slow power-law hole spin decay which is estimated to be on the order of µs. In comparison for zero field or an external field $B_z$ the decay is Gaussian, similar to electrons, with timescales of tens of nanoseconds [19]. Hence, the measured two orders of magnitude longer $T_2^*$ for holes compared to electrons can be explained with the weak coupling of holes to the nuclear spin system in transverse magnetic fields.

Even though the interaction between holes and nuclear spins is weak, a strong amplitude modulation of the Ramsey interference fringes is observed. This is a result of nuclear spin polarization. It changes the optical transition relative to the laser via the Overhauser shift leading to an amplitude modulation and a sawtooth shape of the Ramsey interference fringes [101].

In addition to the results we presented above, our work gives rise to a number of further investigations. For instance, the fact that n-doped samples allow to charge the same QD either with an electron or with a hole could be exploited to directly compare the coherence behavior of both species. This might give new insights into the interaction of electrons and holes with nuclear spins.

For several interesting experiments it is crucial to achieve a long coherence time. In particular we have shown that optically generated holes in QDs are promising to further prolong the spin echo time $T_2$. With their work on quantum teleportation between a photon and a single electron spin qubit, Gao et al. [107] show in which direction research might head. Their experiments in combination with longer coherence time could lead to a realization of spin spin entanglement.
Appendix A

A. Appendix

A.1. Experimental Setup

With a confocal microscope [110] resonant spectroscopy was performed. On the one hand this allows for a small spot size of $\sim 1\mu m$ which is crucial for single QD spectroscopy, on the other hand a large amount of light emitted from the QD can be collected with a large numerical aperture (NA) of the objective. The numerical aperture $NA \propto \sin\theta \sim \theta$ is proportional to the half angle $\theta$ of the light cone from a lens that is illuminated with a plane wave. Therefore a higher NA increases the collection efficiency. We performed all experiments with the lens L1 (see Fig. A.1) that exhibits an NA of 0.68. Additionally, we used a solid immersion lens (SIL) to increase the collection efficiency of the emitted light from the sample surface [111]. A four times increased collection efficiency was experimentally observed by comparing the same sample with and without SIL.

To position the QD into the focal spot, the sample is mounted on piezoelectric xyz-positioners. The described microscope together with the positioners and the sample are placed inside a bath cryostat which reaches temperatures of 1.4K. Such low temperatures are necessary to prevent temperature broadening of the QD resonances. Magnetic fields up to 12T can be applied with a superconducting split coil magnet. This geometry allows for an optical access of the sample along two different axes. Therefore upon rotation of the sample, a magnetic field transverse (shown in Fig. A.1) or along the growth direction can be applied.

For PL measurements the QD is excited with diode laser at 780nm, the emitted photons from the QD are dispersed with a spectrometer (SP) of 75cm focal length and finally analyzed with a liquid nitrogen cooled, back-illuminated CCD array. To perform resonant spectroscopy or PLE, two tunable diode lasers (Toptica DL pro) were available with a tuning range from 915nm to 990nm. The required intensity stabilization for resonant measurement is realized with a double-pass AOM setup [112], before the lasers are coupled into a fiber. While the $dT$ signal is directly measured with the photodiode PD1, the $dR$ signal is first coupled into a single mode fiber (CF) before it is measured on a photodiode. The additional single mode fiber allows for a mode filtering, which is especially needed in RF measurements. In both cases, $dR/R$ and $dT/T$, after passing a preamplifier, the signal is sent to a Lock-
In amplifier. The Lock-In amplifier is synchronized with a function generator that produces a square wave modulation with a DC offset which is applied to the gate of the sample. Because of the Stark-shift, the QD transition is modulated in and out of resonance. This technique is called Stark-shift modulation [31, 71] and allows to suppress noise on all frequencies except the modulation frequency which is usually at 777Hz.

Polarization control is needed to perform resonance fluorescence (RF) measurements. By choosing the polarizer P2 cross-polarized to the polarizer P4 in front of the single mode collection fiber (CF) an extinction ratio of $\sim 10^6$ can be achieved. For RF measurements it is very difficult to optimize the QD position by monitoring the signal, because every change of the sample position leads to an increase of the background. To align the setup and to monitor the QD position a CCD camera (C) was used.

Hanle measurements were performed with a polarization control consisting of the two polarizers P2 and P3 together with the liquid crystal devices (LCW, LCR). The light polarization at the QD is determined by the fixed vertical polarizer P2 and the liquid crystal waveplate LCW, that allows for a retardance between $\lambda/4$ and $3\lambda/4$. The reflected PLE laser photons from the QD experience the same retardance and are therefore linearly polarized after the LCW. To distinguish between co- and cross polarized QD photons a liquid crystal rotator in combination with a linear polarizer P3 were used. To filter out the strong pump laser in PLE experiments a longpass filter (LP) at 950nm is needed. The typical broad lasing background of a diode laser is filtered out with a short pass (SP) at 940nm, such that it is not overwhelming the QD emission at $\sim 960$nm.

For a resonant measurement of the Overhauser shift an independent polarization control of the pump laser La2 and the probe laser La1 is needed, therefore we placed the LCW directly after the resonant laser (see Fig. A.1). In combination with the fixed vertical polarizer P1, a complete polarization control of the resonant laser is achieved. With an additional $\lambda/4$ plate, the pump laser is circularly polarized that allows for polarizing nuclear spins in the QD. Besides the long pass filter, a grating (G) is needed in front of the collection fiber to filter out the strong pump laser.

The creation of the laser pulses necessary for the resonant measurement of the Overhauser shift was performed either with an acousto-optical modulator (AOM) from Crystal Technology, model no.3080-125 or with an electro-optical modulator (EOM) based intensity modulator from Jenoptik AM940b. In both cases the laser pulses were fiber coupled to separate the alignement from the measurement setup presented in Fig. A.1.
Figure A.1.: Measurement Setup. Schematics of the setup used for the experiments in chapter 4. The elements drawn with dotted lines are optional, a detailed description is given in the text. A list of the different components is shown in Table A.1.
<table>
<thead>
<tr>
<th>Item</th>
<th>Description</th>
<th>Manufacturer</th>
</tr>
</thead>
</table>
| Sp   | Spectrometer | Princeton Instruments/Acton Research, SpectraPro 2759, f= 0.750 m  
Princeton Instruments, Spec-10:100BR/LN. 1340×100 pixel back-illuminated CCD array |
|     | Detector    |              |
| C    | CCD camera  | Watec, WAT-120N |
| FM   | Flip mirror | Thorlabs, BB1-E03 |
| P₁, P₂, P₃, P₄ | Polarizer | Thorlabs LPNIR050 |
| LCR  | Liquid crystal polarization rotator | Meadowlark, LPR-200-0915 |
| Sh   | Shutter     | Uniblitz, LS6ZM2 |
| LCW  | Liquid crystal wave plate | Meadowlark, LRC-200-0915 |
| L1   | Achromatic lens | Thorlabs, AC050-008-B-ML, NA=0.68 |
| SIL  | Solid immersion lens | A.W.I. Industries. hemispherical ball lens, ZrO₂, uncoated. |
| S    | Sample      | Described in detail in chapter 2 |
| PP   | Piezoelectric positioners | Attocube, ANPx50/LT and ANPz50/LT. Controller ANC150/3 |
| CR   | Bath cryostat | Oxford instruments, Spectromag with 12T split-coil magnet |
| G    | Grating     | Ibsen Photonics, FSTG-NIR1500-903nm |
| La₁, La₂ | Tunable diode laser 1 (for PLE, dR, RF) | Toptica DL pro, coarse tuning range 915-990nm, mode-hop free tuning range 20GHz, Pₘₐₓ = 40 mW  
Toptica DL pro |
|      | Tunable diode laser 2 (for imaging and PL) | Melles Griot, 56IC5210, λ = 780 nm, Pₘₐₓ = 50 mW |
| CF   | Collection fiber (for RF and dR) | Thorlabs P3-830A-FC-5 |
| SP   | Short pass filter | LC-3RD/940SP |
| LP   | Long pass filter | LC-3RD/950LP |

**Table A.1.** Detailed list of the most important elements used for the setup sketched in Fig. A.1
B. Bibliography


First of all, I would like to thank Atac for giving me the opportunity to do my PhD in his group. He let me accomplish many interesting and challenging experiments, broadened my horizon with several theoretical simulations and let me explore the world of quantum optics with a lot of freedom. I am very thankful also to Prof. K. Ensslin for investing his valuable time in co-examining this work.

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List of Figures

2.1. Growth of self-assembled QDs. .............................................. 7
2.2. Confinement in an InGaAs QD. ........................................... 8
2.3. Field effect structure. ...................................................... 10
2.4. Charging n-doped samples with a single hole. ......................... 11
2.5. Charging diagram measured in PL. ....................................... 12

3.1. dT/T measurement for $X^-$. ............................................. 23
3.2. Optical detection of nuclear spin polarization in $X^+$. ............... 25
3.3. Comparison between dR and $\Delta E_{OS}$. ............................. 27
3.4. Nuclear spin dynamics measured with p-shell emission. ............... 28
3.5. Measurement scheme for pump probe with dR. ......................... 30
3.6. dR measurement with software based Lock-In amplifier. .............. 31
3.7. Pump probe measurement with dR. ....................................... 32
3.8. Pump probe measurement with dR for different $T_{\text{wait}}$............. 33
3.9. Pump probe measurements with RF. ..................................... 35

4.1. Scheme illustrating Hanle measurements. ............................... 39
4.2. Anomalous Hanle effect. .................................................. 41
4.3. Hanle measurements for different nuclear spin polarizations. ....... 42
4.4. Emission at the s-shell. .................................................... 43
4.5. Hanle measurements for different sample structures .................. 44
4.6. RF measurements sample A. .............................................. 46
4.7. Gate modulation at different frequencies. ............................... 47
4.8. CW RF measurement sample B. .......................................... 49
4.9. Extracted positions of the resonances. ................................ 50
4.10. Transition between Overhauser shift and Zeeman splitting. ......... 51
4.11. Transition between Overhauser shift and Zeeman splitting. ......... 52
4.12. Polarization dependence at $B_x=0.45T$. .............................. 53
4.13. Nuclear field changes in $B_x$. ........................................... 56

5.1. Optical line width of $X^+$. ............................................. 61
5.2. $X^+$ in Voigt geometry. .................................................. 62
5.3. Scheme for Rabi oscillations. ............................................ 63
5.4. Rabi oscillations. ............................................. 64
5.5. Ramsey scheme. ............................................. 65
5.6. Delay time control. ........................................... 66
5.7. Ramsey fringes. ............................................. 67
5.8. $T_2^*$ measurement. ........................................ 68
5.9. Ramsey scheme. ............................................. 68
5.10. Spin echo measurements. ................................... 69

A.1. Measurement Setup. ........................................... C