Doctoral Thesis

Spin singlet and triplet states in a quantum dot molecule

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Publication Date:
2012

Permanent Link:
https://doi.org/10.3929/ethz-a-007339310

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Spin singlet and triplet states in a quantum dot molecule

A dissertation submitted to

ETH Zürich

for the degree of
Doctor of Sciences

presented by

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2012
Voll Blüten

Voll Blüten steht der Pfirsichbaum,
Nicht jede wird zur Frucht,
Sie schimmern hell wie Rosenschaum
Durch Blau und Wolkenflucht.

Wie Blüten gehn Gedanken auf,
Hundert an jedem Tag -
Lass blühen! laß dem Ding den Lauf!
Frag nicht nach dem Ertrag!

Es muss auch Spiel und Unschuld sein
Und Blütenüberfluss,
Sonst wär die Welt uns viel zu klein
Und Leben kein Genuss. [1]
Abstract

In this dissertation, resonant laser spectroscopy is employed to study coupled electron spins in vertically stacked self-assembled quantum dots. Quantum dots are semiconductor nanostructures with a quantum confinement potential in all spatial direction, leading to an absorption spectrum consisting of narrow lines, similar to the one of atoms. Therefore, quantum dots are well suited to study quantum mechanics at the single emitter level. But in contrast to atoms, quantum dots are embedded in the solid-state. Hence, the physics is enriched by the non-trivial coupling of single spins and charges to the quantum dot environment. Coupling two quantum dots leads to a subtle energy spectrum, enabling more elaborate applications in the so-called field of solid-state quantum optics.

Coupling between two vertically stacked quantum dots is established via electron tunneling. The tunnel-strength can be varied with an applied gate voltage, which also determines the overall number of charges in the quantum dot molecule. Nevertheless, the sample has to be designed carefully in order to be able to access the desired charging state. A sample structure is developed, which allows for charging each quantum dot of the pair with one electron respectively, yielding spin singlet and triplet ground states.

This dissertation reports two main contributions to solid-state quantum optics, both relying on spin singlet and triplet ground states. First, we realize an optical amplifier by establishing strong coupling between a coupled quantum dot and the back contact, this is a first step towards a single emitter laser. A fast one-directional relaxation rate between the ground states ensures inversion. Operating a coupled quantum dot in the opposite regime, where it is well isolated from the back contact leads to the arguable main result of this thesis; a spin quantum-bit with long spin coherence time. We employ dark-state spectroscopy to determine the coherence time of the system and the noise reservoir limiting it. We find coherence times exceeding 200 nanoseconds for measurements at the sweet spot. Here, the system is rendered to first order insensitive against magnetic and electric field fluctuations, the main dephasing source for single electrons and holes respectively.
Zusammenfassung


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Applications of nano-science become more and more prominent in our daily lives. Recent advances of nano-science in a variety of disciplines have been reported in newspapers. In material science for instance, nano-patterning of batteries could make transparent screens reality [2]. In medicine, nano-diamonds have been employed in cancer treatment of mice [3]. An interesting application of quantum dots (QDs), zero-dimensional nano-objects, are solar cells in the shape of paint [4]. Prerequisite for the development of such applications is a detailed understanding of physical mechanisms on the nanoscale. Pairs of vertically stacked self-assembled QDs are investigated in the framework of this thesis. QDs trap single charges, due to the quantum confinement potential in all spatial directions, allowing the study of physical interaction between the spin of a single electron or hole and the QD environment. The nearest environment are the atoms the QD consists of, whose nuclear spins couple to the electron spin, which is confined in the QD [5, 6, 7, 8]. Furthermore, nearby filled electron [9, 10] or empty hole [11] reservoirs can influence the spin of the trapped charges, thereby offering interesting many-body physics. Last but not least, lattice vibrations (phonons) can lead to spin relaxation [12, 13, 14, 15].

Spin relaxation and dephasing become important in quantum information processing. Here long spin coherence times [16] are prerequisite for quantum information storage. For this purpose QDs are well suited, because they only interact weakly with their environment and exhibit therefore long spin-lifetimes and spin-coherence times [17, 18, 19]. Additionally, self-assembled quantum dots can be probed and manipulated optically, enabling fast operation on a timescale of picoseconds. Moreover, many requirements on a quantum-bit (qubit), the fundamental unit of a potential quantum computer, have been met in self-assembled QDs. First, spin-initialization into a well defined spin-state has been realized in single QDs [20], whereas single-shot spin read-out could be measured in coupled QDs [21]. Ultrafast coherent manipulation of a single spin was shown in single QDs [22, 23]. In coupled QDs two-qubit gates and control of entanglement [21, 25] between two electron or hole spins could be realized. Furthermore, in coupled CQDs a subsystem can be isolated, which is insensitive to nuclear spins [24, 26] and which can be rendered insensitive to electric fluctuations. Nuclear spins are the main dephasing mechanism of electrons [23], whereas electric fluctuations affect the hole spin coherence time strongly [25, 27]. But scalability of qubits [28] is a demand, which can not be met at the moment. Patterning an array of QDs with lateral coupling between the QDs is still under investigation [29].
0. Introduction

The relatively good isolation of QDs from their environment also renders them model systems for quantum optics applications. Well understood phenomena of quantum optics, such as power broadening [56], Autler-Towns-splitting [30, 31] and coherent population trapping [19, 26] were observed.

More concrete applications of QDs and coupled QDs can be found in laser physics and non-linear optics. Single QDs serve as gain medium for lasers [32], whereas a coupled QD could be employed as gain medium for a single emitter laser [33]. In addition, the rich optical spectrum of coupled QDs could allow for parametric down conversion [34, 35].

Coupled QDs in particular enrich the means for the investigation of physical interactions. One QD could serve for instance as probe of the properties of the other QD. First steps in this direction were done with the spectroscopy of high energy states of the bottom QD by employing the top QD [36].

The variety of applications of coupled QDs is reflected in this thesis, which is organized as follows.

At the heart of the present work is resonant laser spectroscopy, used to study the spin properties of self-assembled coupled QDs (CQDs). The first chapter gives an introduction to self-assembled CQDs. The starting point is the growth of the samples and the explanation of the resulting confinement potential. Furthermore, details about possible optical excitations and charge control via embedding the CQDs in a field-effect structure will be given.

Chapter 2 concentrates on the measurement methods. To this end an overview over the different types of spectroscopy, used in the framework of this thesis, and a description of the experimental set-up will be given.

Chapter 3 covers the design and characterization of suitable samples, where each of the QDs of the pair is charged with a single electron. On the way to achieve this so-called (1,1) regime, many other problems were encountered influencing the quality of the sample. Therefore, the most important properties of the sample and methods how to verify the quality of the sample are described in more detail. Chapter 3 concludes with an introduction to the (1,1) regime.

In chapter 4, which is based on publication [33], measurements of sample 2, where the (1,1) regime is accessible will be presented. The CQDs in sample 2 are strongly coupled to the back contact, resulting in a fast one-directional relaxation rate in the ground states. Measurements prove that we can rely on the fast relaxation to create gain.

Chapter 5 is based on publication [26]. Here, the opposite regime, where the CQD is well isolated from the back contact is investigated with sample 3. Dark state spectroscopy (coherent population trapping) is used to measure the coherence time of the system. A detailed investigation of the dependence of the coherence time on electric and magnetic fields shows that the system can be rendered insensitive to both electric and magnetic field fluctuations by tuning it to the so-called sweet spot [26]. Furthermore a model is developed, which allows to determine the character of the noise reservoir and extract dephasing times by studying the dark state. At the sweet spot we find that the $T_2^*$
coherence time exceeds 200 nanoseconds.
In chapter 6, sample 1 is operated in the (3,1) regime, where the bottom QD is charged with three electrons and the top QD with one electron. Here, the lowest energy state of the top QD couples to higher energy states of the bottom QD, enabling spectroscopy of the latter ones. Furthermore by applying a magnetic field, we can manipulate the spin character of the energy states in the bottom QD.
0. Introduction
Chapter 1

Self-assembled quantum dot molecules

The experiments described in this thesis were conducted on single pairs of vertically stacked InGaAs QDs, also referred to as coupled QDs (CQDs) or QD-molecules. After an overview over different types of QDs, I will describe the growth of self-assembled CQDs, their potential and optical excitations. I will conclude with a section about the charge tunability of CQDs.

1.1 Introduction

QDs are zero-dimensional objects, since they have a strong quantum confinement potential leading to quantization in all three dimensions. Therefore, QDs serve as traps for single charge carriers. There exist different types of QDs. Fabricating metal gates on top of a 2D-electron gas allows, by applying gate voltages, for isolated puddles of electrons. They are coupled via tunneling to the electric leads and therefore most easily measured electronically. Colloidal QDs are optically active nano-crystals. Their emission wavelength is determined by their size, which is a growth parameter. Due to their chemical stability they are used as fluorescent markers in biology and medicine. Furthermore, progress in the development of solar cells and displays relying on colloidal QDs has been reported.

Better suited for quantum optics experiments are self-assembled QDs, since their optical resonances exhibit near life-time limited line-widths ($\sim 1.2 \mu$eV). It is also possible to couple two self-assembled QDs vertically as it will be described in the following section.
1. Self-assembled quantum dot molecules

InAs QDs are grown by molecular-beam-epitaxy (MBE) on a (100) semi-insulating GaAs substrate (Fig. 1.1). Strain is induced due to a lattice mismatch of 6.8% between InAs (lattice constant $a = 6.06 \, \text{Å}$) and GaAs ($a = 5.65 \, \text{Å}$). Hence, after a first period of 2D growth (wetting layer), islands form, thereby relaxing the strain. The diameter of these QDs in the plane is about 20 nm and their size in the growth direction is about 10 nm. The QDs nucleate at random positions, though defects can induce nucleation. The QDs also exhibit a spread of shape and size. Via the strength of the confinement potential the size of the QDs directly influences the transition energies of the QDs. We rely on the partially-capped-island (PCI) technique [46] to blue-shift the QD emission wavelength to the NIR, the working wavelength of silicon detectors. The accuracy of the PCI-technique in terms of transition wavelength amounts to ±10 nm. During the PCI-process the QDs are partially capped with a GaAs-layer, annealing leads than to diffusion of In, yielding QDs with reduced height (Fig 1.1b).

The second layer of QDs is grown on top of the first QD-layer, separated by a GaAs spacer-layer, that is locally strained by QDs of the first layer [45]. Therefore, the second layer QDs tend to nucleate directly on top of the QDs in the first layer, leading to the formation of CQDs. Due to the strained capping-layer, QDs in the top layer grow faster than QDs in the bottom layer. Hence, less In is needed for growing the same density of QDs in the second layer as in the first layer. The stacking probability decreases with decreasing QD-density and increasing separation of the QD-layers. One has to keep in mind that the spacer layer also influences the strength of the tunnel-coupling and can therefore not be varied arbitrarily.
1.3 Potential and optical excitations

InAs QDs are surrounded by GaAs, which has a larger bandgap. This leads to a quantum confinement potential in all three dimensions. The potential in the plane perpendicular to the growth direction is plotted in Fig. 1.2. Electrons, holes and excitons (electron hole pairs, experiencing Coulomb interaction) can be trapped in this potential. It is deeper for electrons in the conduction band (CB) than for holes in the valence band (VB), since holes have a larger effective mass. One can approximate the potential harmonically [47]. The discrete energy states of the harmonic potential are labeled as s, p, d, ... in resemblance to the atomic states. Only transitions between states of the same harmonic are allowed. All measurements of this thesis are performed on s-transitions. The p-transitions are split of from the s-transitions by $\sim 35$ meV and have shorter lifetimes than the s-transitions [48], leading to broad line-width.

The optical measurements rely on emission or absorption of a photon. In both cases the optical selection rules have to be fulfilled, meaning that spin is conserved during emission and absorption. Hence a circularly right-handed polarized $\sigma_+$ photon ($J_m = +1$) can be emitted (absorbed) by recombination (creation) of a heavy-hole with spin $J_m = +3/2$ [49] in the VB and an electron with spin $J_m = -1/2$ in the CB. Alternatively, a circularly left-handed polarized $\sigma_-$ photon ($J_m = -1$) can be emitted (absorbed) by recombination (creation) of a heavy-hole with spin $J_m = -3/2$ [49] in the VB and an electron with spin $J_m = +1/2$ in the CB.
1. Self-assembled quantum dot molecules

1.4 Charge tunable device

![Diagram of charge tunable device](image.png)

**Figure 1.3: Sample design.** Shown is the band structure of a CQD sample. The shaded gray area is the band-gap. The material of the different layers is indicated at the bottom of the figure.

Charge control over QDs is established by embedding them into a field effect structure (Fig. 1.3). A GaAs tunnel-barrier separates the first layer of QDs from the negatively Si-doped back contact, whereas a GaAs spacer-layer separates the two layers of QDs. Above the second layer a GaAs capping is grown, followed by a AlGaAs blocking barrier (∼30% Al) and a final GaAs capping. The gate voltage is applied between the back contact and a semitransparent Ti (2 nm) and Au (8 nm) top gate, evaporated after growth. The metal-semiconductor interface results in a offset voltage of ∼−700 mV.

Since the Fermi-energy is fixed to CB edge of the back contact, an applied gate voltage changes the energy of the QD states, depending on the distance of the QD-layer to the back contact. This can be understood similar to a force applied to a lever. Bottom and top QD have different distances to the back contact and therefore their energies tune differently with gate voltage. This can be exploited to bring the electron levels into resonance, enabling tunneling [37]. The strength of the tunneling interaction is determined by the thickness of the spacer-layer.

To prevent that electrons and holes tunnel at the same applied gate voltage, the two QDs deviate in wavelength. The difference in wavelength is indicated as Δλ in Fig. 1.3. Choosing the top QD (QD-R) to have lower transition energy than the bottom QD (QD-B), allows for electron tunneling in the s-shell [53]. The electronic s-states of the two QDs are in resonance at gate voltages for which the hole states are still detuned (Fig. 1.4a). In order to obtain hole tunneling, one can switch the order of the QDs. In Fig. 1.4b the transition energy of the bottom QD (QD-R) is more red detuned than the transition energy of the top QD (QD-B). But one has to keep in mind that due to
the larger effective mass of holes compared to electrons, holes exhibit longer tunneling times. Hence, samples allowing for hole tunneling are grown with thinner spacer-layers to achieve sizable tunnel-coupling. By choosing the thickness of the spacer-layer of the structure in Fig. 1.4, for electron tunneling and $\Delta \lambda \sim 10 \text{ nm}$ rather small, tunnel resonances between electrons in the s-states of the top QD and electrons in the p-states of the bottom QD can be realized.

Another advantage of the gate voltage is the control over the charging state of the QDs. Is the energy of the s or p-states lowered below the Fermi-energy, a single electron tunnels into the QD. In this way the absolute charge of samples with only one QD-layer can be controlled precisely. In CQD-sample only the overall charge, but not the exact charge distribution can be controlled with the gate voltage, since only one top gate is available. Therefore, the charge distribution depends sensitively on growth parameters, especially the spacer-layer and the exact wavelength of the QDs. Depending on the charging state of the CQD, in particular the charge distribution over the two QDs of the pair, the energy states of the CQD tune differently with the applied gate voltage. An example is given in Fig. 1.5a, where the CQDs is charged with two electrons in the ground state. In the lower panel, the energy ground states of the CQD, where two electrons reside in the bottom QD (2,0), one electron in each QD (1,1) and both electrons in the top QD (0,2) are plotted. In the (2,0) regime the energy increases with increasing gate voltage, since both electrons are located far from the top gate. The opposite is true for the (0,2) regime. Here, both electrons sit close to the gate and the energy is reduced with increasing gate voltage. The top panel of Fig. 1.5a shows the energies of the optical excited state, consisting of three electrons and one hole. The tuning behavior of the energy states can be understood in the same way as for the ground state energies. The transition energies can be calculated by taking all possible differences between excited and ground state energies. Some of the resulting transition energies do

![Figure 1.4: Electron tunneling in the s and p-states. a) Shown is the band diagram of a sample which is designed for electron tunneling of electrons sitting in the s-states. b) In this sample structure, electrons in the s-state of the top QD are coupled via tunneling to electrons in the p-states of the bottom QD.](image)
not tune with applied gate voltage and some do. First ones involve the recombination of an electron and a hole which reside in the same QD (direct exciton), latter ones the recombination of an electron sitting in the bottom QD and a hole sitting in the top QD (indirect exciton) or the other way around. Therefore the overlap of electron and hole wavefunction is reduced for indirect excitons, leading to enhanced excitonic recombination times. This effect can be employed for implementation of spin-memories \[54\]. At degeneracy of ground or excited state energies, electron tunneling takes place. Consequently anticrossings evolve with magnitude determined by the strength of the tunnel-coupling (Fig. 1.5).

Last but not least the gate voltage influences the QD-energies via the quantum confined stark effect. The Stark effect stems on the one hand from the permanent dipole moment \(p\) of the exciton and on the other hand from a induced dipole moment (polarizability \(\alpha\)). Therefore, the energy is changed as \(E = E_0 - pV - \alpha V\). Here, \(V\) is the effective electric field, seen by the QDs and \(E_0\) the unperturbed QD-energy. The electron hole-separation in growth direction is larger for indirect than for direct excitons, leading to a larger Stark-effect for indirect than for direct transitions. A Photoluminescence (PL) measurement of the red QD of a single CQD (Fig. 1.6) exhibits both direct and indirect excitons, easily identified by the magnitude of the Stark effect. Moreover, differently charged excitons can be distinguished. The positively charged exciton is labeled as \(X^{1+}\), the negatively charged exciton (trion) as \(X^{1-}\) and so on. Jumps in transition energy are visible between different types of direct excitons, leading to characteristic charging plateaus \[51, 39, 55\]. This can be attributed to Coulomb-blockade; the Coulomb

---

**Figure 1.5:** Dependence of the energy states of a CQD charged with two electron on the gate voltage. **a**) Shown are the energies versus gate voltage of the two-electron ground states (lower panel) and the corresponding excited states (upper panel) consisting of three electrons and one hole. Latter sits in the top QD, only allowing for direct recombination in the upper QD. The quantum confined Stark-effect is neglected. **b**) Same as in a, but including electron tunneling.
1.4 Charge tunable device

Figure 1.6: Plotted is a photoluminescence (PL) measurement of the red QD from a single CQD versus gate voltage. The direct excitons are indicated in the figure. Indirect excitons with weaker oscillator strength are also visible. They exhibit a stronger dipole moment.

The interaction of the participating charges influences the transitions energies.

A complete list of the samples and their important parameters is given in the table below.

<table>
<thead>
<tr>
<th>sample</th>
<th>tunnel-barrier</th>
<th>QD-B λ</th>
<th>QD-R λ</th>
<th>spacer-layer</th>
<th>capping-layer</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 i (ES1366)</td>
<td>30 nm</td>
<td>980 nm</td>
<td>970 nm</td>
<td>9 nm</td>
<td>106 nm</td>
</tr>
<tr>
<td>2 iv (ES1554)</td>
<td>50 nm</td>
<td>970 nm</td>
<td>930 nm</td>
<td>9 nm</td>
<td>212 nm</td>
</tr>
<tr>
<td>3 iii (ES1773)</td>
<td>50 nm</td>
<td>970 nm</td>
<td>940 nm</td>
<td>9 nm</td>
<td>212 nm</td>
</tr>
</tbody>
</table>

Table 1.1: List of samples.
1. Self-assembled quantum dot molecules
Chapter 2

Spectroscopy on quantum dot molecules

This chapter gives an overview of the employed spectroscopy methods and a detailed description of the experimental setup.

2.1 Spectroscopy methods

Resonant differential transmission (dT) and reflection (dR) measurements [56, 57] are at the heart of QD-spectroscopy. dT and dR can detect a change in transmission down to $\sim 0.01\%$ and their resolution can resolve the natural linewidth of QDs ($\sim 1 \mu$eV) [58]. Moreover, spin-states can be probed coherently and the resonant laser does not lead to charge build-up in the QD environment. The resolution of photoluminescence (PL) [59, 60], for instance, used to identify CQDs, is limited by non-resonantly created charge fluctuations.

Photoluminescence

PL relies on a non-resonant laser, creating charges in the GaAs or the wetting layer, which relax non-radiatively into the QD and decay under emission of photons (Fig. 2.1a). PL is employed to fully characterize the charging states of single and coupled QDs [51, 52, 37, 39] and the different types of tunnel resonances for the latter. Furthermore, PL in combination with magnetic fields revealed the spin states of multi-charged excitons [61]. Disadvantages of PL arise from the non-resonant laser excitation, which can charge defects in the QD environment [62]. Hence, the transitions energies of the QDs are shifted and broadened with respect to resonant measurements. Another effect can be ghost resonances and additional jumps in the charging plateaus. All these effects can
produce difficulties in identifying indirect excitons and charging sequences. Another difficulty is found in samples with large tunnel barriers to the back contact (Sample 2, 3). Due to long tunneling times [52] the individual charging regimes overlap (Fig. 1.6). In the present work, PL is used to verify that two QDs belong to a stacked pair by measuring anticrossings and to identify the difference in transition energies between bottom and top QD, which influences the charging sequence.

\[ |E_L + e_s|^2 = |e_s|^2 + 2|E_L e_s| + |E_L|^2. \]

Here, \( E_L \) is the electric field of the laser and \( e_s \) the electric field of the scattered light. Latter one consists of a coherent and incoherent part:

\[ e_s = e_{coh} + e_{incoh}. \]

Resonant spectroscopy

For resonant spectroscopy \[56, 63, 64\] a laser is scanned over the QD-resonance. The measured intensity is given by

\[ |E_L + e_s|^2 = |e_s|^2 + 2|E_L e_s| + |E_L|^2. \]

Therefore, there exist two different ways to measure the response of the QD resonantly. \( dT(dR) \) results from the interference of the coherent part of \( e_s \) with \( E_L \) on and off resonance, given by:

\[ dT/T \approx dR/R = \left< \frac{|E_L + e_s|^2 - |E_L|^2}{|E_L|^2} \right> \approx \left< 2\frac{|E_L e_s|}{|E_L|^2} \right> \]

with \( |e_s|^2 << |E_L|^2 \).

The weakly driven QD scatters photons according to its response function

\[ s(\delta) = E_L \sigma \frac{\delta \Gamma/2 + i (\Gamma/2)^2}{\delta^2 + (\Gamma/2)^2}. \]

\( \sigma \) is the scattering cross-section and \( \Gamma/2 \) the spontaneous emission rate. The absorption is given by the imaginary part of \( s(\delta) \); it is phase shifted by \( \pi/2 \) with respect to the
2.2 Experimental setup

dispersive real part. In addition, the Gaussian laser field acquires a phase shift of \( \pi \) while passing through the focus (Gouys phase). Therefore the absorptive part interferes with the laser field, resulting in a Lorentzian \( dT \)-profile \[63\]. Only photons emitted by the QD which have the same polarization as the laser contribute to the \( dT \) signal. A typical \( dT \) measurement of the neutral exciton in the top QD is shown in Fig. 2.2. The spectrum exhibits the characteristic Lorentzian profile, but is slightly asymmetric \[57\]. This can be attributed to a small admixture of the real part, due to a phase shift of the light emitted from the QD. The phase shift can be either accumulated in the capping layer, should its thickness not be optimized for the QD-emission wavelength, or by passing through the gate material \[63\].

\( dT \)-measurements allow for the observation of typical two-level characteristics such as power broadening \[56\] and saturation \[65\]. In the latter case, with increasing driving laser power the incoherent part of \( |e_s|^2 \) increases. Therefore, the \( dT \)-contrast decreases. Resonant fluorescence (RF) \[66\] is the other resonant spectroscopy method, used in the framework of this thesis. By suppressing the laser one can directly measure \( |e_s|^2 \). The polarization of the measured field is therefore perpendicular to the laser field and the spectrum a symmetric Lorentzian.

![Figure 2.2: \( dT \)-spectrum of the \( X^0 \) in QD-R versus gate voltage, measured with power below saturation, yielding a linewidth of 2 \( \mu \text{eV} \).](image)

2.2 Experimental setup

Resonant spectroscopy requires a small spot size (\( \sim 1 \mu\text{m} \)) and high stability. The first issue is addressed with a confocal microscope \[67\]. The resolution is given by the FWHM of the focus, which is proportional to \( \lambda/\text{NA} \). The numerical aperture of the focusing lens \( \text{NA} = n \sin(\alpha) \) with \( n \) the refractive index describes the half-angle of the maximum cone of light, which can enter or exit the objective. Since the same lens is used to collect photons (PL, RF, dR), the collection efficiency depends on the NA. The higher the NA,
the more photons can be collected. The experiments of this thesis were first performed with a lens of \( \text{NA} = 0.55 \) (chapter 6, 4) and later on with a lens of \( \text{NA} = 0.68 \) (chapter 5).

The CQD-sample is mounted on a three-axis nano-mover stack below the lens (Fig. 2.3). Nano-movers and microscope sit at the end of a \( \sim 1 \text{m} \) steel tube with a diameter of 5 cm. The tube is closed on top with a wedged glass-plate and is filled with He exchange-gas in order to cool the sample to 4.2 K. This is necessary in order to prevent temperature broadening of the resonances. Inserting the tube into a liquid He bath cryostat allows for cooling the sample without introducing vibrations. A superconducting magnetic produces magnetic field up to 7 T parallel to the growth direction of the sample (Faraday-geometry).

For \( \frac{dT}{dT} \) (\( dR \)) the signal is measured with a Si photo-diode, mounted directly underneath the sample (on the optical table). After passing a room-temperature pre-amplifier the signal is fed into a Lock-In amplifier. The Lock-In amplifier is synchronized with a function-generator, which provides the gate voltage superimposed with a rectangular modulation voltage of amplitude \( A = 100 \text{ mV} \) to the sample. The modulation voltage shifts the QD resonance via the Stark effect in and out of resonance (Stark-shift modulation spectroscopy \([68, 69]\)). This allows for Lock-In measurements in order to suppress noise on frequencies other than the modulation frequency (\( \sim 770 \text{ Hz} \)).

For PL, a 780 nm diode laser, filtered with a 800 nm short-pass filter, and for resonant measurements two Sacher TEC-500 external-cavity laser are available. Latter are tunable from \((925 - 980) \text{ nm} \). The tunable lasers are intensity stabilized with a double-pass AOM setup \([70]\) and afterwards coupled into fibers.

The optical setup of Fig. 2.3 is designed very compact to fit on top of the cryostat. It consists of two excitation and one collection arm. All three are fiber-coupled. After the collimating lens linear polarizers define the polarization. They also serve to suppress either a strong pump laser in two-laser experiments or the laser in RF. Here, the polarizer in the excitation arm is cross-polarized with respect to the polarizer in the collection arm. The extinction ratio necessary for RF amounts to \( \sim 10^6 \). Therefore the optical elements have to preserve the polarization. This can only be achieved by using a beam cube to send 50 % of the light, coming from the sample into the collection arm. Wedged beam-samplers, which are used to superimpose the two excitation lasers and to send parts of the light to a CCD camera are not suited. Previously the window closing the steel-tube, was not wedged and oscillations of the laser suppression with period proportional to the window-thickness could be observed. Therefore the straight window was exchanged with a wedged one.

The CCD camera is used for alignment as well as monitoring the QD-density. To this end a 900 nm long-pass filter is mounted in front of the CCD-camera to suppress the non-resonant laser and the wetting-layer emission.

In addition to the detector underneath the sample a detector and a fiber-coupled 750 mm grating-spectrometer are available on the optical table. For PL, the light coming from the collection arm is coupled into the spectrometer, where a 1200 g/mm blaze-grating disperses the light onto a liquid-nitrogen cooled CCD-camera.
2.2 Experimental setup

Figure 2.3: Shown is a scheme of the optical setup with the confocal microscope and the sample mounted in a LHe bath cryostat. Only about one pair of CQDs is in the focal spot of diameter 1 μm (magnified region).
2. Spectroscopy on quantum dot molecules
Chapter 3

Design and characterization of samples with the (1,1) regime accessible

This chapter gives an overview over the steps necessary to develop a CQD-sample where one electron resides in each QD respectively ((1,1) regime, [21, 24]). Furthermore, the sample should be suitable for quantum optics applications, meaning that the spin lifetime of the captured electrons has to remain unaffected by variation of the sample parameters.

3.1 Introduction

One major goal of this thesis was the development of a CQD-sample with the (1,1) regime accessible. The (1,1) regime with eight eigenstates and a subsystem insensitive to nuclear spin interactions offers rich possibilities for spin-manipulation schemes [24, 26], non-linear optics and gain mechanisms [33].

During different steps of the development-process changes in sample parameters affected the quality of the sample. Therefore, requirements on a CQD sample will be explained in the following sections. But first a typical optical characterization process will be outlined, pointing out the important sample properties.

After processing of the sample and verification of the electrical contacts, first optical characterization relies on PL. The sample should exhibit low QD-density with about one CQD in the focus (section 3.2). PL allows furthermore to identify the difference in emission wavelength of the bottom and top QD layers $\Delta \lambda$, which strongly influences the charging sequence (section 3.5). Stacking of two QDs can be verified in PL by measuring the position-dependence of the QD emission and more important anticrossings. Resonant spectroscopy should than be employed on the one hand to determine the spin-lifetime, which is best done by measuring the spin-pumping efficiency of the
3. Design and characterization of samples with the (1,1) regime accessible

$X^{1-}$ of a uncoupled QD (section 3.4). On the other hand resonant spectroscopy allows to identify the charging sequence by measuring the charge stability region of the $X^{1-}$ in both bottom and top QD. In case the (1,1) regime is not accessible, an improved sample has to be grown by taking the knowledge gained from the described characterization into account. The following sections detail the most important requirements on the sample.

3.2 QD-density

A low QD-density is prerequisite to address single pairs of CQDs, since no more than one CQD should be in the focal spot (1 $\mu$m$^2$) in order to prevent charge sensing \[72\] between neighboring QDs and ease identification of pairs of QDs and charging sequences \[39\]. Furthermore, low densities simplify the alignment procedure since single CQD emission can be identified with a CCD camera.

Low densities can be achieved by growing the QD layer with a density gradient over the wafer, thereby increasing the probability of an area on the wafer with the appropriate QD-density. To this end the rotation of the wafer is stopped during the formation of the QDs. Consequently, the part of the wafer that is closer to the In-source exhibits a higher QD-density. An additional challenge offers the stacking probability: similar densities in both QD-layers are required to achieve high stacking probability. One has to keep in mind, that the top QD-layer grows faster due to the strain induced by the first layer. Therefore, less In is needed for the growth of the top-QDs. In order to identify the good-density region, a PL-map of the whole wafer is taken with a 785 nm laser at room temperature. In Fig. 3.1a such a PL-map of sample 3 is shown. The right density can be found at the edge of the dark blue region (piece iii). Here, it is possible to distinguish single pairs of CQDs on an image taken with a CCD camera. Such a CCD image of sample 3 iii, mounted in a bath-cryostat, is shown in Fig. 3.1b.

3.3 Electrical properties

One should also worry about the design of the back contact, since an increased dopant density can decrease the tunnel barrier. The density of the Si-dopant can be controlled with the temperature of the Si-cell in the growth chamber. The temperatures for the measured samples and the resulting resistances of the back contact are given in the table below. The higher temperature of the Si-cell for sample 2 led to a reduction of the tunnel barrier; Si-atoms segregated into the barrier. The higher effective doping density for sample 2 is evidenced in the lower resistance of the back contact, which amounts to 150 $\Omega$ compared to 300 $\Omega$ for sample 3, as well as a smaller effective width of the tunnel-barrier. The effective width of the tunnel barrier is calculated by taking into account the wavelength of various QDs and gate voltage at which one electron tunnels into the respective QD. The segregation of the dopant can be suppressed by decreasing...
3.3 Electrical properties

Figure 3.1: QD density. a) Integrated PL-intensity of sample 3 for emission wavelength between 900 nm and 1200 nm under excitation with a 785 nm laser at room temperature. b) CCD image of sample 3 iii under excitation with a 780 nm laser, where an area with a diameter of approximately 5 µm is illuminated.

<table>
<thead>
<tr>
<th>sample</th>
<th>Si T (°C)</th>
<th>R (Ω)</th>
<th>nominal tunnel barrier</th>
<th>effective tunnel barrier</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 i (ES1366)</td>
<td>1220</td>
<td>350</td>
<td>30 nm</td>
<td>not measured</td>
</tr>
<tr>
<td>2 iv (ES1554)</td>
<td>1270</td>
<td>150</td>
<td>50 nm</td>
<td>37 nm</td>
</tr>
<tr>
<td>3 iii (ES1773)</td>
<td>1210</td>
<td>300</td>
<td>50 nm</td>
<td>53 nm</td>
</tr>
</tbody>
</table>

Table 3.1: Doping properties for the measured samples.

the temperatures at which the barrier is grown. Surprising is the fact that sample 1 exhibits small coupling to the back contact (chapter 6) compared to sample 2, although it was grown with a much smaller nominal thickness of the tunnel-barrier. Possibly, the growth temperature of the barrier of sample 1 and sample 2 deviated significantly.

Electrical connections to the back contact are established by annealing metal into the sample at ∼ 380 °C. One can either use Indium pieces or Au-Ge-Ni layers, evaporated on top of the sample. Oxidation of the metal can prevent contacting. Therefore, the outer parts of the In should be removed and the annealing has to take place in presence of forming gas (Ar/H₂).

To apply a gate voltage, a semitransparent metal gate is evaporated on top of the sample. A suitable combination turned out to consists of 2 nm of Ti and 6-8 nm of Au. Ti sticks to the semiconductor surface, in contrast to Au. But a Au-layer is needed to cover the Ti-layer and prevent its oxidation. The thicker the metal layers are, the lower the resistance becomes, enabling fast gate voltage modulation. Sample 3 (2 nm of Ti, 6 nm of Au) for instance can be modulated with frequencies up to 10 kHz.

On the other hand, the thicker the gate is, the lower the optical transmission through the sample becomes; more light will be reflected. The typical transmission through gate and sample amounts to 30%. High transmission is mainly important for RF measurements, where photons are counted; in resonant differential transmission, the contrast (dT/T)
3. Design and characterization of samples with the (1,1) regime accessible

Figure 3.2: **IV-curve of sample ES1726.** Measurement of the current flowing through the sample versus gate voltage at 294 K (red dots) and at 4.2 K with (gray dots) and without (blue dots) illumination from a 780 nm laser. The shaded gray area in the inset indicates the charge stability regions of the $X^0$ and $X^{1-}$, where the measurements are performed.

is measured, which is independent of the absolute transmission.

A good method to verify that gate and back contact are working properly and that no sizable currents flow at the gate voltages the measurements will be performed at, consists of measuring the current versus applied voltage. Such an IV-curve of sample ES1726 is shown in Fig. 3.2. The shaded gray area indicates the gate voltage range, where the $X^0$ and $X^{1-}$ are stable. Here, strong currents could affect the spin-lifetime. The sample exhibits diode-characteristics because of the AlGaAs blocking barrier. At room temperature (red line), the diode opens at $\sim 50$ mV. The quality of the metal-semiconductor contact (Schottky offset-voltage) determines the value of the gate voltage at which current starts to flow through the sample. At 4.2 K the diode opens later ($\sim 550$ mV) than at room temperature and the resistance is smaller. Shining light on the sample induces negative currents.

In the framework of this thesis, variations of QD-growth and blocking barriers were tested. There are two possibilities to grow the blocking barrier: digital alloy is a succession of AlAs and GaAs, each a view mono-layers thick. The other options is to use a admixture of Al to GaAs of about 30 – 40 %. It turned out that the digital alloy affects the QD-emission, though the mechanism is not understood and many publications of other groups are based on QD samples with digital alloy as blocking barrier [11, 56]. Possibly, contaminations of the MBE-machine led to charge traps in the digital alloy. Fig. 3.3a and b show PL-measurement of an ensemble of QDs for sample ES1516 and ES1514. Sample ES1514 was grown with digital alloy, ES1516 with AlGaAs mixture. Both exhibit strong PL without a top gate and the emission of ES1516 with gate can be recovered by applying gate voltages up 600 mV to compensate the Schottky offset-voltage. An almost complete lack of emission is observed for ES1514 with gate, even
3.4 Spin-lifetime

Spin initialization or spin-pumping [20] is one of the five criteria for quantum computing [28]. Furthermore, the spin-pumping efficiency gives information about the spin-relaxation times. Therefore, QDs with low spin-pumping efficiency are unsuitable for quantum information and most quantum optics applications.

Spin-pumping can be achieved in Λ-type three level systems, consisting of two ground states, which couple to a common excited state. Such a Λ-scheme can be found in the $X^{-1}$ in Faraday-geometry. Here, spin-pumping was observed for the first time in QDs [20]. In Faraday-geometry (Fig. 3.4a), the lower energy transition is optically forbidden, but exhibits a small oscillator strength due to the hyperfine interaction and admixture of light-holes to heavy-holes [74, 75]. Therefore typical branching ratios $b = \Gamma_1/\Gamma_2$ amount to $\sim 250$ [76]. Applying a resonant laser to the spin-up transition brings the population into the excited state, from where it relaxes with rate $\Gamma_1$ either back to the spin-up state or with rate $\Gamma_2$ to the spin-down state. In the latter case the population gets trapped in the spin-down state for characteristic times $1/\gamma$, with $\gamma$ the relaxation rate between the ground states. At zero magnetic field, electron spin flip-flop processes with nuclear...
3. Design and characterization of samples with the (1,1) regime accessible

![a) QD-X 1-; b) CQD-(1,1) regime](image)

**Figure 3.4: Λ-schemes.** a) Energy eigenstates of the $X^{1-}$ of a single QD in Faraday-geometry. The spontaneous emission rate $\Gamma_2$ of the optically forbidden transition is reduced compared to the spontaneous emission rate $\Gamma_1$ of the optically allowed transition. The straight blue arrow indicates a probe laser. b) Energy eigenstates of a CQD in the (1,1)-regime, forming a Λ-scheme. The $S$ to $R_+$ and the $T_0$ to $R_+$ transitions exhibit the same oscillator strength.

... spins lead to a fast $\gamma$, preventing spin-pumping. At finite magnetic fields $\gamma$ is then substantially reduced. The spin-pumping efficiency can be calculated as $\Gamma_2/\gamma$. Hence, spin-pumping is more efficient in Λ-schemes, where $\Gamma_2$ is fast, as in singly charged QDs in Voigt-geometry [73] with the magnetic field applied in the plane. But, Λ-schemes with $b > 1$ can be employed for more efficient spin-read out [21, 76].

Spin pumping on the $X^{-1}$ in Faraday-geometry is measured for two different samples. Sample 3 (Fig. 3.5a) exhibits spin pumping with contrast below the noise level. Only at the edges of the one electron charging regime spin-relaxation due to co-tunneling takes place. Here, one electron tunnels via a virtual state to the back contact and an electron with the opposite spin tunnels back into the QD. Co-tunneling depends on the gate voltage and the thickness of the tunnel-barrier. Sample 2 for instance, displays strong co-tunneling due to its reduced tunnel-barrier to the back contact. Therefore, sample 2 will be employed in the next chapter to create inversion. Additional spin-relaxation was measured in a series of samples, all grown with a contaminated Ga-cell of the MBE. ES1726 (Fig. 3.5b) is one of them; it exhibits residual dT-signal with asymmetric line-shapes at the center of the one electron regime at sizable magnetic fields (1 T). Most probably impurities in the QD environment are responsible for the spin-relaxation. Therefore, the actual cleanliness of the material sources has to be checked after every MBE-opening via measuring the spin-pumping efficiency.

### 3.5 (1,1) charging regime

In case that all the described requirements are met, the next step will be to find a sample structure with the (1,1) regime accessible. The charge stability region of the
3.5 (1,1) charging regime

(1,1) regime strongly depends on the sample design, since only one gate voltage is available to tune the energies of the CQD. Hence, only the overall charge state, but not the charging sequence can be controlled with the gate voltage. An important role in the sample design plays the spacer-layer. Its thickness determines the strength of the tunnel coupling between the QDs. Therefore, it has a strong influence on the charging sequence. The other important parameter is the difference in emission energy of the the two QDs in combination with the tunnel barrier to the back contact. For small tunnel barriers, as in sample 1, one would have to choose the transition energies of the two QDs \(\Delta \lambda \approx 50\) nm apart in order to access the (1,1) regime. This corresponds to the maximum of the laser tuning range and is unfavorable. Therefore, we increased the tunnel-barrier to 50 nm (sample 2, 3) and doubled the thickness of the capping layer. Latter is necessary to achieve reasonable lever arms and symmetric line-shapes in dR-measurements \([57, 63]\). As a result, sample 2 (Fig. 3.6) and 3 both exhibit the (1,1) regime. In Sample 3 the (1,1) regime is accessible in CQDs with a difference in emission-wavelength between \(\Delta \lambda \approx 25\) nm and \(\Delta \lambda \approx 40\) nm.

The spin configurations of the eigenstates of the (1,1)-regime are shown in Fig. 3.7. The three triplet \((T_-, T_0, T_+)\) ground states have spin \(J = 1\) \((J_m = -1, J_m = 0, J_m = +1)\) and the singlet ground state spin \(J = 0\). The energy states of the exciton in the red QD are labeled as \(R_{--}, R_{-}, R_{+}, R_{++}\) and have spin \(J_m = -2, J_m = -1, J_m = +1\) and \(J_m = +2\). The optically allowed transitions are indicated with light \((\sigma^-\) polarized) and dark gray \((\sigma^+\) polarized) arrows in Fig. 3.7. At zero magnetic field the in-plane component of the Overhauser field couples the three T-states as well as the \(R_{--}\) and \(R_{-}\) and the \(R_{+}\) and \(R_{++}\) excited states respectively as indicated with blue arrows. The \(T_0\) and the \(S\) state are to first order insensitive to the hyperfine interaction, only a difference in magnetic field between the locations of the two QDs can influence them. In order to suppress the influence of small magnetic field gradients and second
3. Design and characterization of samples with the (1,1) regime accessible

order processes, a sizable energy separation between $T_0$ and $S$ (exchange splitting $E_{ST}$) is necessary to create a subsystem, insensitive to nuclear spins.

$E_{ST}$ is dependent on the interdot tunneling-strength, since the $S$-state exhibits tunnel resonances and $T$ does not. The (2,0)-$S$ state couples via electron tunneling to the (1,1)-$S$ and the (0,2)-$S$ state. Anticrossings are the result as shown in a simulation in Fig. 3.7c. The (1,1)-$T$ states can only couple to the (2,0)-$T$ state, where one electron sits in the p-orbital, because the spin-state is conserved in tunneling. Consequently, a gate-voltage dependent $E_{ST}$ emerges between the $S$ and $T$ states.

Due to the insensitivity of the $S$ and $T_0$ state to the hyperfine interaction and a sizable $E_{ST}$, spinpumping can be observed in the (1,1) regime even at zero magnetic field. The rich optical spectrum of Fig. 3.7 allows on the one hand for fast spin initialization in a $\Lambda$-scheme with $S$ and $T_0$ as ground states (Fig. 3.4b), on the other hand for efficient spin read-out. Latter can be realized for instance with the $T_{++}$ to $R_{++}$ transition.

Figure 3.6: **Sample design.** a) Shown is a schematic of sample 2. b) Band structure of sample 2 with a gate voltage applied between back contact and top gate.
3.5 (1,1) charging regime

Figure 3.7: **(1,1) charging regime.** a) Spin configuration of the energy eigenstates in the (1,1) regime. Only excitations in the red QD are depicted. b) Optical transitions in the (1,1) regime. Light gray arrow signify left-handed circularly polarized, dark gray arrow right-handed circularly polarized transitions. Blue arrows indicate mixing due to nuclear spins. c) Simulation of ground (bottom panel) and excited state (top panel) energies versus gate voltage. The charging state changes from (2,0) over (1,1) to (0,2) with increasing gate voltage. Possible excitations from the S and T ground states are indicated with blue and purple arrows. The four excited states (X) are degenerate.
3. Design and characterization of samples with the (1,1) regime accessible
Chapter 4

Optical amplification in a quantum dot molecule

This chapter is based on publication [33] and reports the observation of steady-state optical amplification in Raman transitions between the lowest-energy spin states of a single quantum-dot molecule. Absorption and resonance fluorescence experiments demonstrate that the entangled two-electron singlet and triplet states have electric-dipole coupling to a common optically excited state. Fast spin relaxation ensures optical gain on the triplet transition when the singlet transition is driven resonantly. By embedding the quantum-dot molecule in a cavity of modest quality factor, a solid-state single-emitter laser can be realized.

4.1 Introduction

A laser that uses a single quantum emitter as the gain medium [77] can exhibit a plethora of unusual features, including lasing without a well-defined threshold [78, 79] and output intensity fluctuations that remain below the shot-noise limit [78, 80, 81]. For studies of these fundamental issues, single-atom lasers [82, 83, 84] with simple and well-understood level schemes have proven particularly suitable. On the other hand, compact devices capable of continuous-wave operation require monolithic structures involving a solid-state quantum emitter. Although signatures of lasing due to a single quantum dot (QD) in a nanocavity have been reported [85], the nature of the QD lasing states remains unclear, and most probably an intricate non-resonant cavity feeding mechanism is involved [86]. These complications have prevented a detailed understanding of the optical amplification process or the pumping mechanism in single QD lasers.
4. Optical amplification in a quantum dot molecule

4.2 Energy-level diagram

The observation of optical amplification [87, 88] in a single coupled quantum dot (CQD) molecule filled with two separate electrons is observed. This novel solid-state quantum emitter combines an atom-like three-level lambda scheme, which can be fully characterized and driven coherently, with the control and tunability offered by solid-state technology. The optical amplification is a direct result of the interaction between the CQD and its solid-state environment, which induces fast relaxation between the spin singlet and triplet ground states.

The lambda scheme in this work is provided by a pair of vertically stacked self-assembled InGaAs QDs [37, 38, 39, 24, 71], separated by a thin GaAs tunnel barrier and embedded in a GaAs Schottky diode (Fig. 4.1a). When both QDs contain a single electron – a charging regime denoted as (1,1) – the lowest energy levels correspond to spin singlet (S) or triplet (T\(_{-}\), T\(_{0}\), T\(_{+}\)) states (Fig. 4.1b). Electron tunneling between the two dots gives rise to an exchange splitting between the (1,1)S and (1,1)T states (bottom panel in Fig. 4.1c), which allows us to selectively address them optically [24] even without a magnetic field. The size of the exchange splitting depends on the tunneling rate and can be tuned by varying the gate voltage [71]. The lowest-energy optical excitation corresponds to adding an electron-hole pair in the top dot (QD-R), which has a red-shifted transition energy compared to the bottom dot (QD-B). The resulting fourfold degenerate excited states X (top panel in Fig. 4.1c) are labeled by the z-component of the total angular momentum (\(m_z = \pm 1, \pm 2\)); this consists of a contribution from the heavy hole in QD-R (\(m_z = \pm \frac{3}{2}\)) plus the unpaired electron in QD-B (\(m_z = \pm \frac{1}{2}\)). From the associated optical selection rules (inset to Fig. 4.1c), it follows that states S and T\(_{0}\) share two common optically excited states with \(m_z = \pm 1\). At zero magnetic field, the selection rules are modified by the hyperfine interaction with the nuclear spins, which strongly mixes the three degenerate triplet states [40]. Likewise, the four degenerate optically excited states are mixed by both hyperfine interaction and indirect electron-hole exchange [71]. As a consequence, population in any X or T level is efficiently distributed among the entire X or T manifold, so that the full system can be represented by three levels (S, T and X) in a simple lambda configuration, as illustrated in Fig. 4.6b. In this Letter, we use this lambda system to achieve single-pass optical amplification of 0.014 %. All measurements were performed at 4.2 K in a liquid-helium bath cryostat.

4.3 Realization of the (1,1) charging regime

We first perform micro-photoluminescence (PL), in order to select a coupled quantum dot (CQD) pair that exhibits the (1,1) charging regime. As the gate voltage is increased, the number of electrons in the CQD increases one by one. Therefore, the PL spectra in Fig. 4.2 show typical plateaus [51], separated by dotted vertical lines indicating a change in the ground state charge. Each plateau corresponds to emission from the neutral exciton or negatively charged trion located in a particular dot. The detailed shape of the
4.4 Simulation of the charging sequence

We employ numerical simulation to confirm the charging sequence and find an explanation for the large exchange splitting. The numerical simulation diagonalizes the Hamiltonian describing the ground and optically excites states of the CQD as a function of energy level versus gate voltage. State (1,1)S is coupled via electron tunneling to states (2,0)S and (0,2)S, in which both electrons reside in QD-B or QD-R, respectively (as illustrated in the grey boxes, where filled circles depict electrons and open circles depict holes). The coupling gives rise to two anticrossings between the S states that split (1,1)S from (1,1)T, since the latter does not experience tunnel coupling to any of the S states. Dashed lines indicate energy levels not used in the experiment. Inset: optical selection rules for transitions from the (1,1)S and T states to the fourfold degenerate optically excited states X.

plateau for a given QD depends on the number of electrons in its partner dot, due to both tunnel coupling \[37, 38, 39, 24, 71\] and charge sensing \[53, 72\]. From these characteristic PL patterns we identify the CQD charging sequence as \((0,0) \rightarrow (1,0) \rightarrow (1,1) \rightarrow (1,2)\). This sequence is confirmed using numerical simulations in the next section. In the (1,1) regime, we find an exchange splitting between the S and T states of 1.1 meV.

Figure 4.1: **Properties of the (1,1) charging regime.** a) Schematic energy diagram of the molecular-beam-epitaxy-grown device, containing two layers of self-assembled InGaAs QDs, separated by a 9 nm GaAs tunnel barrier and embedded in a GaAs Schottky diode. The voltage \(V\) applied between the Si-doped \(n^+\)-GaAs back contact and the semi-transparent top gate (2 nm of Ti plus 8 nm of Au) controls the CQD charging state and allows both QDs of a pair to be filled with a single electron. b) Spin singlet (S) and triplet (T) states in the (1,1) charging regime. c) Energy level diagram showing the different ground states (bottom panel) and optically excited states (top panel) versus gate voltage. State (1,1)S is coupled via electron tunneling to states (2,0)S and (0,2)S, in which both electrons reside in QD-B or QD-R, respectively (as illustrated in the grey boxes, where filled circles depict electrons and open circles depict holes). The coupling gives rise to two anticrossings between the S states that split (1,1)S from (1,1)T, since the latter does not experience tunnel coupling to any of the S states \[71\]. Dashed lines indicate energy levels not used in the experiment. Inset: optical selection rules for transitions from the (1,1)S and T states to the fourfold degenerate optically excited states X.
4. Optical amplification in a quantum dot molecule

Figure 4.2: PL as a function of gate voltage. a) PL from QD-B. The dotted vertical lines separate regions with different total number of electrons in the CQD; the inferred ground state charge distribution for each region is indicated below the panel. Inside the (1,1) charging region (highlighted by the orange boxes), PL involving the S state is identified by its characteristic curvature, and by its $\sim 3$ times weaker intensity compared to PL involving the threefold degenerate T states. $X^0_B$ ($X^{1-}_B$) indicates emission from the neutral exciton (negative trion) in QD-B. Inset: schematic energy diagram illustrating $X^{1-}_B$ emission in the (1,1) regime. Because holes can tunnel from QD-B to QD-R before recombination, PL from QD-B is much weaker than that from QD-R. b) PL from QD-R. $X^0_R$ ($X^{1-}_R$) indicates emission from the neutral exciton (negative trion) in QD-R. Inset: schematic energy diagram illustrating emission from the optically excited states X to states S or T in the (1,1) regime.

Gate voltage. It then determines the energy and corresponding strength of all possible transitions (i.e. from all optically excited states to all ground states). Relaxation processes are not taken into account, so some of the transitions apparent in the simulations are not observed in the actual measurements.

The large size of the exchange splitting between S and T ($> 1$ meV) is due in part to the sizable tunnel coupling: $t_e = 1$ meV, corresponding to a $(2,0)S - (1,1)S$ anticrossing of $2\sqrt{2}t_e \approx 2.8$ meV (see the bottom panel of Fig. 4.1c). In addition, the large ST splitting reflects the proximity in gate voltage of the $(2,0)S - (1,1)S$ anticrossing ($V \approx -195$ mV) and the $(1,1)S - (0,2)S$ anticrossing ($V \approx -35$ mV). This means that even in the center of the (1,1) regime, the effect of each anticrossing on the ST splitting is substantial.
### 4.5 Lambda-scheme with fast relaxation rate

The proximity of the two anticrossings in turn reflects the similarity of the inter-dot Coulomb energy \( V_{BR}^{ee} = 16.8 \text{ meV} \) to the intra-dot Coulomb energies \( V_{BB}^{ee} = 18 \text{ meV} \) in QD-B and \( V_{RR}^{ee} = 22 \text{ meV} \) in QD-R \([71]\).

**Figure 4.3:** Numerical simulation of CQD PL. **a**) Simulated PL from QD-B versus gate voltage. Parameters used: \( X_0^B = 1360.97 \text{ meV}, X_{1-}^B = 1356.35 \text{ meV}, X_0^R = 1291.37 \text{ meV}, X_{1-}^R = 1287.21 \text{ meV}; V_{BB}^{ee} = 18 \text{ meV} \) (electron-electron Coulomb energy in QD-B); \( V_{RR}^{ee} = 22 \text{ meV} \) (electron-electron Coulomb energy in QD-R); \( V_{BR}^{ee} = 16.8 \text{ meV} \) (electron-electron Coulomb energy between QD-B and QD-R); \( V_{BR}^{eh} = 17 \text{ meV} \) (electron-hole Coulomb energy between QD-B and QD-R); \( t_e = 1 \text{ meV} \) (tunnel coupling of an electron between QD-B and QD-R); \( \delta_0 = 0.2 \text{ meV} \) (dark-bright splitting in each QD). \( X_0^B (X_{1-}^B) \) indicates emission from the neutral exciton (negative trion) in QD-B. The (1,1) charging region is highlighted by the orange boxes. **b**) Simulated PL from QD-R versus gate voltage. \( X_0^R (X_{1-}^R) \) indicates emission from the neutral exciton (negative trion) in QD-R.

### 4.5 Lambda-scheme with fast relaxation rate

To establish the optical connection between the S and T states, we employ resonance fluorescence measurements \([21]\). When resonantly driving the S transition in QD-R (orange arrow in the upper trace of Fig. 4.4a), fluorescence is detected not only at the same energy (Rayleigh scattering), but also at an energy corresponding to the T
transition (coherent and incoherent Raman scattering). Conversely, when driving the T transition in QD-R (orange arrow in the lower trace), additional weaker emission is observed at the S transition. These measurements demonstrate that the (1,1)S and T states indeed share common optically excited states X in which a negative trion is located in QD-R. Moreover, the fact that the T peak in the upper trace is \( \sim 3 \) times stronger than the S peak implies that the fourfold degenerate states X are strongly mixed; without mixing, driving the S transition would only excite the \( m_z = \pm 1 \) subspace, resulting in an equal number of photons emitted on the T and S transitions (see the inset to Fig. 4.1c). Together, these observations provide experimental justification for treating the system of one S, three T and four X states as a simple lambda system, as illustrated in Fig. 4.6b.

It is important to notice that driving the T transition results in much less fluorescence than driving the S transition, although both traces in Fig. 4.4a were taken with identical laser power. We find an S:T fluorescence ratio of \( \sim 8 \) (taking into account the imperfect cancellation of the excitation laser). This surprising asymmetry is also seen in differential transmission (dT) measurements throughout the (1,1) charging regime. On the S-X transition (Fig. 4.4b), scattering of incoming resonant laser photons leads to a dT contrast of \(-0.07 \%\). The dT contrast of the T-X transition (Fig. 4.4c) is only \(-0.011 \%\), i.e. \( \sim 6 \) times smaller. This difference points towards the presence of effective spin relaxation from T to S. When the laser is tuned to the S resonance, relaxation counteracts optical shelving [20] in the T states and thus maintains the photon scattering rate (and therefore the dT or resonance fluorescence signal). In contrast, a laser on the T resonance will quickly drive the system to the S state, where it will remain shelved for a long time, since relaxation from S back to T is impeded by the 1.1 meV S-T energy difference. Thus, the overall photon scattering rate in this case will be strongly reduced. Using a steady-state solution of the rate equations describing the S, T & X populations (section 4.6), we can estimate the relaxation rate \( \gamma \). The measured S:T scattering ratio of \( \sim 6 - 8 \) (obtained from the difference in dT contrast between Figs. 4.4b and 4.4c) or from the difference in fluorescence intensity between the two traces in Fig. 4.4a gives \( \gamma/\Gamma \sim 0.1 - 0.25 \), where \( \Gamma = \Gamma_S + \Gamma_T \sim 1 \mu\text{eV} \) corresponds to the total spontaneous emission rate from X.

4.6 Simulation of level populations

In order to estimate the relaxation rate, the steady-state populations \( \rho_{SS} \) (\( \rho_{TT} \)) of S (T) when driving \( \Omega_S \) (\( \Omega_T \)) are calculated with a three-level rate equation model describing the lambda system as in Fig. 4.6b. We find:

\[
\rho_{SS} = (1 + \bar{n})\gamma_{TS} \cdot \left[ \frac{\Gamma_T + (1 + \bar{n})\gamma_{TS}}{1 + (\Gamma_S/\Omega_S)^2 + (\Gamma_S\Gamma_T/\Omega_S^2)} + \bar{n}\gamma_{ST} + (1 + \bar{n})\gamma_{TS} \right]^{-1}
\]
4.6 Simulation of level populations

Figure 4.4: Identifying fast relaxation rate. a) Resonance fluorescence detected with a spectrometer, when resonantly driving the S-X transition (upper trace) or the T-X transition (lower trace) close to saturation (Rabi frequencies $\Omega_{ST} \sim 1 \mu eV$), at $V = -121$ mV. Orange arrows indicate the excitation energy. The reflected linearly polarized excitation laser is suppressed by a factor of $\sim 10^6$ using a polarizer. Traces have been offset vertically for clarity. b) Differential transmission $dT/T$ (in colorscale) of a laser (with $\Omega_S = 0.5 \mu eV$) scanned across the S-X transition, versus $V$ throughout the (1,1) regime. Inset: schematic energy diagram of the lambda system driven by a laser on the S-X transition. c) Differential transmission $dT/T$ (in colorscale) on the T-X transition, with $\Omega_T = 1.0 \mu eV$; the contrast of the resonance is only $-0.011 \%$. Inset: schematic energy diagram of the lambda system driven by a laser on the T-X transition.

$$\rho_{TT} = (\bar{n})\gamma_{ST} \cdot \left[ \frac{\Gamma_S + \bar{n}\gamma_{ST}}{1 + (\Gamma_T/\Omega_T)^2 + (\Gamma_S\Gamma_T/\Omega_T^2)} + \bar{n}\gamma_{ST} + (1 + \bar{n})\gamma_{TS} \right]^{-1}$$

Here, $\Omega_S$ ($\Omega_T$) is the Rabi frequency of the laser on the S (T) transition; $\Gamma_S$ ($\Gamma_T$) is the total spontaneous emission rate from X to S (T); $(1 + \bar{n})\gamma_{TS}$ is the relaxation rate from T to S; $\bar{n}\gamma_{ST}$ is the relaxation rate from S to T; $\bar{n} = \exp(-E_{ST}/k_BT) \approx 0.05$ is a Boltzmann factor involving the exchange splitting $E_{ST} \approx 1.1$ meV between S and T, Boltzmann’s constant $k_B$ and the temperature $T \sim 4.2$ K of the bath.

To find $\Omega_S$ corresponding to the laser power used in the experiment, we can determine the AC-Stark splitting when pumping the S transition (bottom panel of Fig. 4.6a).
4. Optical amplification in a quantum dot molecule

To find $\Omega_T$, we use the AC-Stark splitting when pumping the T transition (inset to Fig. 4.6c). By comparing the two, we find that for identical laser powers on the S and T transition ($P_S = P_T$), the corresponding Rabi frequency for the S transition is slightly smaller ($\Omega_S \approx 0.9 \Omega_T$). This reflects the fact that the (1,1)S state is admixed with (2,0)S and (0,2)S, reducing the oscillator strength of the S-X transition. We therefore use $\Omega_S = 0.9 \Omega_T$ in the expressions for $\rho_{SS}$ and $\rho_{TT}$ above. In principle, the admixture will also suppress the relaxation rate $\Gamma_S$, while not affecting $\Gamma_T \approx 0.75 \mu eV$. Therefore, we use a ratio $\Gamma_T/\Gamma_S \approx 3/(0.9)^2 \approx 3.7$ in the equations above.

The result of calculating $\rho_{SS}$ and $\rho_{TT}$ and taking the ratio of the two is shown in Fig. 4.5. From the measurements, we find an experimental value for the ratio of $\sim 6 - 8$ (see the main text), which gives a relaxation rate from T to S of $\gamma \sim 0.1 - 0.25$.

![Figure 4.5: Numerical simulation of the steady-state S population when pumping the S-transition, compared to the T population when pumping the T-transition.](image)

The fast spin relaxation is most likely related to the very large 1.1 meV exchange splitting between S and T states and the relatively strong coupling to a degenerate electron gas in our device. Although the distance from the bottom QD layer to the highly doped back contact was designed to be large ($\sim 50$ nm), we have evidence to suggest that dopant segregation has resulted in a much smaller effective distance in our device, drastically increasing the spin-flip cotunneling rate. The mentioned factors enhance the inelastic spin-flip cotunneling rate with the electron reservoir. While fast spin relaxation is undesirable from the perspective of using S and T states to encode a qubit, it is essential for obtaining optical amplification, as we will now demonstrate.

### 4.7 Optical amplification

We drive the system with a strong pump laser that is stepped across the S-X resonance, and probe it by measuring the differential reflection (dR) of a weak probe laser scanned across the T-X resonance. When the pump is far off-resonance and has modest intensity (left and right sides of the top panel in Fig. 4.6a), the probe maps out the unperturbed T-X transition, similar to Fig. 4.4c. As the pump gets closer to the S resonance (middle...
4.7 Optical amplification

of the panel), the sign of the probe dR signal reverses, as indicated by the red color. This signifies that the probe laser actually gains intensity by interacting with the single CQD pair.

Figure 4.6: Optical amplification. a) Differential reflection dR/R (in colorscale) of a weak probe laser (Rabi frequency $\Omega_T = 0.6 \mu\text{eV}$) scanned across the T-transition, in the presence of a strong but undetected pump laser stepped across the S-transition, at $V = -94 \text{ mV}$. The two lasers have orthogonal linear polarizations, allowing the reflected pump light to be extinguished using a polarizer. The strong pump leads to small fluctuations in the exact resonance condition due to the creation of charges around the CQD [72]. The size of the Autler-Townes splitting in the bottom panel allows a calibration of $\Omega_S$ in terms of the pump laser power on the S transition, $P_S$. b) Schematic energy level diagram of the lambda system formed by states S, T & X. $\Omega_S$ and $\Omega_T$ indicate the laser Rabi frequencies; the effective spontaneous emission rate from X to the combined triplet states is about three times faster than to the singlet state ($\Gamma_T \approx 3\Gamma_S$). We observe fast relaxation (with rate $\gamma$) from T to S. c) $dR/R$ (in colorscale) of a weak probe laser ($\Omega_S = 0.5 \mu\text{eV}$) scanned across the S-transition, in the presence of a strong but undetected pump laser ($\Omega_T = 10.4 \mu\text{eV}$) resonant with the T-transition, versus $V$. Inset: $\Omega_T$ as a function of the square root of the pump laser power $\sqrt{P_T}$. $\Omega_T$ is determined from the Rabi splitting in measurements as shown in the main panel.

When the pump detuning is large (compared to the pump Rabi frequency $\Omega_S$), the amplification can be considered as stemming from a stimulated Raman process [31]. Raman gain indeed appears along the diagonal line in each panel in Fig. 4.6, where the two-photon resonance condition is fulfilled (detuning between pump and probe laser equal to S-T splitting). In this limit, there is no significant population in the X state, and therefore no population inversion occurs between the bare X and T states. In the opposite limit of small pump detuning and strong pump intensity, the non-perturbative
coupling of the quantum dot and the pump field leads to the formation of dressed states, which are coherent superpositions of states S and X [30]. Luminescence from these dressed states is predominantly incoherent, and optical amplification here results mainly from population inversion between the T state and either one of the dressed states. This is precisely what we observe in the Autler-Townes anticrossing that is seen in the lower panels of Fig. 4.6a.

As a control experiment, we tune the pump laser to the T transition and probe the S transition (Fig. 4.6c). In this case, a standard (absorptive) Autler-Townes splitting is observed, without any gain even for very high pump powers. This confirms that optical amplification on the S-X transition is prevented by the slow relaxation rate from S to T at low temperatures. Further confirmation of our interpretation comes from numerical simulations of the full 8-level system of S, T, & X states, which show qualitative agreement with the data.

4.8 Simulation of gain process

In order to show that the CQD level scheme exhibits laser amplification under our experimental conditions, we simulate the full 8-level S, T & X system. In the simulation, a probe laser (Ω_T = 0.6 μeV) is scanned across the T-transition, in the presence of a pump laser (Rabi frequency Ω_S) that is stepped across the S-transition. To simplify the modeling (and without affecting the validity of the simulation), both pump and probe lasers are assumed to be not linearly polarized as in the experiment, but right-hand circularly polarized. Pump and probe lasers therefore only couple state S to X(m_z = +1), state T_0 to X(m_z = +1) and state T_+ to X(m_z = +2).

Dissipation is included in the Lindblad approach. The total spontaneous emission rate is Γ = 1μeV; this means that the rate from X(m_z = +2) down to T_+ is Γ; the rate from X(m_z = +1) down to T_0 is Γ/2, and the rate from X(m_z = +1) down to S is a · Γ/2, where a ≈ 0.8 is a numerical factor that takes into account the admixture of (1,1)S with (2,0)S and (0,2)S, which reduces the oscillator strength for the S transitions. Dephasing (with rate 1μeV) is present on the strongly driven T_0-X and T_+X transitions.

The mechanism responsible for spin relaxation between T and S is most likely spin-flip co-tunneling with the nearby electron reservoir. Therefore, we assume relaxation between each of the three T and one S states, plus between X(m_z = +1) and X(m_z = +2). We find that a rate γ = 0.25 μeV for each of these transitions matches the experimental results. This value is within the range 0.1 – 0.25 that was found in connection with Fig. 4.5.

Mixing of the T manifold due to the hyperfine interaction with the nuclear spins is included via a coherent mixing term ω_x = 0.5μeV. In the X manifold, the same mixing term is present between states X(m_z = +1) and X(m_z = +2) and between X(m_z = −1) and X(m_z = −2).
4.9 Conclusions

The simulations qualitatively reproduce the measurement results shown in Fig. 4.6, in particular the occurrence of optical amplification when pumping the S-X transition (red regions in the figure), and the absence of amplification when pumping the T-X transition.

![Numerical simulation of laser amplification](image)

**Figure 4.7:** Numerical simulation of laser amplification. Described is the full 8-level S, T & X system shown in the inset to Fig. 4.6b. Left panels; pumping the S-X transition and probing the T-X transition. Right panel: pumping the T-X transition and probing the S-X transition.

4.9 Conclusions

In summary, we have demonstrated optical amplification in Raman transitions between singlet and triplet states of a single CQD molecule. This scheme is very promising for realizing a fully controllable solid-state single-emitter laser. In contrast to approaches based on the biexciton cascade [87] or p-state pumping [88] in single QDs, the CQD lambda system can be pumped fully coherently, and the depopulation rate between the optical ground states can be controlled using the gate voltage or the tunnel barrier thickness. An alternative lambda scheme is provided by a one-electron QD subjected to an in-plane magnetic field [73]. However, gain in such a system at 4 K would require a very large field on the order of 10 T. In contrast, a CQD in the (1,1) regime can provide gain even in the absence of a magnetic field and at elevated temperatures.

By coupling this new type of solid-state quantum emitter to a micro-cavity, it should be possible to observe laser oscillation. The photon statistics of such a laser are expected to differ from ordinary lasers. From the measured $\sim 0.014\%$ single-pass gain in probe laser
intensity, we estimate that a cavity quality factor of $\sim 7000$ should enable laser oscillation. Employing established techniques such as solid-immersion lenses could increase the gain and thereby reduce the required $Q$-factor by about an order of magnitude, making a practical implementation feasible.

Last but not least, the tunability of the CQD allows it to be tuned to an opposite regime, which will be the topic of the next chapter: if the CQD molecule is sufficiently isolated from the electron reservoir (modest S-T splitting and a small co-tunneling rate to the back contact), the lambda system enables ultrafast optical control of entanglement [24] and coherent dark state spectroscopy [26].
Chapter 5

Optically active singlet-triplet qubits with long coherence times

Atomic transitions that are insensitive to magnetic fields play a fundamental role in precision measurements and metrology. In contrast, most solid-state quantum systems are subject to either strong electric or magnetic field fluctuations that severely limit their $T_2^*$ coherence time. In this chapter, which is based on publication [26], we demonstrate that spin singlet and triplet ground states of an optically active quantum dot molecule can be made robust against both electric and magnetic field fluctuations by adjusting the applied gate voltage and the external magnetic field. Using coherent population trapping on transitions to a common optically excited state, we show that the singlet-triplet $T_2^*$ coherence time is on the order of 200 nanoseconds. The rich optical spectrum of this quantum system, exhibiting recycling transitions for spin measurements and indirect excitons for spin-state dependent long-range dipole-dipole interactions, potentially allows for applications in quantum information processing.

5.1 Introduction

Given the typical presence of proximal charge traps and nuclear spins in the solid state, it is important to identify a quantum system with a ground-state energy splitting that is insensitive to both electric and magnetic field fluctuations. While the coherence time of single confined electron spins in III-V semiconductors is determined by the fluctuating Overhauser field stemming from the nuclear spin environment [89, 90, 23], hole-spin qubits appear to be limited by charge fluctuations [25, 27, 19]. In contrast, a two-electron quantum dot (QD) molecule [24] subjected to an external magnetic field and tuned to its sweet spot where the singlet (S) -triplet (T) splitting is minimal realizes a decoherence-free two-level quantum system. Although groundbreaking experiments based on S-T states in electrically-defined systems have been carried out [91, 40, 18], the coupled QDs in these experiments were operated far from the sweet spot and the minimal exchange
coupling was much smaller than the Overhauser field gradient. Therefore the qubit was exposed to either magnetic or electric field fluctuations.

### 5. Optically active singlet-triplet qubits with long coherence times

### 5.2 S and T states in the (1,1) charging regime

![Diagram](image)

**Figure 5.1:** **Two-electron S and T states in the (1,1) charging regime.** A) Diagram of the device containing two layers of self-assembled InGaAs QDs embedded in a GaAs Schottky diode. B) Ground and optically excited states in the (1,1) regime. Blue arrows indicate electron spins in the bottom (blue) QD, red single (double) arrows indicate electron (hole) spins in the top (red) QD. States with a hole in the blue QD are not shown. We focus on the lambda system formed by states S, T₀ and R± indicated in yellow. C) Schematic energy diagram of the ground and optically excited states versus V. A magnetic field in Faraday geometry leads to Zeeman splittings proportional to the g-factors shown, where \( g_\text{e}^c \) (\( g_\text{e}^b \)) denotes the electronic g-factor in the red (blue) QD, and \( g_\text{h}^c \) the hole g-factor in the red dot. The dashed red line indicates the sweet spot, where \( dE_{ST}/dV = 0 \). **Inset** Dipole-allowed optical transitions between the states shown in (b). Blue and purple (gray) transitions are right-hand (left-hand) circularly polarized.

Our experiments utilize a pair of tunnel-coupled self-assembled InGaAs QDs [39] (Fig. 5.1a). By adjusting the growth parameters we ensure that both QDs are charged with a single electron for a wide range of the applied gate voltage \( V \) [24]. In this so-called (1,1) regime, the S and T ground states (Fig. 5.1b) are split by a voltage-dependent exchange interaction \( E_{ST} \) (see the lower panel in Fig. 5.1b). For a particular gate voltage \( V_0 \) (the “sweet spot”), \( dE_{ST}/dV = 0 \) so that \( E_{ST} \) is to first order insensitive to charge fluctuations. In addition, the large value of \( E_{ST} \) suppresses mixing between the S and T states arising from the Overhauser field gradient. Finally, an external magnetic field (\( B \)) applied along the growth direction \( z \) suppresses hyperfine mixing between the three
5.3 Coherent population trapping

triplets by splitting off $T_{\pm}$ (with spin z-projection $m_s = \pm 1$), while leaving both $S$ and $T_0$ (with $m_s = 0$) unaffected. In our measurements we focus on the lambda system formed by $S$ and $T_0$ plus a shared optically excited state $R_+$ that contains a trion in the red QD (see the upper panel in Fig. 5.1).

Figure 5.2: **Sweet spot of the (1,1) regime.** Differential reflection (dR) measurement of the trion transitions in the red QD versus $V$ at $B = 0.2$ T, measured around saturation power (laser Rabi frequency $\Omega = 0.8 \mu$eV) in the presence of a weak non-resonant (850 nm) laser. Blue (purple) dashed lines indicate the $S-R_+ (T_0-R_+)$ transition energies, extracted from two-laser repump measurements (not shown). The unmarked diagonal feature in the top right-hand corner is due to indirect transitions involving the (1,2) charging ground state. Inset) $E_{ST}$ versus $V$; the red line is a parabolic fit with curvature $s = 2.99 \cdot 10^{-3} \mu$eV/mV$^2$.

First we employ single-laser differential reflection (dR) measurements [57] to map out the optical transitions of the red QD versus $V$. In the (1,1) regime we observe very efficient spin pumping into the S (T) state while probing the T (S) transitions, as evidenced by a vanishing dR contrast (Fig. 5.2). All transitions are only visible in a narrow gate-voltage range at the edge of the (1,1) regime, where spin-flip tunneling processes to and from the back contact lead to spin relaxation between the ground states [33]. By having a resonant laser present on both the $S-R_{\pm}$ and the $T_0-R_{\pm}$ transition simultaneously, the spin pumping is lifted and we can determine the voltage dependence of $E_{ST}$ (inset to Fig. 5.2). From this we find the sweet spot $V_0 = 190$ mV, just outside the (1,1) regime.

5.3 Coherent population trapping

In order to measure the coherence of the two-level system formed by $S$ and $T_0$, we rely on the quantum optical technique of coherent population trapping (CPT) [34, 92]. A weak probe laser is tuned across the $S-R_+$ transition while a non-perturbative coupling laser is incident on the $T_0-R_+$ transition (see the inset to Fig. 5.3). At the
Optically active singlet-triplet qubits with long coherence times

two-photon resonance the population becomes trapped in a dark state consisting of an antisymmetric superposition of S and T₀. Here destructive interference between the two optical transition paths leads to a vanishing scattering amplitude, and thus a dip in the dR spectrum (Fig. 5.3). Since this transparency results from the formation of a coherent superposition of S and T₀, decoherence processes with both slow (yielding T*₂) and fast (yielding Tₙᵣᵢₙ₃₉₅₉₅) decorrelation times lead to suppression of the CPT dip. Therefore, CPT is an excellent technique to reveal the effect of electric and magnetic field fluctuations on the coherence time of the ground states.

5.4 Sensitivity to electric and magnetic field fluctuations

We observe that the S – T₀ coherence is highly sensitive to the external magnetic field and the applied gate voltage. At B = 0.2 T and ΔV = V – V₀ = −8 mV, the CPT dip goes completely to zero for a pump laser Rabi frequency of ΩT₀ = 0.79 μeV (Fig. 5.4a). Tuning V away from the sweet spot to ΔV = −23 mV yields dephasing due to electric field fluctuations, leading to a reduced depth of the CPT dip and a general broadening of the dR spectrum (Fig. 5.4b). The electric field fluctuations are probably due to rapid filling and emptying of charge traps around the QD. We find that they can be reduced by illuminating the sample with a weak non-resonant (850 nm) laser [62]; switching this laser off thus results in a reduced CPT dip even quite close to V₀ (Fig. 5.4c). Most surprisingly, by tuning the magnetic field to a value below that of typical nuclear Overhauser fields (Bₙ ∼ 20 mT), we find that the single dark resonance turns into two shallow transparency dips (Fig. 2e). In this regime the in-plane component of Bₙ ensures
Figure 5.4: Influence of electric and magnetic field fluctuations. a) Measured $dR$ (blue dots) versus probe detuning at $B = 0.2$ T and $\Omega_{T_0} = 0.77 \mu$eV in the presence of a non-resonant (850 nm) laser that reduces the charge fluctuations. The red line is a numerical fit to an eight-level model with fit parameter $\Delta V = -8$ mV; the typical size of the Gaussian voltage fluctuations is known from independent measurements to be $\delta V = 0.6$ mV; $\delta V$ and $\Delta V$ together yield $T_2^* = 32$ ns; $T_2^{\text{markov}}$ is adjusted to 250 ns. The spontaneous emission rate is fixed to $\Gamma = \Gamma_S + \Gamma_{T_0} = 0.8 \mu$eV and the Lorentzian excited-state dephasing to $\gamma = 0.4 \mu$eV. b) Same as in a), but with $\Omega_{T_0} = 0.58 \mu$eV and $\Delta V = -23$ mV (yielding $T_2^* = 11$ ns), $T_2^{\text{markov}} = 1.3 \mu$s, and $\gamma = 0.6 \mu$eV. c) Same as in a), but at $\Delta V = -13$ mV and without the non-resonant laser, which yields $T_2^* = 8$ ns due to increased charge fluctuations $\delta V = 1.5$ mV. d) Same as in a), but at $B = 0$ T, yielding $T_2^* \sim 2$ ns. $T_2^*$ is limited by the coherent hyperfine mixing $\Omega_{HF} = 0.45 \mu$eV.

5.5 Extracting coherence times

To quantify the coherence time and to understand its origin, we measure the CPT dip for different pump laser powers and gate voltage detunings (Fig. 5.5). The results are then analyzed by numerically solving the optical Bloch equations for the full eight-level system shown in Fig. 5.1b. We find that there are two mechanisms that determine the depth of the CPT dip. Far away from the sweet spot, the coherence is limited by charge fluctuations that lead to fluctuations in $E_{ST}$. The magnitude of the Gaussian fluctuations ($\delta V = 0.6$ mV) is known from independent measurements (section 5.6); by
Figure 5.5: Analysis of the coupling to the reservoir. 

a) Series of dR spectra (dots) for various $\Omega_{T_0}$ taken in the same configuration and fitted with the same values as in Fig. 2b. Traces are offset vertically for clarity. 

b) Differential reflection spectra (dots) for various $\Omega_{T_0}$ taken in the same configuration and fitted with the same values as in Fig. 2c. 

c) Close-up on the $\Omega_{T_0} = 0.45$ $\mu$eV spectrum of a. The spectrum is fitted with various $\Delta V$ while keeping $T_2^\text{markov} = 250$ ns constant. From dark to light gray: $\Delta V = 0$ mV, $\Delta V = -8$ mV, $\Delta V = -12$ mV and $\Delta V = -22$ mV, yielding $T_2^* = 2.23 \mu$s, $T_2^* = 32$ ns, $T_2^* = 22$ ns and $T_2^* = 12$ ns respectively. 

d) Same as in c but fitted with various $T_2^\text{markov}$ while keeping $\Delta V = -8$ mV constant. From dark to light gray: $T_2^\text{markov} = 500$ ns, $T_2^\text{markov} = 250$ ns, $T_2^\text{markov} = 170$ ns and $T_2^\text{markov} = 85$ ns.

Treating the exact distance to the sweet spot ($\Delta V$) as a fit parameter we can reproduce the depth of the CPT dip. This procedure gives a value of $T_2^* = 11$ ns for the data in Fig. 5.5b. Moving closer towards the sweet spot, the charge fluctuations become less important. However, spin-flip tunneling with the back contact now becomes stronger, since in this coupled QD pair the sweet spot is located very close to the edge of the (1,1) regime. To reproduce the data in this case (Fig. 5.5c), we have to include the tunnel rate between the S and T ground states, which leads to an additional markovian spin dephasing term ($T_2^\text{markov}$). Its value at a certain $V$ is given by the spin pumping efficiency (section 5.6). By fitting the detailed CPT lineshape using a combination of
charge fluctuations and tunneling, we find a lower bound of \( T_2^* = 32 \) ns (see Fig. 5.5c) and \( T_{2\text{markov}} = 250 \) ns (see Fig. 5.5d). We emphasize that only by using measurements for different coupling laser Rabi frequencies in combination with different gate voltages it is possible to accurately determine the underlying decoherence times by fitting all spectra with the same \( \delta V \). The simulations do not include second-order hyperfine processes, which are expected to yield \( T_2^* \) times on the order of a microsecond.

Figure 5.6: **Long coherence time at the sweet spot.** a) CPT spectrum versus probe detuning of another QD-molecule, measured in dR (blue dots) at the sweet spot (\( \Delta V = 0 \) mV), \( B = 0.2 \) T and \( \Omega_{T_0} = 0.4 \) \( \mu \)eV in the presence of a weak non-resonant (850 nm) laser. For this QD-molecule the typical size of the Gaussian voltage fluctuations amounts to \( \delta V = 0.8 \) mV. Assuming additional second order hyperfine processes, by setting \( s = 1.85 \cdot 10^{-2} \) \( \mu \)eV/mV\(^2\) in the numerical fit (red line), yields \( T_2^* = 200 \) ns. \( T_{2\text{markov}} \) is determined independently to be 2.6 \( \mu \)s. Fixed parameters are \( \Gamma = \Gamma_S + \Gamma_{T_0} = 0.8 \) \( \mu \)eV and \( \gamma = 0.7 \) \( \mu \)eV. b) Same as in a) but fitted with three different values of \( s \) in order to include additional second order Gaussian dephasing processes; for instance second order hyperfine interaction or laser jitter. The resulting dephasing times are: \( T_2^* = 50 \) ns (light gray line), 200 ns (red line) and 1 \( \mu \)s (dark gray line). c) Same as in a) but with \( \Omega_{T_0} = 0.12 \) \( \mu \)eV, yielding a dip with FWHM \( \sim 10 \) MHz. The shown data (blue dots) are averaged over bins with width of \( \sim 1.7 \cdot 10^{-3} \) \( \mu \)eV each containing \( \sim 20 \) points.

To demonstrate such long \( T_2^* \) times, we find another coupled QD pair where the sweet spot is further away from the edge of the (1,1) plateau, so that tunneling-induced spin dephasing is strongly suppressed. Our eight-level fitting procedure yields a \( T_2^* = 1.0 \) \( \mu \)s due to charge fluctuations for this second QD molecule (Fig. 5.6a). However, the finite noise-level of our measurements only allows us to determine that the second-order hyperfine plus charge fluctuation induced \( T_2^* \) is between 200 ns and 1 \( \mu \)s (Fig. 5.6b). The lower bound of \( T_2^* \) is more than two orders of magnitude longer than previously reported values for optically active coupled electron \([24]\) or hole \([25]\) spins away from the sweet spot; in addition, it is more than an order of magnitude longer than the values reported for coupled electron spins in gated QDs \([91, 40]\), and comparable to that of a single hole \([19]\). Our system thus maintains coherence on timescales that could previously be achieved using spin echo techniques; the corresponding reduction in overhead can be very beneficial for applications in quantum information processing. Conversely, the long
$T_2^*$ should improve the effectiveness of a spin echo pulse, and could thus lead to even longer $T_2$ times. Finally, the potential of our system for high-resolution spectroscopy is demonstrated by reducing the pump Rabi frequency to $\Omega_{T_0} = 0.12 \, \mu\text{eV}$, which yields a narrow CPT dip with a full width at half maximum of just $\sim 10\text{MHz}$ (Fig. 5.6c).

### 5.6 Theoretical model

The theoretical model we employ to extract coherence times consists of solving an eight-level master equation for the states of Fig. 5.2b in the steady-state. Lorentzian dephasing is included in the Lindblad form, whereas Gaussian dephasing is taken into account in the following procedure. Solutions of the master equation in steady-state are calculated for a series of shifted exchange energies $E_{ST}$. The energies are related to voltages via the gate dependence of $E_{ST}$, which we extract from fitting a second order polynomial to it, yielding a curvature of $s = 2.99 \cdot 10^{-3} \, \mu\text{eV}/\text{mV}^2$ (Inset Fig. 5.2). Finally, the solution of the master equation is weighed with a Gaussian distribution of width $\delta V$, corresponding to the size of typical voltage fluctuations.

![Figure 5.7: Gaussian fluctuations. a) $\text{dR}$ measurement of the indirect transition in the (1,2)-regime versus gate voltage at $B = 0.2 \, \text{T}$. b) Line cut of a) at 196.2 mV, as indicated by the red line in a. The red line in b is a Gaussian fit with $\delta V = 0.6 \, \text{mV}$ the width of the Gaussian distribution.](image)

The fitting procedure will now be explained in more detail, since it is important to get a reliable value for both Gaussian ($T_2^*$) and Lorentzian ($T_2^{\text{markov}}$) dephasing times, stemming from charge fluctuations and electron tunneling to the back contact respectively. Therefore, we reduce the number of fitting parameters. First, we deduce the size of the typical Gaussian fluctuations $\delta V = 0.6 \, \text{mV}$ by fitting a Gauss to the indirect line in the (1,2) regime (Fig. 5.7). The indirect line exhibits a pure Gaussian line-shape since it is strongly susceptible to charge noise due to its large dipole moment ($s \sim 0.15 \, \mu\text{eV}/\text{mV}^2$).
5.6 Theoretical model

Figure 5.8: **Nuclear spin effects.** $dT$ measurement of blue Zeeman transition of the neutral exciton in QD-R for various B-fields: $B = 0$ T to $B = 0.5$ T. The black arrow indicates the sweep direction of the laser.

To reproduce the overall width of the spectra (Fig. 5.4, 5.5) an additional Lorentzian excited state dephasing rate $\gamma$ is introduced. The Lorentzian ground state dephasing time results from the sum of all rates into and out of $S$, stemming from gate voltage dependent co-tunneling to the back contact [94]. We assume these rates to be equal and refer to the sum over all rates into and out of $S$ as $\gamma_g$ ($T_2 = 1/\gamma_g$). Furthermore we include excited state mixing between $R_-$ and $R_-$ as well as between $R_{++}$ and $R_+$ with rate $\gamma_{es}$, which is larger than $\gamma_g$. This is justified, since the Zeeman splitting for these states only amounts to $g_e \mu_B B = 5.7 \mu eV$ at $B = 0.2$ T and can be further reduced by nuclear spin effects [8]. Nuclear spin effects are evidenced in Fig. 5.8 where the line-shape of the neutral exciton at $B = 0.2$ T clearly deviates from the one at $B = 0$ T. $\gamma_g$ and $\gamma_{es}$ can be determined by the contrast of the $S$ to $R_+$ and $S$ to $R_-$ transitions, shown in Fig. 5.9. For strong excited state mixing (light gray line), the contrast of the $S$ to $R_-$ transition is underestimated, whereas it is overestimated for strong ground state mixing (dark gray line).

Another important factor is the branching ratio between optically allowed (e. g. $R_{++}$ to $T_+$) and optically forbidden transitions (e. g. $R_{++}$ to $T_0$ and $S$). For single QDs the branching ratio was measured [76]. In accordance with these measurements we assume a branching ratio of 250.

The only remaining fitting parameter is the exact distance $\Delta V$ to the sweet spot, which is difficult to determine experimentally, since the charge-environment of the CQD changes over time.

In order to convert $\Delta V$ and $\delta V$, extracted from the simulation, into $T_2^*$, we take the Gaussian distribution of voltage fluctuations with a width $\delta V$, and calculate from this the distribution of $E_{ST}$. This corresponds to a distribution of frequencies with which the coherence between $S$ and $T_0$ would oscillate in time in a Ramsey-measurement. By adding these oscillations with the correct weighing factor, we determine the decay of the oscillation amplitude. $T_2^*$ corresponds to the time it takes until this ”inhomogeneously broadened” coherence has decayed to 1/e of its initial value.
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Figure 5.9: Co-tunneling rates and excited state mixing. Blue dots: dR-spectrum at $B = 0.2 \, \text{T}$ with $\Omega_{T_0} = 0.45 \, \mu\text{eV}$. Red line: fits employing the eight-level simulation with $\gamma_g = 2.5 \cdot 10^{-3} \, \mu\text{eV}$ and $\gamma_{es} = 2.5 \cdot 10^{-2} \, \mu\text{eV}$. Dark gray line: $\gamma_g = 5 \cdot 10^{-3} \, \mu\text{eV}$ and $\gamma_{es} = 2.5 \cdot 10^{-2} \, \mu\text{eV}$; light gray line: $\gamma_g = 2.5 \cdot 10^{-3} \, \mu\text{eV}$ and $\gamma_{es} = 5 \cdot 10^{-2} \, \mu\text{eV}$.

To account for the asymmetry of the spectra, we add $\sim 15\%$ of the dispersive part to the absorptive part of the solution of the master equation. This is justified for an optical interference effect, leading to a more pronounced asymmetry in reflection than in transmission measurements (Fig. 5.10) [57]. A phase shift in the gate metal most probably induces the asymmetry in reflection, since the layer thickness above the QDs is optimized for the emission wavelength we are working at.

To investigate the source of the electric field fluctuations, we compare resonance fluorescence (RF) measurements, while filtering (1 Hz low-pass filter) the applied gate voltage at room temperature, to dR measurements. dR-measurements do not allow for filtering due to the applied voltage modulation. We find no significant difference in CPT dip and the overall width of the spectra for the two different types of measurements. Hence, charged defects in the QD environment are mainly responsible for electric field fluctuations.

It is important to notice that CPT can only be measured in RF in the presence of a magnetic field, which lifts the degeneracy of the excited states. Otherwise the co-polarized pump and probe lasers excite different superpositions of excited states. This is due to the phase difference between the S and $T_0$ states. Co-polarized lasers are necessary in RF-measurements to block both lasers simultaneously.

5.7 Gaussian versus Lorentzian dephasing

In the previous chapter CPT was employed to extract coherence times. Another approach to determine coherence times are time-domain measurements. Here, the decay of Ramsey fringes [24, 91] is given by $T_2^*$, including both Gaussian and Lorentzian dephasing processes with slow and fast decorrelation times of the reservoir respectively.
5.7 Gaussian versus Lorentzian dephasing

Figure 5.10: **Asymmetric line shape.** Shown are \(dT\) (blue trace) and \(dR\) (red trace) measurements of the neutral exciton in the red QD for the same optical alignment at \(B = 0\ T\) with laser power below saturation (\(\Omega = 0.34\ \mu\text{eV}\)).

The spin-echo technique [40, 95] is than often used to filter out Gaussian dephasing and thereby measuring \(T_2^{\text{markov}}\). The spin-echo technique requires pulse-sequences, whereas CPT is performed with continuous-wave lasers. The challenge for the latter one is to resolve narrow CPT-dips and to suppress laser-jitter, introducing dephasing. But CPT also offers the possibility to distinguish between Gaussian and Lorentzian dephasing by studying the CPT-dip for various coupling laser Rabi frequencies. In this section I will investigate the difference between Gaussian and Lorentzian dephasing in CPT-spectra for various coherence times and probe Rabi frequencies necessary to resolve the difference between the two dephasing mechanisms. Furthermore, I will estimate whether it is experimentally feasible to employ CPT to characterize the noise reservoir and determine coherence times at the order of one microsecond.

Two different models are used in order to investigate the effect of dephasing with short or long decorrelation time on the CPT-dip. Solutions for both models are obtained by solving a three-level master equation in steady-state, but with either pure Lorentzian or pure Gaussian ground state dephasing. In the first case, dephasing is included in the Lindblad from (\(\gamma\)); for Gaussian dephasing a model similar to the one described in the previous section is employed. Both models include Lorentzian excited state dephasing \(\gamma = 0.4\ \mu\text{eV}\), but no Gaussian excited state dephasing. The spontaneous emission rate is set to \(\Gamma = \Gamma_S = 0.4\ \mu\text{eV}\) and the states are labeled as in the \(\Lambda\)-scheme of the inset of Fig. 5.3.

The full width at half maximum (FWHM) of the CPT dip is on the one hand determined by the applied Rabi-frequencies, on the other hand by dephasing. The Rabi-frequencies broaden the dip as FWHM= \((\Omega_T + \Omega_S)^2/1\). Dephasing dominates the FWHM of the CPT-dip for small Rabi frequencies. It is important to notice that the dependence on the coupling laser power can be completely different for Gaussian dephasing than for Lorentzian dephasing. Fig. 5.11a shows CPT-spectra with \(T_2^{\text{markov}} = 10\ \text{ns}\) (blue line) = \(T_2^*\) (red line) for \(\Omega_S = 0.03\ \mu\text{eV}\) and various coupling laser Rabi frequencies \(\Omega_T\). For small
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\[ I_m(r_{SR}+) \frac{1}{di 	ext{vision}} \]

Detuning (µeV)

\[ I_m(r_{SR}+) \frac{1}{di 	ext{vision}} \]

Detuning (µeV)

\[ I_m(r_{SR}+) \frac{1}{di 	ext{vision}} \]

Detuning (µeV)

-2 0 2

Im \((r_{SR}+)\)

-2 0 2

Im \((r_{SR}+)\)

-2 0 2

\[ W_{S} \]

0.03 µeV

0.3 µeV

1.3 µeV

0.1 µeV

0.3 µeV

1.3 µeV

Figure 5.11: \( T_2^{markov} = T_2^* = 10 \text{ ns} \). a) Plotted is the solution \((\rho_{SR}+)\) of a three-level master equation including either pure Gaussian (red lines) or pure Lorentzian (blue lines) dephasing for various pump laser Rabi frequencies \( \Omega_T \) and fixed probe laser Rabi frequency \( \Omega_S = 0.03 \text{ µeV} \). From top to bottom: \( \Omega_T = 0.03 \text{ µeV}, \Omega_T = 0.3 \text{ µeV}, \Omega_T = 0.8 \text{ µeV} \) and \( \Omega_T = 1.3 \text{ µeV} \). For both models the Lorentzian excited state dephasing amounts to \( \gamma = 0.4 \text{ µeV} \) and the spontaneous emission rate to \( \Gamma_T = \Gamma_S = 0.4 \text{ µeV} \). The model does not include Gaussian broadening of the optical transition. b) Same as in a), but with \( \Omega_S = 0.1 \text{ µeV} \). c) Same as in a), but with \( \Omega_S = 0.3 \text{ µeV} \).

\( \Omega_T \) the spectrum with Gaussian dephasing (red line) exhibits a reduced depth of CPT-dip with an increased FWHM compared to Lorentzian dephasing (blue line). For high coupling laser powers on the other hand the depth of the dip is increased for the spectrum with Gaussian dephasing. The two spectra are quasi identical for \( \Omega_T = 0.8 \text{ µeV} \). Therefore, the three described regimes are observable for \( T_2^{markov} = T_2^* = 10 \text{ ns} \) not only for \( \Omega_S = 0.03 \text{ µeV} \), but also for \( \Omega_S = 0.1 \text{ µeV} \) (Fig. 5.11b) and \( \Omega_S = 0.3 \text{ µeV} \) (Fig. 5.11c). This is not the case for \( T_2^{markov} = T_2^* = 105 \text{ ns} \) (Fig. 5.12). Due to the longer coherence time the spectra for Lorentzian and Gaussian dephasing exhibit equal FWHM of the CPT-dip already at \( \Omega_T = 0.2 \text{ µeV} \). Hence, the regime where Gaussian dephasing leads to a reduced CPT-dip compared to Lorentzian dephasing is not accessible with \( \Omega_S = 0.3 \text{ µeV} \) without reducing the contrast substantially due to spin pumping for \( \Omega_S < \Omega_T \). Furthermore the difference between the two models for large \( \Omega_T \) is reduced, since small dephasing leads to a deep dip even for Lorentzian dephasing. Finally, the coherence time is increased to \( T_2^{markov} = T_2^* = 1 \text{ µs} \) (Fig. 5.13), leading to a narrow and deep CPT-dip. Hence the difference between the two models is only visible for \( \Omega_S = 0.03 \text{ µeV} \) in the regime of small \( \Omega_T = 0.03 \text{ µeV} \) and \( \Omega_T = 0.1 \text{ µeV} \) (Fig. 5.13a).
5.7 Gaussian versus Lorentzian dephasing

Figure 5.12: $T_2^{markov} = T_2^* = 105$ ns. a) Same as in Fig. 5.11a) but with dephasing times of 105 ns. The coupling laser Rabi frequency is varied from top to bottom: $\Omega_{T_0} = 0.03$ $\mu$eV, $\Omega_{T_0} = 0.1$ $\mu$eV, $\Omega_{T_0} = 0.2$ $\mu$eV and $\Omega_{T_0} = 0.6$ $\mu$eV. b) Same as in a), but with $\Omega_S = 0.1$ $\mu$eV. c) Same as in a), but with $\Omega_S = 0.3$ $\mu$eV.

It is challenging to measure with such small Rabi-frequencies as $\Omega_S = 0.03$ $\mu$eV, since the signal to noise ratio (SNR) decreases with the square root of the applied power or linearly with the Rabi-frequency. This is true as long as the applied laser power is below the power for which the measured transition starts to saturate. To answer the question whether first, the difference between Gaussian and Lorentzian dephasing is measurable for $T_2^* = 1$ $\mu$s ($\Omega_S = 0.03$ $\mu$eV) and secondly whether it is feasible to employ CPT to measure coherence times of $T_2^* = 1$ $\mu$s, an estimation of the integration time, necessary to perform the measurements with the SNR of the current set-up will be given. Furthermore I will compare the integration time to the one of Ramsey fringe experiments similar to [24].

From the simulation in Fig. 5.13a one can deduce the difference in depth of the CPT-dip between the spectra with Lorentzian and Gaussian dephasing to $20\%$ of the peak signal. The SNR should be at least one for the difference in CPT-dips. Therefore the SNR of the peak-signal has to be 5. From measurements we know that the SNR is 29 at $\Omega_S = 0.4$ $\mu$eV with 1 s integration time. Hence a integration time of 5 $s$ is necessary to achieve a SNR of 5 at $\Omega_S = 0.03$ $\mu$eV. Here, we assumed that the SNR increases proportional to the square root of the integration time. To map out the CPT dip with FWHM $\sim 0.003$ $\mu$eV (0.8 MHz) roughly 170 points (step size $\sim 0.0006$ eV) are necessary, resulting in a measurement time of 14 minutes. Such measurement times are still feasible. On the other hand to determine $T_2^* = 1$ $\mu$s with an accuracy of 100 ns even longer integration times are necessary, because the difference in CPT-dips for $T_2^* = 1$ $\mu$s and $T_2^* = 0.9$ $\mu$s only amounts to 2 $\%$ of the peak signal. This is a
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Figure 5.13: $T_2^{\text{markov}} = T_2^* = 1 \mu s$. a) Same as in Fig. 5.11a) but with dephasing times of 1 µs. The coupling laser Rabi frequency is varied from top to bottom: $\Omega_{T_0} = 0.03 \mu eV$, $\Omega_{T_0} = 0.1 \mu eV$, $\Omega_{T_0} = 0.13 \mu eV$ and $\Omega_{T_0} = 0.3 \mu eV$. b) Same as in a), but with $\Omega_S = 0.1 \mu eV$. c) Same as in a), but with $\Omega_S = 0.3 \mu eV$.

factor 10 less than before, resulting in a 100 times longer integration time of 500 s per point. Hence the measurement time for 170 points is about 23 hours. On this time scale, charging events in the QD environment affect the stability of the transition energies. In the next step we will calculate the integration time necessary to perform a Ramsey-type experiment. Now, it is possible to measure at the power with maximum SNR. For sample 3 saturation starts to reduce the dR-contrast above $\Omega_S = 0.4 \mu eV$, thereby reducing the SNR. Performing the measurement on the S to R+ transition at $\Omega_S = 0.4 \mu eV$ with integration time of 1s would yield in the current set-up a SNR of 29. At a time delay of 1 µs the fringe-signal reduces to 1/e of its initial value, reducing the SNR to 11. Under the assumption that a SNR of at least 1 yields reliable results, we can reduce the integration time to 8 ms. One has to keep in mind that spin pumping reduces the contrast. Therefore, $8 \cdot 10^6$ repetitions of the same measurement add up to the contrast, achieved in measurements with equal population of the ground states as in a repump experiment (Fig. 5.2). Hence, the integration time per point with a time delay of 1 µs amounts to 8 s. In order to measure the Ramsay fringes for all possible time delays, again very long measurement times would be needed. But in most experiments only a few points around 1 µs and small time delays are measured. Hence, for long coherence times Ramsay type experiments are more feasible. Especially since in CPT experiments the frequencies of pump and probe laser should be locked to prevent additional dephasing due to laser jitter, which is at the order of 5 MHz for unlocked lasers. In Fig. 5.6b, measured with $\Omega_S = 0.4 \mu eV$, a lower bound of $T_2^* = 200$ ns was found. Reducing $\Omega_S = 0.12 \mu eV$ (Fig. 5.6c) in order to extract the exact coherence time, reduces the SNR.
Quasi-recycling transitions

significantly. The dip with FWHM of 7 MHz can not be resolved. Simulations confirm that the current SNR limits the accurate determination (±10 %) of \( T^*_2 \) to not more than \( \sim 100 \) ns: the difference in the depth of the dip between the spectra with \( T^*_2 = 100 \) ns and \( T^*_2 = 90 \) ns amounts to 3.4 % of the peak signal for \( \Omega_S = \Omega_{T_0} = 0.1 \) \( \mu \text{eV} \). Therefore, the integration time per point amounts to 16.5 s, experimentally feasible compared to the 500 s, necessary to determine \( T^*_2 = 1 \) \( \mu \text{s} \). Furthermore, the higher Rabi frequencies result in a FWHM of 0.03 \( \mu \text{eV} \) (7 MHz), at the order of the laser jitter. Hence, CPT can be employed to measure coherence times at the order of 100 ns in the current set-up. Improvement of the SNR and locking of the laser frequencies are necessary to measure longer coherence times.

The possibility to employ CPT-measurements to characterize the noise reservoir sheds light on the versatility of CPT. Since the model described here assumes a quasi-static reservoir with a decorrelation time going to infinity for the Gaussian fluctuations, a next step could be to develop a more elaborate model including variable decorrelation times.

5.8 Quasi-recycling transitions

Figure 5.14: Separately addressing the \( T_+ \) to \( R_+ \) transition and the \( T_+ \) to \( R_{++} \) transition. a) Probing the \( T_0 \) to \( R_+ \) and \( T_+ \) to \( R_{++} \) transitions with \( \Omega_T = 0.34 \) \( \mu \text{eV} \) while pumping the \( S \) to \( R_+ \) transition with \( \Omega_S = 3.4 \) \( \mu \text{eV} \) at \( B = 2 \) T and \( \Delta V = -15 \) mV. In contrast to the \( T_+ \) to \( R_{++} \) transition, the \( T_0 \) to \( R_+ \) transition is only visible at resonance of the pump laser. b) CPT spectrum of the \( T_0 \) to \( R_+ \) transition at \( B = 2 \) T and \( \Delta V = -11 \) mV with \( \Omega_S = 1.06 \) \( \mu \text{eV} \). At blue detuning the cycling \( T_+ \) to \( R_{++} \) becomes visible. Diagrams of possible relaxation channels from \( R_+, R_{++} \) states under excitations from the \( T \) states are inserted.

In addition to featuring a lambda system that can be robust against electric and magnetic field fluctuations, the two-electron CQD molecule offers additional useful features. We find that the electronic g-factors in both dots are slightly different (\( g_e^{red} = 0.53 \) and 0.55).
5. Optically active singlet-triplet qubits with long coherence times

$g_e^{\text{blue}} = 0.47$, which detunes the $T_+ - R_{++}$ from the $T_0 - R_+$ transition, allowing them to be separately addressed at moderate magnetic fields (Fig. 5.14). To implement single-shot spin read-out [21], which requires recycling transitions, the S-population could be directly transferred to the $R_{++}$ state with a strong laser, and subsequently read out using light scattering on the $T_+ - R_{++}$ transition. The rich optical excitation spectrum of QD molecules in the (1,1) regime thus combines the advantages of both Voigt [73] and Faraday geometries [76].

5.9 Double $\Lambda$-scheme

Moreover, the highly tunable twelve-level spectrum offers rich possibilities for solid-state non-linear optics. A parametric amplifier based on a double lambda scheme for instance, can be implemented by coupling the excited states in the red and blue QD to the $S - T_0$ ground states [35, 96, 97] (Fig. 5.15a). To verify that the excited states of both QDs share common ground states, we perform repump experiments (Fig. 5.15b and c). To this end we probe the $S$ ($T$) transition in the red QD while pumping the $T$ ($S$) transition in the blue QD. When both lasers are on resonance, the spin state is randomized, leading to a sizable $dR$ signal. Another double $\Lambda$-scheme is formed at finite magnetic fields. Here, the $S$ and $T$ ground states couple to $R_+$ and $R_-$ in the red QD ($B_+$ and $B_-$ in the blue QD). The emission energies for both types of double $\Lambda$-schemes are highly tunable by adjusting either growth parameters (QD size, $\Delta E_{ST}$ via interdot tunnel barrier) or the applied magnetic field and bias voltage.

Figure 5.15: **Double $\Lambda$-scheme in the (1,1) regime.** a) Schematic of the double $\Lambda$-scheme, formed by $S$ and $T$ ground states together with the optically excited states in the red and blue QD. A stronger repump laser is applied to the $T$ to $B$ transition of the blue QD and a probe laser to the $S$ to $R$ transitions of the red QD. The corresponding experiment is shown in b), performed in dR with $\Omega_{\text{coupling}} = 0.68 \mu eV$ and $\Omega_{\text{probe}} = 2.0 \mu eV$ at $B = 0$ T and $\Delta V = -93$ mV. c) The probe laser is now swept across the $T$ to $R$ transitions of the red QD, while stepping the coupling laser over the $S$ to $B$ transition in the blue QD. The contrast is therefore three times higher than in b).
Furthermore, one could imagine coupling $S - T_0$-qubits via long-range dipole-dipole interactions, employing indirect excitons [98]. We find such an indirect exciton with a large dc-dipole of size 8 nm at the edge of the (1,1) regime (steep line in the right side of Fig. 5.2). Another very interesting possibility is highlighted in Fig. 5.4e where it can be seen that application of in-plane magnetic field yields two dark resonances [99]: it has been shown theoretically that by adiabatically changing the laser intensity and phase in a three-laser geometry, it is possible to realize a Hadamard Berry-phase gate [100], rotating the system wave-function from one dark state to a coherent superposition of the two dark states. Since the two dark states in this configuration are not protected against nuclear Overhauser field fluctuations along the direction of the external field, it would be necessary to first prepare the QD nuclear states in a well defined state using resonance dragging.

This chapter concludes the investigation of the (1,1) regime and the possible employment of CQDs as ST-qubits in quantum computing.
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Chapter 6

P-state spectroscopy

The experiments in the previous chapters were performed in the (1,1) regime, which proved to be useful for multiple applications in quantum optics and quantum information processing. The focus of this chapter will lie on the (3,1) regime, which exhibits S and T ground states, similar to the (1,1) regime. It will be shown that the (3,1) regime is rendered unsuitable for quantum information processing by fast relaxation of the electron spin, but can be employed for p-state spectroscopy of the bottom QD.

6.1 Introduction

Multiply charged excitons can offer interesting spin physics. The investigation of a triply negatively charged exciton in a single QD for instance [61] revealed a change of the spin configuration of the optically excited state from T to S type and coupling to a continuum of states by applying a magnetic field. Those measurements were performed employing PL. In single QDs multiple spin states are challenging to probe resonantly; p to p transitions for instance are short lived [48] and d to d transitions are in many QDs not stable, because the electron escapes to the back contact. Bracker et al. [36] were the first to use CQDs to probe higher energy hole states of the bottom QD through the $X^0$ of the top QD in PL. In this chapter we gain information about the spin states of electrons in the p-orbitals of the bottom QD via resonant spectroscopy of the top QD. Furthermore we can change the ground state spin character from T to S type by applying a magnetic field [47].

6.2 Resonance of s and p-states

The experiments of this chapter were performed with sample 1. The spacer-layer thickness of sample 1 is optimized for electron-tunneling (9 nm), but the emission wavelength of the QDs is chosen as for hole-tunneling: the top QD has a higher transition energy
than the bottom QD. Therefore, the electronic p-states of the bottom QD can be tuned into resonance with electronic s-states of the top QD (Fig. 6.1a) via the gate voltage, enabling electron tunneling. This is evidenced in the PL-measurement in Fig. 6.1c, where the emission energies and the charging sequence of the two QDs can be identified. The bottom QD emits around 1267 meV, \( \sim 10 \) meV red detuned with respect to the top QD. Consequently, the bottom QD charges first; there are already four electrons captured in the bottom QD, when the ground state of the top QD changes from neutral to singly charged. Two of the four electrons in the bottom QD sit in the p-shell, since the s-shell is fully occupied with the other two electrons. The exact charging sequence is depicted in Fig. 6.1c: the different charging regimes are separated with dashed vertical lines.

Figure 6.1: Tunnel resonances with p-orbitals. a) Shown is a scheme of the band structure of sample 1, where QD-R is the bottom QD and QD-B the top QD. b) Viewgraph explaining the T(4,0) to T(3,1) tunnel-resonance. Violet lines indicate energy states in QD-R, blue lines energy states in QD-B. c) A PL-measurement of both QDs versus gate voltage is shown. The charging states are indicated.

6.3 Properties of the (3,1) regime

Most interesting is the (4,0) to (3,1) ground state tunnel-resonance. On the first glance, the dT-measurement of the top QD (Fig. 6.2a) looks similar to the (2,0) to (1,1) anticrossing. This is not surprising, because the two electrons in the s-orbital of the bottom QD form a spin-singlet \( m_z = 0 \) and do not influence the spin dynamics of the other electrons to first order. Hence, the two remaining electrons form one S and three T eigenstates. But in contrast to the (1,1) regime, the degenerate T-states are lower in
energy than the S-state. This due to the fact, that also the T-states exhibit tunnel-coupling, visible in the curvature of the T-transition in the dT-measurement of the top QD versus gate voltage in Fig. 6.2a. The T-transition is identified by the three times higher contrast compared to the S-transition. The (3,1) T couples to the (4,0) T, where the fourth electron sits in the second p-orbital $p_2$ (Fig. 6.1b). The (4,0) T-state is energetically favorable to the (4,0) S-state, because the energy gained by the S-T exchange interaction $J_{ST}$ is larger than the bare splitting of the two p-orbitals $\Delta_{12}$ for $B = 0$ T [47].

Figure 6.2: **S and T states in the (4,0)-(3,1) regime.** (a) Shown is a dT measurement of QD-B versus gate voltage at $B = 0$ T with saturation power. S and T states are indicated. (b) RF measurements with power below saturation. The photons are detected with a CCD camera after passing a 1200 g/mm grating. Blue trace: the laser is resonant with the S-transition. Red trace: the laser resonant with the T-transition. The yellow arrow indicates the frequency of the laser.

The RF measurement in Fig. 6.2b proves, that the S and T-states in the (3,1) regime form a Λ-scheme together with the degenerate excited states of the top QD. The experiment is performed by either pumping the S (blue trace) or the T-transition (red trace). Emission of both transitions can be detected with a CCD camera after spectrally dispersing the signal with a grating. Although spin-pumping is normally observed in Λ-schemes (section 3.4), both S and T-transitions lack spin-pumping (Fig. 6.2a). This can most probably be attributed to the strong coupling of the p-states to the back contact, because of their high energy compared to the s-states. The sample structure itself allows for spin-pumping, since it was observed for the uncoupled $X^{1-}$ of the bottom QD. Consequently, the (3,1) regime is not suitable for applications in the need of long spin lifetimes, like quantum information processing, but can be employed to probe the spin states of the p-orbitals of the bottom QD.
6.4 Manipulating the p-state spin-character

We have seen, that for $B = 0$ T the lowest energy state of the (4,0) regime is a spin triplet, where one electron resides in the $p_1$ and one in the $p_2$ orbital respectively (Fig. 6.3b). But it is possible to change the spin character of the lowest energy state by applying a magnetic field in Faraday-geometry. Fig. 6.3a shows $dT$-measurements versus gate voltage of the $X_{1-}$ of the top QD for four different magnetic fields. The T state is lower in energy than the S state (the T-transition is higher in energy than the S-transition) for $B < 0.7$ T. Ramping the magnetic field to $B = 0.7$ T leads to degeneracy of the two transitions and for $B > 0.7$ T, the energy of the S-state finally becomes lower than the T-state energy. The mechanism is explained in Fig. 6.3b: for small magnetic fields the energy gained by $J_{ST}$ is larger than the splitting of the $p$-orbitals $\Delta_{12}$. Increasing the magnetic field also increases $\Delta_{12}$; therefore the exchange interaction matches $\Delta_{12}$ at $B = 0.7$ T. Increasing the magnetic field even further finally rises $\Delta_{12}$ above $J_{ST}$ and hence the energy of the S-state below the one of the T-state.

![Figure 6.3: Tuning spin-properties via the B-field. a) $dT$-measurement versus gate voltage with saturation power for various magnetic fields, applied in Faraday-geometry. Top-left to bottom-right panel: $B = 0.4$ T, $B = 0.7$ T, $B = 1$ T and $B = 1.3$ T. b) Scheme of the S and T states in the (4,0) regime. Left panel: the T state is lower in energy for $B < 0.7$ T. Right panel: for $B > 0.7$ T, the S state becomes more favorable.](image)

To conclude, a sample design where the bottom QD is more red detuned than the top QD allows for addressing the (3,1) regime instead of the (1,1) regime. Therefore it is possible to probe and manipulate the p-states of the bottom QD via the s-state of the top QD. One could think of other experiments, where one QD is employed to perform spectroscopy on the other QD; for instance probing nuclear spin polarizations.
Chapter 7

Outlook

This thesis gave a glance on the manifold of experiments and applications possible with CQDs. The singlet and triplet ground states of the (1,1) charging regime can be employed in two different regimes: strong coupling to the back contact in combination with big exchange splitting renders the CQD suitable for optical amplification, whereas to first order decoherence free ST-qubits can be isolated for small coupling to the back contact and operation at the sweet spot. Latter opens the way for applications in quantum information processing. Furthermore, in the previous chapter the top QD was used to probe the p-states of the bottom QD. Not only the p-states, but also other physical properties like nuclear spin polarization can be investigated by using one QD to probe the properties of other QD. In the following paragraphs I will give perspectives for all of the three mentioned applications of CQDs.

7.1 Implementation of a single quantum dot laser

It was stated in chapter 4 that a cavity with modest quality factor \( Q \sim 7000 \) is necessary to implement laser oscillations with sample 2. Different approaches are imaginable in order to embed the CQD into a cavity. One possibility is to grow a DBR mirror underneath the CQD, with wavelength optimized to the \( T_0 \) to \( R_+ \) transition, which will be amplified. The tip of a fiber with dielectrics evaporated on top \[101, 102\], could serve as second mirror of the cavity and photon collector at the same time. Fiber-mirrors are know for their high finesse, exceeding 130000 \[101\]. But the metal on top of the QD-sample would reduce the quality factor of the cavity. Therefore, one could replace the Schottky-diode heterostructure with a \( p - i - n \) structure, thereby rendering a top gate unnecessary. Another difficulty lies in coupling both pump and probe laser into the cavity, because their transition energies deviate by the exchange splitting. A possible solution is to apply the pump laser from the side.

A second option to realize a single QD laser consists of coupling the CQD to whispering gallery modes of a micro-sphere \[103\] or micro-disc cavity \[104\]. The quality factor of
the latter one can exceed 100 million. A fiber-taper \[105\] can be employed to introduce light into the cavity and read-out. Finally the CQD has to be brought close to the cavity to achieve sizable coupling between CQD and cavity. Here, the experimental set-up is challenging, especially to fit the set-up into a cryostat.

7.2 Quantum information processing with a singlet-triplet qubit

Coherent population trapping was employed to extract coherence times of the S and T ground states of a CQD in the (1,1) regime and to characterize the noise reservoir. We showed, that a ST qubit can be rendered insensitive to magnetic and electrical fluctuations by operating the qubit at the sweet spot. A next step could be to utilize the long coherence times of the ST qubit for spin manipulation and implementation of gates.

Stimulated Raman adiabatic passage (STIRAP) \[34\] is one way to transfer population in a Λ-scheme from one ground state to the other. Meaning in our case, rotating the spin-state from S to T type. The experiment has to be performed adiabatically, thereby leaving the excited state unpopulated.

A proposal suggests the implementation of a Hadamar phase-gate in tripod systems \[100\]. Such a tripod system can be found for S and T states in Voigt-geometry \[99\]. Three (new) eigenstates are formed, of which two have strong $T_0$-character: $|T_0\rangle_x = \frac{1}{\sqrt{2}}(|T_+\rangle - |T_-=\rangle)$, $|T_+\rangle_x = \frac{1}{2}(|T_+\rangle + \sqrt{2}|T_0\rangle + |T_-\rangle)$ and $|T_-=\rangle_x = \frac{1}{2}(|T_+\rangle - \sqrt{2}|T_0\rangle + |T_-\rangle)$. Therefore two dark resonances evolve in CPT-experiments; the phase-gate consists of rotating the system wave-function from one dark state to a coherent superposition of the two dark states. Since the experiment makes use of in-plane magnetic fields, the ST qubit is no longer insensitive to magnetic fluctuations. One could employ resonant dragging \[8\] to prepare the Overhauser field in a well defined state.

Resonant dragging was used before to significantly prolong the coherence time of a single electron in a QD \[93\]. Furthermore, theoretical calculations predict dark state dragging \[106\], stabilizing the dark resonance for various probe laser detunings and thereby preparing the nuclear spin environment in a well-defined state. But dark state dragging requires pump and probe laser power levels below saturation, yielding small contrast. Dark-state dragging was not yet observed in single QDs. Since the S and T ground states in CQD are to first order insensitive to the hyperfine interaction, dark state dragging is very unlikely to be observed there. Nevertheless, CPT-measurements at $B = 2$ T show a dark resonance, which is stable over a range of probe laser detunings (Fig. 7.1). The range increases for measurements away from the edge of the (1,1) stability region, where the sweet spot for this QD-molecule is located and co-tunneling is strong. Furthermore, the shape of the CPT-dip depends on the scan direction of the probe laser (gray trace in Fig. 7.1). The mechanism remains an open question; the answer requiring a more detailed theoretical and experimental analysis.
7.3 Probing nuclear spin effects

As described above, resonant dragging can be used to quiet the nuclear spin environment and thereby prolong the electron spin coherence time. Another approach for the realization of long coherence times was proposed in [107, 108], employing the nuclear spin itself for quantum information storage. To this end a more detailed understanding of nuclear spin interactions is required. CQD could prove useful to probe nuclear spin effects. As shown in the previous chapter, the top QD was employed to perform p-state spectroscopy on the bottom QD. In the same way nuclear spin polarization could be probed. Furthermore, the rich optical spectrum in the (1,1) regime with eight optically allowed transitions offers possibilities to create nuclear spin polarization easily. In Fig. 7.2a, a resonant laser probes the T++ to R++ transition while a twenty times stronger pump laser is stepped over the S to R++ and S to R-- transitions respectively, which are optically forbidden. Nevertheless they exhibit a small oscillator strength; \( \sim 250 \) times smaller [76] than for the optically allowed transitions. At resonance of the pump laser, most probably an electron spin flip in the bottom QD (QD-B) is induced. Hence, dynamic nuclear spin polarization takes place, observable in the shift of the T++ to R++ resonance. For pumping the S to R-- transition (positive pump detuning), the nuclear field has opposite sign than for pumping the S to R++ transition (negative pump detuning), corresponding to the direction of the electron flip in QD-B. The pump laser increases the population in the T++-state at resonance to the S to R++ transition and therefore the contrast of the T++ to R++ transition increases as well. Fig. 7.2b shows the same kind of experiment but with the probe laser scanned over the T-- to

Figure 7.1: Dark-state dragging. Shown are CPT-spectra for various detuning from the edge of the (1,1) regime: \( \Delta V = -10 \text{ mV}, \Delta V = -15 \text{ mV}, \Delta V = -20 \text{ mV} \) and \( \Delta V = -25 \text{ mV} \) at \( B = 2 \text{ T}, \Omega_{T_0} = 1.08 \text{ } \mu\text{eV} \) and \( \Omega_{S} = 0.34 \text{ } \mu\text{eV} \). The laser is swept from negative to positive deunings. A reversed sweep is shown at \( \Delta V = -20 \text{ mV} \) (light gray trace).
Figure 7.2: **Dynamic nuclear spin polarization.** a) Probing the $T_{++}$ to $R_{++}$ transition in $dR$ with power below saturation while stepping a twenty times stronger pump laser over the $S$ to $R_{++}$ and $S$ to $R_{--}$ transition respectively. b) Same as in a, but probing the $T_{--}$ to $R_{--}$ transition.

Therefore the transition energy is shifted in the opposite direction than before and the contrast is increased at resonance of the pump laser to the $S$ to $R_{--}$ transition. Depending on how fast the nuclear spin polarization distributes over the two QDs, one could employ the dynamic nuclear spin polarization to locally create magnetic fields. On the other hand one could measure the nuclear spin diffusion time constant by pumping nuclear spins in one QD and probing the polarization in the other QD. Preliminary measurements showed a fast distribution of the nuclear spin polarization created in the top QD over both QDs. This could be due to the fact that the QDs are strongly coupled in the (1,1) regime. Hence, the wavefunction of the QD-molecule is spread over both QDs \[109\]. The fast distribution of the nuclear spin polarization would also complicate experiments investigating the dragging mechanism; it is not clear yet whether electron or hole are involved. Here, one would like to work in a regime, where dragging in one QD would be due to the electron, whereas it would be due to the hole in the other QD. In order to distinguish in which QD the polarization was created in the first place, diffusion should be slow.

Finally I want to emphasize the complexity of CQDs, allowing for a variety of experiments in a bunch of different fields of physics, reaching from classical spectroscopy to realization of single emitter lasers to decoherence-avoiding qubits.
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[1] Hermann Hesse


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5.7 **Gaussian fluctuations.** a) dR measurement of the indirect transition in the (1,2)-regime versus gate voltage at $B = 0.2 \text{ T}$. b) Line cut of a) at 196.2 mV, as indicated by the red line in a. The red line in b is a Gaussian fit with $\delta V = 0.6 \text{ mV}$ the width of the Gaussian distribution.

5.8 **Nuclear spin effects.** dT measurement of blue Zeeman transition of the neutral exciton in QD-R for various B-fields: $B = 0 \text{ T}$ to $B = 0.5 \text{ T}$. The black arrow indicates the sweep direction of the laser.

5.9 **Co-tunneling rates and excited state mixing.** Blue dots: dR-spectrum at $B = 0.2 \text{ T}$ with $\Omega_T = 0.45 \mu\text{eV}$. Red line: fits employing the eight-level simulation with $\gamma_g = 2.5 \cdot 10^{-3} \mu\text{eV}$ and $\gamma_{es} = 2.5 \cdot 10^{-2} \mu\text{eV}$. Dark gray line: $\gamma_g = 5 \cdot 10^{-3} \mu\text{eV}$ and $\gamma_{es} = 2.5 \cdot 10^{-2} \mu\text{eV}$; light gray line: $\gamma_g = 2.5 \cdot 10^{-3} \mu\text{eV}$ and $\gamma_{es} = 5 \cdot 10^{-2} \mu\text{eV}$.

5.10 **Asymmetric line shape.** Shown are dT (blue trace) and dR (red trace) measurements of the neutral exciton in the red QD for the same optical alignment at $B = 0 \text{ T}$ with laser power below saturation ($\Omega = 0.34 \mu\text{eV}$).

5.11 $T^*_2^{\text{Markov}} = T^*_2 = 10 \text{ ns}$. a) Plotted is the solution $(\rho_{SR})$ of a three-level master equation including either pure Gaussian (red lines) or pure Lorentzian (blue lines) dephasing for various pump laser Rabi frequencies $\Omega_{T_0}$ and fixed probe laser Rabi frequency $\Omega_S = 0.03 \mu\text{eV}$. From top to bottom: $\Omega_{T_0} = 0.03 \mu\text{eV}$, $\Omega_{T_0} = 0.3 \mu\text{eV}$, $\Omega_{T_0} = 0.8 \mu\text{eV}$ and $\Omega_{T_0} = 1.3 \mu\text{eV}$. For both models the Lorentzian excited state dephasing amounts to $\gamma = 0.4 \mu\text{eV}$ and the spontaneous emission rate to $\Gamma_{T_0} = \Gamma_S = 0.4 \mu\text{eV}$. The model does not include Gaussian broadening of the optical transition. b) Same as in a), but with $\Omega_{T_0} = 0.1 \mu\text{eV}$. c) Same as in a), but with $\Omega_{S} = 0.3 \mu\text{eV}$.

5.12 $T^*_2^{\text{Markov}} = T^*_2 = 105 \text{ ns}$. a) Same as in Fig. 5.11a) but with dephasing times of 105 ns. The coupling laser Rabi frequency is varied from top to bottom: $\Omega_{T_0} = 0.03 \mu\text{eV}, \Omega_{T_0} = 0.1 \mu\text{eV}, \Omega_{T_0} = 0.2 \mu\text{eV}$ and $\Omega_{T_0} = 0.6 \mu\text{eV}$. b) Same as in a), but with $\Omega_{S} = 0.1 \mu\text{eV}$. c) Same as in a), but with $\Omega_{S} = 0.3 \mu\text{eV}$. 

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5.13 $T_{2}^{\text{markov}} = T_{2}^{*} = 1 \mu s$. a) Same as in Fig. 5.11a) but with dephasing times of $1 \mu s$. The coupling laser Rabi frequency is varied from top to bottom: $\Omega_{Ta} = 0.03 \mu eV$, $\Omega_{Tb} = 0.1 \mu eV$, $\Omega_{Td} = 0.13 \mu eV$ and $\Omega_{Tb} = 0.3 \mu eV$. b) Same as in a), but with $\Omega_{S} = 0.1 \mu eV$. c) Same as in a), but with $\Omega_{S} = 0.3 \mu eV$.

5.14 **Separately addressing the $T_+ \rightarrow R_+$ transition and the $T_+ \rightarrow R_{++}$ transition.** a) Probing the $T_0 \rightarrow R_+$ and $T_+ \rightarrow R_{++}$ transitions with $\Omega_{T} = 0.34 \mu eV$ while pumping the $S \rightarrow R_+$ transition with $\Omega_{S} = 3.4 \mu eV$ at $B = 2 T$ and $\Delta V = -11 mV$. In contrast to the $T_+ \rightarrow R_{++}$ transition, the $T_0 \rightarrow R_+$ transition is only visible at resonance of the pump laser. b) CPT spectrum of the $T_0 \rightarrow R_+$ transition at $B = 2 T$ and $\Delta V = -11 mV$ with $\Omega_{S} = 1.06 \mu eV$. At blue detuning the cycling $T_+ \rightarrow R_{++}$ becomes visible. Diagrams of possible relaxation channels from $R_+$, $R_{++}$ states under excitations from the $T$ states are inserted.

5.15 **Double $\Lambda$-scheme in the (1,1) regime.** a) Schematic of the double $\Lambda$-scheme, formed by $S$ and $T$ ground states together with the optically excited states in the red and blue QD. A stronger repump laser is applied to the $T$ to $B$ transition of the blue QD and a probe laser to the $S$ to $R$ transitions of the red QD. The corresponding experiment is shown in b), performed in $dR$ with $\Omega_{\text{coupling}} = 0.68 \mu eV$ and $\Omega_{\text{probe}} = 2.0 \mu eV$ at $B = 0 T$ and $\Delta V = -93 mV$. c) The probe laser is now swept across the $T$ to $R$ transitions of the red QD, while stepping the coupling laser over the $S$ to $B$ transition in the blue QD. The contrast is therefore three times higher than in b).

6.1 **Tunnel resonances with p-orbitals.** a) Shown is a scheme of the band structure of sample 1, where QD-R is the bottom QD and QD-B the top QD. b) Viewgraph explaining the $T(4,0)$ to $T(3,1)$ tunnel-resonance. Violet lines indicate energy states in QD-R, blue lines energy states in QD-B. c) A PL-measurement of both QDs versus gate voltage is shown. The charging states are indicated.

6.2 **S and T states in the (4,0)-(3,1) regime.** a) Shown is a $dT$ measurement of QD-B versus gate voltage at $B = 0 T$ with saturation power. S and T states are indicated. b) RF measurements with power below saturation. The photons are detected with a CCD camera after passing a 1200 g/mm grating. Blue trace: the laser is resonant with the S-transition. Red trace: the laser resonant with the $T$-transition. The yellow arrow indicates the frequency of the laser.

6.3 **Tuning spin-properties via the B-field.** a) $dT$-measurement versus gate voltage with saturation power for various magnetic fields, applied in Faraday-geometry. Top-left to bottom-right panel: $B = 0.4 T$, $B = 0.7 T$, $B = 1 T$ and $B = 1.3 T$. b) Scheme of the S and T states in the (4,0) regime. Left panel: the T state is lower in energy for $B < 0.7 T$. Right panel: for $B > 0.7 T$, the S state becomes more favorable.
7.1 Dark-state dragging. Shown are CPT-spectra for various detuning from the edge of the (1,1) regime: $\Delta V = -10 \text{ mV}$, $\Delta V = -15 \text{ mV}$, $\Delta V = -20 \text{ mV}$ and $\Delta V = -25 \text{ mV}$ at $B = 2 \text{ T}$, $\Omega_{T_b} = 1.08 \mu\text{eV}$ and $\Omega_S = 0.34 \mu\text{eV}$. The laser is swept from negative to positive detunings. A reversed sweep is shown at $\Delta V = -20 \text{ mV}$ (light gray trace).

7.2 Dynamic nuclear spin polarization. a) Probing the $T_{++}$ to $R_{++}$ transition in dR with power below saturation while stepping a twenty times stronger pump laser over the S to $R_{++}$ and S to $R_{--}$ transition respectively. b) Same as in a, but probing the $T_{--}$ to $R_{--}$ transition.
Acknowledgments

First of all, I would like to thank Atac for letting me join his group and transferring some of his physics knowledge to me. He was always happy to answer questions and gave me the feeling that my work was appreciated. Atac is a nice person, interested not only in physics. He managed to put a group of competent and nice people together. Many thanks go to Jeroen for advice in the lab, help with simulations, many discussions and being a nice person enjoyable to work with.

I want to thank Javier for the effort he put into growing the perfect sample and the insight into growth details he gave me. His everlasting good mood and motivation are inspiring.

I would like to thank the whole group for the good atmosphere and willingness to participate in after-work activities. Special thanks go to Dora for being a great friend, many trips and enjoyable moments. For nice city trips and climbing excursions I would like to thank Andreas, Andres and Susanne. Martin I would like to thank for revising the thesis and many movie and bar evenings. Many thanks go to Mena for being a great office and flat mate, always up for interesting discussions. For many chats I would like to thank Priska.

I want to thank Christian for his interest in physics and his support. Last but not least I want to thank my family for their trust and support.