Optical Study, Preparation and Measurement

of a Single Quantum-Dot Spin

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Preface

When I started my PhD in the Quantum Photonics Group some time ago and the first gatesweeps were completed I thought a little bit naively that this spin quantum jump project that we were working on would actually be finished quite soon; the only problem occupying us was — as I thought — our measurement speed and therefore it was of purely technical nature. In particular, there was no reason why we should not be able to solve it and a period of frustration began after working on this technical problem without success. Luckily, this period passed after we found out about spin pumping, teaching us that physics really works but sometimes it takes its time. After all a spin quantum jump measurement, which is still to be shown, is also a technical challenge but more importantly at that moment we had not yet understood the physics behind. When the insight came, it lead to a possible solution given by Faraday rotation. Looking backwards, from these experiences I have learned to always stay prepared for surprises — and it were in particular these surprises that made these experiments exciting.

As the title suggests, this thesis has its focus on the preparation, the study of the dynamics and the measurement of a single spin confined in a quantum dot. All measurements are performed by optical means which places them at the interface between quantum optics and solid-state physics; further, quantum dot spins have become interesting for the field of quantum information processing [1][2][3][4] due to their potential scalability. 20-msec relaxation times in the same type of quantum dots [5] and 170-msec relaxation and 10-μsec decoherence times in similar dots [6-8] have been demonstrated. Together with the possibility of using ultrashort laser pulses for gate operation this system could allow many operations within a decoherence time. Further, coherent manipulation of singlet-triplet states, on similar quantum dots, has been demonstrated [8, 9].

Chapter 1 is an introduction to quantum dots starting out with the materials and optics aspect and concluding with an experimental as well as theoretical description of the transmission measurement technique used throughout this work. Chapter 2 introduces a method to prepare a single confined quantum dot spin with high fidelity; the underlying physical mechanisms as well as the spin dynamics are discussed and
complemented by further measurements in Chapter 3. Chapter 4 is devoted to the measurement of spin using Faraday rotation and an outlook is given in Chapter 5 which indicates possible experimental paths for the continuation of the presented work, in particular the $N$-shot measurement.
Summary

In this dissertation the fundamental physical system of a single spin confined in a self-assembled InAs/GaAs quantum dot is studied. To achieve control over the number of electrons in the quantum dot, a Schottky heterostructure is used. It is demonstrated that spin pumping into the spin-up or spin-down state using a resonant laser can be achieved with a fidelity exceeding 99.8%. This corresponds to an initialization of the spin state and is therefore an important result for the use of quantum dot spins in the field of quantum information processing. Further, it is shown that the time-averaged spin state can be measured via spin-dependent Faraday rotation of a far-off resonant laser field. For this experiment the spin is fixed in one of its two states using a weak resonant state preparation laser. In addition, the underlying physical mechanisms that determine the dynamics of the single spin are investigated; fundamental spin-reservoir interactions are hyperfine interaction with a nuclear spin ensemble, exchange coupling with a nearby Fermi sea and coupling to the phonon bath.

All experiments are performed using high-resolution transmission spectroscopy. In this scheme, the spin-dependent absorption of laser light due to the quantum dot optical transitions is measured. To study spin dynamics we measure the degree of resonant absorption which results from a competition between optical spin pumping induced by the resonant laser field and spin relaxation induced by the spin-reservoir interactions. We show that the strength of spin relaxation generated by the three fundamental interactions can be changed by up to five orders of magnitude upon varying the applied electric and magnetic fields.

This work paves the way for an all-optical single-shot measurement as well as the coherent manipulation of a single quantum dot spin.

Alle Experimente werden durchgeführt mit Hilfe von hochauflösender Laser-Transmissionsspektroskopie. Dabei wird die durch die spinabhängigen optischen Übergänge des Quantenpunktes induzierte Absorption von Laserlicht gemessen. Insbesondere können hierdurch Informationen über die Spindynamik durch die Stärke der gemessenen Absorption gewonnen werden, die durch ein Gegenspiel von einerseits optischem Spinpumpen und andererseits Spinrelaxation bestimmt wird. Wir demonstrieren, dass die Spinrelaxationszeit, die durch die drei genannten fundamentalen Wechselwirkungen bestimmt wird, bei geeigneter Wahl des angelegten elektrischen Feldes und Magnetfeldes um fünf Größenordnungen verlängert werden kann.

Die hier präsentierten Ergebnisse ebnen den Weg für eine Single-shot Spinmessung sowie für die kohärente Manipulation eines Quantenpunkt-Spins.
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1 Introduction

In the last decades there has been a lot of interest in the study of low-dimensional nanostructures. Enabled by the development of sophisticated growth techniques such as molecular beam epitaxy (MBE) and chemical vapor deposition (CVD), and nano-fabrication commonly used in semiconductor device production, the fabrication and study of quantum well heterostructures (2D), nanowires (1D) and quantum dots (0D) was made possible. In a quantum dot (QD) carriers are confined in three dimensions on nanometer scales. This lab realization of the particle-in-a-box textbook example, which generations of physicists were studying in their undergraduate courses, results, as the example dictates, in a quantization of the particles’ energies.

There are certainly different paths to impose a potential that varies over small enough length scales. The results in this thesis have been obtained with MBE-grown self-assembled InAs/GaAs QDs, which is probably the most studied type of self-assembled QDs. These QDs formed by a strain-induced process and the confinement is determined by the large bandgap difference between InAs and GaAs. In particular, there is confinement for electrons and holes. Among the family of self-assembled QDs, also other materials such as CdSe/ZnSe can be used instead of InAs/GaAs.

An approach of a three-dimensional electron confinement that allows for maximum in-situ control of the QD properties, i.e. shape of the confinement, leads to electrically (lithographically) defined QDs. Here, gates are fabricated on top of a two-dimensional electron gas (2DEG), which can be controllably depleted under the gates when a voltage is applied. By means of these gate electrodes a confinement with a typical quantization energy of \(\sim 1\text{meV}\) is created. This type of QDs is usually studied by transport experiments. Further, there are chemically synthesized colloidal QDs, mostly based on CdSe/ZnS.

In the following we will focus on the basic electronic and optical properties of our QDs.
1.1 Growth of self-assembled quantum dots

As already mentioned, the fabrication of self-assembled InAs/GaAs QDs has been enabled by the development of MBE growth, which allows for controlled deposition on the scale of atomic monolayers. Further, the material composition can be changed with the same resolution.

After such a change in material composition the new material initially grows at the atomic positions i.e. with the same lattice constant as the host material and a wetting layer is formed as depicted in Fig. 1.1 (left). The larger the mismatch of lattices constants between the two materials, the more strain is built up during growth, eventually leading to a release of strain. This means that the atomic positions are reorganized in order to form a configuration with lower energy. In general the strain release process leads to dislocations and defects. In contrast, growth is called coherent if strain is released without abrupt deformations of the lattice. This growth mode is called Stranski-Krastanov growth and produces droplets of InAs on the GaAs substrate after deposition of ~1.6 monolayers [Fig.1.1 (middle)].

In a final step, these islands are partially covered by GaAs before the sample is heated up in order to partially remove In atoms from the top of the islands. This partially covered islands (PCI)-technique leads to a blue-shift of the QD optical transitions to a regime where Silicon detectors are efficient. The QDs are then covered with a ~150nm thick GaAs layer in order to spatially separate the QDs from surface states which are formed at the GaAs/vacuum interface.

As the Indium source is placed at an angle to the substrate, a density gradient of QDs across the wafer is produced. This gradient is quite steep and can be observed in a photoluminescence image, as shown in Fig.1.2. Here, the sample has been moved
in steps of \( \sim 1.5 \text{mm} \). The black area on each of the three snapshots is approximately \( 200\mu\text{m} \times 170\mu\text{m} \).

![Snapshots of QD photoluminescence at different positions along the density gradient, taking steps of \( \sim 1.5 \text{mm} \).](image)

**Figure 1.2:** Snapshots of QD photoluminescence at different positions along the density gradient, taking steps of \( \sim 1.5 \text{mm} \).

The QDs are formed on the substrate at random positions; further, the exact QD properties such as shape and strength of confinement can only be determined with an uncertainty during the growth process; this randomness leads to an inhomogeneous broadening of \( \sim 40 \text{meV} \) in the QD emission wavelength, which is huge compared to the narrow radiative linewidths of \( \sim 2 \mu\text{eV} \). There are efforts to control the QD nucleation points; however, to date the best spectral properties (such as narrow lifetime-limited linewidths) have so far only been achieved with QDs grown at random sites.

### 1.2 Confinement and Coulomb renormalization

Since the QDs are lens-shaped with height \( \sim 5 \text{nm} \) and diameter \( \sim 20 \text{nm} \), they exhibit strong confinement along the growth direction and weaker confinement in the plane. Within a harmonic oscillator approximation the \( z \)-quantization energy largely exceeds the in-plane quantization energy such that the QD can be considered to be always in the state with the lowest principal quantum number. For the QDs used in our experiments, the quantization energy of the weaker in-plane confinement for electrons (holes) is \( \hbar \omega _ z = 15 \text{ to } 20 \text{ meV} \) (\( \hbar \omega _ h = 10 \text{ to } 15 \text{ meV} \)).

Information about the lowest-lying optical transitions in a QD can be obtained experimentally using photoluminescence (PL) techniques, which have been widely used in the field of QD spectroscopy within the last decade. In this measurement, a pump laser typically generates free electrons and holes in the bulk semiconductor matrix. Part of these carriers see the QD potential and relax to lower energy levels on timescales \( \tau \sim 40 \text{ps} \) due to carrier-phonon and carrier-carrier interaction. After reaching the QD ground state, further relaxation i.e. electron-hole recombination
can only take place after a rather long radiative lifetime \( \tau_{rad} \sim 0.8 \text{ns} \), which in turn determines the linewidth of the photon that is emitted. For this to happen, all processes that lead to non-radiative decay of the confined exciton i.e. a bound electron-hole pair have to be absent. This is not the case in general, but essentially holds for our type of QDs.

The previously described PL process is depicted in the upper sequence of Fig. 1.3. If, in addition to an exciton, another carrier e.g. an electron is present in the QD, a charged complex is formed as shown in the lower sequence of Fig. 1.3. This is referred to as singly-charged exciton, trion or \( X^{1-} \). The energy of this three-particle state then becomes renormalized due to Coulomb interaction, which leads to a shift of the energy of the emitted photon with respect to the neutral exciton \( X^0 \) [10]. In particular, the emission energies of the charged excitonic complexes are unique for each configuration. These Coulomb effects are on the energy scale of \( \sim 1 \text{ to } 5 \text{ meV} \) i.e. weaker than the quantization energy due to the harmonic confinement. However, the energy shift in emission energy is still considerable, e.g. for \( X^{1-} \sim 1000 \times \) the radiative linewidth.

![Figure 1.3: Processes in a photoluminescence experiment with an empty QD (upper sequence) and a singly-charged QD (lower sequence).](image)

In bare QD samples the number of charges present in the dot can only partially be controlled by adjusting the pump intensity leading to a change in the rate of optical charging of carriers given that there is typically a preference for the capture of either electrons or holes. For true deterministic charging, a different approach has to be followed.
1.3 Deterministic Charging

In order to gain control over the number of excess electrons the QDs are embedded in a heterostructure similar to a Schottky-diode \[11\]. Here, a gate voltage \( V_g \) is applied between a top (Schottky) gate and a back contact which is formed by a n-doped layer below the QDs, creating an electric field along the growth direction. Fig. 1.4 shows a gatesweep i.e. PL as a function of gate voltage.

![Graph showing PL as a function of gate voltage]

**Figure 1.4:** In a gatesweep, discrete emission energies reveal different excitonic states, continuous shifts are due to DC Stark shift.

The discrete PL emission energies can be attributed to different excitonic charging states denoted as \( X^0 \), \( X^{1-} \) etc. which refer to a neutral exciton (bound electron-hole pair), negatively charged exciton (two electrons and a hole) etc. At certain gate voltages the charging state switches abruptly. This can be understood from the band structure diagram given in Fig. 1.5.

A built-in electric field due to the Schottky contact leads to a tilt of conduction and valence band within the intrinsic region. This tilt can be decreased (increased) by applying positive (negative) gate voltage, thus lowering (raising) the QD energy levels with respect to the Fermi energy \( E_F \) of the back contact. When the lowest QD energy level drops below the Fermi energy, the QD becomes singly charged (in...
Figure 1.5: Heterostructure and band diagram. The width of the tunnel barrier is 25nm. At a gate voltage $V_g = V_1$ the QD is empty. At a different gate voltage $V_g = V_2$ an electron is loaded into the QDs from the nearby n-doped GaAs Fermi sea.

Fig.1.5(right): empty QD at $V_g = V_1$, singly-charged QD at $V_g = V_2$). It then requires an additional increase of gate voltage to load a second electron into the QD. This phenomenon is referred to as Coulomb blockade [10]; it means that for the addition of a second electron a charging energy ($\sim 20\text{meV}$) is needed due to the fact that the QD acts as a small capacitor.

Besides changing the QD charging state, the electric field further perturbatively alters the wavefunctions of the confined carriers, leading to a DC Stark shift similar to the Stark shift observed in atomic physics. However, a term linear in electric field appears due to a built-in permanent excitonic dipole moment oriented along the growth direction; the weaker, quadratic term is related to the polarizability of the QD $\beta_{qd}$. The total shift of the optical transitions is then given by

$$\Delta E = -d_{\text{perm}} \cdot E + \beta_{qd} E^2$$  \hspace{1cm} (1.1)$$

Finally, a magnetic field along the growth (z) direction also leads to a shift of the energies of the optical transitions by

$$\Delta E = \pm \frac{1}{2} \mu_H (g_e - g_h) B_z + \left(\frac{\hbar e^2}{8\pi m^* c^2 \omega^2}\right) B_z^2$$  \hspace{1cm} (1.2)$$

with $\hbar \omega$ the quantization energy of the harmonic potential and $g_e$ ($g_h$) the electron (hole) g-factor. The first term linear in $B$ corresponds to Zeeman splitting and the second term quadratic in $B$ is the diamagnetic shift. Fig. 1.6 shows a magnetic-field
dependent PL measurement on the singly-charged exciton $X^{1-}$ where two branches appear, revealing the Zeeman-split optical transitions.

![Figure 1.6: Magnetic field response of the singly-charged $X^{1-}$ (trion) state.](image)

### 1.4 Spin and Pauli Blockade

The confined energy level scheme of a QD containing a single excess electron in the presence of external magnetic field is depicted in Fig. 1.7. The lowest conduction band states (s-shell) have spin projection $S_z = \pm \frac{1}{2}$. In the valence band, the bulk heavy-light hole degeneracy at the $k = 0$ point is lifted due to confinement in conjunction with the different heavy and light hole masses, giving rise to a heavy-light hole splitting $\Delta_{hh}$. As a consequence, the highest energy valence-band states are heavy hole states with spin projection $J_z = \pm \frac{3}{2}$. From selection rules it follows that there can only be two optical transitions which couple the electronic and heavy-hole states and have orthogonal circular polarization.

We neglect the four optical transitions coupling the two light hole states with spin projection $J_z = \pm \frac{1}{2}$ to both electronic states. They are not addressed by a laser with frequency in the vicinity of the heavy-hole transitions during transmission measurements due to large heavy-light hole splitting $\Delta_{hl} > 10\text{meV}$ which greatly exceeds all other relevant energy scales such as Zeeman splitting ($\sim 100\mu\text{eV}$) and radiative linewidth of the optical transitions ($\sim 1$ to $2\mu\text{eV}$). However, in the presence
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Figure 1.7: QD level scheme with the relevant heavy-hole optical transitions in the case of a single excess conduction-band electron. Depending on the electron spin state, one of the transitions is Pauli-blocked.

of heavy-light hole mixing the QD spin dynamics can be altered due to the presence of the light-hole transitions; this issue is discussed in Chapter 3.

When assuming perfect optical selection rules, the QD can only be excited with either $\sigma^+$ or $\sigma^-$ light depending on the spin state of the excess electron as it can be seen in Fig. 1.7: it is forbidden by the Pauli principle to excite a second electron having quantum numbers identical with the excess electron that is already present. Exploiting this so-called Pauli blockade in an optical measurement sensitive to the absence or presence of the corresponding spin-selective optical transitions e.g. by detecting resonance fluorescence or absorption, a spin quantum jump measurement could be realized [4]. However, Fig. 1.7 describes the selection rules of the QD transitions in an idealized limit without imperfections such as heavy-light hole mixing and interactions with nuclear spins, the Fermi sea and phonons.

1.5 Exciton Fine Structure

As discussed in section 1.2 the energies of the confined carriers become renormalized due to Coulomb interaction leading to the appearance of distinct emission energies in photoluminescence gatesweeps.

Further, electron-hole exchange interaction gives rise to a fine structure splitting [12–14] by mixing the spin of the electron and hole of the neutral exciton $X^0$. An
Excitonic basis can be constructed from the corresponding spin states \( \{S, S_z\} = \{\frac{1}{2}, \pm \frac{1}{2}\} \) for the electron and \( \{J_h, J_{h,z}\} = \{\frac{3}{2}, \pm \frac{3}{2}\} \) for the (heavy) hole. Again, light-hole states are neglected due to large heavy-light hole splitting. The excitonic basis is then given by the angular momentum projections of the total excitonic spin \( M = S_z + J_{h,z} \). Optical selection rules dictate that radiative decay of states with \( M = \pm 1 \) is allowed; therefore these are referred to as bright excitons whereas states with \( M = \pm 2 \) are dark.

The spin part of the electron-hole exchange interaction is given by

\[
H_{e-h} = -\sum_{i=x,y,z} (\alpha_i J_i S_i + \beta_i J_i^2 S_i)
\]

and can be written in Matrix form using the exciton basis introduced beforehand with \( M = \{+1, -1, +2, -2\} \)

\[
H_{e-h} = \frac{3}{4} \begin{bmatrix}
\alpha_x + 2.25\beta_z & 0.5(\beta_x - \beta_y) & 0 & 0 \\
0.5(\beta_x - \beta_y) & \alpha_x + 2.25\beta_z & 0 & 0 \\
0 & 0 & -\alpha_z - 2.25\beta_x & 0.5(\beta_x + \beta_y) \\
0 & 0 & 0.5(\beta_x + \beta_y) & -\alpha_z - 2.25\beta_z
\end{bmatrix}
\]

Hence, electron-hole exchange interaction lifts the degeneracy of dark excitons due to the terms \( (\beta_x + \beta_y) \). In contrast the bright excitons remain degenerate when the term \( (\beta_x - \beta_y) \) is zero which is the case of a QD with perfect in-plane symmetry. If this symmetry is broken the degeneracy is lifted, yielding a linearly polarized doublet in the absence of magnetic field, the \( x-y \) splitting. This zero-field splitting ranges from 0 to \( \sim 40 \mu\text{eV} \), depending on the QD.

In the case of the negatively charged exciton \( X^{-1} \), there are two possible excitonic states (both are bright) characterized by the hole spin projections \( \{J_{h,z} = \pm \frac{3}{2}\} \); the two electrons form a singlet state with \( S = 0 \). In consequence the spin Hamiltonian (1.3) vanishes leading to the absence of a splitting without external magnetic field.

Whereas it is difficult to observe the small \( X^0 \) splitting by common PL techniques, it can be clearly resolved in high-resolution transmission spectroscopy. Here, a narrow-band laser is scanned over the excitonic transitions while the transmitted light intensity is recorded. The transitions then appear as a dip in transmission due to absorption of light by the QD [13, 15–19]. This technique will be explained in more detail in the following sections. Fig. 1.8(left) shows such a laser scan of the \( X^0 \) transition at 0 Tesla revealing an \( x-y \) splitting of \( \sim 3 \text{ GHz} \). A scan of the \( X^{-1} \) is shown on the right; here only one transition can be observed in the absence of
magnetic field.

Figure 1.8: Fine structure splitting of the neutral exciton (left); on the singly charged exciton (right) only a single transition can be observed.

1.6 Light scattering by a single quantum dot

We will now discuss the response of a single QD to monochromatic excitation light described by a focused Gaussian beam. We note that this calculation is based on the treatment given in [18]. The geometry is sketched in Fig. 1.9: a focused Gaussian wave propagating along the optical axis $z$ with waist $w_0$ and Rayleigh range $z_R$ is impinging on the QD which is located in the focus at $r = 0$. The detector is centered on the $z$-axis at position $z_{\text{det}} \gg z_R$ and has radius $R_{\text{det}}$.

We will show that under these circumstances the incident field is attenuated through interference with the QD dipole field; it is the well-known problem of scattering in forward direction and closely linked to the optical theorem which relates the attenuation of a scatterer to its forward scattering amplitude [20].

In general, the operator for the total field can be written as

$$ E_T(r, t) = E^+_{q}(r, t) + E^-_{q}(r, t) $$

The total field is the sum of laser field and scattered field

$$ E^\pm_T(r, t) = E^{\pm}_{q}(r, t) + E^{\pm}_{L}(r, t) $$

(1.6)
1.6. Light scattering by a single quantum dot

The excitation field in the focal plane i.e. at position \( z = 0 \) (Fig. 1.9) can be described by a plane phase front and Gaussian intensity distribution

\[
E_L^\pm(x, y, t) = \pm \frac{i}{2} A_0 \exp(-\frac{x^2 + y^2}{w_0^2}) \exp(\pm \omega t) \cdot e_L
\]

where \( e_L \) is a polarization vector and the beam waist is denoted as \( w_0 \). \( A_0 \) describes the maximum field amplitude on the optical axis. It follows that the total time-averaged laser power is

\[
\langle I(r, t) \rangle = \varepsilon_0 c \langle E_T^-(r, t) E_T^+(r, t) \rangle
\]

\[
= \langle I_L(r, t) \rangle + \langle I_{sc}(r, t) \rangle + \varepsilon_0 c 2 \text{Re} \langle E_L^+(r, t) E_{qd}^-(r, t) \rangle
\]

where \( \langle I_L(r, t) \rangle = \varepsilon_0 c \langle E_L^-(r, t) E_L^+(r, t) \rangle \) is the laser intensity and \( \langle I_{sc}(r, t) \rangle = \varepsilon_0 c \langle E_{qd}^-(r, t) E_{qd}^+(r, t) \rangle \) refers to the total intensity (coherently and incoherently) scattered by the QD. This term describes the light observed in other, resonance fluorescence-type experiments when only detecting at angles larger than the excitation beam opening angle. We neglect this term since only a relatively small part of the excitation light is scattered (\(|\langle E_{qq}^- \rangle| \ll |\langle E_L(r, t) \rangle|\)), at least for numerical apertures within the paraxial approximation.
After propagation to a distance much larger than \( z_R \), the excitation field can be written as [21]

\[
E_I^\pm(r, t) = \pm i\phi_G b_0 \frac{z}{r^2} \exp(\pm i(kr - \omega t)) \exp \left( -\frac{1}{c_0} \frac{x^2 + y^2}{r^2} \right)
\]

(1.10)

where \( \phi_G \) describes a phase shift and \( b_0 \) and \( c_0 \) are fixed beam parameters given by \( b_0 = \frac{A_0 \pi w_0^2}{\lambda} \) and \( c_0 = \frac{4}{k^2 w_0^2} \).

It is important to consider the Gouy phase shift, which has been discovered in 1890 [22][23]

\[
\phi_G = -\arctan \frac{z}{z_R}
\]

(1.11)

This phase shift describes the relative phase between a focussed Gaussian beam and a plane wave, both propagating along the z-axis. For distances much larger than the Rayleigh length i.e. \( z > z_R \), \( \phi_G = -i \).

The field that is radiated by the QD, located at the origin \( r = 0 \), is described by the source-field expression [24]

\[
\tilde{E}_{q_d}(r, t) = \tilde{E}_d^\pm(r, t) = y_0 \frac{1}{|r|} i\sigma_{eg}(t - \frac{|r|}{c})
\]

(1.12)

Here, the dipolar emission in the far field paraxial limit is described as a spherical wave. The factor \( y_0 \) is given by

\[
y_0 = \frac{\varepsilon_0 \omega_0^2 D_{12}}{4\pi \varepsilon_0 c^2}
\]

(1.13)

where \( D_{12} \) is the dipole moment of the relevant optical transition of the QD and \( \omega_0 \) its frequency. The background intensity i.e. the far-off resonant intensity when the QD response can be neglected is given by

\[
I_{\text{off}}(r, t) = \varepsilon_0 c|E_L(r, t)|^2 = \langle I_L(r, t) \rangle
\]

(1.14)

The Poynting vector of the total field is directed away from the origin due to the nature of the spherical wave. Its time-averaged value is given by
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\[ \langle S(r) \rangle = \frac{1}{2} \varepsilon_0 c \langle |I(r)| \rangle \frac{r}{|r|} \]  

(1.15)

The total power on the detector is calculated by integration over the projection of the Poynting vector \( S(r) \) onto the detector surface normal \( n \).

\[ P_{\text{det}} = \int_{\text{Det.Area}} dA \overline{S(r) \cdot n} \]  

(1.16)

Before proceeding we note that in the experiment we are interested in normalized transmission

\[ \frac{T}{T_{\text{off}}} = \frac{P_{\text{det}}}{P_{\text{det,off}}} = \frac{\int dA \cos \theta(I_L(r)) + \int dA \cos \theta \, 2\text{Re} \langle \overline{E_x(r,t)E_y(r,t)} \rangle}{\int dA \cos \theta(I_L(r))} = 1 - \Theta \]  

Again, the scattered intensity \( \langle I_{\text{sc}} \rangle \) from equation (1.7) has been neglected. In order to calculate the relative absorption \( \Theta \) we replace the denominator in (1.17) by \( dA \cos \theta(I_L(r)) = P_L/(\varepsilon_0 c) \) given in (1.9), assuming that the detector aperture is large enough to collect the entire excitation light. We are then left with the evaluation of the integral over the real part of the interference terms. Therefore we use [24]

\[ \langle \sigma_{eg}(t) \rangle = \bar{\rho}_{ge}(t) \exp(i\omega t) \]  

(1.18)

For times \( t \to \infty \), \( \rho_{ge} \) can be split into a part that reaches steady-state and a term oscillating at the excitation frequency [24]

\[ \bar{\rho}_{ge}(\infty) \exp(i\omega t) = -\frac{\Omega}{2} \left[ (\omega - \omega) - i\frac{\Gamma}{2} \right] \exp(i\omega t) \]  

(1.19)

Now the integral appearing in the numerator of equation (1.17) can be rewritten using cylindrical coordinates \( x = r' \sin \phi, y = r' \cos \phi, |r|^2 = r'^2 + z_{\text{det}}^2, \cos \theta = \)
\[
\Theta = - \int dA \cos \theta \ 2 \text{Re}(\hat{E}_d^+(r, t) \hat{E}_d^-(r, t))
\]
(1.20)
\[
= - \int_0^{2\pi} d\phi \int_0^{R_{det}} dr' \frac{z_{det}}{\sqrt{r'^2 + z_{det}^2}} 2 \text{Re} \left[ i\phi c b_0 \frac{z_{det}}{r'^2} \exp \left( - \frac{1}{c_0^2} \frac{r'^2}{r^2} \right) \frac{y_0}{r} \tilde{\mu}_{ye}(\infty) \right]
\]

Here, the oscillating terms of type \( \exp(i(\kappa r - \omega t)) \) have cancelled. In order to simplify the evaluation of the integral over \( r' \) we make the paraxial approximation, \( r^2 = z_{det}^2 + r'^2 \approx z_{det}^2 \) which, together with (1.19) results in
\[
\Theta = 4\pi b_0 y_0 \frac{\Omega_R \Gamma}{4(\omega_0 - \omega)^2 + \Gamma^2 + 2\Omega^2} \frac{1}{z_{det}^2} \frac{c_0^2 z_{det}^2}{2} \int_0^{R_{det}} dr' \frac{r'^2}{r^2} \exp \left( - \frac{1}{c_0^2} \frac{r'^2}{r^2} \right)
\]
(1.21)

We again assume that the detector aperture is greater than the aperture of the focusing lens, i.e. \( R_{det} \to \infty \) and the \( r' \) integral can easily be solved
\[
\Theta = 4\pi b_0 y_0 \frac{\Omega_R \Gamma}{4(\omega_0 - \omega)^2 + \Gamma^2 + 2\Omega^2} \frac{1}{z_{det}^2} \frac{c_0^2 z_{det}^2}{2}
\]
(1.22)

Now we insert \( y_0 \) and \( b_0 \) and further include the spontaneous emission rate \( \Gamma = \frac{\varepsilon_0^2 \omega_0^3 D_{12}^2}{3\pi\varepsilon_0 h e^2} \) to get
\[
\Theta = 4\pi A_0 \frac{\pi \omega_0^2}{\lambda} \frac{\omega_0^2 D_{12} A_0 D_{12} \Gamma}{4 \frac{\pi \varepsilon_0 c^2}{h} \frac{4h}{4(\omega_0 - \omega)^2 + \Gamma^2 + 2\Omega^2}} \frac{1}{\gamma^2}
\]
(1.23)
\[
= \frac{3 \pi^2 A_0^2}{2} \frac{\gamma^2}{k^3 \lambda} \frac{\omega_0^2 D_{12} A_0 D_{12} \Gamma}{4 \frac{\pi \varepsilon_0 c^2}{h} \frac{4h}{4(\omega_0 - \omega)^2 + \Gamma^2 + 2\Omega^2}}
\]

After further simplification and taking the size of the laser spot to be \( A_L = \pi w_0^2 \) we obtain for the relative absorption
\[
\Theta = \frac{3 \pi^2 A_0^2}{2} \frac{\omega_0^2 D_{12} A_0 D_{12} \Gamma}{2\pi A_L} \frac{\Gamma^2}{P_L/(\varepsilon_0 c)}
\]
(1.24)
\[
\text{The power removed from the beam, i.e. the total scattered power is}
\]
\[
P_{sc} = \Theta \cdot P_L = \frac{\sigma}{A_L} P_L
\]
(1.25)

Hence, the QD scattering cross-section is
\[
\sigma = A_L \Theta = \sigma_0 \frac{\Gamma^2}{4(\omega_0 - \omega)^2 + \Gamma^2 + 2\Omega^2}
\]
(1.26)
which includes the well-known textbook result for the on-resonance low-power cross-section of a two-level system $\sigma_0 = \frac{3A^2}{2\pi}$.

In order to estimate the experimentally expected relative absorption $\Theta$ we take into account that the QD is located in a high-refractive index solid-state matrix and therefore replace $\lambda$ by $\lambda/n$ in the cross-section $\sigma_0$. For a numerical aperture of $NA = 0.55$ and a wavelength of $\lambda = 950\text{nm}$, the diffraction limited spot size is $A_L \sim 0.59\mu m^2$. For $\sigma_0$ we obtain $0.033\mu m^2$ which yields

$$\Theta_{\text{theory}} = \frac{\sigma_0}{A_L} \sim \frac{0.033\mu m^2}{0.59\mu m^2} \sim 5.6\%$$

Typical values achieved in an experiment are $\sim 0.6\%$ (for a numerical aperture of 0.55). In these experiments, a lock-in detection scheme has been used. This causes the absolute absorption to drop by a factor measured to be $S_{\text{lock-in}} = 2.25$ whereas the background power used for the normalization is not modified; hence, this factor has to be taken into account for $\Theta$ for comparison with equation (1.27). Furthermore, typical experimental linewidths (300 to 500 MHz) exceed the purely radiative linewidth (200 MHz for a radiative lifetime of $\tau_{\text{rad}}=0.8\text{ns}$ as measured in [25]), which implies broadening mechanisms such as dephasing or spectral diffusion to be present. Both lead to a drop of peak absorption. In addition, the diameter of the focal spot was $\sim 1\mu m$, which makes the estimate in (1.27) decrease to 4.2% whereas we obtain an experimental value of 3.4% when taking into account lock-in and line broadening.

The theory presented in the previous section determines the dependence of relative absorption on intensity and laser detuning. Fig. 1.10 shows absorption $\Theta$ observed on one of the neutral exciton transitions for different incident excitation powers using a transmission measurement. The points are raw uncorrected data.

Expression (1.24) indicates that the power dependence of on-resonance absorption can be described by a function of type

$$\Theta(I_L) = \alpha_0 \cdot \frac{1}{1 + I_L/\alpha_1}$$

(1.28)

Where $\alpha_0$ takes into account the spatial mismatch factor $\alpha_0 = \sigma/A_L$ and $\alpha_1$ is a proportionality factor linking the power observed on the detector to the effective power driving the QD optical transition.

The solid line in Fig. 1.10 is a fit using equation (1.28) which yields $\alpha_0 = 0.76\%$ and $\alpha_1 = 6.3$. The value of excitation intensity (x-axis) at which the relative absorption
drops to one half of its low-power initial value (i.e. $I_L = \alpha_1$) marks the point where the relation $2\Omega_R^2 = \Gamma^2$ is satisfied. The upper dashed curve has been compensated for the loss of signal due to lock-in.

1.7 Experimental Setup

All experiments described here are carried out with a confocal microscopy setup immersed in a liquid helium bath cryostat at a temperature of 4.2 Kelvin (see Fig. 1.11). The numerical aperture of the microscope is 0.68 resulting in a spot size of $\approx 1\mu m$. Area density of QDs in our sample is low enough to have $\leq 3$ dots in the focal spot simultaneously. Different QDs can then separated spectrally separated due to their inhomogeneous broadening. A magnetic field of up to 10 Tesla along the z-axis can be applied by a superconducting magnet. Piezo-electric nanopositioners allow us to move the sample in XYZ-space. Transmitted light is collected and sent to a circular polarization analyzer which distributes the light on two photodetectors [Fig. 1.11(right)].

The initial characterization of a QD is performed with a gatesweep (section 1.3). Hereafter, the differently charged excitonic complexes can be identified by their characteristic emission energy and voltage dependence [11]. From then on we apply resonant excitation by using polarization-selective differential transmission (DT).
1.7. Experimental Setup

Figure 1.11: Experimental setup. Laser light is focussed onto a QD sample, then split according to its polarization and directed to photodetectors.

Again, all aspects of this technique are covered in Refs. [13, 15–19]. The drop in transmitted light i.e. the QD-induced absorption is given by equation (1.24) which yields a Lorentzian lineshape when a laser is scanned across the optical transition.

In order to improve the signal-to-noise ratio a lock-in detection scheme is used; therefore, a small square-wave modulation of frequency $f_{\text{lock-in}}$ typically in the range of $\sim 3\text{kHz}$ is added to the DC gate voltage. The photocurrent output of the transmission detectors is then passed through current-to-voltage converters of gain $10^7$ to $10^9$ V/A and demodulated by a lock-in amplifier.
2 A Spin in a Quantum Dot: State Preparation

We demonstrate the high-fidelity preparation of a QD spin-state, via laser cooling [optical pumping [26]]. Using the Pauli-blockade strength of the corresponding optical transitions as a means to infer the electron-spin state, we show that spin cooling due to spontaneous spin-flip Raman scattering can dominate over the heating introduced by hyperfine-induced spin-flip or cotunnclng events. This allows us to cool the spin temperature of an electron from 4.2 K (determined by the heat bath) down to 20 mK. By controlling the relative strength of these processes via gate voltage and magnetic field, we can tune the system from the regime of an isolated artificial atom to that of a quantum-confined solid-state system coupled either to a charge or a spin reservoir. Within the framework of quantum information processing, this corresponds to a spin-state preparation with a fidelity exceeding 99.8%.

This chapter is based on publication [2].
2.1 Background

All experiments presented in the remaining chapters were realized with a slightly modified heterostructure. In particular, the tunneling barrier was increased from 25nm to 35nm in order to reduce co-tunneling effects. In addition, an AlGaAs barrier was introduced above the QD plane to prevent holes from tunneling out. The distance between the barrier and the QDs is kept minimal to reduce coupling of confined holes to hole continuum states [27]. Fig.2.1 shows a scheme of this heterostructure.

2.2 Weak diagonal and strong vertical transitions

The QD level scheme shown in Fig.1.7 can be reformulated using the trion four-level description as illustrated in Fig.2.2(left); here, the two ground states (|↑⟩ and |↓⟩) correspond to the spin of the excess conduction-band electron being either up or down. According to optical selections rules, two excitonic states (|↑↓⟩ and |↑↓⟩) formed out of two ground-state electrons in a spin singlet and a ground-state heavy-hole can be optically excited using circularly polarized light. Pauli blockade as demonstrated in Fig.1.7 is again reflected by the spin-selectivity of the optical transitions here: conditional on the electron spin-state only one of the optical transitions is active. Optical coupling to electronic triplet states can be neglected since due to the spatial confinement, the energies of these states are much higher than that of the singlet.

In addition to the strong vertical transitions |↑↓⟩ ↔ |↓⟩ and |↑↓⟩ ↔ |↑⟩, weak diagonal transitions |↑↓⟩ ↔ |↑⟩ and |↑↓⟩ ↔ |↓⟩ are induced by inherent heavy-light hole mixing, or magnetic field that is not parallel to the strong confinement (z) axis as well as spin state mixing induced by the randomly fluctuating hyperfine field, yielding $\Gamma \gg \gamma \neq 0$, where $\Gamma$ and $\gamma$ are the allowed and forbidden spontaneous
emission rates, respectively, as indicated in Fig.2.2(left). The strong vertical transitions leave the resident electron spin unaltered, while the weak diagonal transitions lead to a net spin-flip of the resident electron.

In addition to these optical transitions, the strong hyperfine interaction of the resident electron spin with the QD nuclear spin ensemble leads to random spin-flip events at rate $\xi_{11}$. Previous studies on similar structures have shown that $\xi_{11}(B = 0) < \Gamma$ in the absence of a magnetic field, but is strongly suppressed even under relatively weak magnetic fields ($B \sim 0.1$ T) due to incommensurate electron and nuclear Zeeman energies [28][5]. This is the case depicted in Fig. 2.2(middle) where $\xi_{11}(B > 0) \ll \gamma$. When a second re-pump laser is resonant with the other Zeeman-split trion transition $| \uparrow \downarrow \uparrow \rangle \leftrightarrow | \uparrow \rangle$ (Fig. 2.2(right)), the $| \uparrow \downarrow \rangle \leftrightarrow | \downarrow \rangle$ transition will also take place via spontaneous spin-flip Raman scattering, with a rate proportional to the laser intensity.

\[ \]

2.3 Observation of spin pumping/cooling

Fig.2.3(left) is the absorption analog of Fig.1.4, showing the expected $X^{1-}$ plateau at $B = 0$ Tesla, where the QD is single-electron charged for gate voltages ($V_g$) in the 320 mV to 424 mV range. The probe laser is scanned across the $| \uparrow \downarrow \rangle \leftrightarrow | \downarrow \rangle$ transition, and has the corresponding circular polarization $[\sigma^-]$ as determined by the optical selection rules. Fig.2.3(middle) shows a suppression of the $X^{1-}$ plateau center at 0.2 Tesla magnetic field: the QD becomes transparent for gate voltages in the 344 mV - 396 mV range. Given that the corresponding $X^0$ plateau remains unaffected under all magnetic fields, this strong suppression of the signal in the $X^{1-}$

\footnote{We note that a full theoretical model will be developed in Chapter 3.}
plateau center is a signature of optical electron-spin pumping into $|\uparrow\rangle$ state due to the uni-directional spontaneous Raman scattering process ($\gamma$) that dominates over the bi-directional spin-flip process ($\xi_{ij}(B > 0)$). In this case, we can confirm that the electron remains in the spin-up state 98.5% of the time, as the laser is resonant with the Pauli-blocked [29][4] $|\uparrow\downarrow\rangle \leftrightarrow |\downarrow\rangle$ transition of the electronic spin-down state. Similar measurement at 0.3 T already shows that the electron is in the $|\uparrow\rangle$ state 99.8% of the measurement time. To date, this value is the highest state-preparation fidelity reported in a solid-state system [8][30], and is achieved for both electron-spin states. We note that the verification of higher fidelity values is not limited by the involved physical mechanism, but rather by our signal-to-noise level.

Figure 2.3: Single-electron plateau laser scans showing full plateau and no spin pumping at 0 Tesla (left), spin pumping and co-tunneling regimes at 0.2 Tesla (middle), and the same but with re-pumping laser at 0.2 Tesla (right).

To prove that the electron is shelved in the $|\uparrow\rangle$ state by the probe laser at the $|\uparrow\downarrow\rangle \leftrightarrow |\downarrow\rangle$ transition, we simultaneously apply a re-pump laser on the $|\bar{\uparrow}\downarrow\rangle \leftrightarrow |\uparrow\rangle$ transition with orthogonal circular polarization $|\sigma^+\rangle$. Fig.2.3(right) shows the resulting gate sweep where an absorption peak at $V_g = 372$ mV appears. The linewidth of this peak is equal to that obtained for the trion transition at 0 Tesla and it is observed when the re-pump laser is detuned from the probe laser

\[2\text{We emphasize that all these measurements are performed on time scales faster than significant dynamical nuclear-spin polarization [31][32], so as to avoid any alteration of the trion absorption spectrum. This time scale of } \sim 300 \text{ ms, which corresponds to how long the laser remains resonant with the transition, in turn, defines our detection-noise level.}\]
by exactly 6 GHz, i.e. the independently measured Zeeman splitting of the $X^1-$
transition. The fact that we recover the probe laser absorption only when both
lasers are resonant with the corresponding trion transitions indicates that the re-
pump laser now allows for bi-directional spin-flip spontaneous Raman scattering and
prohibits any net spin-shelving; the system is now described by the illustration in
Fig. 2.2(right). When we keep the frequency of the re-pump laser unchanged, but
increase the magnetic field to 0.3 Tcsla, we observe that the absorption peak on
the transparent section of the plateau shifts in accordance with the corresponding
9-GHz Zeeman splitting.

2.4 State-Preparation Fidelity

Fig.2.4(left) shows the magnetic-field dependence of the absorption displaying the
Pauli-blockade strength in the middle of the charging plateau, normalized to the
maximum absorption at 0 T. The red line is a theoretical curve obtained from rate
equations by taking into account both photon-assisted hyperfine-induced spin-flip
events and spontaneous spin-flip Raman transition. The parameters used to simulate
the experimental results are $\Gamma = 300 \text{MHz} \times 2\pi$, $\gamma = 100 \text{kHz} \times 2\pi$, $B_{\text{nuc}} = b_0/\sqrt{N} = 12.5 \text{mT}$ [7], and $g_e = -0.6$. The spin cooling rate (below saturation) is independent
of laser power for the single-$\Lambda$ system, further supporting our assumption that both
cooling and spin relaxation are (linearly) proportional to the probe laser intensity.
We emphasize however that an exponential fit seems to be in better agreement with
the data, indicating that our simple theoretical model may not be capturing other
relevant processes such as phonon-assisted spin-flips or spin-orbit coupling.

The triangles at 0.2Tesla in Fig.2.4(left) correspond to the partial (full) recovery
of the absorption as the bi-directional optical spin pumping is realized using the
re-pump laser that is of weaker (comparable) intensity with respect to the probe
laser. The state-preparation fidelity for the electron spin as a function of magnetic
field is plotted in Fig.2.4(right). Already at 0.3Tesla the electron is in the spin-up
state with a fidelity exceeding 99.8%. With each data point, the corresponding spin
temperature is provided as obtained from the state occupancies and a net cooling
from 4.2 K to 20 mK is achieved. As a token of our cooling efficiency, we emphasize
that such state-preparation fidelity can only be achieved at a 62-Tesla external field,
when relied solely on thermal equilibration at 4.2 K.

$\Gamma = 300 \text{MHz} \times 2\pi$ and $g_e = -0.6$ are average values measured on numerous QDs within this
sample. $B_{\text{nuc}} = 12.5 \text{mT}$ implies $10^7$ nuclei within the QD which, in turn, is consistent with
transmission electron microscope images of similar QDs.
Another striking feature of the gate sweeps shown in Fig.2.3 is that the absorption remains essentially unaffected for gate voltages that define the edges of the plateau. Spin-cooling is ineffective in this regime despite the fact that hyperfine induced electron spin-flip rate should not depend on the gate voltage. On the other hand, it has been shown that the cotunneling-induced spin-flip rate varies across the absorption plateau by a factor as large as $10^6$ [33], for 20-meV electron charging energy\(^4\). We expect the electron spin-flip rate $\xi_{i1}(B)$ to be dominated by cotunneling at the edges of the plateau, leading to annihilation of spin-pumping. In more general terms, the spin cooling dynamics is determined by interplay between spontaneous spin-flip Raman scattering, hyperfine-induced electron-spin-flips and electronic cotunneling processes. In the absence of magnetic field and at a gate voltage within the plateau middle QD spin strongly interacts with the QD nuclei. Alternatively, at finite magnetic field and at a gate voltage within the edges of the plateau, QD spin strongly interacts with the back-gate electron reservoir. In both regimes, the electron spin cannot be considered as an isolated quantum system anymore due to

\(^4\)The absorption linewidth at the edge of the plateau is not broader than that at the center suggesting a tunnelling rate less than 300 MHz, which puts an upper limit of 0.3 KHz for the cotunneling rate. Further, the absorption peak in the plateau center does not saturate above our detection-noise level [Fig.2.4(left)], indicating that the cotunneling rate must be at least $10^9$ slower than $\gamma$. This, in turn, marks a more accurate upper bound of 0.1 KHz.
its dominant coupling to a spin or a charge reservoir.

2.5 Outlook

These results constitute the first step towards stimulated Raman transition [34] on a single quantum-dot electron for coherent preparation of an arbitrary superposition of spin states, as well as cavity-assisted spin-flip Raman transitions as a source of indistinguishable single photons with near-unity collection efficiency [35].
3 Optical study of QD spin dynamics

We have performed all-optical measurements of spin relaxation in single self-assembled InAs/GaAs quantum dots (QD) as a function of static external electric and magnetic fields. To study QD spin dynamics we measure the degree of resonant absorption which results from a competition between optical spin pumping induced by the resonant laser field and spin relaxation induced by reservoirs. Fundamental interactions that determine spin dynamics in QDs are hyperfine coupling to QD nuclear spin ensembles, spin-phonon coupling and exchange-type interactions with a nearby Fermi sea of electrons. We show that the strength of spin relaxation generated by the three fundamental interactions can be changed by up to five orders of magnitude upon varying the applied electric and magnetic fields. We find that the strength of optical spin pumping that we use to study the spin relaxation is determined predominantly by hyperfine-induced mixing of single-electron spin states at low magnetic fields and heavy-light hole mixing at high magnetic fields. Our measurements allow us to determine the rms value of the hyperfine (Overhauser) field to be \( \sim 15 \) mTesla with an electron g-factor of \( g_e=0.6 \) and a hole mixing strength of \( |\varepsilon|^2 = 5 \times 10^{-4} \).
3.1 Introduction

A single quantum dot (QD) electron spin is a fundamental physical system which allows for a controlled study of confined spin dynamics in the solid-state. In contrast to higher-dimensional semiconductor structures QD spins have been demonstrated to possess long relaxation and coherence times exceeding 20 msec and 10 μsec respectively. These findings along with demonstration of single spin manipulation and read-out have strengthened the proposals for using QD spins as physical representation of qubits in quantum information processing [3, 4, 36]. The prolongation of spin relaxation times for QD spins stems from a drastic reduction in spin-phonon coupling mediated by a combination of electron-phonon and spin-orbit interactions and suppressed by strong quantum confinement of electrons. As a consequence additional spin-reservoir interactions such as hyperfine coupling to QD nuclear spins and exchange-type (co-tunneling) coupling to a nearby Fermi-sea become prominent in determining the spin dynamics in QDs.

Here we study the dynamics of an electron spin confined in a self-assembled InAs/GaAs QD which is in turn embedded in a Schottky heterostructure. In order to assess the relative importance and external field dependence of the three elementary spin-relaxation mechanisms we use the degree of resonant absorption as a measure: since the degree of absorption i.e. strength of the trion-resonant light scattering is determined by competing optical spin pumping (OSP) [37] and spin relaxation induced by spin-reservoir interactions, the strength of spin relaxation can be inferred from the absorption measurement. First we demonstrate that at low magnetic fields (up to 1 Tesla) spontaneous spin-flip Raman scattering that allows for one-way pumping into the optically-uncoupled spin-state is predominantly mediated by a mixing between the electronic spin states induced by the fluctuating hyperfine nuclear (Overhauser) field. Next, we show that upon varying the external gate voltage by about 50 mV the spin relaxation due to exchange coupling to the nearby Fermi-sea of electrons can be changed by as much as five orders of magnitude. Finally, we show that at the high magnetic field regime (1 to 10 Tesla) spin pumping is due to heavy-light hole mixing and spin relaxation is dominated by phonons in conjunction with spin-orbit interaction.

Before proceeding we note that major advances in understanding relaxation and decoherence of single confined electron spins have already been achieved in electrically defined QDs. By implementing a single-shot electrical read-out of a QD spin Elzerman et al have shown that spin lifetimes in electrically defined QDs can reach up to ~1 msec even at elevated magnetic fields of 8 Tesla [28] and very recent
measurements have revealed a relaxation time of 170ms at 1.75 Tesla [6]. Similarly in double QDs Johnson et al have investigated hyperfine-induced triplet-singlet relaxation [7] Petta et al have demonstrated coherent manipulation of singlet-triplet states [8] and Koppens et al have shown detection and control of hyperfine-induced singlet-triplet mixing [38] and Rabi oscillations using microwave pulses [9]. Further measurements on InAs/GaAs self-assembled QD ensembles have revealed $T_1$ times exceeding 20 msec at a magnetic field of 4 Tesla and a temperature of 1 Kelvin [5].

This exhaustive chapter is organized as follows: In Section 3.2 we introduce the coupling of the localized spin to nuclear spins charge reservoir and phonons. Section 3.3 then theoretically describes the QD spin dynamics in the framework of the trion four-level system with spin-reservoir coupling. In Section 3.4 we present our experimental results obtained with single QD absorption spectroscopy in distinct regimes of external electric and magnetic fields where different interactions dominate. Finally Section 3.5 gives an overview on the above-mentioned interactions together in a self-contained picture before the conclusions in Section 3.6. For information about experimental techniques we direct the reader to the introductory chapter 1; an illustration of the sample is given in 2.1.

3.2 Interactions of a Single Confined Spin

3.2.1 Nuclear Spins

The interaction of a localized electron spin with a surrounding nuclear spin ensemble can be written in the form of the Fermi contact interaction which yields [39]

$$
\hat{H}_{\text{hyp}} = \frac{\nu_0}{8} \sum_i A_i |\psi(R_i)|^2 (\hat{I}_i \cdot \hat{\sigma})
$$

(3.1)

The sum runs over all nuclei $i$ in the lattice. $\nu_0$ is the volume of an InAs unit cell $\psi(R_i)$ the electron envelope wavefunction at the $i$th nucleus and $\hat{I}_i$ and $\hat{\sigma}$ are the spin operators of nuclear and electron spin. $A_i = (2\mu_0 g_0 \mu_B \mu_i / 3I_i) |u_e(R_i)|^2$ is the hyperfine coupling strength and reflects the electron density described by the electron Bloch wavefunction $u_e(R_i)$ at the site of the nuclei. $\mu_B$ is the Bohr magneton and $\mu_i$ the nuclear magnetic moment $\mu_0$ is the permeability of vacuum and $g_0$ the free-electron g-factor.

In order to estimate the total number of nuclei within the spread of the electron wavefunction we use the dimensions of the QD: InAs and GaAs have Zincblende-
type lattice with a lattice constant of 6.06 Å and 5.65 Å. There are four Arsenic (nuclear spin $I_{\text{As}} = 3/2$) and four Indium ($I_{\text{In}} = 9/2$) or Gallium ($I_{\text{Ga}} = 3/2$) atoms in a fcc unit cube. Taking this into account and assuming that the QD creates a box-like confinement that equals the dimensions of the QD ($\approx 20\text{nm} \times 20\text{nm} \times 5\text{nm}$) the number of As atoms is on the order of $4 \times 10^4$ and one has to add the same number of In or Ga atoms depending on the composition of the QD such that the total number of nuclei interacting with the QD spin can be taken to be $N = 10^4$ to $10^5$.

We note that the Fermi contact interaction relies on a finite value of the Bloch wavefunction at the sites of the nuclei. Due to the s-like symmetry of their wavefunction electrons are susceptible to this interaction whereas the p-symmetric holes are not. For holes it is only possible to interact with nuclear spins via the much less efficient dipole-dipole interaction.

The hyperfine interaction constitutes a special case among the three spin-reservoir interactions discussed in this work as our experiments suggest that its dominant effect is not described by a textbook system-reservoir interaction in the Born-Markov approximation as it is the case for phonon and exchange coupling. In the optical measurements presented here nuclear spins mainly act by exerting a quasi-static magnetic field (Overhauser field) with rms-value on the order of $B_{\text{nuc}} = 15$ mTesla. This field leads to a Rabi-type slowly varying coherent mixing of the spin ground states which can also be understood as precession of the electron spin in the nuclear magnetic field; in this context slowly varying means that the correlation time of the hyperfine field fluctuations ($\sim 1\text{ms}$) is much longer than the precession time ($\sim 1\text{ns}$) of the electron spin in this field.

In contrast to the ground states the excited states remain unchanged as the hole is not susceptible to the nuclear magnetic field and the two electrons form a singlet which is immune to magnetic field variations too. The mixing of the ground states if strong enough would thus lead to a fluctuating observable splitting of the excitonic transitions however it turns out that its magnitude is less than the broadening of these transitions due to their radiative lifetime and therefore cannot be resolved in laser scanning absorption measurements even with experimental resolution much better than the transition linewidth.

In order to understand the effect of the hyperfine field in optical experiments we consider two regimes: First an external magnetic field with strength smaller than the hyperfine field is applied or the external field is completely absent. Hence the direction of the total magnetic field seen by the electron spin is fully random after a nuclear field correlation time. As we will see in section 3.3 fast bidirectional OSP
3.2. Interactions of a Single Confined Spin

will be the consequence inducing efficient spin relaxation dominating over other mechanisms as the one predicted in Ref. [40]. In the second regime the applied external field is much stronger than the hyperfine field. In this case the electron spin mainly sees the external magnetic field along the z-axis and the hyperfine field only leads to small fluctuations of the nuclear field vector. In this regime the light-induced spin relaxation is slow and other mechanisms such as phonons are dominant.

We will now analyze the effective hyperfine field in detail. In the absence of dynamical nuclear spin polarization (DNSP) scenarios the action of nuclear spins upon the localized spin dominates the reverse action due to a much larger number of degrees of freedom on the side of the nuclear spins ensemble. Therefore the Hamiltonian (3.1) is reduced to an effective magnetic field seen by the QD spin which is commonly referred to as Overhauser field

\[ B_N = \frac{\nu_0}{g_e} \frac{\bar{A}}{\mu_B} \left( \sum_i \hat{I}_i \right) \]  

(3.2)

where \( \bar{A} \) is an average spin-nuclei coupling constant and \( g_e \) is the QD electron g-factor.

For simplicity we will in the following treat the hyperfine field as a purely classical field \( B_N(t) \) with correlation time \( \tau_{corr} \sim 1 \text{ms} \). The correlation time is expected to be similar to the decay time of nuclear spin polarization in the presence of a QD electron and absence of external magnetic field, as measured in [41]. \( \Delta_{Bn} \) refers to the rms-value of the Gaussian distribution as defined by

\[ f(B_N) = \frac{1}{B^3_{\text{nuc}}(2\pi)^{3/2}} \exp(-|B_N|^2/2B^2_{\text{nuc}}) \]  

(3.3)

which yields

\[ \langle B_N(t) \rangle = 0 \]  

(3.4)

\[ \langle |B_N(t)|^2 \rangle = 3B^2_{\text{nuc}} \]

Here \( \langle \cdot \rangle \) denotes the time average over many correlation times.

As our \( B_N(t) \) is quasi-classical, we treat the \( B_{N,i}(t) \) with \( i = x, y, z \) as independent random variables each one following a Gaussian distribution function given by
and \( f(B_{N,i}) \) is then given by the product of the Gaussian distributions of its three spatial components

\[
f(B_{N}) = \prod_{i=x,y,z} f(B_{N,i})
\]

Clearly

\[
\langle B_{N,i}(t) \rangle = 0
\]
\[
\langle |B_{N,i}(t)|^2 \rangle = B_{\text{nuc}}^2
\]

\( B_{\text{nuc}} \) can be written in the form (similar to [42][39])

\[
B_{\text{nuc}} = \frac{b_0}{\sqrt{N}}
\]

with \( b_0 \) a parameter characterized by the species of nuclei and the composition of the QD \(^1\) and \( N \) the number of nuclear spins interacting with the QD spin.

The QD composition is taken to be 90\% InAs and 10\% GaAs yielding \( I(I + 1) = 13.2 \) when averaging over the different nuclear species [43]. Similarly we obtain \( \overline{A^2} = 2500 \mu eV^2 \) which yields \( b_0 = 3.0 \) Tesla. Using (3.8) with \( N = 10^4 \) to \( 10^5 \) nuclear spins we obtain for our QDs

\[
B_{\text{nuc}} = 9.5 \text{ mT to 30 mT}
\]

We can now rewrite (3.1) as

\[
\hat{H}_{\text{Overh}} = g_e \mu_B B_N(t) \cdot \hat{\sigma}
\]

Here the component of the nuclear field along the z-axis \( B_{N,z}(t) \) only leads to Zeeman splitting whereas the in-plane components induce a mixing of the \( | \uparrow \rangle \) and \( | \downarrow \rangle \) states.

\[
\frac{1}{2} \frac{b_0^2}{N} = \frac{1}{3} \left( \frac{b_0}{s_{n,\mu n}} \right)^2 \sum_i (A_i)^2 |\psi(R_i)|^4 (I_i^2) = \frac{1}{2N} (\overline{A^2} \overline{I(I + 1)}) / (g_e \mu_B)^2 \text{; this result is obtained by assuming a box-like confinement potential transforming the sum into an integral and using the localization volume of the QD electron given by } V_L^{-1} = \int d\mathbf{r} |\psi(R)|^4 = 8/(N \nu_0).
3.2. Interactions of a Single Confined Spin

The in-plane hyperfine field is

\[ B_{N,xy}^2(t) = B_{N,x}^2(t) + B_{N,y}^2(t) \]  

and we define

\[ \hbar \Omega_H(t) = \frac{g_e \mu_B B_{N,xy}(t)}{2} \]

We note here that our measurement time (typically 10 to 100 ms) is longer than the correlation time of the nuclear field; i.e. for each measured data point we expect that we average over many configurations of the nuclear magnetic field.

3.2.2 Coupling to electron spin reservoir

![Figure 3.1](image)

**Figure 3.1:** (a) Stability diagram of the QD ground states, neglecting spin. Energies of the zero, one and two-electron QD as a function of gate voltage. Crossover points are marked A and B. \( E_{12} \) denotes the gate voltage-dependent energy difference between the singly-charged and the doubly-charged state, or charging energy. (b to d) The QD can exchange its single electron with the charge reservoir via a virtual empty or two-electron (shown here) state. When one of the two singlet-electrons tunnels out, it leaves the remaining QD spin in a mixed state, equivalent to spin relaxation. \( \Delta = \Gamma_{tunnel} \) marks the tunneling rate through the 35-nm GaAs barrier, \( \varepsilon \) the detuning from the Fermi energy \( \varepsilon_F \), \( E_Z \) is the Zeeman splitting and \( E_{12} \) the energy required to charge a second electron.

Another interaction mechanism arises from the presence of the Fermi sea in the back contact (see Section 1.3) that couples to the QD via the tunneling barrier. It is well known that exchange interaction of a confined spin with an electron spin

---

2In this context we note that other experiments suggest that the dynamics of the nuclear spin ensemble could be altered by the measurement itself and the hyperfine field stays locked for a time on the order of seconds [38] Fig.4(b). In our measurements there is some evidence for alteration of dynamics at large magnetic fields and large gate voltage detunings (not shown in this work); however at low magnetic fields these locking effects do not seem to play a dominant role.
reservoir leads to co-tunneling \(^3\) [44] and at temperatures lower than the Kondo temperature \(T_K\) to the formation of a Kondo singlet [45–48].

This interaction can be written as

\[
\hat{H}_{\text{charge}} = \sum_{k,k'} \hbar g_{t,k}(\epsilon_{QD,\sigma}^\dagger \epsilon_{QD,\sigma}^1 e_{QD,\sigma}^1 e_{QD,\sigma}^1 + \text{c.c.})
\]

(3.13)

where \(\epsilon_{QD,\sigma}^\dagger\) and \(\epsilon_{QD,\sigma}\) are the creation and destruction operators for an electron with spin \(\sigma\) in the QD and similarly in the reservoir. \(g_{t,k}\) is the tunneling matrix element, which is linked to the tunneling rate \(\Gamma_{\text{tunnell}}\) by Fermi’s Golden rule \(\Gamma_{\text{tunnell}} = \frac{2\pi}{\hbar} |g_{t,k}|^2 \rho(E)\) with \(\rho(E)\) being the density of states in the back contact.

Fig. 3.1(a) shows the energies of the empty, singly and doubly charged QD state as a function of gate voltage [27]. Which state has lowest energy obviously depends on the gate voltage and the QD attempts to reach it by either attracting or repelling electrons from or into the reservoir. Clearly there is a range of voltages (single electron charging plateau) where it is energetically favorable for the QD to accommodate a single electron, marked by the shaded region in the figure. At the points A and B two charging levels are degenerate and fast exchange of the QD electron with the reservoir can take place, only limited by the tunneling rate. The real gate voltages \(V_A\) and \(V_B\) that need to be applied in order to reach points A and B can vary from dot to dot depending on its confinement properties.

In other words the QD is singly-charged for \(V_A < V_g < V_B\) with \(V_g\) the gate voltage. We define the plateau center \(V_c\)

\[
V_c = \frac{V_B - V_A}{2}
\]

(3.14)

The gate voltage detuning is

\[
\Delta V_g = V_g - V_c
\]

(3.15)

The schematic co-tunneling process is depicted in Fig. 3.1(b-d). The initial state is characterized by a QD with a single spin-down electron and Coulomb blockade prohibits tunneling of further electrons into the dot (b). Together with a spin-up electron from the reservoir a virtual spin singlet (c) is formed at energy difference

\(^3\)The type of co-tunneling relevant for us is inelastic co-tunneling as it leads to a transition between the spin ground states.
3.2. Interactions of a Single Confined Spin

\[ \Delta E = \varepsilon + E_{12} \] where \( \varepsilon \) is the detuning from the reservoirs' Fermi energy \( \varepsilon_F \). Finally the QD returns to the singly-charged state with a spin-up electron (d).

\[ E_{12} \text{ and } E_{01} \text{ are given by} \]

\[ E_{12} = E_2 - E_1 = \frac{e(V_B - V_g)}{\lambda} \]

\[ E_{01} = E_1 - E_0 = \frac{e(V_g - V_A)}{\lambda} \]

with \( E_i \) the energy of the QD charged with \( i \) electrons and \( \lambda \) a constant describing the geometric lever arm of the heterostructure.

Using (3.13) one obtains for the cotunneling rate in second-order [33, 44]

\[ \kappa_{\text{cotunnel}} = \frac{\hbar}{\Gamma_{\text{tunnel}}} \int \frac{1}{\varepsilon(V_B - V_g) - \varepsilon + \frac{1}{2} \hbar \Gamma_{\text{tunnel}}} \left| \frac{1}{\varepsilon(V_B - V_g) - \varepsilon + \frac{1}{2} \hbar \Gamma_{\text{tunnel}}} \right|^2 f(\varepsilon)[1 - f(\varepsilon)] d\varepsilon \]

The integral is the sum over all second-order transitions with different detunings \( \varepsilon \) from the Fermi energy according to Fig. 3.1(b-d). In addition the term with \( e(V_g - V_A)/\lambda = E_{01} \) describes the related process where the virtual state is an empty QD. \( f(\varepsilon) \) is the Fermi function \( f(\varepsilon) = \frac{1}{1 + \exp(\varepsilon/kT)} \). Expression (3.17) is valid under the condition \( E_{Z,\varepsilon} \ll kT \) i.e. for low magnetic fields. To obtain the exact expression for all magnetic fields the Fermi function terms in the integral have to be modified 4.

The imaginary part of the denominator introduces a finite lifetime to the electronic states limited by the tunneling rate \( \Gamma_{\text{tunnel}} \), implying that the main cause for broadening of the spin ground states is tunneling. This is relevant for elements of the integral with vanishing real part.

In order to obtain an estimate for the cotunneling times in our structure we use results obtained on samples with 25nm tunneling barrier where in certain gate voltage regimes tunneling rate is larger than radiative recombination rate i.e. \( \Gamma_{\text{tunnel}} > \Gamma \), leading to broadening in the linewidths observed in photoluminescence measurements [33]. Then from a Wentzel-Kramers-Brillouin (WKB) estimation of the two different tunneling barriers together with the measured tunneling rate we estimate

4With Zeeman splitting \( \kappa_{\uparrow \rightarrow \downarrow} \neq \kappa_{\downarrow \rightarrow \uparrow} \) i.e. the rate flipping the spin down is different from the rate flipping it up. The term \( f(\varepsilon)[1 - f(\varepsilon)] \) has to be replaced by \( f(\varepsilon \pm \hbar \omega_z/2)[1 - f(\varepsilon \pm \hbar \omega_z/2)] \) for \( \kappa_{\downarrow \rightarrow \uparrow} \) and \( \kappa_{\uparrow \rightarrow \downarrow} \) respectively.
the tunneling rate $\Gamma_{\text{tunnel}}$ to be on the order of 0.02 to 0.1 ns$^{-1}$ in our structure. We take it to be independent of the gate voltage within the single electron regime. Fig. 3.2 shows the calculated cotunneling rate obtained with expression (3.17) using two different tunneling rates of $\Gamma_{\text{tunnel}}=0.02$ ns$^{-1}$ and $\Gamma_{\text{tunnel}}=0.1$ ns$^{-1}$ representing the minimum and the maximum cotunneling rate we expect in our experiments respectively. Co-tunneling rate is characterized by its strong voltage dependence. When close to the crossover points $V_A$ and $V_B$ it exhibits an ultra-steep slope; in contrast, the voltage-dependence is weak in the plateau center $V_C$.

**Figure 3.2:** Expected cotunneling rate obtained using expression (3.17) with the parameters $\Gamma_{\text{tunnel}}^{-1}=10$ns (solid curve) and $\Gamma_{\text{tunnel}}^{-1}=50$ns (dashed curve), $V_A=-50$ mV, $V_B=+50$mV, $kT = 300 \mu$eV, $\lambda=5.3$.

### 3.2.3 Spin - Phonon interaction

Spin relaxation in higher-dimensional systems is mainly due to spin-orbit (SO) interaction in conjunction with phonons [49, 50]. Still, SO interaction is the enabling mechanism for phonon-assisted spin flips in QDs and a considerable amount of theoretical work has been done on this spin-relaxation mechanism [50–54]. SO coupling is a well-known phenomenon in atomic physics as well as in semiconductors and is in general characterized by an interaction term of type $H_{SO} = \sum_{i,j} a_{ij} \hat{l}_i \hat{\sigma}_j$ with $\hat{l}$ the angular momentum operator and $\hat{\sigma}$ the spin operator of the electron; the sum runs over all pairs $i, j = x, y, z$. In the case of a crystal with bulk inversion asymmetry (BIA) such as GaAs and InAs SO coupling is of Dresselhaus type [55]. Similarly
Rashba SO coupling results from asymmetry along the z-direction (SIA, structural inversion asymmetry) [56]. For a 2DEG the spin-orbit coupling can then be written as

$$H_{SO} = \beta(-p_x\sigma_z + p_y\sigma_y) + \alpha(p_x\sigma_y - p_y\sigma_x)$$ (3.18)

where \(\beta\) reflects the strength of the Dresselhaus SO coupling and \(\alpha\) the Rashba SO interaction. In [54] the difference between the effect of Dresselhaus and Rashba SO coupling on QD spin relaxation are discussed.

SO interaction leads to spin-orbital admixed states which can weakly couple to phonons leading to an effective spin-phonon reservoir coupling of type

$$\hat{H}_{\text{ph,eff}} = \hbar \sum_q g_{\text{phon}}(\hbar \epsilon_Q D_{q\downarrow}^\dagger c_{QD,\downarrow} + c.c.)$$ (3.19)

Here \(\omega_s = c_s|q|\) with \(c_s\) the speed of sound. The effective spin-phonon coupling \(g_{\text{phon}}\) depends on the strength of SO interaction, the electron-phonon coupling strength as well as the phonon density of states at the Zeeman energy.

The resulting spin relaxation rate is a function of magnetic field and is given by

$$\kappa_{\text{phonon}} = \frac{(g_e \mu_B B)^5}{\hbar(\hbar\omega_0)^4} \Lambda_p$$ (3.20)

where \(\hbar\omega_0\) is the quantization energy for electrons and \(\Lambda_p\) a dimensionless constant describing the strength of the piezoelectric coupling. The \(B^5\) dependence valid for \(E_{Z,e} \gg kT\) becomes replaced by \(B^4 \cdot kT\) when \(E_{Z,e} \ll kT\) due to the Boltzmann factor in (3.32), [50, 52].

In addition to this first mechanism there are ways of direct spin-phonon coupling which turn out to be orders of magnitude weaker than the admixture mechanism described above [50]. In [51] the spin-flip rates due to different other phonon related mechanisms are estimated which all depend on electronic Zeeman splitting i.e. magnetic field as \(\sim B^5\).

Beyond spin relaxation by emission or absorption of a single phonon, two-phonon processes in conjunction with SO interaction are predicted to dominate at small magnetic fields. In that case a phonon with wavevector \(p\) is scattered into a phonon with wavevector \(q\) with energy conservation \(\hbar c_s|p - q| = E_{Z,e}\). These two-phonon rates have characteristically strong temperature dependence, estimated to be in the
range T^7 to T^11 [51, 52].

Alternatively it has been proposed that phonons together with the hyperfine-induced mixing of the Zeeman s-levels lead to relaxation of the QD spin. As already mentioned in the hyperfine subsection this mechanism is inefficient and the resulting rate is predicted to depend on the external magnetic field as \( \sim B^3 \) [40]; according to the calculations presented in this reference the rate will be less than \( \kappa \sim 1 \text{s}^{-1} \) at a magnetic field of 1 Tesla when considering the larger quantization energy in our QDs.

### 3.3 Ground-state optical transitions of the singly-charged dot

#### 3.3.1 Four-level model

![Figure 3.3](image)

**Figure 3.3:** (a) Four-level system in magnetic field along the growth direction. (b) After transformation an effective \( \Lambda \)-system becomes visible.

A singly-charged QD is described as a four-level system with two ground states and two excited states, coupled by two vertical optical transitions, as shown in Fig.3.3(a). The ground state \(| \uparrow \rangle \ (| \downarrow \rangle \) with angular momentum projection \( m_z = +1/2 \) \((m_z = -1/2)\) is coupled to an excited state (trion state) formed out of two electrons in a singlet and a heavy hole \(| \uparrow \downarrow \rangle \ (| \downarrow \uparrow \rangle \) with spin projection \( m_z = +3/2 \) \((m_z = -3/2)\), according to optical selection rules by \( \sigma^+ \) (\( \sigma^- \)) polarized optical transitions. The states are defined as
3.3. Ground-state optical transitions of the singly-charged dot

\[ |\uparrow\rangle = e_{QD,+1/2}^{\dagger}|0\rangle \]
\[ |\downarrow\rangle = e_{QD,-1/2}^{\dagger}|0\rangle \]
\[ |\uparrow\rangle = h_{QD,+1/2}^{\dagger}|0\rangle \]
\[ |\uparrow\downarrow\rangle = e_{QD,-1/2}^{\dagger}e_{QD,+1/2}^{\dagger}h_{QD,+3/2}^{\dagger}|0\rangle \]

where \( e_{QD,\sigma}^{\dagger}\) (\( h_{QD,\sigma}^{\dagger}\)) is the operator that creates an electron (hole) in the QD with spin \( \sigma \) along the z-axis and \( |0\rangle \) is the vacuum (empty dot) state.

All four states undergo different Zeeman shifts when an external DC magnetic field along the z-axis is applied, leading to Zeeman splitting of the optical transitions. A \( \sigma^+ \) polarized laser field is introduced at Rabi frequency \( \Omega_R \) and detuning \( \Delta \omega = \omega_0 - \omega_L \) with \( \omega_0 \) the frequency of the trion transition and \( \omega_L \) the laser frequency. If only a \( \sigma^+ \) polarized laser field is present, the trion state with \( m_s = -3/2 \), i.e. \(|\uparrow\downarrow\rangle\), is inactive since the coupling strength is reduced by a factor exceeding \( 10^3 \) at magnetic fields larger than 60mTesla, due to a combination of selection rules and, in the presence of a magnetic field, optical detuning. As we shall discuss shortly, the weak spontaneous emission to the other spin ground state cannot be neglected due to its long lifetime.

Thus, the system reduces to three levels; its quantum dynamics is fully described by the corresponding optical Bloch equations. These are obtained from a density matrix approach.

The system Hamiltonian reads

\[ \hat{H} = H_{\text{Zeeman}} + H_{\text{int.,rad}} + H_{\text{spin-reservoir}} \]  

Then by tracing over the reservoir we obtain the master equation for the system (reduced) density operator \( \dot{\rho} \)

\(^5\)From the measured light polarization in our experiment we obtain, that the unwanted \( \sigma^- \) component is suppressed by a \( 25\times \) factor. An additional factor comes from the optical detuning with respect to the diagonal transition in magnetic field, with the laser being on the strong transition. At \( B =60\text{mTesla} \), the detuning leads to a further suppression \( \sim 1/50 \), altogether suppressing the unwanted excitation of the weak transition by more than \( 1200\times \), increasing with magnetic field. At fields less or on the order of the hyperfine field, spin ground states are strongly mixed leading to all possible (vertical as well as diagonal) couplings in the four-level scheme, thus creating two differently polarized \( \Lambda \)-systems with equal decay rates to both ground states. The presence of the (unwanted and mainly suppressed) \( \sigma^- \) polarized light would just increase the bidirectional optical spin pumping, which in any case takes place at roughly equal rates, by a small amount. In these considerations, heavy-light hole mixing has been neglected; for more details we refer to Section 3.3.4.
\[
\frac{d}{dt} \hat{\rho} = \frac{1}{i\hbar} \left[ \hat{H}_0, \hat{\rho} \right] + \hat{L}_{\text{relaxation}} \tag{3.23}
\]

The term \( \hat{H}_0 = H_{\text{Zeeman}} + H_{\text{int,rad}} \) describes the unitary dynamics and \( \hat{L}_{\text{relaxation}} \) results from the interactions with reservoirs.

In the following we will discuss the different ingredients of this master equation. With a magnetic field along the z-axis \( \mathbf{B}_{\text{ext}} = \mathbf{B}_z = (0, 0, B_z) \), the total magnetic field at the QD is

\[
\mathbf{B} = \mathbf{B}_z + \mathbf{B}_N \tag{3.24}
\]

where the nuclear magnetic field (second term) is only seen by the electron spin, but not the hole spin. The Zeeman Hamiltonian then reads using (3.10)

\[
H_{\text{Zeeman}} = H_{Z,e} + H_{Z,h} \tag{3.25}
\]

\[
H_{Z,e} = g_e \mu_B \hat{\mathbf{B}} \cdot \hat{\mathbf{\sigma}} = \hbar \Omega_H(t) \hat{\sigma}_z + \hbar \omega_z \hat{\sigma}_z
\]

\[
H_{Z,h} = g_h \mu_B B_z \cdot \hat{\mathbf{\sigma}}
\]

with \( \Omega_H(t) \) as defined in (3.12). In addition,

\[
\hbar \omega_z = g_e \mu_B (B_z + B_{N,z}(t)) \tag{3.26}
\]

The interaction with the radiation field in semi-classical form is

\[
H_{\text{int,rad}} = \hbar \Omega_R \left( e^{i\Delta \omega \tau} \epsilon_{QD,-1/2}^\dagger \epsilon_{QD,+3/2} + \text{h.c.} \right) \tag{3.27}
\]

Here, the laser detuning is \( \Delta \omega = \omega_0 - \omega_L \) and the Rabi frequency is \( \Omega_R \). For simplicity we use the notation
\[ |1\rangle = |\downarrow\rangle \]
\[ |2\rangle = |\uparrow\rangle \]
\[ |3\rangle = |\uparrow\downarrow\rangle \]

\( H_0 \) can be written as

\[
\hat{H}_0 = \hbar \begin{pmatrix}
\omega_z & \Omega_H(t) & 0 \\
\Omega_H(t) & 0 & \Omega_R \\
0 & \Omega_R & \omega_0 - \omega_1
\end{pmatrix}
\] (3.29)

We note here that a separation of timescales, i.e. \( \Omega_H(t) \approx \Omega_H \) can be done, since the time evolution of \( \Omega_H(t) \) is much slower than all the timescales over which the system reaches steady-state.

\( \hat{H}_{\text{spin-reservoir}} \) refers to the interaction of the QD spin with the thermal reservoirs of electron spins and phonons.

\[
\hat{H}_{\text{spin-reservoir}} = \hat{H}_{\text{charge}} + \hat{H}_{\text{ph,eff}}
\] (3.30)

In our three-level model, each spin-reservoir coupling is treated as an incoherent relaxation rate \( \kappa_i \), coupling states \( |1\rangle \leftrightarrow |2\rangle \) bidirectionally. The total rate of spin relaxation, identical with the inverse of the spin \( T_1 \) time (\( \kappa^{-1} = T_1 \)), is the sum of all contributions \( \kappa_i \). We note that all \( \kappa_i \) depend on external magnetic and/or electric field.

\[
\kappa = \sum \kappa_i = f(V_g, B_{ext}) = \kappa_{\text{colunnel}}(V_g, B_{ext}) + \kappa_{\text{phonon}}(B_{ext}) + \kappa_{\text{exp}}
\] (3.31)

Here, we have included a term that describes an experimentally induced spin relaxation rate \( \kappa_{\text{exp}} \). Unless specified otherwise, this relaxation is absent, but can be invoked by large-amplitude gate voltage modulation in electron cycling experiments as discussed in Section 3.4.

After adding the relaxation terms due to the coupling to the thermal bath of radiation field modes (spontaneous emission terms) at rate \( \Gamma \), the relaxation terms
in the Lindblad form are [57]

\[
\hat{L}_{\text{relaxation}} = \frac{\Gamma}{2} \left( 2\hat{\sigma}_{23} \hat{\rho}_{32} - \hat{\sigma}_{33}\hat{\rho} - \hat{\rho}\hat{\sigma}_{33} \right) \\
+ \frac{\kappa}{2} \hat{n} \left( 2\hat{\sigma}_{12} \hat{\rho}\hat{\sigma}_{21} - \hat{\sigma}_{22}\hat{\rho} - \hat{\rho}\hat{\sigma}_{22} \right) \\
+ \frac{\delta}{2} \left( \hat{n} + 1 \right) \left( 2\hat{\sigma}_{21} \hat{\rho}\hat{\sigma}_{12} - \hat{\sigma}_{11}\hat{\rho} - \hat{\rho}\hat{\sigma}_{11} \right)
\]  

(3.32)

Here, \( \hat{\sigma}_{ab} = \langle a \rangle \langle b | \) is the projection operator, \( \Gamma \) is the spontaneous radiative decay rate of the optical transition and \( \kappa \) the total spin relaxation rate. At temperatures smaller or comparable to the electronic Zeeman splitting \( kT < E_{Z,e} \), a Boltzmann factor \( \hat{n} = 1/(\exp(\hbar\gamma B/kT) - 1) \) needs to be taken into account which leads to thermalization of the electron spin, i.e. in the absence of light \( \rho_{11}/\rho_{22} = \exp(-E_{Z,e}/kT) \), where \( E_{Z,e} \) is the electronic Zeeman energy. In the case of exchange coupling, the \( \hat{n} \) terms cannot be regarded as an occupancy; it can however be shown that a similar factor appears in the co-tunneling rate (3.17) when Zeeman splitting is taken into account.

We note, that coupling of the trion states due to hole spin relaxation has been neglected here. This issue is discussed in Section 3.4.7.

The optical Bloch equations are derived from the master equation (3.23); including rotating-wave approximation and taking the limit of \( E_z \ll kT \) which eliminates the Boltzmann factors, the optical Bloch equations read using the basis states introduced in (3.28)

---

\( ^6 \) Including Zeeman splitting, relation (3.17) can be written in the form \( \kappa_{\uparrow\downarrow} = \exp(-\hbar\omega_z/(2kT)) \int \text{d}A(c)\tilde{f}(c) \) and similarly \( \kappa_{\downarrow\uparrow} = \exp(\hbar\omega_z/(2kT)) \int \text{d}A(c)\tilde{f}(c) \) with \( \tilde{f}(c) = ([1 + \exp(c + \hbar\omega_z/(2kT))] + 1 + \exp(c - \hbar\omega_z/(2kT))]^{-1} \). In (3.32), the \( \kappa\hat{n} \) term has to be replaced by \( \kappa_{\uparrow\downarrow} \), whereas the \( \kappa(\hat{n} + 1) \) is replaced by \( \kappa_{\downarrow\uparrow} \). As a consequence, the ratio of spin-up versus spin-down state occupation is governed, as one would expect, by the Boltzmann factor \( \kappa_{\uparrow\downarrow}/\kappa_{\downarrow\uparrow} = \exp(-\hbar\omega_z/(kT)) \)
3.3. Ground-state optical transitions of the singly-charged dot

\[
\begin{align*}
\frac{d}{dt} \rho_{11} &= i\Omega_H (\rho_{12} - \rho_{21}) + \gamma_{hm} \rho_{33} - \kappa (\rho_{11} - \rho_{22}) \\
\frac{d}{dt} \rho_{22} &= i\frac{\Omega_R}{2} (\rho_{23} - \rho_{32}) + i\Omega_H (\rho_{21} - \rho_{12}) + \Gamma \rho_{33} + \kappa (\rho_{11} - \rho_{22}) \\
\frac{d}{dt} \rho_{33} &= i\frac{\Omega_R}{2} (\rho_{32} - \rho_{23}) - (\Gamma + \gamma_{hm}) \rho_{33} \\
\frac{d}{dt} \rho_{12} &= i\frac{\Omega_R}{2} \rho_{13} + i\Omega_H (\rho_{11} - \rho_{22}) - \kappa \rho_{12} \\
\frac{d}{dt} \rho_{13}' &= i\frac{\Omega_R}{2} \rho_{12} - i\Omega_H \rho_{23}' + \left( -\frac{\Gamma + \gamma_{hm} + \kappa}{2} - i\delta \omega \right) \rho_{13}' \\
\frac{d}{dt} \rho_{23}' &= i\frac{\Omega_R}{2} (\rho_{22} - \rho_{33}) - i\Omega_H \rho_{13}' + \left( -\frac{\Gamma + \gamma_{hm} + \kappa}{2} - i\delta \omega \right) \rho_{23}'
\end{align*}
\]

with

\[
\rho_{13} = \rho_{13}' e^{i\omega_L t} \\
\rho_{23} = \rho_{23}' e^{i\omega_L t}
\]

We have \( \rho_{31}' = \rho_{13}' \), \( \rho_{21}' = \rho_{12}' \), \( \rho_{32}' = \rho_{23}' \), and \( \rho_{11} + \rho_{22} + \rho_{33} = 1 \).

3.3.2 Dressed states and rate equation description of spin pumping

In order to gain a better understanding of our 3-level system we apply a dressed-state formalism. Hereafter, we will see that the 3-level system eventually is a \( \Lambda \)-system; with some approximations it allows us to capture the main features of the spin dynamics in the form of rate equations. We choose a new basis

\[
\begin{align*}
|\tilde{1}\rangle &= |\tilde{1}\rangle = \cos \phi |1\rangle - \sin \phi |2\rangle \\
|\tilde{2}\rangle &= |\tilde{2}\rangle = \sin \phi |1\rangle + \cos \phi |2\rangle \\
|\tilde{3}\rangle &= |\tilde{3}\rangle = |3\rangle
\end{align*}
\]

with \( \phi = \Omega_H / \omega_\phi \). In order to simplify the calculations we put the constraint \( \phi \ll 1 \) and only take into account first-order terms in \( \phi \). The transformation which diagonalizes the coupling to the quasi-static nuclear (Overhauser) field can then be written as
The off-diagonal terms due to $\Omega_H$ have been eliminated and it is obvious from the Hamiltonian (3.36) that both ground states couple to the excited state via an optical transition. The spontaneous emission terms are given by

$$S_{\text{res}} = \hbar \left( \begin{array}{ccc} \omega_z & 0 & \Omega_{R,1} \\ 0 & 0 & \Omega_{R,2} \\ \Omega_{R,1} & \Omega_{R,2} & \Delta \omega \end{array} \right)$$

(3.36)

For the new projection operator $S\sigma_{23}S^\dagger$ we obtain

$$S\sigma_{23}S^\dagger = \phi \sigma_{ij} + \sigma_{ij}$$

(3.38)

and the conjugate relation. Here, $\sigma_{ij} = \langle i | j \rangle$. Using this with the previous relation, we obtain
\[ S_{\text{relaxation,relaxations}} = \frac{\gamma}{2} (2\sigma_{13} \tilde{\rho} \sigma_{31} - \sigma_{33} \tilde{\rho} - \tilde{\rho} \sigma_{33}) \]

where \( \gamma = \phi^2 \Gamma \) and \( \Gamma = \Gamma \). \( \sigma_{ij} = |i\rangle \langle j| \) is the projection operator acting on \( \tilde{\rho} \). The first, \( \gamma \) term corresponds to relaxation via a weak optical transition induced by the hyperfine field, allowing for spin-flip Raman events and the second, \( \Gamma \), describes relaxation via the strong optical trion transition.

The last term describes coherence induced by the spontaneous relaxation into a superposition of dressed-basis ground states at a rate proportional to the occupation of the excited state \( \tilde{\rho}_{23} \). When multiplying with \( \langle 2 \rangle \) from the left and \( |1\rangle \) from the right we obtain

\[ \frac{d}{dt} \tilde{\rho}_{21} = -2\phi \Gamma \tilde{\rho}_{33} \]

The same relation is obtained for \( \tilde{\rho}_{21} \) when multiplying with \( \langle 1 \rangle \) and \( |2\rangle \) respectively. The transformed \( \kappa \) terms keep the Lindblad form and we obtain for \( E_{Z,c} \ll kT \)

\[ \tilde{L}_{\text{relaxation,relaxations}} = \frac{\kappa}{2} \left[ (2\tilde{N}_{21} \tilde{\rho} \tilde{N}_{12} - \tilde{M}_{11} \tilde{\rho} - \tilde{\rho} \tilde{M}_{11}) \right. \]

\[ + (2\tilde{N}_{12} \tilde{\rho} \tilde{N}_{21} - \tilde{M}_{22} \tilde{\rho} - \tilde{\rho} \tilde{M}_{22}) \]

with \( \tilde{N}_{21} = S \sigma_{21} S^\dagger, \tilde{N}_{12} = \tilde{N}_{21}^\dagger, \tilde{M}_{ii} = S \sigma_{ii} S^\dagger \) and \( \kappa = \kappa \).

The result of the transformation is shown in Fig.3.3(b): A single laser that interacted with the \( \sigma^+ \) trion transition is now represented by two laser fields coupling states \( |\tilde{1}\rangle \) and \( |\tilde{3}\rangle \) (|2\rangle and |3\rangle, respectively). Effectively, the system can be decomposed into two quasi-two-level systems with its own spontaneous emission rates, Rabi frequencies and effective laser detunings. We summarize for the \( |\tilde{1}\rangle \leftrightarrow |\tilde{3}\rangle \) subsystem:

\[ \tilde{\gamma} = \phi^2 \Gamma \]

\[ \tilde{\Omega}_{R,1} = \phi \Omega_R \]

\[ \Delta \omega_1 = \Delta \omega + \omega_z \]
And for the $|2\rangle \leftrightarrow |3\rangle$ subsystem:

$$\Gamma = \Gamma$$

$$\tilde{\Omega}_{R,2} = \Omega_R$$

$$\tilde{\Delta}\omega_2 = \Delta\omega$$

(3.43)

In this first order approximation, the couplings of the dressed $|2\rangle \leftrightarrow |3\rangle$ system are the same as for the bare $|2\rangle \leftrightarrow |3\rangle$ transition.

In the following we will discuss the properties of the transformed three-level system with a resonant laser on the $|2\rangle \leftrightarrow |3\rangle$ subsystem, i.e. under the condition $\Delta\omega = 0$: We start out with the system being in state $|2\rangle$ and we ignore state $|1\rangle$. After an intermediate time $t_0$ given by $\tilde{\gamma}^{-1} \gg t_0 \gg \Gamma^{-1}$, the laser field induces a steady-state occupation of the excited state $\tilde{\rho}_{33}(t_0)$. Further, for times much longer than $(\tilde{\rho}_{33}(t_0)\tilde{\gamma})^{-1}$ the system can also be found in state $|1\rangle$.

The net effect of this spin-flip Raman process is a transfer of occupation from state $|2\rangle$ to state $|1\rangle$. We will refer to this process as optical spin pumping (OSP), due to its similarity to experiments performed with atoms [26]. Further, we note that a scheme that uses OSP for spin state preparation had been proposed in [58], and state preparation has been experimentally demonstrated in [37].

In order to obtain the OSP rate that transfers the system from state $|2\rangle$ to state $|1\rangle$ under the presence of a resonant laser ($\Delta\omega = 0$), we assume the system is in state $|2\rangle$. As already mentioned, $\tilde{\gamma}$ represents a weak escape channel only, and we can work with the steady state occupation for the trion state $|3\rangle$, which is [24]

$$\tilde{\rho}_{33}(t = \infty) = \frac{\Omega_{R,2}^2}{\Gamma^2 + 2\Omega_{R,2}^2}$$

(3.44)

The OSP rate from state $|2\rangle$ to $|1\rangle$ then is the trion steady state occupation times the spontaneous emission rate into the weakly allowed channel:

$$R_{2 \rightarrow 1} = \tilde{\rho}_{33}(t = \infty) \cdot \tilde{\gamma}$$

(3.45)

The $|1\rangle \leftrightarrow |3\rangle$ sub-system has a spontaneous emission rate of $\tilde{\gamma}$, however the relaxation of its excited state is governed by a strong escape channel which is determined by the $\Gamma$ rate. The rate of transfer from $|1\rangle$ to $|2\rangle$ is [24]
\[ R_{1\rightarrow 2} = \frac{\Omega_{R,1}^2 \tilde{\Gamma}}{4\omega_z^2 + \tilde{\Gamma}^2} \approx \frac{\Omega_{R,1}^2 \tilde{\Gamma}}{4\omega_z^2} \] (3.46)

Using (3.42) to (3.46), the time-averaged ratio of ground state occupations is obtained under the condition that \( \kappa \ll R_{1\rightarrow 2}, R_{2\rightarrow 1} \)

\[ \frac{\tilde{\rho}_{22}}{\tilde{\rho}_{11}} = \frac{R_{1\rightarrow 2}}{R_{2\rightarrow 1}} = \frac{\tilde{\Gamma}^2 + 4\Omega_R^2}{4\omega_z^2 + \tilde{\Gamma}^2} \approx \frac{\tilde{\Gamma}^2}{4\omega_z^2} \] (3.47)

The approximation on the right hand side is valid in the limit of a weak incident beam \( (\Omega_R \ll \tilde{\Gamma}) \) and a Zeeman splitting largely exceeding the trion decay rate \( (\omega_z \gg \tilde{\Gamma}) \). Under these conditions, from (3.44) follows that \( \tilde{\rho}_{33}(\infty) \ll 1 \), such that together with \( \text{tr}(\tilde{\rho}) = 1 \) we can safely assume

\[ \tilde{\rho}_{11} + \tilde{\rho}_{22} \approx 1 \] (3.48)

and we obtain

\[ \tilde{\rho}_{22}(t = \infty) \approx \frac{1}{1 + \frac{4\omega_z^2}{\tilde{\Gamma}^2}} \] (3.49)

In the case \( R_{1\rightarrow 2} \ll \kappa, R_{2\rightarrow 1} \) the ratio of ground state occupations is

\[ \frac{\tilde{\rho}_{22}}{\tilde{\rho}_{11}} = \frac{\kappa}{R_{2\rightarrow 1} + \kappa} \] (3.50)

and together with (3.48) we obtain

\[ \tilde{\rho}_{22}(t = \infty) \approx \frac{1}{2 + \zeta} \] (3.51)

\[ \zeta = \frac{\tilde{\Gamma}}{\kappa} \cdot \frac{\Omega_R^2}{\tilde{\Gamma}^2 + 2\Omega_R^2} \]

Hence in the case of fixed laser intensity i.e. constant \( \Omega_R^2 \), the spin-state occupations are determined by the ratio of OSP rate versus spin relaxation rate.
3.3.3 Numerical Studies

The derived formalism considers a static randomly oriented nuclear field. Within a measurement time $B_N$ changes $\sim 100$ times. In order to calculate measurable quantities such as linewidths and peak heights as functions of electric and magnetic field we have thus performed numerical simulations: For a given set of parameters the steady-state solutions of the optical Bloch equations as given in Section 3.3.1 are numerically evaluated, in particular $\text{Im}(\rho_{23}(\infty))$ is linked to the absorption (details in Section 3.4). A fluctuating hyperfine field is implemented by pulling three random numbers $B_{N,i}$ following (3.5). From $B_{N,xy}$, using (3.11), state-mixing strength $\Omega_H$ (3.12) and pure Zeeman splitting $\omega_z$ are calculated before evaluating the density matrix steady-state. This procedure is repeated in order to average over $\sim 100$ random settings of the hyperfine field. In the cases the simulation could not be performed throughout the whole parameter space, we confirmed in key regimes that results agree well with that of a static Overhauser field with equal magnitude in $x, y, z$: From (3.5) and (3.12) we obtain for the rms-value of $\Omega_H(t)$

$$
\langle \Omega_H^2(t) \rangle = \left( \frac{g_e \mu_B}{\hbar} \right)^2 \frac{\langle B_{xy}^2(t) \rangle}{4} = \left( \frac{g_e \mu_B}{h} \right)^2 \frac{B_{\text{muc}}^2}{2}
$$

(3.52)

Here, the assumption for the observed absorption $\Theta$ to be made is

$$
\langle \Theta (B_{N,xy}^2(t)) \rangle \approx \Theta (\langle B_{N,xy}^2(t) \rangle)
$$

(3.53)

i.e., the averaging over the absorption strength for different settings of the hyperfine field approximately equals the strength of absorption for the average field magnitude, equal in $x, y, z$.

3.3.4 Hole mixing

Valence-band mixing, as described by the Luttinger Hamiltonian [59], is a well-known feature in quantum wells. Similarly, it is expected to play a role in quantum dots. With valence-band mixing, a heavy hole acquires a small contribution of light holes and vice versa such that the effective hole state as it was defined in (3.21) has the form

$$
| \tilde{h} \rangle_{\text{mix}} = (h_{QD,+3/2}^\dagger + \varepsilon_+ h_{QD,+1/2}^\dagger + \varepsilon_- h_{QD,-1/2}^\dagger) |0\rangle
$$

(3.54)
with $|\varepsilon_{\pm}| \ll 1$. Pseudopotential calculations for self-assembled InAs QDs yield admixtures on the order of a few percent [60]. As it has been pointed out in [4] valence-band mixing would have a major impact upon the effective optical selection rules by introducing a diagonal relaxation channel between states $|3\rangle$ and $|1\rangle$ due to the admixed light hole component of state $|3\rangle$. Two cases have to be distinguished: First, the mixing contribution associated with $\varepsilon_+$ further leads to an effective coherent laser coupling in addition to the coupling induced by hyperfine interaction (\(\propto \frac{\mu_i \omega}{\omega_2} \Omega_2\)) at a detuning $\Delta \omega + \omega_2$ as shown in Fig. 3.3(b). Second, the $\varepsilon_-$ part essentially only appears as a relaxation channel without coherent laser coupling, as the dipole moment of this linearly polarized transition lies along the propagation axis of the laser beam and therefore cannot be excited. Hence the following diagonal relaxation terms are added to equation (3.32) \(^7\)

$$\tilde{L}_{\text{relax, hm}} = \frac{\gamma_{hm}}{2} (2\sigma_{13}\rho \sigma_{31} - \sigma_{33}\rho - \rho \sigma_{33})$$

(3.55)

with $\gamma_{hm} = |\varepsilon|^2 \Gamma = (|\varepsilon_+|^2 + |\varepsilon_-|^2) \Gamma$.

This diagonal rate leads to OSP in a way similar to the $\tilde{\gamma}$ channel enabled by hyperfine interaction. The main difference between hyperfine-induced OSP and valence-band mixing induced OSP is that the first one is magnetic field dependent as discussed previously, and the latter is not: the valence-band mixing strength $\varepsilon$ is expected to be independent of magnetic field, as long as the Zeeman splitting is much smaller than the heavy-light hole splitting ($\Delta_{hl} > 10\text{meV}$), which is true for all realistic experimental magnetic fields. Since the hyperfine-induced OSP rate drops with magnetic field $\tilde{\gamma} \propto B^{-2}$, hole-mixing induced OSP should dominate at high fields. From our measurements at high magnetic fields (Fig.3.9(b)) we extract a $\gamma_{hm}^{-1}$ of $2 \pm 0.8 \mu$s which yields a hole mixing strength of $|\varepsilon| \sim 2.2\%$. This $|\varepsilon|$ value is indeed much smaller than 1, but we also expect the exact value to vary from one QD to another.

We note that a slightly tilted external magnetic field would yield identical dynamics in the absence of any hole mixing since it would lead to mixing of electronic states induced by the in-plane component of the applied field. These two fundamentally different mechanisms are experimentally indistinguishable for a fixed magnetic field orientation. Therefore we repeated our experiments as a function of sample tilt under a magnetic field. For a $\pm 1.5^\circ$ coverage of tilt in all directions our measurements yielded no observable change in the measured quantity $\gamma_{hm}$. Hence, we can safely

\(^7\)This can be seen from $\langle \uparrow \downarrow |\text{int mix} \mid H_{\text{int, rad}} \mid \downarrow \rangle \neq 0$ as opposed to $\langle \uparrow \downarrow |H_{\text{int, rad}} \rangle = 0$. 

3.3. Ground-state optical transitions of the singly-charged dot 49
state that the inherent hole mixing in our QDs indeed dominates over small-angle tilt-induced mixing of electronic spin states.

### 3.4 Single dot absorption spectroscopy with resonant laser

#### 3.4.1 Experimental method

The experimental and theoretical aspects of the transmission measurements have already been discussed in Section 1.6 and the absorption of a QD two-level system has been given by equation (1.24). In order to link this to the absorption of the 3-level system of the singly-charged QD we use the effective A-system picture as described in the previous Section. With a resonant laser and large external magnetic field along the z-axis i.e. \( \Delta \omega = 0, \omega_z \gg \Gamma \gg \dot{\gamma} \), the \( \tilde{2} \leftrightarrow \tilde{3} \) subsystem with strong spontaneous emission rate \( \bar{\Gamma} \) acts as the main scattering source. In the \( B \to \infty \) limit only the strong transition contributes to light scattering; since in our dressed-state transformation we assumed \( \phi \ll 1, \Omega_H \ll \omega_z \), we neglect the contribution of the \( \tilde{1} \leftrightarrow \tilde{3} \) transition.

Since in this limit light can only be absorbed if the \( \tilde{2} \leftrightarrow \tilde{3} \) transition is possible, the amount of time-averaged absorption is proportional to the time-averaged occupation of the bright spin state \( \tilde{p}_{22}(\infty) \). Hence we can perform a relative measurement of \( \tilde{p}_{22}(\infty) \) for varying magnetic field and gate voltage but keeping laser power and detuning constant.

For an absolute measurement of \( \tilde{p}_{22}(\infty) \) we still need a calibration point, i.e. an experimental value of \( \Theta \) for a known \( \tilde{p}_{22}(\infty) \). In the absence of an external magnetic field the spin ground states can be considered to be fully mixed due to the in-plane part of the Overhauser field leading to \( \tilde{\gamma} \sim \Gamma \) and a branching ratio of \( \eta = 1 \). As a consequence the \( |11\rangle \leftrightarrow |33\rangle \) and the \( |22\rangle \leftrightarrow |33\rangle \) transitions equally contribute to light scattering and fast back and forth OSP takes place, leading to a fully randomized spin i.e.

\[
\tilde{p}_{11}(t = \infty, B = 0) = \tilde{p}_{22}(t = \infty, B = 0) = \frac{1}{2}
\]

and we obtain

\[
\tilde{p}_{22}(t = \infty, B, V_g) = \frac{1}{2} \frac{\Theta(B, V_g)}{\Theta(B = 0, V_g = V_0)}
\]

(3.56)

(3.57)
3.4.2 Optical spin pumping

Fig. 3.4(a) shows absorption on resonance on the blue Zeeman transition as a function of magnetic field normalized to on-resonance absorption at 0 Tesla i.e. $\Theta(B_{\text{ext}})/\Theta(B_{\text{ext}} = 0)$. The gate voltage was kept in the plateau center i.e. in a regime where $\kappa_{\text{tunnel}}$ is minimal. The inset shows the corresponding raw laser scans for 0 T (top) to 300 mT (bottom). The zero positions of the probe laser detuning has been readjusted in the graphs to compensate the Zeeman splitting.

Absorption drops by nearly two orders of magnitude over the plotted range of $B_z = 0$ to 300 mT. With a resonant laser in the weak excitation limit and Zeeman splitting much larger than the trion transition linewidth i.e. $\Delta \omega = 0$, $\omega_z \gg \tilde{\Gamma} \gg \tilde{\gamma}$ equation (3.49) yielded

$$\tilde{\rho}_{22}(t = \infty) \approx \frac{1}{1 + \frac{4\omega_z^2}{\tilde{\Gamma}^2}}$$

all provided that the spin relaxation rate $\kappa \ll R_{2\rightarrow1}, R_{1\rightarrow2}$ which we can safely assume for low magnetic fields [40][51] and strongly suppressed exchange coupling in the gate voltage plateau center. Consistent with (3.49), drop of absorption follows a $B^{-2} \propto \omega_z^2$ law indicated by the dashed line. For fields less than 100 mT (see Fig.
3.4(a)) the approximations included in (3.49) do not hold any more and $\tilde{\Gamma} \sim \tilde{\gamma}$. Without any approximation the steady-state solutions of the optical Bloch equations are evaluated (solid line) numerically; they are in excellent agreement with our data at all magnetic fields. The Rabi frequency $\Omega$ in units of $\Gamma$ for a given incident laser power can be independently determined by saturation spectroscopy and power broadening measurements. The radiative lifetime $\Gamma^{-1} = 0.8$ns used in our simulation is based on a measurement in as-grown dots [25].

### 3.4.3 Electron cycling

Given that the OSP rates $R_{1-2}$, $R_{1-2}$ and the spin relaxation rate $\kappa$ are unknown the experimental data shown in Fig. 3.4(a) do not reveal direct quantitative information about $\tilde{\gamma}$. However, the branching ratio can be extracted using an rms-coherent coupling $\langle \Omega_H^2(t) \rangle$ given in (3.52)

$$\eta = \frac{\tilde{\gamma}}{\Gamma + \tilde{\gamma}} = \frac{\langle \Omega_H^2(t) \rangle}{\omega_2^2} = \frac{B_{mic}^2}{2B_z^2}$$

(3.58)

$\eta$ is equivalent to the probability that the system decays via the $\tilde{\gamma}$-channel when excited into a trion state.

To determine $\eta$ we applied a large square-wave modulation (Amplitude 80mV peak-peak) at different frequencies to the gate which in every cycle first loaded another electron of opposite spin into the QD forming a singlet together with the QD electron. Then one of the electrons was forced to leave, and as the tunneling probability for each of the two electrons is equal the remaining QD spin was fully randomized. The advantage of this technique which we will refer to as electron cycling leads to enforced spin relaxation at a known rate $\kappa_{exp}$ (also refer to equation (3.31)). In the case $\kappa \approx \kappa_{exp} \gg R_{1-2}$ i.e. enforced spin relaxation rate exceeds the optical back-pumping rate $R_{2-1}$ and $\tilde{\gamma}$ can be determined by a fit using equations (3.50, 3.51).

Fig. 3.5(a) shows absorption normalized to on-resonance absorption in the cotunneling regime; the data was obtained with electron cycling for different modulation frequencies and laser powers at a fixed external magnetic field $B_z = 300$ mT. The upper, green points correspond to $\kappa = 54.3$kHz, the middle, red point to $\kappa = 19.3$ kHz, and the lower, blue points to $\kappa = 3.3$ kHz.

Using (3.56) and (3.51) the absorption ratio shown in the Figure can be written as
3.4. Single dot absorption spectroscopy with resonant laser

Figure 3.5: (a) Electron recycling measurements at three different frequencies. (b) Check experiment as described in the text. (c) Intensity dependence at $B_z=0\text{T}$ and $B_z=100\text{ mT}$ without electron recycling.

\[
\frac{\theta_{\text{in plateau}}}{\theta_{\text{cotunnel}}} = \frac{\rho_{22,\text{in plateau}}}{\rho_{22,\text{cotunnel}}} = \frac{2}{2 + \zeta} \tag{3.59}
\]

The grey lines are fits using this expression. Best match with the data can be obtained with a total OSP rate $\gamma_{\text{tot}}(300\text{mT})=\gamma(300\text{mT})+\gamma_{hm}=1.6\mu\text{s}^{-1}$, where the two contributions stem from nuclear spins and hole mixing, respectively. The hole mixing-induced contribution can be independently determined from high-magnetic field measurements to be $\gamma_{hm}^{-1} = 2\mu\text{s}$. Using the branching ratio (3.58) with $\tilde{\gamma}^{-1} = 0.8\text{ns}$ we then solve for the rms-nuclear magnetic field and obtain $B_{\text{nuc}} = 15\text{mT}$ which is in good agreement with (3.9).

Fig. 3.5(b) shows a measurement that demonstrates the difference between electron cycling (large amplitude modulation) and in-plateau (small amplitude) modulation: For in-plateau modulation we do not observe absorption (the noise level is marked by the horizontal dashed line). In contrast, when large amplitude modula-

---

8Due to our square-wave modulation scheme inducing a non-exponential spin decay the conversion from the known modulation frequency to $\kappa_{\text{exp}}$ rate does not need to include a factor of $2\pi$. This has been confirmed by comparing the result of a numerical simulation using square wave-shaped spin relaxation to the outcome of a simple rate equation model including exponential decay of spin.
tion is applied absorption is partially recovered due to forced spin relaxation at a controlled rate $\kappa_{\text{opt}}$, as shown by the red peak in the figure. Fig.3.5(c) was obtained in the plateau center without electron cycling technique, showing on-resonance absorption as a function of incident laser power. At 100 mT (blue squares) relative absorption is one order of magnitude weaker than at 0 mT (red circles) due to OSP. In both cases absorption exhibits weak dependence on laser power. This dependence arises from being in the vicinity of laser power required for saturation, hence a deviation from the assumptions of (3.49).

### 3.4.4 Peak shift in the plateau center

Fig.3.6 shows laser scans obtained at $B_z=150$mT on the blue (a) and red (b) Zee-man transition throughout the whole single-electron plateau. Absorption strength is color-coded. The line tilt is due to the quantum confined Stark effect and pixelization is due to experimentally limited voltage resolution. At gate voltages 395 mV and 480 mV the two co-tunneling regimes show strong absorption when spin relaxation is fast due to charge reservoir coupling (for details we refer to Section 3.2.2). In the plateau center hyperfine interaction dominates and leads to spin pumping.
3.4. Single dot absorption spectroscopy with resonant laser

and drop of absorption as already discussed. Here, we further observe a shift of the spectral position of the absorption peak in the plateau center as compared to the co-tunneling regime. This shift is directed to the red (blue) for the blue (red) Zeeman transition. This resembles effects one might expect for dynamical nuclear spin polarization (DNSP) [32, 61]; these effects can however be excluded. Our numerical simulation is able to reproduce this behaviour without taking into account DNSP as shown in Fig.3.6(c) and (d), using the parameters $\Omega = 0.6 \Gamma$, $\Gamma^{-1} = 0.8 \text{ ns}$, $B_{\text{m}} = 15 \text{ mT}$, $\gamma_{\text{m}}^{-1} = 2 \mu\text{s}$. The lineshift in the plateau center is $\pm 0.9 \text{ GHz}$ for both red and blue transition which is close to the electronic Zeeman splitting at $150 \text{ mT}$, $E_{Z\pm} \approx 1.3 \text{ GHz}$. At the co-tunneling edges $\kappa$ is large and maximum of absorption is observed when the laser is exactly on resonance with the transition. When $\kappa$ is small as it is the case in the plateau center absorption of a strictly resonant laser is suppressed due to spin pumping depending on the external magnetic field. When the laser frequency is moved towards the center between the strong $\tilde{\Gamma}$ and the weak $\tilde{\gamma}$ transitions i.e. the spectral detuning with respect to the $\tilde{\gamma}$ transition is reduced, the back-pumping at rate $R_{1 \to 2}$ becomes more efficient and maximum of absorption will be reached for a spectral detuning that fulfills the condition $R_{1 \to 2} = R_{2 \to 1}$. As a consequence both transitions contribute to absorption which leads to a shift of the absorption maximum towards the weak $\tilde{\gamma}$ transition i.e. a blueshift when the red line is observed and vice versa.

3.4.5 Peak broadening at plateau edges

Fig. 3.7(a) shows example laser scans for different magnetic fields $B_z$ ranging from 0 to 1 Tesla, obtained in the co-tunneling regime where spin relaxation is fast. The scans have been laterally shifted in order to eliminate the Zeeman shift. In (b) the measured linewidths are plotted as a function of magnetic field (red circles) along with a calculated curve (solid line). A broadening to almost double the zero-field linewidth appears at magnetic fields between 60 mT and 80 mT; at higher fields linewidth becomes as narrow as in the case $B_z = 0$.

As a Gedanken experiment we assume transfer of polarization from light to nuclei via the electron spin under a static positive magnetic field. Then, due to negative electron g-factor the spin-down state has higher energy than spin-up state. The laser is resonant with the red (first case) or the blue (second case) Zeeman transition. In the first case electron spin is pumped to the spin-up state which leads to nuclear spin-up polarization after an electron-nuclei flip-flop interaction. As the hyperfine constant $A$ is positive the energy of the electron spin-down state is consequently lowered while the trion states remain untouched, leading to a blue-shift of the red Zeeman optical transition which is being observed. The same happens in the second case where spin-down nuclear polarization is created and the energy of the spin-up state is lowered, again leading to a blue-shift of the observed transition. This scenario is clearly opposite of what we observe, thus ruling out the presence of an efficient DNSP.
The physical reason for the observed broadening is very similar to that described in the previous Section 3.4.4: Both $\gamma$ and $\Gamma$ transitions contribute in a non-negligible way to absorption and maximum is observed when $R_{1,2} = R_{2,1}$ condition is fulfilled. Consistently the linewidth increases as much as the electronic Zeeman splitting initially but drops at magnetic fields where $\Gamma(B) \gg \gamma(B)$ and a single transition is established. In contrast to Section 3.4.4, $\kappa$ is large here due to co-tunneling and annihilates the absorption drop caused by spin pumping hence making the transition visible at all magnetic fields. The solid line is a calculated curve using a randomly fluctuating Overhauser field with $B_{\text{over}} = 15\text{mT}$, well reproducing this feature. Other calculation parameters were $\Omega = 0.6\Gamma$, $B_{\text{ext}} = 15\text{mT}$, $\Gamma^{-1} = 0.8\text{ns}$, $\gamma_{\text{hm}}^{-1} = 2.0\mu\text{s}$, $\kappa^{-1} = 2.5\mu\text{s}$. We note that in order to put as many constraints as possible on the choice of simulation parameters we have used the maximum co-tunneling-induced spin relaxation rate extracted from the data shown in Fig.3.8(b). The effect of $\gamma_{hm}$ on the simulation is negligible, advocating that the dominant OSP mechanism is hyperfine interaction.
3.4. Single dot absorption spectroscopy with resonant laser

3.4.6 Coupling to electron spin reservoir

We have performed laser scans as a function of gate voltage throughout the whole single-electron plateau as defined in Section 3.2.2. The measured on-resonance absorption signal for each laser scan is plotted in Fig. 3.8(a). The data has been obtained at an external magnetic field of $B_z=300 \text{ mT}$ taking coarse voltage steps.

At gate voltages lower than 540 mV and higher than 625 mV as marked by the shaded regions absorption drops below noise level indicated by the horizontal dashed line. At these voltages the QD either becomes empty (left of point A) or doubly charged (right of point B). Absorption then vanishes since in those cases the QD is not described by the trion level system any more; the optical transitions for these gate voltages are not observed within our scanning window of 30 GHz around the trion transitions. The unshaded part indicates the region where the QD contains a single electron and as it has been mentioned before the co-tunneling rate is maximum when gate voltage is at the crossover points A or B. Here, relaxation via co-tunneling is faster than the optical pumping rates $\kappa_{\text{column}} > R_{12}, R_{21}$ leading to thermalization of the electron spin and thus strong absorption. The scenario drastically changes when gate voltage is tuned to the center of the plateau. Here, co-tunneling rate $\kappa_{\text{column}}$ reaches its minimum where our numerical calculation...
predicts a drop of as much as five orders of magnitude (also see Fig. 3.2) compared to the crossover points such that \( \kappa_{\text{co-tunnel}} \ll R_2 \). Consequently the occupation of the spin states is governed by OSP (equation (3.49)) rather than Boltzmann factor meaning that the spin is predominantly in the dark state and vanishing absorption is observed.

The semilogarithmic plot in Fig. 3.8(b) shows a voltage fine scan of the low voltage plateau edge around the A crossover point obtained at \( B_z = 300 \text{mT} \). The gate voltage for point A is different from Fig. 3.8(a) as this data was taken on another QD. The observed absorption drops by half within a gate voltage detuning of \( \pm 5 \text{mV} \) from the maximum position. This data demonstrates the enormous gate-voltage dependence of this spin relaxation mechanism. The gray solid line is a best-fit numerical simulation using expression (3.17) as spin relaxation rate showing good accordance with the data. The co-tunneling rate at the maximum of the peak as determined from the fit is \( \kappa_{\text{max}}^{-1} = 2.5 \mu \text{s} \). The noise level is indicated by the dashed line; it deviates from the one shown in (a) due to different experimental settings such as lock-in time constants and filter slopes. Again on the left side of the peak the QD is empty, yielding vanishing absorption below the noise level. The gradual decrease of absorption is due to finite temperature. On the right side the spin pumping regime is located; here some weak absorption remains according to the occupation of the observed spin state, revealing the strength of spin pumping.

### 3.4.7 Coupling to phonon reservoir

Based on the theoretical estimates of Section 3.2.3 we now seek for signatures of SO-phonon induced spin relaxation at high magnetic fields. Fig.3.9(a) shows 2D plots of color-coded absorption strength as a function of laser detuning and gate voltage for four different magnetic fields obtained for the red Zeeman transition. The scans cover the whole single-electron plateau; excitonic Zeeman shift has been eliminated by shifting the y-scale for each 2D graph separately. The linear dependence of the excitonic transition energy on gate voltage is due to the quantum confined stark shift.

At 0 Tesla absorption is clearly visible throughout the whole plateau due to fast spin flips with the neighboring nuclear spins. When a small magnetic field (\( B = 0.1 \text{T} \)) is applied, absorption in the plateau center drops because of hyperfine-induced OSP as discussed in the previous sections. Close to the plateau edges absorption still remains due to fast co-tunneling. At 0.5 Tesla increasing OSP leads to further drop of absorption. These absorption characteristics in the plateau center remain the
same up to 5 Tesla, however absorption starts to come back at even higher fields: when the magnetic field is raised up to 9.9 Tesla a significant recovery of plateau-center absorption is observed. This effect cannot be explained by OSP which only causes monotonous decrease of absorption nor by co-tunneling which is negligible in the plateau center and hardly shows any magnetic field dependence. Owing to its $B^5$ dependence however, phonon-assisted spin relaxation is a good candidate to be responsible for the observed effects in the context of spin relaxation mechanisms.

Fig.3.9(b) shows the quantitative evolution of normalized absorption with magnetic field i.e. the ratio of absorption in the plateau center versus co-tunneling regime (black data points). Further, the solid red line along with the gray shaded region indicate calculated strength of absorption for $\gamma_{hm}^{-1} = 2\mu$s with an uncertainty of $\pm0.8\mu$s; the phonon-induced spin relaxation rate was $\kappa_{\text{phonon}} = \alpha_0 B^5$ with the coefficient $\alpha_0 = 0.031$ in units of $[T^{-5}\text{sec}^{-1}]$. Whereas $\kappa_{\text{phonon}}$ is strongly $B$-dependent, the hole mixing contribution $\gamma_{hm}$ has no $B$-field dependence within the magnetic field range considered here. Therefore these two mechanisms have distinguishable effects on Fig.3.9(b) and thus can be identified independently. The good agreement with the experimental data strongly suggests that in this regime of electric and magnetic fields the dominant spin relaxation is indeed phonon-assisted. Fur-
ther, within our uncertainty $\kappa_{\text{phonon}}$ matches well with the results that have been previously obtained on an ensemble of self-assembled InAs/GaAs QDs [5].

There are two fundamentally different mechanisms which employ holes to yield OSP: First, hole mixing of strength $\varepsilon$ leads to an admixture of the light hole states to the trion states as discussed in section 3.3.4. Second, hole-spin relaxation leads to an incoherent coupling of the trion states contributing to OSP. In [62] hole spin relaxation rate is predicted to be below $10^3$/sec and monotonically increase with magnetic field which suggests that it is not the main mechanism responsible for OSP. We therefore neglect hole spin-flips, further assuming that there is no other efficient hole spin flip mechanism at low magnetic fields. In the first mechanism, OSP is independent of magnetic field and the strength is equal to the hyperfine-induced OSP rate at $\sim$1 Tesla. At higher magnetic fields the hyperfine-induced OSP rate drops with $B^{-2}$, therefore hole mixing becomes the dominant OSP mechanism here.

### 3.5 Full interaction map

![Figure 3.10](image)

**Figure 3.10:** Calculated absorption maxima for the whole single-electron plateau, plotted as a function of magnetic and electric field. (a) Laser resonant with red Zeeman transition. (b) Same, but for blue Zeeman transition.

In the previous sections the three dominant mechanisms acting on the confined spin have been identified separately along with the two mechanisms for OSP. In this final part we will present results of our numerical simulation based on parameters
that have been measured or estimated before. The calculations have been performed within a parameter space approximately overlapping with the full scale of our experimental tuning ability of the static electric and magnetic fields. For details of the simulation we refer to Appendix 3.3.3.

Fig. 3.10 shows calculated maximum values of absorption for the red (a) and the blue (b) trion transition with a laser having the corresponding circular polarization. Absorption strength is color-coded in logarithmic scale as a function of gate voltage detuning and external magnetic field.

All of the following points have been discussed in the previous chapters; here we mention them briefly as a key to the plots: the necessary conditions for observing strong absorption are either $\kappa \gg R_{1-2}, R_{2-1}$ or $R_{1-2} \sim R_{2-1}$. Further, at large magnetic fields $B > 8$ Tesla when $E_{Z,E} \sim kT$ the Boltzmann factor leads to a difference of the spin ground state occupations and thus a difference between absorption strength on the red and blue Zeeman transition.

In the plot we distinguish three different regimes of strong absorption:

1. **Magnetic fields lower than the fluctuations of the hyperfine field ($B \leq 15$ mTesla).** Here, fast bidirectional OSP due to hyperfine-induced state mixing leads to strong absorption.

2. **High magnetic fields ($>5$ Tesla).** Here, $\kappa_{\text{phonon}}$ induces fast thermalization i.e. $\kappa_{\text{phonon}} \gg R_{1-2}, R_{2-1}$. The spin ground state occupation is mainly determined by the Boltzmann factor leading to a lowering (increase) of absorption on the higher (lower) energy spin state occupation (a) ((b)).

3. **Large gate voltage detunings from the plateau center ($\pm 40 mV$).** Here, cotunneling ($\kappa_{\text{cotunnel}}$) is responsible for fast spin relaxation and appearance of absorption.

An intriguing feature that becomes apparent now is the blue *island* in the center of the color-coded spin relaxation plot. It marks the regime where absorption (i.e. all reservoir interactions) is suppressed by five orders of magnitude, or in other words the localized spin becomes maximally isolated hence the frequently used concept of an artificial atom is meaningful. Within the scope of quantum information processing this indicates the relevant regime of operation.
3.6 Summary and Conclusion

We have investigated the dominant interactions of a confined electron spin in a single self-assembled QD by optical means and demonstrated the regimes where each reservoir coupling becomes important. For magnetic fields $B \lesssim 1$Tesla, the dominant contribution to OSP stems from the fluctuating hyperfine field mixing the electronic spin states and creating a weak channel for diagonal relaxation in the trion four-level picture. Exchange and phonon-induced spin-flip processes dominate over hyperfine-induced spin pumping and establish a thermal steady-state at large gate-voltage detunings and/or large external magnetic fields; in the plateau center at intermediate magnetic fields the situation is reversed and spin pumping dominates, strongly altering the state occupations away from thermal equilibrium values. Signatures of heavy-light hole mixing dominated spin cooling can be observed for fields $\gtrsim 5$Tesla.

From a quantum control perspective these results demonstrate that the quantum dynamics of a single confined spin can be significantly altered by externally controlled parameters such as electric and magnetic fields. A natural extension of this study would be the investigation of spin decoherence in a single QD using similar optical techniques. These measurements would require more advanced schemes such as electromagnetically-induced transparency (EIT). Further, knowledge gained on single-electron spin dynamics can be utilized in the resonant optical study of more complex systems such as coupled QDs or QDs with a single excess heavy-hole.
4 Faraday Rotation from a single spin

Ability to read-out the state of a single confined spin lies at the heart of solid-state quantum information processing [1]. While spin measurements using Faraday rotation (FR) of light polarization have been implemented in semiconductor spin ensembles [63][64][65], single spin read-out has only been achieved using transport measurements[28][9]. Here, we demonstrate an all-optical dispersive measurement of the time-averaged spin state of a single electron in a quantum dot (QD). We obtain information on the spin state through conditional FR of a spectrally detuned laser, induced by the polarization- and spin-selective trion (charged QD) transitions. To assess the sensitivity of the technique, we use an independent resonant laser for spin-state preparation [37]. We infer that there are ~10 spin-flip Raman scattering events (i.e. back-action) within our measurement timescale. Therefore, straightforward improvements such as incorporating solid-immersion lenses [66][67] and higher efficiency detectors would allow for back-action evading spin measurements, without the need for a cavity.

This chapter is based on publication [3]
4.1 Motivation and Background

Theoretical proposals based on FR from a microcavity-embedded QD have suggested that QND [68] measurement of a single spin could be implemented [69][70]. Remarkably, our observations suggest that the back-action evading spin measurement in the basis defined by the Pauli operator \( \sigma_z \) (the strong confinement axis) could be realized even in the absence of an optical cavity enhancing the FR. We estimate that the QD scatters a photon every 2.2 microseconds; the role of these events is to leak state information into the field reservoir, thus, inducing back-action on the \( \sigma_z \) observable and collapsing the state into a \( \sigma_z \) eigenstate. The spin-flip Raman scattering events, which lead to back-action on the measured observable (\( \sigma_z \)), occur once every 2 to 20 milliseconds. Even though it is impossible to avoid Rayleigh scattering within a measurement time yielding a signal-to-noise (SNR) ratio exceeding unity, spin-flip scattering can be negligible if the ratio of the peak absorption cross-section to the laser area (\( \sigma_0/A_L \)) considerably exceeds the ratio between the rates of allowed versus forbidden transitions i.e. the branching ratio \( \eta \) (given in (3.58)) into the spin-flip scattering channel. We have determined that the branching ratio in self-assembled QDs is between \( \eta \approx 10^{-3} \) to \( 10^{-4} \) and is primarily due to hyperfine-induced state mixing.

Spin measurements could also be carried out using resonance fluorescence or absorption experiments. However due to background Rayleigh scattering and limited collection efficiency, the timescales needed to measure the spin state via resonance fluorescence technique exceed the spin-flip timescales. Resonant absorption measurements are likewise limited by electrical noise that dominates over the shot noise for laser intensities that do not saturate the QD transitions.

Figure 4.1 shows noise and signal-to-noise ratio (SNR) for a DT experiment on resonance as a function of laser intensity. At low powers the noise is limited by the preamplifier stage \( n_p \) indicated by the green line. In contrast, for high laser power > 500nW, photocurrent shot noise \( n_s \) (blue line) dominates and the total noise given by \( n_{tot}^2 = n_p^2 + n_s^2 \) is indicated by the yellow curve. The SNR curve (dashed, gray line) has been obtained using the total noise together with a typical saturation curve similar to that shown in Fig.1.10.

In order to reach shot noise limit, power has to be much higher than the power needed to achieve maximum SNR. In other words: at the point where SNR is maximum and hence highest measurement speed is achieved the total noise has still a large contribution from the constant preamplifier noise. A potential way to overcome this limitation is the use of large off-resonant laser powers where shot noise
4.2. Polarization-dependent response from a QD

Consider a polarization-selective detection scheme similar to the one shown in figure 4.2 detecting in a linear basis $e_x, e_y$. It is convenient to work with Jones vectors, i.e.

$$E = \begin{bmatrix} E_x \\ E_y \end{bmatrix}$$ (4.1)

The response of a QD with a spin-up electron to an excitation field polarized along the x-direction is $\sigma^+$ polarized.

Figure 4.1: Noise and signal-to-noise for the resonant DT experiment as used in the previous chapters.

clearly dominates over electronic preamplifier noise. This situation can be realized in a Faraday rotation experiment all provided that additional laser fluctuations can be neglected.

4.2 Polarization-dependent response from a QD

...
After rotation by 45° we obtain
\[
E_L = E_L \begin{bmatrix} 1 \\ 0 \end{bmatrix} \quad \text{and} \quad E_{\text{qd}} = E_{\text{qd}} \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ i \end{bmatrix}
\] (4.2)

Hence the QD fields' x- and y-polarized components carry different phase factors which enter in the term with \( R_c \) of (1.20). Both the x and y polarized total fields thus carry an equal mixture of dispersive (real part of \( \sigma_{ge}(\infty) \)) and absorptive (imaginary part) QD response and the corresponding term in (1.20) reads after removing all non-phase factors and taking \( \phi_G = -i \)

\[
R_c \left[ \frac{1 \pm i}{2} \sigma_{ge}(\infty) \right] \quad \text{instead of originally} \quad R \left[ i \sigma_{ge}(\infty) \right]
\] (4.4)

where the \(+\) (\(-\)) sign appears in the x(y)-polarized component of the total electric field. The power on the two detectors can then calculated from the total field \( E_{T,x} \) and \( E_{T,y} \) in the x- and y-polarized channel. Similar to section 1.6 the normalized transmission for both polarizations \( T_x/T_{x,\text{off}} = 1 - \Theta_x \) and \( T_y/T_{y,\text{off}} = 1 - \Theta_y \) have

**Figure 4.2:** Polarization-sensitive detection scheme for Faraday rotation experiment.
4.2. Polarization-dependent response from a QD

to be evaluated:

\[
\Theta_x = \frac{\sigma_0}{A_L} \frac{\Gamma^2/4 + \Gamma(\omega_0 - \omega)/2}{4(\omega_0 - \omega)^2 + \Gamma^2 + 2\Omega^2} \\
\Theta_y = \frac{\sigma_0}{A_L} \frac{\Gamma^2/4 - \Gamma(\omega_0 - \omega)/2}{4(\omega_0 - \omega)^2 + \Gamma^2 + 2\Omega^2}
\]

(4.5)

Here a factor of 1/2 has been added to account for the mismatch between the linear excitation and the QD circular polarization. The sum then leads to the well-known absorptive result whereas the difference is dispersive

\[
\Theta_+ = \Theta_x + \Theta_y = \frac{\sigma_0}{A_L} \frac{\Gamma^2/2}{4(\omega_0 - \omega)^2 + \Gamma^2 + 2\Omega^2} \\
\Theta_- = \Theta_x - \Theta_y = \frac{\sigma_0}{A_L} \frac{\Gamma(\omega_0 - \omega)}{4(\omega_0 - \omega)^2 + \Gamma^2 + 2\Omega^2}
\]

(4.6) (4.7)

The difference corresponds to a rotation of the linear polarization with the angle given by

\[
\beta[\text{rad}] = \frac{1}{2} \Theta_- = \frac{\Delta T_x - \Delta T_y}{T_{x,\text{off}} + T_{y,\text{off}}}
\]

(4.8)

Hence, when the excitation polarization is chosen properly, absorption and dispersion coexist in an optical field’s response to a spectrally detuned optical transition. While these responses are of comparable strength for small detunings, dispersive response dominates over the absorptive part as the detuning is increased. Measurement of the dispersive response provides information about the ground (spin) state, if the transition is spin-selective with optical selection rules, as is the case for a QD confining a single excess electron [37][34]. In this Chapter, we demonstrate a measurement of a QD spin by detecting this dispersive response through FR of a far-detuned linearly polarized laser.

A single electron confined in a QD presents a four-level system in the trion representation as it had been introduced in 3.3. The ground state consists of an electron in | \uparrow \rangle (| \downarrow \rangle ) state. The excited state | \uparrow \downarrow \rangle (| \uparrow \uparrow \rangle ) corresponds to the QD with two electrons forming a singlet and a hole with angular momentum projection \( J_z = +3/2(-3/2) \) along the growth direction. The trion transitions are circularly polarized as determined by the optical selection rules. If a linearly polarized laser, e.g. \( \sigma^\pm \), feels this transition, the \( | \sigma^- \rangle (| \sigma^+ \rangle ) \) polarization component acquires a phase shift rotating the laser’s polarization by an angle \( \beta (-\beta) \) in the linear basis. Owing to Pauli Blockade [4], only one of these transitions is available at any given time and the laser polarization is rotated in positive or negative angular direction.
conditional on the electron's spin state. In our results we exploit precisely this spin-state-dependence of the laser polarization.

Figure 1b illustrates our detection scheme. A polarized laser propagates through a gated heterostructure incorporating QDs. A polarizing beam splitter distributes the transmitted light into two linear polarizations in the rectilinear basis of \((X, Y)\) and directs each arm to a photodiode. Along with each detector's output \((T_x \text{ and } T_y)\), such a configuration allows us to measure their sum and difference simultaneously. The sum \((\Theta_+)\) normalized to the total incident laser intensity indicates a change of photon number in the total light, yielding the absorptive response, while the difference of the two normalized signals \((\Theta_-)\) is linked to the polarization rotation arising from the dispersive response.

In the absence of external magnetic field, the trion transition has twofold spin degeneracy. Therefore, while we fully observe the absorptive response in the summed detector signals, the dispersive response is exactly cancelled due to the fast spin-flips induced by the hyperfine interaction [8]. Under an external magnetic field however, the Zeeman splitting \((E_Z)\) lifts the spectral degeneracy of the two spin-selective trionic transitions. Consequently, the dispersive response is expected to appear along with the absorptive part in the spectral vicinity \((\Delta = \omega_0 - \omega \ll E_Z)\) as it had been shown in figure 4.3.

Figure 4.3 shows how the sum \(\Theta_+/2\) (black circles) and difference \(\Theta_-/2\) (red circles) signals from one of the Zeeman-split transitions behave as a function of laser polarization basis, when an external magnetic field of 1 Tesla is applied along the strong confinement axis of the QD. The transitions are split by 26 GHz, which is \(\sim 60\) times larger than the total transition linewidth \(\Gamma\), and the laser’s response in the near vicinity of the \([\sigma^-]\)-polarized transition linked to spin-down electronic state is displayed. When the laser is also circularly polarized (left plot), it acquires an overall phase that can not be detected leading to a purely absorptive signal (black circles). Same scan with a linearly polarized laser (right plot) displays the dispersive response (red circles) alongside the absorptive (black circles): since the acquired phase is now relative, it leads to a linear polarization rotation. The solid gray lines are fits using (4.7) for the dispersive QD response. Both curves in the case of linear excitation have \(\sigma_0/A_L = 0.14\%\), \(\Omega^2_R\), and \(\Gamma = 466\text{MHz} \times 2\pi\), as obtained from line-broadening in saturation spectroscopy, where FWHM = \(2\sqrt{(\Gamma/2)^2 + \Omega^2_R/2}\). At such small detuning with respect to Zeeman splitting, the laser experiences FR primarily due to one Zeeman transition nearby. The data presented here is obtained in the cotunneling regime to avoid spin pumping [37]. We note here that similar dispersive response signatures of single QDs have recently been reported [16]. In contrast to the
work presented here, the dispersive signals observed there are polarization and spin independent and arise from an interference effect that depends on the QD position with respect to the laser focal spot in a planar Fabry-Perot cavity.

4.3 Off-resonant Spin Measurement

We now consider probe-laser detunings that are larger than Zeeman splitting ($\Delta \gg E_Z$), where the difference signal arises from a competition between the two transitions. Figure 4.4 shows the difference signal when the probe laser is blue-detuned by 20 GHz to 40 GHz with respect to the $|\sigma^+\rangle$-polarized transition obtained in a 60-sec measurement timescale per point. The black circles display the difference (offset) signal when the preparation laser is left detuned from the two Zeeman transitions: in this case, no state-preparation is implemented and the electron spin-state is effectively thermalized being close to a completely mixed state; since spin-flip events occur on timescales short compared to our measurement timescale.

As gate voltage is increased, the Zeeman-split optical transitions experience an equal strength of DC-Stark shift. Consequently, the detuning of the probe laser with respect to the two transitions is also decreased, creating an offset signal in accor-
dance with the incommensurate detunings and partial cancellation of the Faraday rotations. The red circles display the difference signal when the weak preparation laser is in the near-vicinity of the $|\sigma^-\rangle$-polarized Zeeman transition. At 415 mV gate voltage the preparation laser hits resonance and the electron is spin cooled to the spin-up state with near-unity fidelity due to state-mixing induced spin-flip Raman transitions. Therefore, we no longer observe the difference of two dispersive responses, rather the full signal due to one spin-state. Figure 2b is a similar measurement when the preparation laser is tuned to resonance with the $|\sigma^+\rangle$-polarized Zeeman transition again at 415 mV gate voltage, preparing the electron in the spin-down state. The full dispersive signal from a single electron spin is recovered, but now with opposite sign indicating FR in the opposite direction. The amplitude of this signal is less than that of figure 4.4(left) in accordance with the additional detuning of $E_Z = 26\text{GHz}$ due to Zeeman splitting. Beyond 450 mV gate voltage, the QD charging state switches from one excess electron to two electrons forming a spin-singlet ground state. Since the trionic transitions are no longer present beyond this point, the laser experiences no dispersive response. Figure 4.5 shows the 60-second time-averaged FR angle obtained from the dispersive signal for various probe laser and preparation laser detunings. The red (blue) circles correspond to the FR angle in response to the spin-up (spin-down) prepared state at 30 GHz (56 GHz) detuning. The red (blue) squares correspond to the FR angle for a probe detuning of 66 GHz (92 GHz). The corresponding grey circles (squares) indicate the FR angle when the electron spin is left in a thermal mixed state. The white circles indicate the signal

**Figure 4.4:** Faraday rotation for spin-up state (red circles) and spin-down state (blue circles) averaged over a probe laser detuning of 30 GHz to 45 GHz. Black circles: Preparation laser far-off resonance.
Figure 4.5: FR angle obtained for various probe- and preparation-laser detunings.

level when the QD has a spin singlet of two electrons. The dashed curves indicate the theoretically expected FR angle for each case studied experimentally. The hatched areas mark the gate voltage values for which the quantum dot is charged with two electrons. The yellow and orange areas are a guide to the eye. As expected, the FR angle reaches positive (negative) maximum value when pure spin-up (-down) state is enforced. When the electron spin is left in a thermal mixed state, the value of FR angle is reduced for probe-laser detunings much larger than Zeeman splitting (56 GHz and 92 GHz vs. 26 GHz, respectively).

Figure 4.6(a) displays a full map of the dispersive signal at a 100-msec measurement timescale per data point, plotted as a function of gate voltage and probe-laser detuning when the preparation laser is $\sigma^+$-polarized. The center frequency of the probe laser is detuned 92 GHz ($\sim$200 times $\Gamma$) for the top figure and 56 GHz for the bottom figure. Once again, 415 mV-gate voltage marks the preparation of spin-
Figure 4.6: Dispersive signal as a function of gate voltage and probe laser detuning. At 415 mV the electron is prepared in the spin-down state (left plot) and spin-up state (right plot), respectively. The Faraday rotation angle at 415 mV is displayed in the middle plot along with the theoretically expected behavior.
ratio of $\eta = 10^{-4}$ and $10^{-3}$, respectively). This limitation arises from the fact that our optical system has a numerical aperture of 0.5 and a photo-detector efficiency of 10\%: under these conditions, $(\sigma_0/A_L)$ can not safely exceed the branching ratio. By using a combination of index-matched solid-immersion lenses ($n_{\text{GaAs}}^2 \sim 12$) and commercially available higher efficiency photo-detectors [66], achieving a $\sim$100-fold improvement of $(\sigma_0/A_L)$ is plausible. It would then be possible to effectively eliminate spin-flip Raman scattering from the probe laser within the anticipated measurement time of $\sim$10 msec. In fact, based on the predicted phonon [53][5] and co-tunneling [33] limited spin-flip times, we could envision resolving spin quantum jumps with these improvements. Alternatively, incorporating gated structures into photonic crystal nano-cavities [71] is demanding, but the existence of a far-detuned cavity mode can well be the way to obviate measurement back-action, at the same time allowing for coherent resonant manipulation of spins. We note that a similar time-averaged single-spin measurement has been performed, where partial spin preparation is achieved via an above-bandgap laser [72].
Faraday Rotation from a single spin
5 Outlook

5.1 Towards $N$-shot measurement of a single spin

A major challenge would be the realization of an all-optical single-shot readout similar to the experiment performed on lithographically defined QDs in [28]. Here, the findings about the Faraday rotation measurement as demonstrated in chapter 4 can be used; the condition to be satisfied is that the measurement time must be shorter than the spin relaxation time; the spin relaxation time in turn has a contribution from the natural spin relaxation determined by phonons and cotunneling as discussed in chapter 3 as well as a contribution that stems from the measurement-induced spin-flips (back-action). The most intriguing feature of such a single-shot measurement would be the appearance of spin quantum jumps i.e. instantaneous switches between two levels of the detected signal corresponding to the electron being in the spin-up or spin-down state. While it is commonly accepted that this kind of experiment could be realized incorporating a cavity to enhance the effective spin-light interaction, the challenge is to realize it without the help of complex nanostructures introducing their own technical burden. Using solid-immersion lenses (SILs)

Figure 5.1: Double-SIL assembly developed in collaboration with Boston University.

the laser spot area can be reduced and the same signal i.e. absolute rate of scattered photons can be achieved with a lower incident laser power which in turn results in a
reduction of shot noise and laser fluctuations. As a consequence a shorter measurement time $\tau_m$ is possible whereas the back-action time $\tau_{\text{back-action}}$ remains constant. In the case $\tau_m < \tau_{\text{back-action}}$ a single-shot measurement is possible; if $\tau_m \sim \tau_{\text{back-action}}$, still an $N$-shot measurement can be performed allowing for the extraction of the total spin relaxation time. In collaboration with Boston University (BU) a double-

![Figure 5.2: Power dependence using a GaAs-SIL, with and without correction for the drop of signal due to the lock-in detection scheme.](image)

...SIL assembly has been developed as shown in Fig.5.1. The hemispheric SILs are made from GaAs ($n = 3.5$) in order to achieve a small focal spot and a theoretical improvement of up to $(\times 3.5^2 = 12.25)$ is expected. Fig.5.2 shows a power sweep similar to Fig.1.10 but using a GaAs hemisphere mounted on the top surface. The contrast i.e. relative absorption is enhanced by a factor $> \times 4$ which represents the average experimental improvement for the investigated QDs, compared to the case without SIL. The improvement in contrast shows that we are on the right path, but it is still below the required level for a single-shot spin measurement.

It is technically easier and nevertheless appealing to realize a so-called $N$-shot readout (with $N > 1$). Here, a protocol as shown in Fig.5.3(left) is needed which is composed of a spin state preparation part and a consecutive measurement period where the signal is recorded as a function of time. Averaging separately every single point in time over $N$ sequences should then exhibit an exponential decay yielding the effective spin relaxation time $\tau$. At low probe laser intensities, $\tau$ should remain constant as sketched in Fig.5.3(right). In this regime it is only determined by the natural spin relaxation. In contrast, at high probe laser powers measurement-induced spin-flips should dominate. The $N$-shot experiment would complement the
indirect measurements of relaxation time shown in chapter 3 and further allow for the observation of spin relaxation at intermediate magnetic fields where relaxation time is at its maximum.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.3.png}
\caption{Left: Timing protocol for N-shot measurement. Right: Expected dependence of relaxation time on probe laser intensity.}
\end{figure}

5.2 Going further

As already mentioned, the incorporation of an off-resonant cavity is expected to reduce the number of back-action events that occur during a spin measurement down to below unity, allowing for a spin quantum jump measurement or single-shot spin readout. Further the Faraday rotation measurement can be used to create entanglement between two spins when applied to a double QD e.g. two QDs located in two separate stacked layers. The double-spin system is projected to an entangled state if the measurement outcome for the total angle of rotation due to both spins is exactly zero.

A next step following our study of spin relaxation presented in chapter 3 would be the investigation of spin decoherence in a single self-assembled QD using similar optical techniques. These measurements would require more advanced schemes e.g. using electromagnetically-induced transparency (EIT) or coherent population trapping (CPT) [73–75]; here, an external magnetic field in the QD plane would be needed to create the necessary \(\Lambda\)-system. Further, EIT could lead to understanding and control of the dynamics of the nuclear spin ensemble: the observation of a spectrally narrow EIT dip would project the quantum state of the nuclear field ensemble onto a subset leading to a narrowing of the nuclear field distribution, similar as discussed in [76].

In the context of coherent spin manipulation the findings presented here pave the way for electron-spin resonance (ESR) measurements, as well as for stimulated
Raman transition. Further, arbitrary rotations on the Bloch sphere should be possible using microwave pulses and laser pulses, when operating with repetitive timing protocols such as preparation $\rightarrow$ manipulation $\rightarrow$ detection.
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