Doctoral Thesis

Optical investigation of many-body effects in single quantum dots

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Optical Investigation of Many-Body Effects in Single Quantum Dots

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Summary

This thesis discusses optical signatures induced by the interplay between a well-defined few-degree system and many body interactions with the complex environment of a solid-state system. In particular, the interactions of a single confined QD electron with the $10^4 - 10^5$ nuclear spins of the host-material and the interaction with a Fermionic reservoir (FR) are the subjects of interest.

The first part of the thesis, focuses on the coupling a single confined electron to an ensemble of $10^4 - 10^5$ nuclear spin of the host material. A first experiment demonstrates that for a wide range of parameters, it is not possible to isolate elementary QD excitations from the strong influence of nuclear spins: the absorption lineshape at magnetic fields exceeding 1 Tesla indicates that the nuclear spins polarize by an amount that ensures locking of the QD resonance to the incident laser. In stark contrast to earlier experiments, this nuclear spin polarization is bi-directional, allowing the electron plus nuclear spin system to track the changes in laser frequency dynamically on both sides of the resonance. We find that the confluence of laser excitation and nuclear spin polarization suppresses the fluctuations in resonant absorption. In a second experiment, time-resolved absorption spectroscopy is used to investigate the dynamics of the coupled electron-nuclear spin system at an external magnetic field of 5 T. This experiment reveals that in the absence of co-tunneling, the system dynamics is dominated by hyperfine interaction: indirect nuclear spin interaction mediated by the electronic spin leading to redistribution of nuclear spin polarization within the QD.

The second part of the thesis deals with the interactions between a single confined QD spin and the spins of a Fermionic reservoir (FR). This interaction leads to one of the most spectacular phenomena of many body physics – the Kondo effect. Here the observation of Kondo correlations in optical absorption measurements on a single semiconductor quantum dot is reported. In contrast to transport experiments, absorption of a single photon leads to an abrupt change in the system Hamiltonian and a quantum quench of Kondo correlations. This quantum quench leads to so-called orthogonality catastrophe, induced by a vanishing overlap between the initial and final many-body wavefunctions. The signature of the latter is characteristic power-law tails in the experimental absorption lineshapes.

We show that the power law exponent that determines the degree of orthogonality is tunable by an external magnetic field, which unequivocally demonstrates that the observed absorption lineshape originates from Kondo correlations. Our experiments demonstrate that optical measurements on single artificial atoms offer new perspectives on many-body phenomena previously studied exclusively using transport spectroscopy. Moreover, they initiate a new paradigm for quantum optics where many-body physics plays a crucial role.
Zusammenfassung

Im Rahmen dieser Arbeit werden die optischen Signaturen behandelt, welche durch das Zusammenspiel eines wohldefinierten Systems weniger Freiheitsgrade und den Vielteilchenwechselwirkungen der komplexen Umgebung in einem Festkörper entstehen. Im Detail gilt das Interesse den Wechselwirkungen zwischen einem einzelnen Elektron, gebunden in einem Quantenpunkt, mit einem Kernspinreservoir und einem fermionischen Reservoir.

Im ersten Teil dieser Arbeit wird die Kopplung eines einzelnen gebundenen Elektrons an die $10^4 - 10^5$ Kernspins des Quantenpunktkristalls untersucht. Wir demonstrieren in einem Experiment, dass es über einen weiten Parameterbereich nicht möglich ist, elementare Quantenpunkübergänge vom starken Einfluss der Kernspins zu isolieren: In Absorptionsspektren bei Magnetfeldern, welche grösser sind als 1 T, werden Kernspins in einem Masse polarisiert, dass die sich die Übergangsfrequenz exakt um die Laserverstimmung ändert. Die Absorptionslinien bei Magnetfeldern, welche grösser sind als 1 T, deuten an, dass eine Kernspinpolarisation aufgebaut wird, welche die Verstimmung des einfallenden Lasers kompensiert. In starkem Kontrast zu früheren Messungen ist diese Kernspinpolarisation bidirektional, was dem Elektron-Kernspin-System erlaubt, Änderungen in der Laserenergie dynamisch zu folgen. Wir zeigen, dass die Konfluenz von Laseranregung und Kernspinpolarisation Fluktuationen in der resonanten Absorption unterdrückt. In einem weiteren Experiment wird mittels zeitaufgelöster Absorptionsspektroskopie die Dynamik des gekoppelten Elektron-Kernspin-Systems bei einem externen Magnetfeld von 5 T untersucht. Dieses Experiment zeigt, dass die Dynamik bei fehlender Kotunnelkopplung durch Hyperfeinwechselwirkung dominiert ist: die indirekte, diffusionsartige Kopplung zweier Kernspins über das Elektron führt zu einer Umverteilung der Kernspinpolarisation im Quantenpunkt.


In unseren Experimenten zeigen wir, dass die Abhängigkeit der Flanken gegeben ist durch den Grad der Orthogonalität, welche durch ein externes Magnetfeld
verändert werden kann.

Unsere Experiment demonstrieren, dass optische Messungen an einem einzelnen Quantenpunkt neue Möglichkeiten bieten, Vielteilchenphänomene zu erforschen, die bisher exklusiv Transportexperimenten vorbehalten waren. Ferner leiten sie ein neues Paradigma für die Quantenoptik ein, in der nun Vielteilchenphysik eine entscheidende Rolle spielt.
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1. Introduction

The enormous progress in nano-fabrication within the last decades enabled scientists to tailor matter at nanometer length scale. This progress led to the emergence of many new research areas, all exploring the physics of low dimensional structures and their applications. Generally speaking, nanotechnology is an umbrella term referring to the research of fabrication and manipulation of objects wherein at least one spatial dimension extends only a few nanometer. In most solids at temperatures of interest, this is the length scale of the electron’s de Broglie wavelength and hence when objects are made smaller and smaller, a classical description fails to describe the physics at this scale and quantum mechanical effects become important.

With respect to the electronic and optical properties, an important role is played by the confinement of charge carriers in one or more dimensions. In this thesis we restrict ourselves to semiconductor structures with confinement in all three dimensions: so called quantum dots (QDs). QDs are zero-dimensional synthetic materials made of thousands of atoms and have the ability to confine charge carriers on their length scale [1]. Due to their discrete energy spectrum and optical properties, semiconductor QDs are similar to atoms, and therefore sometimes referred to as ‘artificial atoms’. With respect to their optical emission wavelength, the term ‘artificial atom’ could not be more accurate. QD are particularly significant for optical applications due to the tunability of their emission wavelength. In an ensemble, QDs could be used as a broad-band gain medium in lasers with a high output power and low threshold currents, in optical detectors and white-light sources [2]. In photovoltaic devices they may be able to increase the efficiency. A particular kind of QDs made of cadmium and selenium have been proven to be a very useful tool in biotechnology, where they are utilized in medical research and diagnostics for labeling and observation of biological processes. The prominent luminescence properties can be used as a tool for selective imaging of tumor cells in living organisms [3].

Besides their applications as an ensemble, the study of single QDs lies at the heart of nano-technology where confined single-electron spins in III-V semiconductor QDs are considered to be promising candidates for the implementation of devices in quantum communications and quantum information processing. A huge research field has emerged which solely addresses the problem of initialization, manipulation, and readout of single electron spins [4–7]. An obvious advantage of using such ‘artificial atoms’ over real atoms is scalability - a complex “quantum circuitry” could in principle be packed on a small chip. The price one needs to pay in return is the rapid destruction of quantum coherence in solid-state as opposed to atomic systems. Indeed, the electron in a QD sees a variety of complex environments and interactions intrinsic to the host material. Interactions with such environments are a major source for interrupting the desired quantum evolution in the solid state. For example, the electron spin is coupled to an ensemble of $10^4$ nuclear spins of the QD atoms via hyperfine interaction. For an unprepared nuclear spin state each
nuclear spin is randomly oriented giving rise to a fluctuating effective magnetic field seen by the electron. It is now well-known that this field (Overhauser field) is the main source of electron spin decoherence [8]. Although, this may appear at first as a drawback, one could imagine preparing the nuclear spins in specific states in order to prolong electron spin coherence. Furthermore, by controlling the nuclear spins, one could envision manipulating the spin of the electron [9].

In addition to the quantum information perspective, QDs can be used as a sensitive and well-controlled probe of interesting and rich many-body physics which lies at the heart of aforementioned interactions in the solid state. As an example, consider the coupling of a single QD electron spin with the spins of a nearby Fermionic reservoir (FR) arising from the exchange interaction which can be controlled by sample design and experimental parameters. This constitutes a realization of the Kondo problem using a QD spin as the impurity. In the Kondo effect, a localized magnetic impurity (here a single electron) is screened by FR electron within a certain energy range (the Kondo temperature $T_K$) determined by the system parameters [10]. The screening occurs via a many-body effect, by the built-up of strong correlations between the impurity spins and the FR spins. This effect was first explained by Jun Kondo in 1964 in the context of “resistance minimum problem” in metals with magnetic impurities. Recently, the Kondo effect has been studied extensively in electrically confined QDs using transport measurements [11–13].

**Scope of this thesis**

The study of such many-body physics through optical spectroscopy of self-assembled QDs constitutes the main theme of this thesis.

In Chapter 2 we start by giving an introduction to the physics of self-assembled QDs. We describe the fabrication of samples, where QDs are embedded in a Schottky structure which enable deterministic charging of a QD with single electrons. Furthermore, we discuss the various couplings of electron to the environment. In the last part of chapter 2, we introduce the basic experimental concepts of QD optical spectroscopy: The non-resonant $\mu$-photoluminescence and resonant, high-resolution laser scattering spectroscopy.

Chapters 3 and 4 delve into the coupling of a single QD electron to the $10^4 - 10^5$ nuclear spins of the host material.

In chapter 3 we present a novel effect, where dynamic nuclear spin polarization (DNSP) leads to “rectangular” absorption lineshapes: when a laser is scanned across a resonance at moderate magnetic fields, DNSP ensures locking of the QD transition energy to the incident laser frequency. Remarkably, the DNSP is bidirectional with only one circular laser polarization. We show that this locking leads to a suppression of fluctuations in the transition energy. We present a model, based on an effective non-collinear dipolar hyperfine interaction between the electron and the nuclear spins.

When DNSP builds up in a QD this leads to an effective magnetic field, the Overhauser (OH) field, seen by the electron. In chapter 4, we study the decay of DNSP by monitoring the OH field at 5 T in the absence of laser excitation for different temperatures and exchange couplings to a Fermionic reservoir (FR). We
find that at 200 mK and the absence of co-tunneling the OH field lifetimes are up to $10^5$ s. Indeed, the electron-nuclear spin system provides a near-perfect realization of the central spin problem.

In chapter 5, we study the effect of Kondo correlations on the optical lineshape and demonstrate that the absorption lineshape contains a vast information about the system in various regimes of interaction which can be tuned externally. In contrast to electrical measurements, optical excitation allows one to realize a “quantum quench” where the many-body interactions can be turned on or off at a rate much larger than an intrinsic energy gap which may be present in the system. Unlike the case of slow turn on of interactions where adiabatic continuity is generally obeyed, here the system finds itself in a superposition of the new eigenstates and eventually evolves into one of them. Interestingly, a quantum quench of Kondo correlations manifests itself in an orthogonality catastrophe with drastic consequences for in the absorption lineshape.
2. Self-assembled quantum dots

In this chapter, we discuss the main material system of this thesis work viz., InGaAs self-assembled quantum dots (QDs). Owing to their relatively large band gap and high sample quality, InGaAs QDs lend themselves for optical investigation via spectroscopic means. In addition, the large spin-orbit interaction in such systems allows one to create, control and manipulate spins by light. This fact makes them attractive for applications in spintronics and quantum information science. In order to realize these applications, it is of paramount importance to understand the dynamics of QD spins due to various interactions intrinsic in the solid state. A thorough understanding of such processes will not only help us to control their detrimental effects on the spin dynamics but also allow us to envision new ways of manipulating single electron spins.

We first begin with a discussion of the electronic and material properties of InGaAs QDs used in our experiments followed by a detailed discussion of their optical properties. This is followed by a brief introduction to the various experimental techniques that are used in this work. Finally, we mention the interactions of the QD spin with a few “reservoirs” that are studied in this thesis. This chapter sets up the stage for further discussion of the rich physics emerging from such system-reservoir interactions, pursued in the following chapters.

2.1. Band structure of III-V quantum dots

In order to understand the electronic structure in a QD, we shortly summarize the most important properties of III-IV semiconductors. The band structure arises from the hybridization of the s and p orbitals and the character of the orbitals is retained in the crystal. In III-IV bulk semiconductor the valence band (VB) predominantly consists of bonding p-states and the conduction band (CB) of anti-bonding s-states. The electronic spin interacts with the orbital motion of the electron via spin orbit interaction. Since the s-states have zero orbital momentum, spin orbit interaction is much less important in the conduction band. Therefore the lowest lying discrete energy states in the VB of the QD are spin-1/2 states. The dispersion around the Γ-point is parabolic with an effective mass $m^*_{c,\text{GaAs}} \approx 0.06 m_e$ for GaAs and $m^*_{c,\text{InAs}} \approx 0.023 m_e$ for InAs respectively, where $m_e$ is the free electron mass[14].

Spin-orbit interaction splits the $J = 1/2$ band by around 300 meV from the $J = 3/2$ band in the valence band. The remaining two bands with $J_z = 3/2$ and $J_z = 1/2$ have a different dispersion around the Γ-point, leading to different effective masses of the hole states. The different bands are therefore called heavy holes ($m^*_{hh} \approx 0.41 m_e$ for InAs) and light holes ($m^*_{lh} \approx 0.026 m_e$ for InAs), respectively[14]. In bulk, both bands are degenerate at the Γ-point. In the presence of quantum confinement, the difference of the effective masses lifts this degeneracy and splits the light holes ($J_z = 1/2$) by 20 meV from the heavy holes ($J_z = 3/2$). Effectively,
Figure 2.1.: Schematic illustration of the QD growth process. InAs is deposited on top of a GaAs substrate by molecular beam epitaxy. After a critical thickness of \( \sim 1.7 \) ML strain due to the different lattice constants leads to the formation of QD on the wetting layer. A further overgrowth of the QDs with GaAs completes the growth process.

in QDs the confined hole states are treated by a pseudo-spin 3/2 particle with a small admixture of light-hole character. The confinement potential in lateral direction is sketched in figure 2.2a.

2.2. Growth and fabrication of QDs

Self-assembled InGaAs quantum dots (QDs) are nanometer-sized InGaAs islands embedded in GaAs, which have the ability to confine charge carriers in all directions. Typically the size of the InGaAs islands is around 20-30 nm in lateral direction and 10 nm in growth direction. The band gap alignment in a InAs/GaAs structure forms a type-I heterostructure. This means that the conduction band edge of GaAs lies above the one of InAs and the valence band edge of GaAs lies below the one of InAs. Due to the small size and band alignment, InGaAs QDs provide a three dimensional confinement for both the electron in the conduction band and the holes in the valence band. A schematic of the growth sequence is sketched in figure 2.1.

The QDs used in this thesis work were grown by strain driven molecular beam epitaxy in Stransky-Krastanov growth mode. When InAs is deposited on a [001] GaAs surface, only a thin layer InAs can be grown, the so-called wetting layer. While GaAs has a lattice constant of 5.65 Å, the lattice constant of InAs is 6.8% larger (6.06 Å). This leads to build-up of strain in the InAs layer. After approximately 1.7 monolayers, the strain relaxes leading to the spontaneous formation of small InAs islands on top of the wetting layer: the QDs. They have a diameter of \( \sim 20-30 \) nm and a height of \( \sim 10 \) nm. The growth is then continued by capping the QD’s with GaAs to isolated them from surface states.

For QDs which are grown as described above, the typical transition energies are around 1300 nm (0.95 eV), an inconvenient wavelength range because silicon based photo-detectors lose their sensitivity above 1100 nm. In order to circumvent this issue a more advanced growth technique is used. Instead of completely overgrowing the QDs with GaAs, they are only partially covered, followed by an annealing step. During the annealing the top part diffuses out of the islands into the GaAs, thereby reducing the height of the QDs to \( \approx 5 \) nm. Subsequently, the growth of GaAs is continued. In this way, the confinement in the growth direction becomes stronger,
2.3. Charge tunable quantum dots

One of the major advantages of QDs is the ability to exactly control their charge state. By doing this, one not only suppresses the random charge fluctuations which might occur in the sample but this also opens up ways to study and even control the interaction of single charge or spin with various reservoirs by having a specific number of carriers in the QD. In order to deterministically charge a QD on a single electron level, they are embedded in a Schottky structure, sandwiched between two metallic plates. One plate consists of a 40 nm thick n++ GaAs layer below the QD layer and is referred to as the Fermionic reservoir (FR) or electron gas. In all our experiments, we assume a two-dimensional character of the FR. The QDs are separated by a layer of intrinsic GaAs, which acts as a tunnel barrier from the FR. The thickness of the tunnel barrier depends on the particular experiment and varies between 15 and 35 nm within this work. The other metallic plate is a thin, semitransparent film of either 5 nm NiCr or 8 nm TiAu processed on the sample surface.

The structure between the QD layer and the sample surface contains an AlGaAs super-lattice which prevents current through the sample and also the formation of a hole gas in the proximity of the QD [15]. This super-lattice is usually separated by 10 nm the QDs. A thin layer of GaAs (10-40 nm) is grown on top of the super-lattice to prevent oxidation of the Al on the surface. As will be discussed in section 2.5.2, the thickness of the sample between the QD’s and the sample surface is of great importance for resonant scattering experiments where the actual absorption lineshapes could be distorted due to an optical interference effect. A schematic of the sample structure is depicted in figure 2.2b.

Figure 2.2.: a) Sketch of the QD confinement potential in the growth plane. The bandgap of the surrounding GaAs is $\approx 1.519$ eV. The confinement is $\approx 300$ meV for electrons and $\approx 150$ meV for holes. b) Schematic of a typical sample structure. By applying a voltage $V_g$ to the top-gate the QD levels can be tuned with respect to the Fermi energy $E_F$. which shifts the transition energy from 1.3 $\mu$m to 950 nm. The method is referred to as partly covered island (PCI) technique.
Figure 2.3.: Photoluminescence (PL) versus gate voltage ($V_g$) from a single QD at 4.2 K at zero external magnetic field. The PL changes abruptly at particular gate voltages where the charging state of the QD is changed.

The Schottky voltage on the order of 1 V drops linearly between the sample surface and the electron gas, which is contacted by a GeAu alloy. A gate voltage $V_g$, superimposed on the Schottky voltage changes the alignment between the electronic states in the QD and the Fermi energy. Suppose that the lowest conduction band state lies above the Fermi energy $E_F$ for a particular gate voltage $V_g$. When $V_g$ is raised, at some point the QD level will cross $E_F$ and an electron tunnels into the quantum dot. The large Coulomb repulsion $U_{ee}$, which is 7-25 meV, depending on the QD size, prevents a second electron from tunneling into the QD. In this way the QD can be deterministically charged with single electron from the back contact. This process is schematically depicted in figure 2.2b.

2.4. Optical properties of QDs

Optical excitation leads to the formation of localized excitons in QDs. For an uncharged exciton formed from an electron in the conduction band and the heavy-hole in valence band, there are four different possible combinations coming from two spins states for each band. Given that the electron spins are either +1/2 or -1/2 and the hole spins are +3/2 and -3/2, neutral excitons ideally have a total angular momentum $L$ of ±2 or ±1. Only excitons with total angular momentum of ±1 are optically active due to optical selection rules. The excitons with $L = ±2$ are called dark excitons as they are one-photon forbidden. For the neutral exciton, anisotropic exchange interaction causes a mixing of ±1 excitons [16]. In the case of a single electron charged quantum dot, optical excitation leads to the formation of so-called trion states: two electrons in a singlet state and a hole with spin $±3/2$. During a resonant excitation process, the angular momentum of the photon is transferred to the exciton. For example if the QD is charged with a ↑-electron, absorption of a right-hand circularly polarized photon ($\sigma^+$) excites a trion with spin $+3/2$. Due to conservation of angular momentum a $\sigma^-$ polarized photon cannot be absorbed (Pauli-blockade) if the QD is charged with a $|\uparrow\rangle$ electron.
A powerful tool to optically characterize a QD sample is $\mu$-photoluminescence spectroscopy. For this purpose, a QD sample is placed inside a fiber-based confocal microscope at cryogenic temperatures [17, 18](see section 2.5). A laser with an energy above the band-gap of GaAs creates free carriers in the bulk. Typically we used an excitation wavelength of 780 nm. Within time-scales of a few ten picoseconds, the free carriers relax via phonon emission to the band-edges. Some of the carriers are then trapped by the QDs and form excitons. After 2 ns corresponding to the lifetime of excitons [19], they recombine by emission of a photon. Spectral analysis, such as emission energy and polarization of the emitted photons, provide us with information about the recombination process. For our sample, the characterization is performed by monitoring the emission spectrum for low excitation powers (10-500 nW over 1 $\mu$m$^2$ spot) as a function of applied gate voltage $V_g$. In such a PL-gateweep one can observe distinct plateaus which can be attributed to different excitonic transitions. The transitions are characterized by the charge and spin configuration in the ground and excited state. For example the $X^{-1}$ transition has a single electron charged ground state and a excited trion state. A detailed description of excitonic lines can be found in [17, 18, 20].

Figure 2.3 shows gatesweeps of two different QD sample. The QDs in figure 2.3a are separated by a nominally 25 nm thick tunnel barrier from the FR. Due to the very small tunnel coupling, the different plateaus ($X^{+1}, X^0, X^{-1}$) have a substantial overlap in gate voltage. However, all lines exhibit a dependence of the transition energy on the applied gate voltage. This is due to the quantum confined Stark effect, which is given by [21]:

$$E = E_0 - pF + \beta F^2,$$

(2.1)

where $p$ is the permanent dipole moment of the exciton and $\beta$ its polarizability. For our purpose the shift can be considered to be linear with a magnitude of $-2\mu$eV per applied mV in gate voltage. The linear Stark shift plays an important role in resonant absorption experiments (see section 2.5.1), where it is used to bring a particular transition in and out of resonance with an incident laser field.

The gatesweep of the QD in figure 2.3b was taken on a sample with a nominally 15 nm tunnel barrier. The comparison with figure 2.3a makes clear how the PL can qualitatively and quantitatively differ, depending on the particular sample structure. For example, the emission spectrum exhibits a strong broadening at the edges of the plateaux arising from the interactions of a quantum dot electron with FR. The detailed form of the spectrum is discussed in chapter 5.

### 2.5. Experimental Setup

All experiments in this work, except the ones in chapter 3 were carried out in a dilution refrigerator. The experiments in chapter 3 were carried out in a helium bath cryostat. Such a setup is slightly different to operate than the one described in this section. A detailed description of the bath cryostat setup is given in the references [17, 18, 22].

In order to perform high resolution laser scattering spectroscopy at milliKelvin temperatures, a fiber-based confocal microscope was attached to the cold-finger of a dilution refrigerator with a base temperature of 10 mK in the mixing chamber,
measured by a calibrated resistor (give type). A picture and a detailed schematic of the microscopy is shown in figure 2.4. An optical single mode fiber is guided all the way to the end of the cold-finger, where it is coupled to an objective, which is mounted on a stack of low-temperature xyz-piezoelectric-positioners. The fiber core with a diameter of 5µm acts as a pinhole of the confocal system. The numerical aperture (NA) of the objective lenses are chosen such that one matches the NA of the single mode fiber. The NA of the focussing lens was 0.68, desirably high to provide a large collection efficiency and high resolution [17, 23]. The sample was mounted with silver paint on a gold plated sample holder below the objective, thermally connected to the cold-finger by the microscope housing. During the measurements, the transmission(section 2.5.1) through the sample was measured with a p-i-n-photon-diode, mounted on the bottom side of the sample holder, just below the sample.

All parts in the microscope were covered with shields to protect them against 4 K radiation from the liquid helium bath within the inner vacuum chamber of the insert. Furthermore all parts of the microscope housing were made of gold-plated copper for maximum thermal conductivity without oxidation.

Some of the experiments required large magnetic fields, which were generated by a 8.5 T superconducting magnet.

2.5.1. Resonant Rayleigh Scattering

Resonant Rayleigh scattering is a powerful tool to resonantly probe the QD dynamics with high resolution. The principle is simple: A narrow band laser is tuned across
an excitonic transition of interest while the transmitted intensity though the sample is recorded with a Si-based photo-diode \(^1\). It should be noted that this is not really an absorption experiment, but rather an interference experiment, where the laser interferes with the scattered photons of QD. We consider the following scenario (figure 2.6a.): The QD is placed in the focal spot of a laser whose field at a position \(x\) is denoted by \(E_L(x)\). The response of the QD to the incident laser field is

\[
E_{\text{QD}} = \chi(\delta)E_L(0),
\]

where \(\chi(\delta) = \chi'(\delta) + i\chi''(\delta)\) is the susceptibility of the QD with a dispersive part \(\chi'(\delta)\) and an absorptive part \(\chi''(\delta)\).

The measured intensity at the position \(x_{\text{det}}\) of the detector below the sample is then proportional to the time averaged square modulus of the superposition of the laser and the QD field:

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

(2.3)

Since the laser intensity is kept constant during a measurement, the first term in equation 2.3 just contributes a constant offset to the measured signal. Furthermore, the laser field is orders of magnitude stronger than the QD field. Therefore the last term in 2.3, the QD resonance fluorescence, is negligible, such that the signal which we detect is \(\propto \text{Re}\langle E_L(x_{\text{det}})E_{\text{QD}}(x_{\text{det}}) \rangle\).

For a simple two-level system, the absorptive and dispersive part of the QD susceptibility are given by \(\chi'(\delta) \propto \frac{\gamma\delta}{\delta^2 + \Gamma^2}\) and \(\chi''(\delta) \propto \frac{\gamma^2}{\delta^2 + \Gamma^2}\), respectively, where \(\gamma = \Gamma/2\) with the linewidth \(\Gamma\). The QD field, generated in the focal spot of the laser is then

\[
E_{\text{QD}} = E_L(0)(\chi' + i\chi'').
\]

(2.4)

In the most simple case with no reflecting surfaces in the proximity of the QD, the detected intensity at the detector is

\[
I_{\text{det}} = \text{Re}\langle |E_L(0)\chi + E_L(0)e^{i\frac{\pi}{2}}|^2 \rangle.
\]

(2.5)

The second term is the laser field at the detector position, which features a Gouy phase (an additional phase shift occurring in the propagation of focused Gaussian beams [24]) of \(\pi/2\) relative to the QD field as it propagates from the QD position to the detector position. The measured quantity in the experiments is the transmission contrast \(\Delta T/T\), defined as the ratio of the change in transmitted intensity \(\Delta T\) and the transmission \(T\), corresponding to the laser intensity. Using the above expressions for the optical susceptibility for a two-level system on can show that the differential transmission as a function of laser detuning \(\delta\) is given by

\[
\frac{\Delta T}{T} = \frac{\sigma_0 \gamma^2}{A \delta^2 + \gamma^2}
\]

(2.6)

where \(A\) is the spot size of the laser and \(\sigma_0 = \frac{3\lambda^2}{2\pi}\) is the scattering cross-section. A detailed derivation of this equation based on the optical theorem is given in references [17, 25]. Figure 2.5a shows a typical differential transmission signal of a \(X^-\)

\(^1\)An alternative to achieving a detuning between the incident laser and the transition is to shift the transition via the Stark shift by sweeping the gate voltage.
signal of a sample with a 25 nm tunnel barrier. For many experiments it is important that the laser has only a perturbative influence on the dynamics. Figure 2.5b and c show a laser saturation [26] experiment where the peak contrast and the linewidth are plotted versus the laser intensity. The laser intensity is then chosen such that it does not broaden the resonance or lead to a drastic decrease in the peak contrast.

2.5.2. Influence of optical interference on measured lineshapes

So far we have neglected the presence of the sample surface. As mentioned above, interference of scattered QD photons within the sample structure lead to a distortion of the measured spectral lineshapes. Some of the resonantly scattered photons towards the sample surface are reflected and interfere with the forward scattered photons [25]. The experimental situation is depicted in Fig. 2.6a. The light field at the detector has now three contributions: one is the laser field, the second one is field of photons scattered from the QD into the forward direction. The third contribution is the field of photons scattered from the QD into the backward direction, which is then reflected from the sample surface (combination of top gate and dielectric interface) and thereby redirected into the forward direction. This field acquires an additional factor \(re^{i\phi}\), with the surface reflectivity \(r\), and a phase \(\phi = 2\pi n/\lambda L\), where \(L\) is the distance from the QDs to the sample surface. \(n\) is the refractive index of the GaAs and \(\lambda\) the wavelength of the photons. Putting everything together, one obtains the following expression for the total field at the position of the detector:

\[
E_{\text{tot}} = iE_L(1 - i\chi(\delta)(1 + re^{i\phi})).
\]  

Hence, the differential transmission signal \(\Delta T/T\) is given by

\[
\frac{\Delta T}{T} \propto \text{Re}(i\chi(\delta)(1 + re^{i\phi})).
\]

The reflection at the sample surface can therefore cause a mixing of the absorptive and the dispersive part of \(\chi(\delta)\) if the distance \(L\) is chosen such that the two QD field do not interfere constructively or destructively.
2.5. Experimental Setup

Figure 2.6.: A) Surface effect in differential transmission experiments. The QD field at the detector position has two contributions: a forward scattered field \( E_{\text{QD,f}} \) and a backward scattered field, which is reflected at the sample surface \( E_{\text{QD,b}} \). B) The interference effect leads to a mixing of the absorptive and dispersive part of the QD response.

In the case of the 15 nm tunnel barrier structure used in chapter 5 \( L = 90 \) nm, \( \lambda = 904 \) nm and \( r = 0.67 \) and \( n = 3.5 \), so that \( \phi = 1.4\pi \). If the absorptive part \(-\chi''(\nu)\) was purely Lorentzian, the interference effect would modify the lineshape in a way depicted in Fig. 2.6b: the mixing of the absorptive and dispersive part of \( \chi(\nu) \) leads to a shift of the absorption peak to blue laser detunings and causes the signal to become negative for small red detunings. Moreover, tails that decay faster than \( 1/\nu \) are changed by the interference effect to decay as \( 1/\nu \), as discussed later.

If the sample structure was exactly known, the interference-free lineshape could be simply obtained by the following procedure: The real and imaginary part of \( \chi(\omega) \) are linked by the Kramers-Kronig relation:

\[
\chi'(\omega) = -\frac{1}{\pi} P \left( \int d\omega' \frac{\chi''(\omega')}{\omega' - \omega} \right) \equiv \hat{K}(\chi''(\omega)) \quad (2.9)
\]
\[
\chi''(\omega) = \frac{1}{\pi} P \left( \int d\omega' \frac{\chi'(\omega')}{\omega' - \omega} \right) \quad (2.10)
\]

where \( P(.) \) denotes the principle value part of the integral. Since the real and imaginary part of \( \chi(\omega) \) are linked, the detected signal \( S(\omega) \) is only a function of the absorptive part \( \chi''(\omega) \):

\[
S(\omega) = \text{Re} \left( -i (\chi'(\omega) + i\chi''(\omega)) \right) (1 + re^{i\phi}) \\
= \text{Re} \left( -i \left( \hat{K}(\chi''(\omega)) + i\chi''(\omega) \right) \right) (1 + re^{i\phi}) \\
= f(\chi''(\omega)) \quad (2.11)
\]

We define a new function \( F(\chi''(\omega)) \) and search for its zeros:

\[
F(\chi''(\omega)) = f(\chi'(\omega)) - S(\omega) = 0. \quad (2.12)
\]
This equation can in principle easily be solved numerically. However a practical implementation as required for example in chapter 5 is difficult due to the low signal to noise in the tails of the absorption lineshape.

2.6. Coupling of a single QD electron to different environments

Although QDs are often referred to as “artificial atoms” they can not be completely isolated from the solid-state environment in which they are embedded in. Therefore they have properties which are fundamentally different from that of atoms. In the following, we will discuss the electronic system and its coupling to the nuclear spins of the host material and the Fermi reservoir.

2.6.1. The electronic system

All phenomena discussed in the following chapters originate from many-body interactions of a single confined QD spin with its environment. Let us first discuss the properties of electron in the QD before moving on to the nuclear spin bath to which it is coupled.

An external magnetic $B_{\text{ext}}$ field along the growth direction (Faraday geometry) leads to Zeeman splitting of the electron:

$$\hat{H}_Z^e = g_e \mu_B \hat{s} B_{\text{ext}},$$

(2.13)

where $\hat{s}$ is the electron spin operator. The electron g-factor $g_e$ varies between 0.4 and 0.6.

The heavy holes with spin $\pm 3/2$ are treated as pseudo-spins with $\pm 1/2$. Its g-factor $g_h$ is positive and varies between +1 an +2 [27, 28].

2.6.2. Nuclear spins

The exact composition of the QD is unknown but it consists of InAs with an admixture of Ga due to the diffusion of Ga into the QD during growth. Therefore, we deal with the three different nuclear species with their naturally occurring isotopes [29]: $^{115}\text{In}$ (95.3%), $^{113}\text{In}$ (4.7%), $^{75}\text{As}$, $^{69}\text{Ga}$ (60.1%), $^{71}\text{Ga}$ (39.9%). Indium has a spin of 9/2, while all other species have spin 3/2. Each nuclear spin is characterized by its spin $\hat{I}_i$ and its gyromagnetic ratio. As the electron spin, an external magnetic field leads to a Zeeman splitting:

$$\hat{H}_{N,Z} = \sum_i \gamma_i \hat{I}_i \cdot \vec{B}_{\text{ext}},$$

(2.14)

where $\hat{I}_i$ is the $i$-th nuclear spin-operator. The gyromagnetic ratios for are $\gamma_{115}\text{In} = 3.910^{-2} \mu\text{eV/T}$ and $\gamma_{75}\text{As} = 310^{-2} \mu\text{eV/T}$ [29].

At low magnetic fields, which are not relevant for the experiments in this work, nuclear dipole-dipole interaction cause nuclear spin diffusion and nuclear spin decay. The strength of the interaction is characterized by the local magnetic field produced
2.6. Coupling of a single QD electron to different environments

by a nuclear spin an seen by another one. Typical coupling strengths are on the order of $10^{-1}$ mT in GaAs. External magnetic fields exceeding the coupling strength render the decay induced by the non-secular terms on the dipolar interaction ineffective. From experiments in chapter 4 we find that also dipolar spin-flip events between nuclear spins are suppressed. We attribute this feature which is unique in strained QDs to strong inhomogeneous quadrupolar fields which can exceed 200 mT (see section 2.6.5).

2.6.3. Fermi-contact hyperfine interaction

The dominant interaction between the electron spin and the nuclear spins is the Fermi-contact hyperfine interaction \[8, 30–32\], which is given by the following Hamiltonian:

$$\hat{H}_{hf} = \sum_i A_i |\psi(\vec{R}_i)|^2 \hat{s} \cdot \hat{I}_i$$ \hspace{1cm} (2.15)

where $\psi(\vec{R}_i)$ is the magnitude of the electron envelope at the position of the the $i$-th nucleus. The hyperfine coupling constant is $A_i = \frac{2}{3} \mu_0 \mu_B \hbar \gamma_i |u(\vec{R}_i)|^2$, where $g_0$ is the free electron g-factor. Importantly, the coupling constant depends on the the magnitude of the Bloch wavefunction at the position of the nucleus. This means that the coupling depends on the parity of the wavefunction. As mentioned before, QD electrons have an s-type symmetry and therefore couple to nuclear spins via 2.15. Due to the p-type symmetry of the QD holes, the Fermi contact hyperfine interaction vanishes. The coupling constants $A_i$ are on the order of $10^{-2}$ μeV.

The Hamiltonian 2.15 can be rewritten as $\hat{H}_{hf} = \sum_i A_i (\hat{s}_z \hat{I}_z + \frac{1}{2} (\hat{I}_+ \hat{s}_- + \hat{s}_+ \hat{I}_-))$, an expression which contains a static part $\propto \hat{s}_z \hat{I}_z$ and a dynamic part $\propto \hat{I}_+ \hat{s}_- + \hat{s}_+ \hat{I}_-$. The static part constitutes and effective magnetic field seen by the electron spin: the Overhauser(OH) field:

$$B_{OH} = \sum_i A_i \langle \hat{I}_z \rangle$$ \hspace{1cm} (2.16)

The OH field will be subject of chapters 3 and 4. For a fully polarized nuclear spin bath, the OH is $\approx 170 \mu eV$ which corresponds to a magnetic field of 5 T. Conversely, each nuclear spin sees an effective spatially varying Knight-field, produced by the electron spin \[33\].

The dynamic part $\propto (\hat{I}_+ \hat{s}_- + \hat{s}_+ \hat{I}_-)$ enables transfer of the the electron spins to a nuclear spins \[33\] via a flip-flop process.

2.6.4. Anisotropic dipole-dipole hyperfine interaction

In addition to Fermi-contact hyperfine interaction the electron couples to nuclear spins via anisotropic dipolar hyperfine interaction \[34\]:

$$\hat{H}_{d-d} = -\frac{\mu_0}{4\pi \gamma_N \gamma_e} \sum_i r_i^2 \hat{s} - 3r_i (\hat{s} r_i) \hat{I}_i.$$ \hspace{1cm} (2.17)

The latter expression has to be averaged over the electron wavefunction. It can be shown, that in the case of an s-type wavefunction, electron-nuclear dipole interaction is much smaller than Fermi contact hyperfine interaction. However this interaction
could still have a small, but finite coupling strength. In fact, the experimental results in chapter 4 suggest, that the non-collinear dipolar coupling could be present.

2.6.5. Quadrupole interaction

The strain due to the different lattice constants of InAs and GaAs leads to strong electric field gradients in self-assembled QDs. When considering a nucleus, it is important to take into account its finite dimension. In fact, the asymmetric part of nuclear charge distribution couples to an electric field gradient present at the nucleus. Importantly the spin and charge distribution are related. It can be shown that nuclei with \( I > \frac{1}{2} \) have a non-spherical charge distribution and therefore a quadrupole moment.

A derivation of the quadrupole interaction can be found in \([35]\). The interaction is given by

\[
E^{(2)} = \frac{1}{6} \sum_{\alpha\beta} V_{\alpha\beta} Q_{\alpha\beta} \tag{2.18}
\]

where

\[
Q_{\alpha\beta} = \frac{1}{3} \int \rho(\vec{r})(3x_\alpha x_\beta - \delta_{\alpha\beta} \vec{r}^2) \tag{2.19}
\]

is the nuclear quadrupole moment. So far this is a pure electrostatic interaction. The determination of the values of this quantity in a particular state of a system requires an averaging of the operator 2.18 over the corresponding wavefunction. For a nuclear spin in a state \(|I, m\rangle\) the operator can be expressed as:

\[
H_{\text{quadr}} = \frac{eQV_{zz}}{4I(2I-1)} \left(3(I_{\text{principal}}^2) - \hat{I}(\hat{I} + 1)\right) \tag{2.20}
\]

where

\[
I_{\text{principal}} = \hat{I}_z \cos(\theta) + \hat{I}_y \sin(\theta) \tag{2.21}
\]

is the principle axis which is rotated with respect to an external magnetic field by an angle \( \theta \).

The fact, that the nuclear spins are aligned with the quadrupole moment leads to an interaction of the nuclear spin with the electric field gradients in the crystal lattice. The gradients provide an effective in-plane magnetic field for the nuclear spins which couples different spin states. For a spin 3/2 nucleus and a small angle \( \theta \), the quadrupolar Hamiltonian has the following matrix representation:

\[
\begin{pmatrix}
E_Q & \sqrt{3}E_Q\theta & \frac{\sqrt{3}}{2}E_Q\theta^2 & 0 \\
\sqrt{3}E_Q\theta & -E_Q & 0 & \frac{\sqrt{3}}{2}E_Q\theta^2 \\
\frac{\sqrt{3}}{2}E_Q\theta^2 & 0 & -E_Q & -\sqrt{3}E_Q\theta \\
0 & \frac{\sqrt{3}}{2}E_Q\theta^2 & -\sqrt{3}E_Q\theta & E_Q
\end{pmatrix} \tag{2.22}
\]

2.6.6. Exchange interaction with a Fermionic reservoir

The QD is tunnel-coupled to the back contact\([36]\), which is a Fermionic reservoir. Tunnel coupling is well described by the Anderson Hamiltonian which is discussed in detail in chapter 5.
2.6. Coupling of a single QD electron to different environments

In the case of a single electron charged QD in the Coulomb blockade regime first order tunneling is suppressed. When no charge exchange with the FR takes place, the Anderson Hamiltonian can be projected into the \( n_e = 1 \) subspace by a unitary transformation\[37\] yielding the so-called s-d or Kondo Hamiltonian:

\[
\hat{H}_{s-d} = \sum_{\vec{k}, \vec{k}' } J_{\vec{k}\vec{k}'} \left( \hat{c}^\dagger_{\vec{k}, \uparrow} \hat{c}_{\vec{k}', \downarrow} \hat{e}^\dagger_{\downarrow} + \hat{c}^\dagger_{\vec{k}', \downarrow} \hat{c}_{\vec{k}, \uparrow} \hat{e}^\dagger_{\uparrow} \right) + \frac{1}{2} \left( \hat{c}^\dagger_{\vec{k}, \uparrow} \hat{c}_{\vec{k}, \uparrow} - \hat{c}^\dagger_{\vec{k}', \downarrow} \hat{c}_{\vec{k}', \downarrow} \right) \left( \hat{e}^\dagger_{\uparrow} \hat{e}^\dagger_{\downarrow} \right).
\]  

(2.23)

where the coupling constant is given by:

\[
J_{\vec{k}\vec{k}'} = V_{\vec{k}}^2 \left( \frac{1}{-\varepsilon_d + i\gamma/2 + \varepsilon_k} + \frac{1}{-\varepsilon_d + i\gamma/2 + \varepsilon_{k'}} \right) + V_{\vec{k}}^2 \left( \frac{1}{\varepsilon_d + U_{ee} + i\gamma/2 + \varepsilon_k} + \frac{1}{\varepsilon_d + U_{ee} + i\gamma/2 + \varepsilon_{k'}} \right).
\]  

(2.24)

(2.25)

The Hamiltonian 2.23 describes an effective spin exchange of the QD electron with the FR. The corresponding co-tunneling rate \( \kappa \) can be calculated in Born approximation by standard perturbative system reservoir theory:

\[
\kappa(t - t') = \int_{-D}^{D} d\omega_k D(\omega_k) \int_{-D}^{D} d\omega_{k'} |J_{\vec{k}\vec{k}'}|^2 f(\omega_k)(1 - f(\omega_{k'})) e^{-i(\omega_k - \omega_{k'})(t - t')}.
\]  

(2.26)

In the most simple case possible, the reservoir is 'memoryless', or Markovian i.e., the system-reservoir coupling does not depend on the the system dynamics in the past. The (time-independent) co-tunneling rate is then \( \kappa = \kappa(t - t')\delta(t - t') \). A closed analytical expression cannot be obtained for \( \kappa \) in general. However it is possible for the following parameters: \( \varepsilon_d = -\frac{U_{ee}}{2} \) and \( V_{\vec{k}} = V \). We then have

\[
\kappa = \left( \frac{8V^2}{U_{ee}} \right)^2 D^2(\varepsilon_F) \int_{-D}^{D} d\omega f(\omega)(1 - f(\omega)) \]  

(2.27)

\[
= \left( \frac{8V^2}{U_{ee}} \right)^2 D^2(\varepsilon_F) k_B T \tanh \left( \frac{D}{2k_B T} \right) \to_{D \to \infty} \left( \frac{8V^2}{U_{ee}} \right)^2 D^2(\varepsilon_F) k_B T. \]  

(2.28)

Here, one can see that the co-tunneling rate increases linearly with the temperature. However, if \( k_B T \) approaches 0, \( \kappa \) vanishes in expression 2.28. The reason is that co-tunneling is highly non-Markovian at low temperatures. Therefore, the Markov approximation breaks down at temperatures below an energy scale \( T_K \), the so-called Kondo temperature, as will be treated in chapter 5 in detail. The typical co-tunneling rates in QD samples depend strongly on the sample design and experimental parameters, and can be tuned over several orders of magnitude ranging from \( 10^5 \text{s}^{-1} \) in a sample with a 35 nm tunnel barrier (see chapter 4) to \( 10^{11} \text{s}^{-1} \) in a 15 tunnel barrier sample(see chapter 5).
3. Bi-directional dragging of quantum dot optical resonances by dynamic nuclear spin polarization

Experiments using transport [7] as well as optical spectroscopy [38] revealed that the nature of hyperfine interactions in QDs is qualitatively different than that of atoms: coupling of a single electron spin to the mesoscopic ensemble of $\sim 10^5$ QD nuclear spins results in non-Markovian electron spin decoherence [31] and presents a major drawback for applications in quantum information science. Nevertheless, it is still customary to refer to QDs as artificial atoms; i.e. two level emitters with an unconventional dephasing mechanism. In this chapter, we present resonant absorption experiments demonstrating that for a wide range of system parameters, such as the gate voltage, the thickness of the tunnel barrier that separates the QDs from the back contact and the external magnetic field, nuclear spins strongly modify the signatures of elementary optical excitations. We determine that the striking locking effect of any QD transition to an incident near-resonant laser, which we refer to as dragging, is associated with dynamic nuclear spin polarization (DNSP); in stark contrast to previous experiments [6, 33, 39–43] the relevant nuclear spin polarization is bi-directional and its orientation is determined simply by the sign of the excitation laser detuning. We find that fluctuations in the QD transition energy, either naturally occurring [44] or introduced by externally modulating the Stark field, are suppressed when the laser and the QD resonances are locked. We also find that when the exchange interaction between the confined QD electron and the nearby electron Fermi-sea that leads to incoherent spin-flip co-tunneling [36] is sufficiently strong, it can suppress the confluence of laser and QD transition energies by inducing fast nuclear spin depolarization [45]. We first give a detailed qualitative overview of the observed effect. Then we will consider a model, which describes our observations. In the end, we discuss the parameter dependence and compare with the theory.

3.1. Locking of quantum dot resonances to an incident laser

For a single-electron charged QD, the elementary optical excitations lead to the formation of trion states (X$^-$) which are tagged by the angular momentum projection (pseudo-spin) of the optically generated heavy-hole (figure 3.1A). In the absence of an external magnetic field ($B_{\text{ext}} = 0$ T), the absorption lineshape associated
Figure 3.1: (A) Energy-level diagram of a single-electron charged quantum dot: external magnetic field enables spin-selective excitation of Zeeman split trion ($X^-$) states. Right-hand circularly polarized laser with Rabi frequency $\Omega_L$ (blue arrow) and detuning $\Delta \omega = \omega_X - \omega_L$ couples the higher energy Zeeman branch between the spin-up electron ground state $|\uparrow\rangle$ and the trion state $|\uparrow \downarrow \uparrow\rangle$. The relevant decoherence processes (radiative decay $\Gamma$, co-tunneling $\kappa$ and heavy-hole/light-hole mixing $\gamma_{hh-lh}$) are indicated by grey arrows. (B), (C) Trion absorption spectra at zero magnetic field (Lorentzian fit with a linewidth of 2 $\mu eV$) and at 4.5 T, respectively. For magnetic fields exceeding 1 T, the on-resonance scattering is maintained over many natural linewidths to both sides of the bare resonance depending. Red (blue) spectra show data obtained by tuning the laser from an initial blue (red) to a final red (blue) detuning from the quantum dot resonance. The green trace shows the energy range over which resonant absorption is recovered in fixed laser detuning experiments (see figure 3.3C). (D) The dragging effect is even more prominent in the absorption spectra of the blue Zeeman branch of the neutral exciton $X^0$. In all experiments, the temperature was 4.2 K.
3.1. Locking of quantum dot resonances to an incident laser

Figure 3.2.: Dependence of dragging on system parameters. Two dimensional absorption maps of the blue trion Zeeman branch as a function of gate voltage and laser energy. Red (blue) arrows indicate the scan direction with decreasing (increasing) laser detuning $\Delta \omega = \omega_N - \omega_L$. (a) Absorption map recorded at 4.5 T for a quantum dot in sample A with a 25 nm tunneling barrier: the data was obtained by keeping the laser energy fixed and scanning the gate voltage. The inset illustrates the exchange coupling to the Fermi reservoir that flips the quantum dot electron spin via co-tunneling events. The co-tunneling rate is minimum in the center and maximum at the edges of the stability plateau; the dragging width scales inversely with the co-tunneling rate $\kappa$. (b) Absorption maps of a quantum dot with a 35 nm tunneling barrier that were obtained by scanning the laser at a fixed gate voltage in external magnetic fields of 5 T. Finite absorption contrast is restricted to the edge of the single-electron charging plateau due to electron spin pumping. The set of data is complementary to (a) and shows that dragging is independent of whether the laser or the gate voltage is scanned.

close to its peak value for laser frequencies that are blue detuned from the bare trion resonance by as much as $7\Delta \nu$. The absorption scans of figure 3.1C show that the trion resonance locks on to the laser frequency and can be dragged to either higher or lower energies by tuning the laser frequency, provided that the scan frequency-step size is small. We observe this dragging effect for a wide range of laser Rabi frequencies $\Omega_L$ ranging from $\sim 0.3 \Gamma$ to $3 \Gamma$. We also emphasize that dragging is not a simple line-broadening effect: the area of the absorption curve is an order of magnitude larger than its $B_{\text{ext}} = 0$ T counterpart.

It is well known that the optical response of a neutral QD for $B_{\text{ext}} < 1$ T is qualitatively different, owing to the role played by electron-hole exchange interaction [28]. To assess the generality of the dragging phenomenon, we investigated the response of a neutral QD for $B_{\text{ext}} \geq 2$ T: despite an energy level diagram that is substantially different than that of a single-electron charged QD, the bright exciton transitions of a neutral QD exhibit absorption lineshapes (figure 3.1D) that are qualitatively similar to that of a trion. In fact, we observe that typical dragging widths for neutral QD exciton transitions are significantly larger than that of trion
Further insight into the locking phenomenon depicted in figures 3.1C-D can be gained by studying its dependence on basic system parameters. Figure 3.2 shows the two-dimensional (2D) map of resonant absorption as a function of laser frequency and gate voltage $V_g$ for two different sample structures exhibiting radically different ranges of the co-tunneling rate. Figure 3.2A shows the 2D absorption map of a QD that is separated from the Fermi sea by a 25 nm GaAs barrier (sample A): each horizontal cut is obtained by scanning the gate voltage for a fixed laser frequency. Red (blue) bars show data obtained by scanning the gate voltage such that the detuning $\Delta \omega = \omega_X - \omega_L$ decreases (increases). We estimate the co-tunneling rate $\kappa$ for this sample at the center of the absorption plateau ($V_g = 200$ mV) to be $1 \times 10^6$ s$^{-1}$ from electron spin pumping experiments carried out at $B_{ext} = 0$ T [46]. We observe that the bi-directional dragging effect is strongest in the plateau center and is completely suppressed at the edges ($V_g \sim 160$ mV and $V_g \sim 260$ mV).

Figure 3.2B shows absorption maps for a QD that is separated from the Fermi sea by a 35 nm tunnel barrier (sample B) for $B_{ext} = 5$ T: each vertical cut is obtained by scanning the laser energy for a fixed gate voltage. The finite contrast in figure 3.2B is obtained for a narrow range of $V_g$ near the edges of the charging plateau: the large tunnel barrier drastically suppresses the tunnel coupling, such that the highest $\kappa$ (obtained at the plateau edges) coincides with the lowest rate obtained for sample A. Consequently, bi-directional dragging in sample B extends all the way out to the edge of the charging plateau, while in the plateau center absorption disappears completely due to electron spin pumping into the $|\downarrow_z\rangle$ state [5]. The overall range for dragging is $\sim 30$ $\mu$eV for $B_{ext} = 5$ T. Experiments carried out on QDs in all samples showed that the dragging width increases sub-linearly with $B_{ext}$ beginning at $\sim 1$ T. Further experiments carried out on sample C (not shown) with QDs separated by a 15 nm tunnel barrier from an electron reservoir and exhibiting $\kappa > \Gamma$ throughout the plateau, did not show dragging effects. These results show that locking of QD resonance to the incident laser energy is possible provided that the co-tunneling rate of the QD electron satisfies $\kappa < 10^8$ s$^{-1}$. We observed that the neutral exciton transition of sample B QD exhibits dragging even at the center of the plateau where we expect $\kappa \leq 1$ s$^{-1}$ (not shown). This observation suggests that dragging of the QD resonances takes place even for QDs that are completely isolated from a back contact. Finally, we note that bi-directional dragging is also observed when the red-trion or the red-Zeeman neutral exciton transitions are driven by a resonant laser field; in contrast to the blue-trion (figure 3.1C) and high-energy neutral exciton (figure 3.1D) transitions, the forward and backward scans in this case are highly asymmetric.

3.2. Build-up and decay of nuclear spin polarization

The disappearance of dragging with increasing co-tunneling rate or vanishing external magnetic field strongly suggest that locking of the QD optical transition to the laser frequency is associated with DNSP. Recent studies carried out with laser fields resonant with the excited state transitions of QDs have shown that the nuclear spin depolarization rate has a strong dependence on the electron spin co-tunneling
3.2. Build-up and decay of nuclear spin polarization

Figure 3.3.: Absorption signal (light blue/red: no absorption; dark blue/red: large absorption) as a function of laser detuning and time. (A) Decay: The nuclear spins are polarized by dragging the resonance by 10 $\mu$eV $\simeq 5\Delta\nu$ (arrow 1). The detuning condition is instantly changed to $| \omega_X - \omega_L | \leq 8 \mu$eV (arrow 2). The recovery of the absorption signal after a time $\Delta t$ indicates that the nuclear spin polarization has decayed. (B) Time-trace of the resonance signal along the dashed line in A (C) Buildup: After completely depolarizing nuclear spins, the laser is set to a finite detuning (arrow). The recovery of the absorption indicates a build-up of nuclear spin polarization. (D) Simulation of the quantum dot level population in steady state for the experiment described in C (E) Probability distribution $p(I_z)$ for obtaining a value $I_z$ of the Overhauser field for a detuning range ($-10\Delta\nu, 10\Delta\nu$). The simulation shows bidirectional nuclear spin polarization as well as a reduction in the Overhauser field variance when the system is locked on to resonance. (F) $p(I_z)$ for two detunings marked by the dashed lines in E.

rate [45]; these experiments also demonstrated that when the QD is neutral, the nuclear spins do not depolarize even on time-scales exceeding an hour [47]. To confirm that DNSP indeed plays a key role in our experimental findings, we have carried out experiments to determine the relevant time scales for the buildup and decay of the locking phenomenon. We note here, that the decay discussed in this chapter, it to a large extent induced by the laser, due to the applied measurement technique. The details of nuclear spin decay are extensively studied and described in chapter 4. We anticipate though that nuclear spin polarization can be completely erased by the presence of an electron which undergoes strong exchange interaction with a nearby FR. Details about the decay of nuclear spin polarization are given in chapter 4.

Figure 3.3A presents a set of experiments that reveal the timescales associated with the decay of the DNSP generated during dragging, for the trion transition in sample A at $B_{ext} = 4.5$ T and $V_g$ that minimizes the coupling to the Fermi sea. The procedure used in these experiments is to first drag the QD trion transition by about $5\Delta\nu$ to the red side of the bare resonance $\omega_X$, and then to abruptly change the detuning between the trion and the laser field by a millisecond voltage ramp. The effect of this ramp is to set a new detuning condition $\omega_X - \omega_L \leq 4\Delta\nu$, which in turn results in an instantaneous loss of the absorption contrast. We observed that after a waiting time on the order of seconds, the initially vanishing absorption strength recovered its maximum value (figure 3.3A, red-coded data). By repeating
this experiment for a set of final detunings ranging from $4\Delta\nu$ to 0, we determined the characteristic exponential timescale for contrast recovery to be $\tau_d = 4.9 \pm 0.9$ sec. When we repeated the same experiment by first dragging the trion resonance to the blue side of the bare resonance and monitoring absorption for a final set of detunings satisfying $\omega_X - \omega_L \geq -4\Delta\nu$, we determined a contrast recovery time of $\tau_d = 3.7 \pm 0.7$ sec (figure 3.3A, blue-coded data). These results provide information about the decay time of the DNSP that is built up during the dragging process; as DNSP decays in the absence of a resonant laser, the Zeeman shifted trion resonance frequency changes until it once again reaches resonance condition with the laser field. Since the presence of a laser field that is detuned by less than $2\Delta\nu$ would speed up also the recovery of resonant absorption (see figure 3.3C), the timescales we obtain for the decay of the nuclear spin polarization in this experiment could be regarded as an upper bound on the nuclear spin depolarization rate. The DNSP decay times that we determine are in agreement with the values one would extrapolate from earlier experiments where $\kappa \sim 10^8$ s$^{-1}$ resulted in DNSP decay times on the order of few milliseconds at $B_{\text{ext}} = 0.2$ T [41]. When we repeated the experiment of Fig. 3 for the neutral QD exciton transition, we observed that the absorption contrast for $|\omega_X - \omega_L| < 4\Delta\nu$ always remained zero, indicating that the DNSP decay time was much longer than our measurement time of $\sim 1$ hour; this observation is in perfect agreement with earlier experiments [45, 47]. Finally, we remark that the bistable behaviour of resonant absorption contrast that is evident in vertical line cuts taken from the data in figure 3.3A (shown in figure 3.3B) is very characteristic of nonlinear nuclear spin dynamics in QDs [41, 42].

Figures 3.1 and 3.1 demonstrate that the response of a QD to a given laser detuning strongly depends on how the system reaches that particular detuning. To determine the QD optical response in the absence of such memory effects, we have carried out a set of experiments, where we first set the laser frequency to a large negative detuning with completely negligible excitation of the trion and kept the QD in a parameter regime with a strong co-tunneling rate for several seconds. This procedure allows the QD nuclear spins to thermalize with the lattice, which in turn ensures vanishing nuclear spin polarization. We then abruptly changed the voltage in milliseconds timescale, thereby instantaneously establishing $|\omega_X - \omega_L| \leq 7\Delta\nu$. Subsequently, we observed the time-dependence of the absorption signal at this fixed detuning. figure 3.3C shows that for a detuning range of $3\Delta\nu \geq |\omega_X - \omega_L| > \Delta\nu$, the absorption strength grows from zero to its maximum value on a timescale of a few seconds while within $|\omega_X - \omega_L| \leq \Delta\nu$ the on-resonance condition is established on a timescale below the temporal resolution limit of a few milliseconds in our experiment. Even though the frequency range over which the QD is able to lock on to the laser field is identical for red and blue detunings, the absorption recovery time is a factor of 2 slower for a laser that is tuned to the blue side of the bare trion resonance. Experimentally we find $\tau_b = 3.6 \pm 0.8$ sec for blue, and $2.1 \pm 0.7$ sec for red laser detunings (solid lines in figure 3.3C). We also note that the frequency range over which full absorption is recovered in these fixed laser frequency dragging experiments (green curve in figure 3.1C and figure 3.3C) is narrower than that of dynamical dragging bandwidth obtained by tuning the laser across the resonance (red and blue curves in figure 3.1C).
3.3. Bi-directional nuclear spin polarization

Perhaps the most unexpected feature of our experiments is the remarkably symmetric dragging effect that indicates bi-directional DNSP. Particularly interesting is the fact, that bi-directional DNSP is achieved with only a fixed circular polarization. This is in stark contrast to previous reports on DNSP where the angular momentum of a photon is transferred to the nuclear spins via the residual electron.

Also recent experiments in transport have demonstrated only unidirectional dragging of the electron (microwave) spin resonance and the bi-stability of the coupled QD electron-nuclei system [48–50]

3.4. Suppression of fluctuations in transition energy

The experiments depicted in figures 3.1-3.3 demonstrate the existence of a feedback mechanism that polarizes the necessary number of nuclear spins needed to shift the transition energy in a way to maintain resonance with the excitation laser. Consequently, fluctuations in the QD transition energy that would normally lead to a fluctuating absorption signal should be suppressed by such a compensation mechanism provided that the fluctuations occur within the effective feedback bandwidth.

Reference [51] presents a measurement where externally introduced gate-voltage fluctuations within the bandwidth of the feedback are suppressed when dragging condition is satisfied.

Even in the absence of an external perturbation, most QDs exhibit time-dependent fluctuations in $\omega_X$ [44]: these fluctuations could arise either from the electromagnetic environment of the QD or fluctuating nuclear Overhauser field. Figure 3.4 (upper panel) shows a typical time record of the resonant absorption signal of sample A QD together with the corresponding distribution function for $\Omega_L \sim \Gamma$: we observe up to 100% fluctuations in the resonant absorption signal, which is in turn a factor
Bi-directional dragging of quantum dot optical resonances by dynamic nuclear spin polarization

of 3 larger than our noise floor. In contrast, for $B_{\text{ext}} = 4.5 \text{T}$ (figure 3.4 lower panel), we find that the fluctuations in the absorption signal are reduced to the noise-floor. The absorption signal occasionally drops to a low value, indicating bistability in the response of the coupled electron-nuclei system to the resonant laser field. The frequency of jumps in absorption strength depends strongly on the system parameters.

3.5. Theoretical model

In this section, we present a model which captures the observed effect. It is based on an $\hat{I}_z \hat{s}_z$ type interaction. In general, such terms are contained in the non-collinear dipole-dipole hyperfine interaction between the electron and the hole, or they could originate from a combination of hyperfine and quadrupolar interaction. We want to point out that recently, attempts have been made which utilize the non-collinear hole-nuclear dipolar hyperfine interaction \[52\] to explain the dragging effect. Since the exact coupling strengths are unknown and under debate at the moment, we restrict ourselves to quadrupolar iteration. However, all mechanisms, qualitatively lead to a similar model.

If such an interaction is present a nuclear spin flip can occur simply by the presence of an electron in the QD. However, for sufficiently large external magnetic fields, such that the nuclear Zeeman splitting exceeds the coupling strength, these nuclear spin flips are suppressed. Therefore an additional processes is required. Here, we derive a model where nuclear spin flips are assisted by laser absorption followed by a spontaneous emission event. We find that this leads to an incoherent nuclear spin polarization rate with a dependence on the sign of the laser detuning.

Since it is not obvious how quadrupolar interaction leads to an $\hat{I}_z \hat{s}_z$ coupling, we sketch a derivation of an effective Hamiltonian. The Fermi contact hyperfine and quadrupolar Hamiltonians $\hat{H}_{\text{hyp}}$ and $\hat{H}_{\text{quad}}$ are given by the following expressions:

\[
\hat{H}_{\text{hyp}} = \sum_i A_i (\hat{I}_z \hat{s}_z + \frac{1}{2} (\hat{I}_+ \hat{s}_- + \hat{s}_+ \hat{I}_-))
\]

\[
\hat{H}_{\text{quad}} = \frac{e Q V_{zz}}{4I(2I-1)} (3(\hat{I}_{\text{principal}})^2 - I(I+1)).
\]

The flip flop terms in the hyperfine Hamiltonian are strongly suppressed at high magnetic fields due to the large difference in electron and nuclear Zeeman splitting such that we only have to take into account $\hat{s}_z \hat{I}_z$. The majority of nuclei (Ga,As) in the vicinity of the QD electron are spin-3/2. Expanding the Hamiltonian for such nuclei the interaction couples the $|\pm 3/2\rangle$ and $|\pm 1/2\rangle$ spin states (see section 2.6.5). The positive and negative spin manifolds are not coupled. We simplify the model by only considering one spin manifold. Assuming that the angle $\theta$ of the principle axis to $z$ is small, we find that the $| + 3/2\rangle$ and $| - 1/2\rangle$ are mixed. The $\hat{I}_z$ then transforms to:

\[
\hat{I}_z \rightarrow \hat{I}_z + \lambda \hat{I}_x
\]

with

\[
\lambda = \frac{3}{2} \frac{\sqrt{3} E_q \theta}{\Delta_N + 2 E_q}
\]
where \( E_q = eQV_{zz}/4 \). This leads to a dressing of the spin-conserving terms \( \hat{I}_z \hat{s}_z \) in the hyperfine interaction, producing terms like \( \hat{s}_z \hat{I}_z \) which allow for a nuclear spin flip without the requirement of flipping the electron spin. Importantly, \( \lambda \) depends on the magnetic field. In this way we can obtain an effective Hamiltonian \( \hat{H}_Q \):

\[
\hat{H}_Q = \sum_i B^i_\lambda \hat{I}_z \hat{s}_z
\]  

(3.5)

where \( B^i_\lambda = A_i \lambda \).

In the following we focus on \( X^− \) and simplify the physical system by considering only the blue trion transition. Then the ground state, where the quantum dot contains a single electron, interacts with the surrounding nuclear spin via the previously described process.

For this system, we can write down an effective Hamiltonian:

\[
\hat{H} = \delta \hat{s}_t + \Delta_N \hat{I}_z + \sum_i A_i \hat{I}_z \hat{s}_z + \frac{\Omega}{2} (\hat{\sigma}_{\uparrow \uparrow} + \hat{\sigma}_{\downarrow \downarrow}) + \sum_i B_i (\hat{I}_+ + \hat{I}_-) \hat{\sigma}_{\uparrow \uparrow}.
\]  

(3.6)

where \( \delta = \omega_t - \omega_L \) is the laser detuning with respect to the bare resonance, \( \Delta_N = gN\mu_B B_{ext} \) is the nuclear Zeeman splitting, \( A_i \) is the hyperfine coupling constant to the i-th nucleus and \( \Omega \) is the Laser Rabi frequency. As mentioned above the last term is ineffective due to the strong applied magnetic field and we eliminate it to first order by a Schrieffer-Wolff transformation. After the transformation we obtain laser-absorption assisted nuclear spin flips which are described by:

\[
\hat{H}_{flip} = \sum_i \frac{\Omega B_i}{2(\Delta_N + A_i)} \left( (\hat{\sigma}_{\uparrow \uparrow} - \hat{\sigma}_{\downarrow \uparrow}) (\hat{I}_+^i - \hat{I}_-^i) \right).
\]  

(3.7)

\( \hat{H}_{flip} \) couples the ground and the excited state with a Rabi frequency \( \Omega' = \frac{\Omega B_i}{2(\Delta_N + A_i)} \), where during the absorption process a nuclear spin is flipped. In the following we treat the nuclear spins as as a number \( I_z = \langle \hat{I}_z \rangle \) quantifying the nuclear spin polarization along the z-axis. We also make the assumption that the electron couples equally to all nuclear spins with a coupling strength \( A/N \), where \( A \) corresponds to an OH field for a fully polarized nuclear spin bath consisting of \( N \) nuclear spins. It is convenient to think about a manifold of two-level systems, where each system belongs to a certain nuclear spin polarization (figure 3.5). For a nuclear spin polarization \( I_z \) we can label the two level system by the states \( |\uparrow, I_z \rangle \) and \( |t, I_z \rangle \).

The excited states \( |t, I_z \rangle \) do not couple to the nuclear spins and differ by an energy \( \Delta_N(<0) \). The ground states which couple to the nuclear spins differ by an energy \( \Delta_N + A/N \). Let us assume the scenario, where \( I_0 = 0 \) and the excitation laser is slightly red detuned \( (\delta = \omega_t - \omega_L > 0) \) from the spontaneous emission and laser broadened trion resonance (figure 3.5): the effective detuning of \( \hat{H}_{flip} \) is then \( \Delta_N + \delta \) for the \( |\uparrow, I_z \rangle \rightarrow |t, I_z + 1 \rangle \)-transition and by \( \delta - \Delta_N \) from the \( |\uparrow, I_z \rangle \rightarrow |t, I_z - 1 \rangle \)-transition. Since \( |\delta - \Delta_N| > |\delta + \Delta_N| \) a transition event is more likely to occur on the \( |\uparrow, I_z \rangle \rightarrow |t, I_z + 1 \rangle \)-transition, causing a pump rate \( R_+ \) which flips nuclear spins up.
Figure 3.5.: Ladder diagram showing that a red detuned laser ($\delta > 0$) causes a nuclear spin polarization rate $R_+$ which leads to a polarization along the $+z$-axis until resonance condition is satisfied again. The details are described in the text.

Conceptually, the situation is similar to ion side-band cooling [53–56]. Whenever a nuclear spin is flipped the OH field changes by an amount $\pm A/N$, thereby shifting the energy of the ground state and decreasing the trion transition energy until locking condition $\omega_t - A/NI_z - \omega_L = 0$ to the laser frequency is satisfied again. In the case of a blue detuned laser ($\delta < 0$) the same mechanism leads to a built-up of a negative nuclear spin polarization with a rate $R_-$. At zeros laser detuning, both rates cancel. In this way, dragging effect of the blue trion resonance is achieved, where nuclear spins act as a feedback to ensure that the locking condition is satisfied for a wide range of laser detunings.

We can calculate the corresponding rate $R_+$ and $R_-$ at which nuclear spins flips occur using Fermi Golden Rule. In the case where the laser induces a nuclear spin flip from $I_z$ to $I_z + 1$. The possible transition of interest in this case is then $|\uparrow, I_z\rangle \rightarrow |t, I_z + 1\rangle$:

$$R_+ = \left( |\langle t, I_z + 1 | \hat{H}_{flip} |\uparrow, I_z\rangle|^2 + |\langle \uparrow, I_z + 1 | \hat{H}_{flip} |t, I_z\rangle|^2 \right) \rho_+ N_-$$

where $\rho_+$ denotes the density of available final states and $N_-$ is the number of nuclear spins which are polarized along the $-z$ axis. The density of state is simply given by a spontaneous emission and laser broadened Lorentzian, with a dependence on the laser detuning and the nuclear Zeeman splitting:

$$\rho_+ = \frac{\Gamma/2}{(\delta + \Delta_N - AI_z)^2 + \Gamma^2/4 + \Omega^2/4}$$

Similarly, we obtain a rate $R_-$:

$$R_- = \left( \frac{\Omega B}{2(\Delta_N + A_i)} \right)^2 \rho_- N_+$$
3.5. Theoretical model

Figure 3.6.: a) Ladder diagram showing the diffusive nuclear spin flip processes as described in the text. b) Reverse OH process: Nuclear spins can be flipped via spontaneous emission assisted hyperfine interaction.

With

$$\rho^- = \frac{\Gamma/2}{(\delta - \Delta_N - AL)^2 + \Gamma^2/4\Omega^2/4}$$

Both rates only differ by the sign of $\Delta_N$ in the denominator and the number of available nuclear spins $N_-$ and $N_+$, such that the total rate $R = R_+ - R_-$ of nuclear spin polarization exhibits a dependence on the laser detuning.

In addition to laser-assisted processes, nuclear spins can flip assisted by spontaneous emission. Such processes have a diffusive character since they only depend on the population in the trion state $\rho_{tt}$ are most probably the limiting factor for the dragging width:

$$\Gamma_{\text{diff}} = \left(\frac{B}{\Delta_N + A}\right)^2 \Gamma \rho_{ee}(N_+ - N_-),$$

with

$$\rho_{tt}(\delta) = \frac{\Omega^2/4}{\delta^2 + \Gamma^2/4 + \Omega^2/2}.$$ (3.13)

A last contribution to nuclear spin flips arises from spontaneous emission assisted hyperfine interaction. We claimed before that the flip-flop terms in the Fermi-contact hyperfine interaction are suppressed by the large difference in nuclear and electronic Zeeman energy. Spontaneous emission can take care of this difference. This so-called reverse Overhauser process is given by[51]:

$$\Gamma_{\text{RO}} = \left(\frac{A}{\Delta}\right)^2 \Gamma \rho_{tt} N_-.$$ (3.14)

In the case of a coupling of the excited state to the nuclear spins via $I_x s_z$ a similar model can be obtained. This is the case for the neutral exciton.

A comparison between theory and experiments on $X^0$ is shown in figure 3.7. The following data are all measured on QDs from sample A (25 nm tunnel barrier). Figure 3.7a shows two laser scans across the blue Zeeman line of $X^0$ at 4.5 T with opposite sweep directions. For both scans, DNSP was erased by waiting at a gate voltage of strong co-tunneling for 10 s. A calculation of the DT signal is plotted...
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Figure 3.7.: a (c): Example scans of the blue (red) trion resonance at 4.5 Tesla. b and d show the calculated scans using the model described in the text.

In figure 3.7b for the following parameters: $N = 3 \cdot 10^4$, where $N$ is the number of nuclear spins, $A = 170/N$, $B_{\text{ext}} = 4.5$ T, $B = 10^{-2} A, Q = 0.1$, $\Gamma = 0.66$, step size $\Delta \delta = 0.2 \mu$eV and dwell time $t_c = 0.2$ s. The experiments are qualitatively reproduced well. The calculation only differs in

So far, we have not discussed the red trion transition. While a laser scan across the blue transition leads to a positive feedback of the nuclear spins to ensure locking condition, a scan across the red Zeeman line will cause an anti-dragging effect. By anti-dragging, we mean, that the nuclear spin polarization causes a pushing of the resonance away from the laser. Changing to the red Zeeman line is equivalent to reversing the sign of the external magnetic field in the derivation of the model.

Figure 3.7c shows two scans of the red Zeeman transition of $X_0$ in opposite direction. The corresponding calculated spectrum is shown in figure 3.7d. The parameters except for the sign of the magnetic field were chosen as for the blue transition.

Again, the qualitative agreement is good.

In both figure 3.7a and 3.7b an asymmetry to positive laser detunings is obvious. We would argue that the reverse Overhauser rate could cause such an asymmetry. However for realistic parameters in the model, the asymmetry cannot be reproduced well. To obtain better insight, more experimental and theoretical investigations are necessary.

3.6. Dependence of laser dragging of quantum dot resonances on experimental parameters

Figure 3.9b shows the maximum dragging width at 4.5 T as a function of the laser Rabi frequency $\Omega_L$ for the neutral ($X^0$)exciton of a QD from sample A. The maxi-
3.6. Dependence of laser dragging of quantum dot resonances on experimental parameters

![Figure 3.8: Calculation of the static-dragging experiment shown in figure 3.3b.](image)

The maximum dragging width is plotted versus the laser Rabi frequency $\Omega_L$, normalized with respect to the linewidth $\Gamma$ at 0 T. All three sets of data peak around the same value, where $\Omega \simeq \Gamma$. Figure 3.9a depicts the maximum dragging measured for the neutral exciton as a function of external magnetic field. The sub-linear dependence is apparent. The electron $g$-factor for this quantum dot ($g_e = -0.67$) was measured by directly pumping the weakly-allowed trion transition ($\downarrow \leftrightarrow |\uparrow \downarrow \uparrow\rangle$) [57].

The absorption, or equivalently, differential transmission (DT) measurements [58] were performed by setting the gate voltage/laser energy to a specific detuning, waiting for a time $T_{dwell}$, and monitoring the output of the lock-in amplifier. After each measurement, the voltage/laser energy was changed by a detuning step $\Delta$. Figure S1 shows the dependence of the measurement on the parameters $T_{dwell}$ and $\Delta$. The presented data have been taken on the blue Zeeman transition of a neutral sample A QD.

Figure 3.10a and 3.10b show DT measurements where the energy detuning was varied in discrete steps of $\Delta = 0.4 \ \mu eV$ and $\Delta = 1.2 \ \mu eV$, respectively. The dwell time $T_{dwell}$ after each step was 200, 1000 and 5000 ms (light to dark grey) corresponding to the lock-in time constant. The energy range where the dynamical nuclear spin polarization (DNSP) effectively compensates for the detuning varies drastically with both the step size and the dwell time constant. The dependence of the dragging width on the dwell time is presented in Fig. 3.10c for fixed detuning steps of $\Delta = 0.2, 0.4, 1.6$ and $2.0 \ \mu eV$ (from top to bottom). In two limiting cases, namely for short and long time constants, the dragging width becomes independent of $T_{dwell}$ and saturates at minimum/maximum values. The solid lines are saturation fits to each set of data with a common fitting parameter $\tau = (1.0 \pm 0.3)$ s, a time scale consistent with the characteristic buildup time for DNSP. The minimum (down-triangles) and maximum (up-triangles) values of the dragged resonance are plotted in Fig. 3.10d along with exponential growth/decay fits as grey solid lines. The fitting parameter here was the critical step size, $\Delta \omega_c = (0.75 \pm 0.05)$ $\mu eV$, indicating that the DNSP range reduces exponentially when $\Delta \omega_c \geq \Gamma$. 
Figure 3.9.: Dependence of the maximum dragging width on external magnetic field and laser power: (a) Sub-linear growth of the maximum dragging width (in $\mu$eV) for a fixed laser Rabi frequency $\Omega_L = \Gamma$ as a function of external magnetic field (filled circles) plotted along with the calculated dragging width for the parameters given in the text and $\Omega = 0.1 \ast \Gamma$. The calculated width is smaller than the measured one but qualitatively agrees well. (b) Maximum dragging width (in units of the saturated linewidth $\Gamma$) as a function of laser Rabi frequency (normalized by $\Gamma$) for $T_{\text{dwell}} = TC = 200$ ms and $\Delta = 0.2 \mu$eV measured for a quantum dots in sample A at 4.5 T. The solid line is a calculation with the parameters in the text.
3.6. Dependence of laser dragging of quantum dot resonances on experimental parameters

Figure 3.10.: Dragging of the quantum dot resonance as a function of integration time and detuning step size. Absorption scans for different dwell times $T_{\text{dwell}} = 200$, 1000 and 5000ms (light to dark gray) at a given detuning step size of (a) $\Delta = 0.4 \, \mu\text{eV}$ and (b) $\Delta = 1.2 \, \mu\text{eV}$. (c) Dragging width as function of dwell time $T_{\text{dwell}}$ for different detuning steps $\Delta = 0.2, 0.4, 1.6, 2.0 \, \mu\text{eV}$ (from top to bottom). (d) Minimum (down-triangles) and maximum (up-triangles) values of the dragging width as a function of detuning step size.
4. Hyperfine interaction dominated dynamics of nuclear spins in self-assembled quantum dots

It is well known that co-tunneling of the electron leads to a decay of nuclear spin polarization in a self-assembled QD. In fact, we have used this mechanism in the last chapter to erase previously built-up nuclear spin polarization to avoid possible memory-effect in the measurements. In this chapter, we measure the dynamics of nuclear spins in a self-assembled QD at a magnetic field on 5 Tesla. We identify two distinct mechanisms responsible for the decay of the Overhauser field. We attribute a temperature-independent decay which lasts \( \sim 100 \) seconds to intra-dot diffusion induced by hyperfine-mediated indirect nuclear spin interaction. In addition, we observe a gate-voltage and temperature dependent decay stemming from co-tunneling mediated nuclear spin flip processes. By adjusting the gate-voltage and lowering the electron temperature to \( \sim 200 \) milliKelvin, we prolong the corresponding decay time to \( \sim 30 \) hours. Our measurements indicate possibilities for exploring quantum dynamics of the central spin model using a single self-assembled QD.

Hyperfine interaction between a single quantum dot (QD) electron and the nuclear spin ensemble defined by the nano-scale confinement provides a realization of the central spin problem [59–64]. This model has attracted considerable attention recently since the correlations between the confined electron and the nuclear spin ensemble induced by hyperfine coupling constitute the principal decoherence mechanism for the electron spin [31]. It has been recognized in this context that an enhancement of electron coherence time could be achieved by preparing nuclear spins in eigenstates of the Overhauser (OH) field operator [43, 65]: for this approach to be effective, it is essential to understand and characterize the dynamics of prepared (polarized) nuclear spin states.

In this chapter, we present measurements of nuclear spin dynamics in a single electron charged self-assembled QD. In contrast to prior work in self-assembled and gate-confined QDs [45, 51, 66, 67], we probe nuclear spin dynamics when both the exchange coupling to a Fermionic reservoir (FR) and the dipolar interaction between nuclear spins are vanishingly small. Our observations reveal a spatially limited, temperature-independent, nuclear spin diffusion originating from electron mediated nuclear spin interactions in addition to co-tunneling mediated, temperature dependent, decay of the OH field approaching \( 10^5 \) s. Remarkably, the diffusion induced reduction in the OH field taking place on \( \sim 100 \) s timescale can be strongly suppressed by repeating the preparation cycle consisting of polarization (pump) and free-evolution (wait).

The QDs in this sample are separated by a 35 nm GaAs tunnel barrier from a doped n++-GaAs layer. A bias voltage applied between a top semi-transparent
Ti/Au Schottky gate and back contacts allows to control the charging state of the QD and the relative alignment of its electronic levels with the Fermi energy of the FR [20]. High-resolution resonance-scattering spectroscopy [44] was performed on a single QD in a fiber-based confocal microscope incorporated in a dilution refrigerator [68]. The electron temperature was varied between 200 mK and 4 K while the applied magnetic field in the Faraday geometry was kept constant at 5 Tesla. We adopted a modulation-free measurement to keep the energy of the electron fixed during its interaction with the nuclear spins.

4.1. Experiment

We used a “pump-probe” technique to investigate the OH field dynamics. In the first step, the QD nuclear spins were polarized by slowly scanning a single-mode laser across the blue detuned Zeeman resonance of the neutral exciton (X₀): as was shown in Ref. [51], the magnitude of the OH field obtained in such a “dragging” experiment is given precisely by the detuning of the applied laser field from the bare resonance. A typical dragging process is shown in figure 4.1a. After generating an OH field of ~20 µeV with ~40 seconds of dragging, the gate voltage is abruptly changed (in < 1 msec) to a value V_wait that results in the injection of an electron into the QD from the FR (figure 4.1b). Due to the large trion (X⁻) energy-shift of ~5 meV, the laser field is far off-resonance during the waiting time τ_wait in which the coupled electron-nuclear system evolves freely. As a last step, the magnitude of the remaining OH field is measured after removing the electron from the QD and rapidly scanning the laser across the transition in ~50 ms. This probing is fast enough not to cause any appreciable dynamic nuclear spin polarization and simply reveals the resonance energy at the time of the measurement. We also confirmed that no appreciable change in OH field takes place during the time needed to switch the gate voltage between V_pump and V_wait. The “pump-wait-probe” sequence is then repeated for different τ_wait. A typical OH decay curve obtained using this procedure is shown in figure 4.1c.

In perfect agreement with earlier measurements [47, 51], we found no measurable decay of the OH field for an empty QD up to 1000 s. This result re-confirms that the OH field in self-assembled QDs is stationary in the absence of a confined electron (figure 4.2b black triangles). A possible explanation for this observation is the presence of large and inhomogeneous quadrupolar shifts within the QD that render dipolar-interaction mediated nuclear spin diffusion largely ineffective. Conversely, the non-trivial OH field dynamics in the presence of an electron that we discuss below demonstrates that the QD electron-nuclei system is a near-perfect realization of the central spin problem where the (nuclear) spins only interact with the central (electron) spin [64].

4.2. Decay in the presence of co-tunneling

When we choose V_wait such that the single-electron charged QD is in the co-tunneling regime[5] and the exchange coupling to the FR is strongest, we find that the OH
4.2. Decay in the presence of co-tunneling

Figure 4.1.: a) Build up of nuclear spin polarization by resonant optical dragging of the neutral exciton at 5 T. b) Schematic of “pump-probe” technique to measure the OH field decay: (1) A nuclear spin polarization is built up at a voltage $V_{\text{pump}}$ by slowly scanning the laser energy (full circle to open circle and back). (2) Gate voltage is set to $V_{\text{wait}}$ for a time $\tau_{\text{wait}}$ keeping the laser energy fixed. (3) Gate voltage is set to $V_{\text{pump}}$ again, followed by a fast (50 ms) laser scan to measure the OH field. c) A typical measurement of the OH field decay at 200 mK in the presence of a resident electron in the strong co-tunneling regime. The solid black line is calculated using the model (see text).

The measured OH field exhibits a fast decay \[45, 51\] on the order of a few seconds. The observed decay is clearly temperature dependent (see figure 4.2a).

In contrast, the OH field dynamics for $V_{\text{wait}}$ that corresponds to negligible electron co-tunneling shows decay on two distinct timescales (figure 4.2). The initial decay now takes place on a timescale of $\sim 100$ s and is temperature independent. Within this time, only a fraction of the OH field decays; for a single polarization cycle the decaying fraction is approximately 50% (figure 4.2b inset). This initial decay is followed by a temperature-dependent slower decay which varies from 3500 s at 4 K (figure 4.2b red dots) to $10^5$ s at 200 mK 1 (figure 4.2b blue squares).

The presence of two different OH field decay time scales with different temperature dependence points to two independent electron mediated mechanisms. To get further insight, we measured the gate voltage dependence of the temperature dependent decay rate across the single-electron charging plateau: in figure 4.3, squares (circles) denote the experimentally measured values of the temperature dependent decay time at $T = 200$ mK ($T = 4$ K). The values are extracted by fitting an exponential to this decay; the full (open) squares or circles indicate that the measured rate is the faster (slower) component of the OH field decay. The solid blue (red) curves show the gate voltage dependence of the co-tunneling time at $T = 200$ mK ($T = 4$ K) scaled by a (common) constant factor. The measured temperature dependent decay rates follow the gate voltage and temperature dependence of the co-tunneling rate across the charging plateau. In fact, we use the expected linear temperature dependence of the depicted co-tunneling rate and the good agreement with the experimentally measured decay times to determine our electron temperature to be

1Extrapolated from data taken up to $10^4$ s.
4.3. Electron-mediated nuclear spin diffusion

To investigate the temperature-independent initial decay of the OH field, we polarized the QD nuclear spins successively in four steps with a waiting time of 200 s in the presence of an electron between each step. The magnitude of the OH field at the end of each polarization cycle was kept the same. As shown in figure 4.4b, with successive polarization we find that the initial decay of the OH field is practically eliminated. This observation strongly suggests that the temperature independent component of decay arises due to “spatially limited diffusion” of nuclear spin polarization. Indeed, with such a polarization scheme, the nuclear spin polarization could diffuse within the QD in each dwell time leading to an overall increase in the polarization. As a result, to reach the same magnitude of the OH field in later steps, progressively smaller nuclear polarization is required during dragging. As the diffusion process just redistributes the excess polarization created during each step, one expects to see smaller decay of the OH field with each step, eventually leading to a complete suppression of the diffusion induced decay of the OH field.

4.4. Theoretical model

Our measurements thus indicate the presence of two qualitatively different mechanisms determining nuclear spin dynamics: temperature dependent decay and temperature independent diffusion of nuclear spin polarization, both mediated by the electron and leading to a decay of the OH field. To explain our findings, we use

\[ T \approx 200 \text{ mK}[68] \]

The ratio of the fast decay rates at the plateau edges for \( T = 4 \text{ K} \) and \( T = 200 \text{ mK} \) is 8; this discrepancy could arise due to gate voltage fluctuations that act as an effective finite temperature when \( V_{\text{wait}} \) is in the co-tunneling region.
a model which includes Fermi-contact hyperfine interaction (\(H_{\text{hyp}}\)) describing the coupling of the electron spin \(\hat{S}\) to \(\sim 10^5\) nuclear spins \(\hat{I}\) of the QD host material, exchange interaction between the QD electron and the electrons in the FR (\(H_{\text{exch}}\)), and an effective non-collinear dipolar hyperfine interaction \(H_{\text{dip}}\). The total Hamiltonian can then be written as

\[
\hat{H} = \hat{H}_0 + \hat{H}_{\text{hyp}} + \hat{H}_{\text{exch}} + \hat{H}_{\text{dip}} \tag{4.1}
\]

where, \(\hat{H}_0 = \Delta \hat{S}_z + \sum_i \Delta_i^N \hat{I}_i^z\) is the Zeeman-Hamiltonian with \(\Delta = g_e \mu_B B \approx 180 \mu eV\) and \(\Delta_i^N\) denoting the electronic and nuclear Zeeman energies, respectively. Here we have incorporated the inhomogeneous quadrupolar interaction induced energy shifts of each nucleus in \(\Delta_i^N\); since typical quadrupolar fields (0.3 Tesla) are much smaller than the external field (5 Tesla), we have \(\Delta_i^N \simeq \Delta_N = g_N \mu_N B \approx 0.1 \mu eV\).

\(\hat{H}_{\text{exch}} = \sum_{k,k'} J_{k,k'} \hat{s}_{k,k'} \cdot \hat{S}\) describes the exchange coupling between the QD electron and the FR with \(\hat{s}_{k,k'}\) denoting the spin operator of the FR at the position of the QD and \(J_{k,k'}\) denoting the exchange interaction strength. This interaction leads to an incoherent electron spin flip rate \(\kappa\) which in our QD is \(\sim 10^{-7} \mu eV\) at 4 K in the center of the charging plateau where the co-tunneling is smallest. The Fermi-contact hyperfine interaction is given by \(\hat{H}_{\text{hyp}} = \sum_i A_i (\hat{I}_i^z \hat{S}_z + \frac{1}{2} (\hat{I}_i^+ \hat{s}_- + \hat{s}_+ \hat{I}_i^-))\), where \(A_i \propto |\psi(\vec{r}_i)|^2 \approx 10^{-2} \mu eV\) is the hyperfine constant of the \(i\)-th nucleus with \(\psi(\vec{r}_i)\) denoting the QD electron wave-function. The first term in \(\hat{H}_{\text{hyp}}\) is the OH (Knight) field seen by the electron (nuclei). For large magnetic fields used in our experiments, the flip-flop terms in \(\hat{H}_{\text{hyp}}\) are ineffective due to the large difference in the electron and nuclear Zeeman energies. Eliminating these terms in Eq. (3) using a Schrieffer-Wolff transformation [69], we obtain new terms describing electron mediated spin flip of two spatially separated nuclear spins:

\[
\hat{H}_{\text{ind}} = \sum_{i \neq j} \frac{2 A_i A_j}{\Delta - \Delta_N} \hat{I}_i^x \hat{I}_j^+ \hat{s}_z . \tag{4.2}
\]
This indirect, coherent long-range interaction leads to a diffusion of nuclear spin polarization within the region where the electron wave-function is non-vanishing (figure 4.4a). Although the total magnitude of QD nuclear spin polarization does not decrease due to this diffusion process, the OH field seen by the electron decays partially due to a redistribution of the nuclear spin polarization within the QD. We attribute the temperature independent decay of the OH field to such an electron mediated diffusion.

The last term in the Hamiltonian describes an effective non-collinear dipolar hyperfine interaction between the electron and the $i$-th nucleus with coupling constant $B_i$:

$$\hat{H}_{\text{dip}} = \sum_i B_i \hat{I}_i \hat{s}_z.$$  \hspace{1cm} (4.3)

Such terms could appear due to small but non-zero dipolar hyperfine interaction between the QD electron and nuclei. Alternatively, they could be induced by quadrupolar axes of nuclear spins that are non-parallel to the external field [70]. These terms induce nuclear spin flips that lead to a decay of the nuclear spin polarization. In fact, the temperature-dependent decay of the OH field can be explained by a second order process originating from $\hat{H}_{\text{dip}}$. The energy conservation in this irreversible nuclear spin flip process is ensured by the coupling of the QD electron to the FR (figure 4.3a); the corresponding OH field decay rate is then $(B_i/\Delta_N)^2\kappa$. The explains the temperature and gate voltage dependence of the decay shown in figure 4.3. Since $\Delta_N \ll T$, we expect this rate to be linearly proportional to the electron temperature $T$. We rule out any contribution of co-tunneling assisted direct hyperfine flip-flop processes, since the corresponding rate can be shown to be four-orders-of-magnitude slower than the rates that we measure in our experiments.

### 4.5. Numerical calculation based on semi-classical rate equations

We model the nuclear spin dynamics using semi-classical rate equations, taking into account the diffusion and decay processes arising from $\hat{H}_{\text{ind}}$ and $\hat{H}_{\text{dip}}$, respectively. For simplicity, we assume a two-dimensional QD with $N = 10^4$ spin 1/2 nuclei. The rate equations describing the change in time of the probability $P_{i\uparrow}(t)$ that the $i$-th nucleus is in the $|\uparrow\rangle$ state become:

$$\frac{dP_{i\uparrow}(t)}{dt} = \left(\frac{B_i}{\Delta_N}\right)^2 \kappa (1 - 2P_{i\uparrow}(t)) + \sum_j \left(\frac{2A_i A_j}{\Delta}\right)^2 \rho_{ij}(P_{j\uparrow}(t) - P_{i\uparrow}(t)).$$  \hspace{1cm} (4.4)

The first term on the RHS of Eq. 4.4 represents the temperature dependent decay while the second term represents the decay induced by the spatially limited diffusion. To obtain Eq. 4.4, we assume that the coherent coupling of two distant nuclear spins with similar energies via $\hat{H}_{\text{ind}}$ is interrupted by a pure dephasing process with rate $\gamma_{\text{deph}}$. The Lorentzian factor $\rho_{ij} = \gamma_{\text{deph}}/(\delta_{ij}^2 + \gamma_{\text{deph}}^2)$ describes the effective density
4.5. Numerical calculation based on semi-classical rate equations

Figure 4.4.: a) Schematic of the electron mediated nuclear spin diffusion: due to the inhomogeneous Knight shifts and quadrupolar fields, only nuclear spins with a small energy difference can interact which is depicted by the dashed circles. Two nuclear spins can flip without flipping the electron spin. b) Demonstration of the spatially limited nuclear spin diffusion: By sequential polarization of the nuclear spins, the polarization can be saturated, suppressing further nuclear spin diffusion (see text for details).

of states for the flip-flop process between two (distant) nuclear spins with energy difference $\delta_{ij} = \Delta^i_N + A_i - \Delta^j_N + A_j$.

A possible source of $\gamma_{deph}$ is the intrinsic gate voltage fluctuations in our experimental setup; such fluctuations would influence the electron wave-function giving rise to an effective broadening of the Knight field experienced by the nuclei. As the bandwidth of this noise is limited by the bandwidth of the gate in our sample ($\sim 50$ kHz), these fluctuations should not affect the decay process which is accompanied by a nuclear spin flip and requires an energy exchange of $\sim \Delta_N \approx 0.1 \mu eV$. Eq. 4.4 can be solved for a given initial distribution of nuclear polarization which we assume is proportional to $\psi(\vec{r})$; with the knowledge of $P^i_1(t)$ for all nuclear spins, one can easily get the OH field as $OH(t) = \sum_i A_i (P^i_1(t) - 0.5)$. For the calculations, we used $\Delta=174 \mu eV$, $\Delta_N=0.1\mu eV$, $\sum A_i=174 \mu eV$ (5 T) and $B_i \sim 10^{-2} A_i$. We first fix the parameters for the case of smallest co-tunneling at 4 K ($\kappa = 10^{-7} \mu eV$) and then use calculated $\kappa(V_{wait}, T)$ to obtain $OH(t)$ for different temperatures and gate voltages. We obtain a value of $\sim 2$ kHz for $\gamma_{deph}$ which is well below the bandwidth of the gate. The results of the calculations plotted in FIGS. 4.1-4.4 with solid lines, show good agreement with the experiment.

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$^3$Charge fluctuations near the QD can also induce fluctuating electric field gradients and cause a broadening of nuclear spin energies; this process will also contribute to $\gamma_{deph}$. 
Signatures of Kondo correlations in optical absorption

5. Signatures of Kondo correlations in optical absorption

5.1. Introduction

The interaction between a single confined spin and the spins of a Fermionic reservoir leads to one of the most spectacular phenomena of many body physics – the Kondo effect [10, 71]. It was unintentionally discovered in 1934 by de Haas and van der Berg [72]. They measured the resistance of metals which they assumed to be pure and found that the resistance exhibited a minimum around 10 K. Later it turned out that these metal were not pure but rather contained small concentrations of magnetic impurities. This was very unexpected because the resistance was considered to be well understood: in a metal it usually decreases when the temperature is lowered and then saturates. At high temperatures, the resistance originates from electron-phonon scattering with a $T^5$ dependence. A reduction in temperature leads to freezing out of phonons and at some point electron-electron scattering starts to be important. This scattering has a $T^2$ dependence and vanishes as $T \to 0$. At very low temperatures the resistance is due to potential scattering which arises from imperfections in the crystal lattice. The increase was an open question for 30 years until the Japanese physicist Jun Kondo noticed that not potential scattering has to be considered but also spin-spin scattering between electron spins and impurity spins. Even for low concentrations of impurities, spin-spin scattering is important. Kondo calculated perturbatively [71] that spin-flip scattering leads to a logarithmic divergence of the resistance $\propto \ln(T_K/T)$ at low temperature. The quantity $T_K$ is called the Kondo temperature: a characteristic energy scale, determined by the system parameters, at which this effect starts to become important. With this previously missing contribution, the resistance could now be well reproduced by the theory. Almost 40 years later, the Kondo effect experienced a revival, when it was demonstrated in single gate defined QDs [10–13, 73]. When a QD, tunnel coupled to a Fermionic reservoir (FR), is charged with an odd number of electrons from the leads, it behaves like a magnetic impurity. In order to study the Kondo effect, the QD is coupled two the leads, a source and a drain with a small voltage applied. The presence of the Kondo effect leads to an increased forward scattering through the QD, measurable as an increased tunnel current though the dot. This occurs when the single occupied electron level in the QD is not aligned with the Fermi energy. Here the great advantage compared to metals lies in the tunability of the parameters.
5.1.1. The Anderson Hamiltonian

The tunnel interaction between a localized magnetic impurity, like a quantum dot and a sea of Fermions is well described by the Anderson Hamiltonian [74]. The Hamiltonian can be written as

\[ \hat{H} = \hat{H}_{QD} + \hat{H}_{FR} + \hat{H}_{tunnel} \]

\[ \hat{H}_{QD} = \sum_{\sigma} \varepsilon_{\sigma} \hat{e}_{\sigma}^\dagger \hat{e}_{\sigma} + U_{ee} \hat{e}_{\uparrow}^\dagger \hat{e}_{\downarrow}^\dagger \hat{e}_{\downarrow} \hat{e}_{\uparrow} \]

\[ \hat{H}_{FR} = \sum_{\vec{k},\sigma} \varepsilon_{\vec{k},\sigma} \hat{c}_{\vec{k},\sigma}^\dagger \hat{c}_{\vec{k},\sigma} \]

\[ \hat{H}_{tunnel} = \sum_{\vec{k}} V_{\vec{k}} (\hat{e}_{\vec{k},\sigma}^\dagger \hat{e}_{\sigma} + \hat{e}_{\sigma}^\dagger \hat{c}_{\vec{k},\sigma}) \]

\( \hat{H}_{QD} \) describes the QD where \( \hat{e}_{\sigma}^\dagger (\hat{e}_{\sigma}) \) creates(annihilates) an electron with spin \( \sigma \). We assume that only the lowest QD level with energy \( \varepsilon_{\sigma} \) is important here. The second term is the on-site Coulomb repulsion with energy \( U_{ee} \). If \(-U_{ee} < \varepsilon < 0\) this term prevents a second electron from tunneling into the QD.

\( \hat{H}_{FR} \) describes the a non-interacting reservoir of electron FR, where \( \hat{c}_{\vec{k},\sigma}^\dagger (\hat{c}_{\vec{k},\sigma}) \) creates(annihilates) an electron with k-vector \( \vec{k} \) and spin \( \sigma \).

\( \hat{H}_{tunnel} \) describes the tunnel coupling between the two systems with a coupling constant \( V_{\vec{k}} \equiv V \) which we assume to constant for all \( \vec{k} \)’s. The tunneling processes are determined by the density of states \( \rho \) and \( \Gamma \). \( \Gamma \) is the level broadening and related to \( V \) by \( V = \sqrt{\Gamma / \pi \rho} \).

5.1.2. The Kondo Hamiltonian

The Anderson Hamiltonian describes the physics of a QD coupled to a FR, if the QD is uncharged, single charged or double charged. The interesting physics happens, when the QD contains exactly one electron. One can project the Anderson Hamiltonian into this subspace by the Schrieffer-Wolff transformation [37], thereby obtaining an effective spin-exchange Hamiltonian:

\[ \hat{H}_{Kondo} = -J \hat{s} \hat{S}(0), \]

where \( \hat{s} \) is the QD electron spin operator and \( \hat{S}(0) \) is the collective spin operator of all FR electrons at the position of the QD spin. Eqn. 5.5 describes spin-flip processes, where the QD electron spin is flipped with a spin from a FR electron via an intermediated virtual state. A schematic of two lowest order processes is shown in figure 5.1: the gate voltage \( V_g \) is adjusted such that the electronic level is far below the Fermi energy. The energy required to charge the QD with a second electron is \( U_{ee} \). In this so called local-moment regime, the QD is in the Coulomb blockade regime. The costs of permanently adding an electron from the FR to the QD is \( \Delta E = U_{ee} - E_F \) and is not available. However an electron can hop in and out of the QD within a time \( \Delta t \leq \hbar / 2 \Delta E \). This is called co-tunneling. If the electron which tunnels out of the QD has opposite spin than the one which tunneled in, the QD electron spin has effectively flipped. Similarly, the QD electron can tunnel out
of the dot within a time $\Delta t \leq \hbar/2|\varepsilon|$ and is replaced by an electron with opposite spin from the FR.

A projection of the Anderson Hamiltonian in the local-moment regime yields additional terms which describe an energy-renormalization of both, the QD electron and the FR. This arises from virtual tunneling of the QD electron to empty states above the FR and is usually neglected. However, as we will see in section 5.7, we measure a strong renormalization of the trion transition energy, which enables us to extract the relevant system parameters, $\Gamma$ and $U_{ee}$ in our QD.

### 5.1.3. Excitonic Anderson Model (EAM)

In the case of optical absorption, where the excited state contains a hole, an extension of the Single Impurity Anderson Model is required, which we call this the Excitonic Anderson Model (EAM) [75]: The Hamiltonian is given by $H = H_{QD} + H_{FR} + H_{tunnel}$, where

$$H_{QD} = \sum_\sigma (\varepsilon_{e\sigma} n_{e\sigma} + \varepsilon_{h\sigma} n_{h\sigma}) + U_{ee} n_{e\uparrow} n_{e\downarrow} - \sum_{\sigma \sigma'} U_{eh} n_{e\sigma} n_{h\sigma'}$$  \hspace{1cm} (5.6)

describes the dot, with electron number $n_{e\sigma} = e_{\sigma\dagger} e_{\sigma}$ and hole number $n_{h\sigma} = h_{\sigma\dagger} h_{\sigma}$. The Coulomb repulsion $U_{ee}$, excitonic attraction $U_{eh}$ and hole energy $\varepsilon_{h\sigma}$ are taken to be positive. The energies $\varepsilon_{e\sigma}$ and $\varepsilon_{h\sigma}$ both shift linearly with gate voltage $V_g$, with a slope of opposite sign, but same magnitude,

$$\Delta \varepsilon_{e\sigma} = -\frac{|e|}{\alpha_g} \Delta V_g,$$
$$\Delta \varepsilon_{h\sigma} = \frac{|e|}{\alpha_g} \Delta V_g,$$  \hspace{1cm} (5.7)

where $|e|$ is the unit of charge and $\alpha_g$ the lever arm. For dot 1, whose gate-voltage dependence we studied in detail, the lever arm is given by $\alpha_g = 7$. The terms $\hat{H}_{FR}$ and $\hat{H}_{tunnel}$ are defined as above introduced.
When an incident photon is absorbed by the quantum dot, it creates a particle-hole pair. The interaction between photon and dot is given by
\[ H_L \propto (e^\dagger_{\sigma} h^\dagger_{\bar{\sigma}} e^{-i\omega_L t} + \text{h.c}), \]
where \( e^\dagger_{\sigma} \) and \( h^\dagger_{\bar{\sigma}} \) create a QD electron and a hole with well defined spins \( \sigma \) and \( \bar{\sigma} = -\sigma \). In our case, we are interested in the \( \text{X}^- \) transition, where the initial state contains a single electron and the final state contains two electrons in a singlet and a hole. It is therefore convenient to define two different Hamiltonians, \( H^{i/f}_c = H^{i/f}_e + H_c + H_t \), which describe the system before and after absorption, where
\[ H^a_e = \sum_{\sigma} \epsilon^a_{\sigma} n_{e\sigma} + U_{ee} n_{e\uparrow} n_{e\downarrow} + \delta_{af} \epsilon_{h\bar{\sigma}} \quad (a = i, f). \] (5.8)
These Hamiltonians differ (i) in the position of their \( e \)-levels (\( \epsilon^i_{\sigma} \) and \( \epsilon^f_{\sigma} = \epsilon^i_{\sigma} - U_{eh} \)), where the \( e \)-level of the final Hamiltonian is pulled down by the excitonic Coulomb attraction and (ii) in the term \( \delta_{af} \epsilon_{h\bar{\sigma}} \) which accounts for the energy of the hole.

### 5.2. Numerical renormalization group approach

In this section we sketch the concept of the numerical renormalization group (NRG), an iterative numerical method to diagonalize quantum impurity models such as the Anderson Hamiltonian and the Kondo Hamiltonian. A more detailed and excellent description can be found in references [76–82]. The coupling of a quantum dot electrons to the Fermi sea is well described by the Anderson Hamiltonian as discussed in section 5.1.1. We will assume a constant tunnel coupling for all the calculations in this work. The QD electron couples to a FR with a bandwidth \( 2D \) (from \(-D\) to \(D\)) where we assume for simplicity a constant density of states \( \rho = 1/2D \). In the following \( D = 1 \) will serve as an energy unit. Obtaining a simple numerical solution is difficult, due to the large size of the Hilbert space. The QD can be empty, singly occupied with either a spin-up or a spin down electron or doubly occupied. The same applies to each state in the Fermi sea. For \( N^{20} \) electron in the FR, the total Hilbert space is \( 2^{10^{30}} \) and it is impossible to diagonalize the system Hamiltonian numerically. In the 1970’s K. G. Wilson developed a scheme to solve the Kondo problem non-perturbatively. Generally, the problem is that in the case of the Kondo model, all energy scales have to taken into account. The properties of the system are determined by energy scales that are much smaller than the energy scales of the Hamiltonian. In the case of Kondo, the relevant energy scale is the Kondo temperature \( T_K \) which is orders of magnitude smaller than the Coulomb energy \( U_{ee} \) and tunnel coupling \( \Gamma \). Wilson’s idea was the following: in a first step the FR is logarithmically discretized around the Fermi energy. The FR is divided into intervals \( \pm [\Lambda^m, \Lambda^{m+1}] \), where \( \Lambda > 1 \) is the discretization parameter. Typically, \( \Lambda \) is chosen between 1.5 and 3 (we typically use \( \Lambda = 1.8 \)). In a second step a Fourier transformation of the operators in each interval yields a complete set of orthonormal function which span the whole \( k \)-space. The set of each interval can be approximated by a single state which couples to the QD electron spin. A schematic is depicted in Fig. 5.2.

In a third step, the system is mapped on a semi-infinite chain, where the QD electron couples only to the first site of the chain. The first chain site couples to the
5.2. Numerical renormalization group approach

The resulting Hamiltonian describes a nearest neighbor hopping:

$$\hat{H} = \hat{H}_{\text{QD}} + \sqrt{\frac{\Gamma}{\pi}} \sum_{\sigma} (\hat{f}_{0,\sigma}^{\dagger} \hat{c}_{\sigma} + \hat{c}_{\sigma} \hat{f}_{0,\sigma}^{\dagger}) + \frac{1}{2} (1+\Lambda^{-1}) \sum_{\sigma} \sum_{n=0}^{\infty} \Lambda^{-n/2} \xi_n (\hat{f}_{n,\sigma}^{\dagger} \hat{f}_{n+1,\sigma} + \hat{f}_{n+1,\sigma}^{\dagger} \hat{f}_{n,\sigma})$$

where $\xi_n = (1 - \Lambda^{-n-1})(1 - \Lambda^{-2n-1})^{-1/2}(1 - \Lambda^{-2n-3})^{-1/2}$. The matrix elements of the hopping Hamiltonian decrease exponentially with $\Lambda^{-n/2}$. Due to the separation of the energy scales on each chain site, the problem can be solved recursively, where

$$\hat{H}_0 = \hat{H}_{\text{QD}}$$

$$\hat{H}_{N+1} = \sqrt{\Lambda} \hat{H}_N + \xi_N (\hat{f}_{N+1,\sigma}^{\dagger} \hat{f}_{N,\sigma} + \hat{f}_{N,\sigma}^{\dagger} \hat{f}_{N+1,\sigma})$$

Now it is possible to calculate the eigenstates and eigenvalues of $\hat{H}_{N+1}$ if the ones from $\hat{H}_N$. This is exact in the case of $\Lambda = 1$ and $N \to \infty$. For an implementation one first starts with the QD site, which can be diagonalized. Then the first site of the chain is added via the above recursion relation and the new Hamiltonian is diagonalized again. More chain sites are then added by successive diagonalization and adding, while each new chain site can be considered as a perturbation on the existing chain. Again, the Hilbert space grows exponentially with the length of the chain as $4^N$, and after a few iterations, a numerical diagonalization takes a very long time or is not possible any more. Therefore, a truncation scheme is necessary where the high energy states are discarded. In this work typically, 1024 states are kept in each iteration step. The energy resolution which is achieved after the $N$-th step is $\delta \propto \Lambda^{-n/2}$. To reach the energy scale of interest, the chain length is usually $N \approx 60 - 80$.

To get insight into the physics which dominate the processes at certain energy scales it is instructive to plot the energies of the lowest lying eigenstates for each

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**Figure 5.2.:** Schematic of the discretization.
iteration step (corresponding to an energy scale). Since the energy scales decrease exponentially at each iteration, it is convenient to renormalize them by multiplying them with a factor $\Lambda^{-(n-1)/2}$. Such a plot is called a flow-diagram. Whenever a change in the physical processes occurs, it leads to a change in the rescaled eigenvalues. The flow diagram has certain regimes where the rescaled eigenstates do not change over many iteration steps. The regimes are called fixed-point. The Anderson Hamiltonian has 3 fixed-point: A free-orbital regime, a local moment regime and the strong-coupling regime. The free orbital regime is characterized by charge fluctuations in the QD due to tunneling. In the local moment regime, charge fluctuations are frozen out and the physics is dominated by spin-fluctuations. At even lower energy-scales, the strong exchange interaction leads to Kondo correlations, where the QD spin is screened by the FR electrons. Figure 5.4 shows the 100 lowest lying renormalized Eigenenergies of the system as a function of iteration step.

5.2.1. Calculation of the absorption spectrum

The absorption spectrum for the $X^-$ transition can then be calculated according to Fermi’s golden rule

$$A_\sigma(\delta) = 2\pi \sum_{mm'} \rho^\dagger_m |\langle m'|e^\dagger_\sigma|m\rangle|^2 \delta(\omega_L - E^f_{m'} + E^i_m)$$

(5.12)

where $|m\rangle_a$ and $E^a_m$ are the eigenstates and -energies of $H^a$. The detuning $\delta = \omega_L - \omega_{th}$ is defined relative to the threshold frequency $\omega_{th} \equiv E^f_G - E^i_G$ below which at $T = 0$ no photons can be absorbed, which is given by the difference of the ground state energies $E^G_a$ of $H^a$. The positive hole is taken as a static spectator, interacting with the dot through Coulomb interaction $U_{eh}$ only. We remark that the definition of $A_\sigma(\delta)$ in equation 5.12 contains a creation operator $e^\dagger_\sigma$ of opposite spin (in contrast to the convention used in Ref. [75] for the $X^0$ transition, which contains $e^\dagger_{\bar{\sigma}}$). The convention used here ensures that $A_\sigma(\delta)$ describes the transition with highest weight for an initial configuration containing a spin-$\sigma$ electron ($n^1_{e\sigma} \simeq 1$), which requires the added electron from the exciton to have opposite spin, $\bar{\sigma}$.

5.2.2. Numerical Renormalization Group approach for spectral functions

In order to calculate dynamical quantities, such as the absorption lineshape a more advanced NRG method has to be applied. In this work we made use of a highly
5.2. Numerical renormalization group approach

When the matrix elements of in Eqn. 5.12 are calculated, it is important to take into account eigenstates of all energies. Since in each NRG step, the high energy states are discarded, at the end of the calculation one is left with only a set of eigenstates describing to low energy physics. In the case of the absorption lineshape, we compare the laser detuning to the Kondo temperature. Even when $T_K$ small compared to a blue detuned laser, Kondo correlation still modify the lineshape. In this section we sketch the procedure how to obtain correct absorption lineshapes.

The quantities $|m⟩_a$ and $E^a_m$ occurring in Eq. 5.12 can be calculated using the numerical renormalization group [77] (NRG). By combining NRG data from all iterations, it is possible to construct a complete set [83] of approximate many-body eigenstates of the full Hamiltonian. These can be used to evaluate equilibrium spectral functions via their Lehmann-representations; at finite temperatures, this can be done using the full density matrix (FDM)-NRG [84].

Equation 5.12 contains matrix elements between initial and final states that are eigenstates of different Hamiltonians, $H^i$ and $H^f$. Hence, two separate NRG runs are required to calculate these (similar to what is done for time-dependent NRG [83]).

The strategy is then as follows: In a first step an NRG run generates a complete set of approximate eigenstates $|m⟩_i$ and eigenvalues $E^i_m$ for the initial Hamiltonian $H^i$, describing a single electron in the quantum dot. In a second run, we generate a complete set of approximate eigenstates $|n⟩_f$ and eigenvalues $E^f_m$ for the final trion state. The double sum in Eq. (5.12), over all initial and final eigenstates, is performed in two steps: First we perform a backward run, with site index $k$ running from the end to the beginning of the Wilson chain [83], and calculate for each shell $k$ the contribution $ρ^k_i$ towards the initial density matrix from that shell (obtained using data from NRG run #1). This is followed by the usual forward run, in which the matrix elements $|⟨n|e^σ_0|m⟩|^2$ between shell-$k$ eigenstates from NRG runs #2 and NRG #1 are calculated, combined with $ρ^k_i$, and binned according to the corresponding frequency difference $E^f_n - E^i_m$. The $T = 0$ threshold frequency

---

**Figure 5.4.**: NRG flow diagram showing the 100 lowest lying NRG Eigenenergies as a function of iteration step, renormalized by $Λ^{-(n-1)/2}$. Such a plot shows the fixed-points of the system.
for the onset of absorption is given by the difference of ground state energies of the first two NRG runs, \( \omega_{th} \equiv E^f_G - E^i_G \). The absorption spectrum is expected to have divergences at the threshold \( \omega_t \), hence all frequency data are shifted by the overall threshold energy \( \omega_{th} \) prior to binning. (For finite temperature, the sharp onset is broadened and divergences are cut off.) The discrete eigenvalues of shell \( k \) are spread over an energy range comparable to the characteristic energy \( \Lambda^{-k/2} \) scale of that iteration, which decreases exponentially with \( k \). Thus, the bins used for collecting the discrete data are likewise chosen to have widths decreasing exponentially with decreasing energy. The discrete, binned data are subsequently broadened using a log-Gaussian broadening scheme, characterized by a broadening parameter \( \alpha \) as described in Ref. [84], here taken as \( \alpha = 0.4 \).

### 5.3. Structure of the Fermi reservoir

The ground state of the Kondo Hamiltonian 5.1.2 is the interesting “Kondo singlet”. Due to the presence of exchange interactions between the magnetic impurity and the FR, the system lowers the total energy by forming a singlet, where the Kondo temperature \( T_K \) can be envisaged as the binding energy of the state. One can write down an expression for this state:

\[
|\psi_{\text{Kondo}}\rangle = (\hat{e}^+_\uparrow \hat{a}^+_0 \uparrow - \hat{e}^+_\downarrow \hat{a}^+_0 \downarrow) \prod \hat{a}^+_n \uparrow \hat{a}^+_n \downarrow |\text{VAC}\rangle. \tag{5.13}
\]

The operators \( \hat{e}^+_\sigma \) create a QD electron with spin \( \sigma \). The operators \( \hat{a}^+_n,\sigma \) are a superposition of the “original” FR operators:

\[
\hat{a}^+_n,\sigma = \sum \xi_n \hat{c}^+_k \sigma. \tag{5.14}
\]

\( n = 0 \) corresponds to the singlet cloud of FR electron, which screen the local impurity. The is a set of most localized electrons, where the localization is related to the bandwidth \( D \) of states that the QD electron couples to. For \( D \to \infty \) the cloud would be localized on a point. In reality, the finite bandwidth leads to a finite localization of the Kondo cloud. This object, a many body wavefunction acts as a strong scattering potential for other FR electron close to the Fermi energy. In the absence of a Kondo cloud, their wavefuncton extends over the whole crystal. However, when the strongly correlated ground state is occupied at very low temperatures. These electrons are scattered of the Kondo cloud and experience scattering phase shifts. Figure 5.5 shows a schematic in the case of a single or double electron charged quantum dot: when the quantum dot contains a single electron a Kondo singlet form, which lead to strong phase shifts. If there a two electron in a local singlet in the QD, this objects only constitutes a very weak or negligible scattering potential for FR electrons.

When this happens, the QD electron and the FR cannot be considered as separate systems which are coupled by an interaction. They rather have to been treated as one big system. It turns out that the structure of the FR is strongly modified in the ground state. Qualitatively one can explain the physical situation as follows: forming a singlet lowers the total ground state energy by \( T_K \). The way this happens is that all single-particle states in the FR experience small scattering phase shifts, say \( \varepsilon_k \to \varepsilon_{k'} \) which give rise to energy shifts. But the shifts are not uniform: levels
close (within $T_K$) to the Fermi energy experience strong phase-shifts, by almost half a level spacing, those beyond $T_K$ from the Fermi energy get only weakly shifted. The sum of all energy shifts is on the order of $T_K$ justifying the term binding energy. To summarize: by forming a singlet, the system lowers its energy by an amount $T_K$, which is stored in the form of energy phase-shifts in the FR within an energy band $T_K$ around the Fermi energy. Or more accurately: scattering phase shifts can be associated with single-particle level shifts, and the binding energy is stored in the level shifts.

Excitations above the ground state behave like free electron excitations, except that they have an extra phase shift $\delta E$, which depends on the distance $\varepsilon$ to the Fermi surface. The phase shift $\delta E$ smoothly crosses over from $\pi/2$ to 0 as $\varepsilon$ changes from $\varepsilon \ll T_K$ to $\varepsilon \gg T_K$.

It is a difficult task to experimentally observe these phase shift. As we will see, a quantum quench of the Kondo Hamiltonian by optical absorption provides a powerful tool to directly access them.

### 5.4. Kondo in optical absorption

Here we report the observation of Kondo correlations in optical absorption measurements on a single semiconductor quantum dot tunnel-coupled to a degenerate electron gas. In stark contrast to transport experiments [11–13], absorption of a single photon leads to an abrupt change in the system Hamiltonian and a quantum quench of Kondo correlations. By inferring the characteristic power law exponents from the experimental absorption lineshapes, we find a unique signature of the quench in the form of an Anderson orthogonality catastrophe [85, 86], induced by a vanishing overlap between the initial and final many-body wave-functions. We show that the power law exponent that determines the degree of orthogonality is tunable by an external magnetic field [75], which unequivocally demonstrates that the observed absorption lineshape originates from Kondo correlations. Our experiments demonstrate that optical measurements on single artificial atoms offer new perspectives on many-body phenomena previously studied exclusively using transport spectroscopy. Moreover, they initiate a new paradigm for quantum optics where many-body physics influences electric field and intensity correlations.

We present differential transmission (DT) experiments [44] on a single charge-tunable QD that reveal optical signatures of the Kondo effect [71]. In contrast to
prior experiments [5, 87], the tunnel coupling between the QD and a nearby degenerate electron gas, which we refer to as the Fermionic reservoir (FR), is engineered to be so strong that the resulting exchange interactions cannot be treated within the framework of a perturbative system-reservoir theory: In the initial state, the “system” (QD spin) is maximally entangled with the FR, forming a screened singlet. While various settings have been proposed for finding optical signatures of Kondo physics [75, 88–91], our work is most closely related to the theoretical investigation of Refs. [75, 91].

Optical spectroscopy of single quantum dots (QD) has demonstrated its potential for applications in quantum information processing, particularly in the realization of single and entangled photon sources [92, 93], coherent spin qubits [94, 95] and a spin-photon interface [96, 97]. Even though recent experiments have established this system as a new paradigm for solid-state quantum optics, all of the striking experimental observations to date could be understood within the framework of single- or few-particle physics enriched by perturbative coupling to reservoirs involving either phonons, a degenerate electron gas [87, 98], or nuclear spins [51, 99]).

The fundamentally new feature that differentiates the results we present from all prior transport based investigation of the Kondo effect [11–13], is the realization of a quantum quench of the local Hamiltonian; in our experiments, photon absorption abruptly turns the exchange interaction between the QD electron and the FR off, leading to the destruction of the correlated QD-FR singlet that otherwise acts as a local scattering potential for all FR electrons. As was shown by Anderson [85, 86], the overlap between N-electron FR states with and without a local scattering potential scales as $N^{-\alpha}$ with $\alpha > 0$. This reduced overlap, termed Anderson orthogonality catastrophe (AOC), leads to a power-law tail in absorption if the scattering potential is turned on or off by photon absorption. Here, we determine the AOC induced power-law exponents in absorption lineshape that uniquely characterize the quench of Kondo correlations. Moreover, by tuning the applied laser frequency, we observe both the perturbative and non-perturbative regimes of the Kondo effect in one absorption lineshape, without having to change the FR (electron) temperature $T_{FR}$.1

5.5. Quantum quench

As discussed above, we investigate Kondo correlations by quenching the system Hamiltonian. The general idea is that due to an event, which could be a photon absorption process, the Hamiltonian is suddenly changed, such the the generated final state is not an eigenstate of the final Hamiltonian. A prominent example are X-ray absorption spectra in metals, which exhibit strong power-law tails to blue photon detunings originating from an Anderson orthogonality catastrophe. In this section we give a qualitative picture, why a quantum quench in optical absorption can lead to a strong modification of the lineshape. For the beginning, we assume a single electron charged QD which is completely decoupled from the FR. Laser

1The AOC after a Kondo quench can, in principle, also be probed by core-level X-ray absorption spectroscopy of suitable bulk materials [100], but optical studies of QDs offer higher resolution and a tunable local Hamiltonian.
excitation leads to the formation of a trion state. Both, the initial and the final state are eigenstates of the system. This means that right after absorption, the system is in one of its eigenstates. The absorption spectrum is given by the following expression [75]:

$$A(\delta) \propto \text{Im} \int dt e^{-i\delta t} \langle \psi_i | \hat{e} e^{-i\hat{H}t} \hat{e}^\dagger | \psi_i \rangle$$

(5.15)

This is in just the Fourier transform of the correlation function $\langle \psi_f(0) | \psi_f(t) \rangle$. When the final state $\psi_f$ is an eigenstate of $\hat{H}$, the square modulus of this correlation function is constant in time. This in turn means that the absorption spectrum is then (if we neglect spontaneous emission and other broadening effects) simply given by a $\delta$-function (figure 5.6a).

Now, if the quantum dot is not isolated from the FR, but strongly coupled via exchange interaction, the situation is different. The initial state is now strongly correlated with the FR electron, whose dynamics is determined by the Kondo Hamiltonian. We can assume that the final state with two electron in a local singlet is to a large extent decoupled from the FR. The absorption process immediately projects the QD part of the system in a local singlet, while the correlations in the FR still exist.

In the case of the $X^-$ transition, the physics is well described by the excitonic Anderson Hamiltonian (section 5.1.3). However the parameters are different in the initial and the final state. The important thing is that the QD contains only a single electron in the ground state, which is strongly correlated with the FR electrons. The final state contains two electron which are to a large extend decoupled from the FR. The absorption of a photon leads to an immediate charge in the parameters of the Hamiltonian, such that the final state just after the absorption process is not an eigenstate of the final Hamiltonian. If we let the system evolve, the final state will relax into an eigenstate of the final Hamiltonian. Due to the time dynamics, the
Figure 5.7.: A single quantum dot strongly coupled to a Fermionic reservoir. a, Band structure of the device. The QDs are separated by a 15 nm tunnel barrier from a n++-doped GaAs layer (Fermi sea). A voltage $V_g$ applied between the electron gas and a semi-transparent NiCr gate on the sample surface controls the relative value of the QD single-particle energy levels with respect to the Fermi energy $E_F$. b, Low temperature (4 K) photoluminescence spectrum of a single QD, dot 1, as a function of $V_g$. The interaction of the QD electron with the Fermi sea leads to a broadening of the photoluminescence lines at the plateau edges (yellow arrows) and indirect recombinations of a QD hole and a Fermi sea electron (red arrow). Indirect transitions are identified by the stronger $V_g$ dependence of the transition energy compared to direct transitions. A detailed discussion of the origin of various photoluminescence lines can be found in Ref. [87].

spectrum is not a $\delta$-function any more; the absorption is now distributed over a range of frequencies.

In the case of absorption from a Kondo state into a non-Kondo state the situation is even more interesting: it turn out that at low energies, which corresponds to small laser detunings, the FR’s in the initial and final state are orthogonal. This is analogous to X-ray absorption spectra in metals where the sudden generation of a core-hole and an electron on top of the Fermi sea cause shake-up processes which lead to a similar orthogonality catastrophe.

Another way to think about the modification of the absorption spectrum is that when a scattering potential is generated or destroyed, the system evolves for $t \to \infty$ to an orthogonal state, with no matrix element to the initial state. This leads to a suppression of the absorption at the threshold frequency: for a step-function, the absorption is suppressed at the onset and a $\delta$-singularity is transformed into a weaker kind of singularity, a power-law singularity.

5.6. Experimental setup

All experiments were carried out in a dilution refrigerator with a base temperature of 8 mK in the mixing chamber. From fits to experimental data, we find, that the (relevant) electron temperature is around 180 mK (see section 5.8.4). The schematic of the QD sample we study is shown in Figure 5.7a: a gate voltage $V_g$ applied between a top Schottky gate and the degenerate electron gas allows us to
tune the charging state of the QD \[20\]. Figure 5.7b shows the photoluminescence (PL) spectrum of a particular QD (dot 1), as a function of \(V_g\), where different discrete charging plateaux are clearly observable. The dependence of the PL energy on the QD charging state originates from a Coulomb renormalization of the optical transition energy. In addition to PL lines (e.g. \(X^0\)) associated with a fixed charging state (e.g. neutral) of the QD, we also observe spatially indirect transitions with a strong dependence on \(V_g\) (see Figure 5.7b, red arrow) \[87\]. In this chapter we focus on the \(X^-\) plateau, for which the QD is single-electron charged and the influence of the FR on the QD PL dispersion and linewidth is strongest. The \(X^-\) optical transition couples the initial configuration, containing on average one electron in the QD, to a final configuration, containing on average two electrons and a valence-band hole (a negatively charged trion).

This transition can be described within the framework of an excitonic Anderson model (EAM) \[75, 91\], depicted schematically in Figure 5.8c (and described explicitly in section 5.1.3). It is parametrized by the energy \(\varepsilon\) of the QD electron level with respect to the Fermi level, the on-site Coulomb repulsion \(U_{ee}\), the tunnel rate \(\Gamma\) between QD and FR, the half-bandwidth \(D\) of the FR, and the electron-hole Coulomb attraction \(U_{eh}\). The latter is relevant only in the final configuration, where it effectively lowers the electron level energy to \(\varepsilon - U_{eh}\), thus ensuring the double occupancy of the electron level in the final configuration. A Hartree-Fock estimate from the PL data in Figure 5.7b yields \(U_{eh} \approx U_{ee} + 4\) meV.

### 5.7. Renormalization of the transition energy

The inset of figure 5.8a shows high resolution laser absorption spectroscopy on dot 1 across the \(X^-\) single electron charging plateau. Here, we parametrize \(V_g\) in terms of \(\varepsilon\), normalized and shifted such that \(\varepsilon = -\frac{1}{2}U_{ee}\) for the gate voltage where the absorption contrast is maximal. Instead of the usual linear DC-Stark shift of the absorption peak that is characteristic of charge-tunable QDs, we find a strongly non-linear \(\varepsilon\)-dependent shift of the \(X^-\) transition energy\[87, 98\], which measures the energy difference between the final and initial ground states.

As depicted in Fig. 5.8c and, this energy shift arises from a renormalization of the initial state energy \[74\] due to virtual tunneling between the singly-occupied QD and FR (analogous to the Lamb shift of atomic ground states). The initial configuration features a single electron in the QD, whose energy is lowered by virtual tunneling between QD and FR. Since virtual excitations with energy \(\Delta E\) contribute a shift proportional to \(-\Gamma/\Delta E\), the total shift (involving a sum over all possible \(\Delta E\)), is strongest near the edges of the \(X^-\) plateau. Toward the right edge (\(\varepsilon\) near 0), the dominant contribution comes from virtual tunneling of the QD electron into the FR (as depicted); toward the left edge (\(\varepsilon\) near \(-U_{ee}\)), it comes from virtual tunneling of a FR electron into the QD (not depicted). This renormalization-induced red-shift of the initial state is strongest at the plateau edges and leads to an \(\varepsilon\)-dependent blue-shift of the optical resonance frequency.

The final trion state energy, on the other hand, is hardly affected by virtual tunneling processes, due to large \(U_{eh} - U_{ee}\). In the final configuration, the QD contains two electrons and a hole. The electron-hole Coulomb attraction \(U_{eh}\) effectively low-
Figure 5.8.: The gate voltage dependence of the peak absorption strength of the negatively charged exciton $X^−$ for dot 1, measured at 180 mK. a, Inset: absorption as a function of the gate voltage. Main figure: Experimental data (symbols) for the $\varepsilon$-dependence of the shift in the resonance energy $\Delta E_{\text{transition}}$ (blue, left axis) and the absorption contrast (red, right axis) are well reproduced by NRG calculations (solid lines) for the following parameters: $U_{ee} = 7.5$ meV, $\Gamma = 0.7$ meV, $D = 3.5$ meV, $U_{eh} = 11$ meV, $T_{FR} = 180$ mK. b, Lower panel: NRG results for the occupancy of the QD electron level in the initial and final ground states. c, Schematic of the energy renormalization process (see text for details).

ers the QD electron level energy to $\varepsilon - U_{eh}$. This raises the energy costs $\Delta E$ for virtual excitations by $U_{eh} - U_{ee}$ (which is $\gg \Gamma$), so that final state energy renormalization is negligible. The renormalization of the transition energy, probed by a weak laser, is thus mainly due to initial state energy renormalization.

A perturbative expression for this self-energy corrections $\Sigma_{X^−}^i$ of the initial state can be found for the local moment regime:

$$\Sigma_{X^−}^i = \sum_k V^2 \left( \frac{1}{\varepsilon_k - \varepsilon} + \frac{1}{\varepsilon + U_{ee} - \varepsilon_k} \right). \quad (5.16)$$

However, due to the strong tunnel coupling in this sample, expression 5.16 cannot be applied here: the occupation of the initial state not close to one for most gate voltages. We therefore use a different approach to determine the EAM parameters.

To obtain the values of $\Gamma$, $U_{ee}$ and $D$, we fit the numerical predictions for the gate-voltage dependence of the transition energy $\Delta E_{\text{transition}}$ to the experimental data (Figure 5.8a, blue symbols, which give the frequency where the absorption spectrum reaches its maximum).

Within our model the transition energy $E_{\text{transition}} = \omega_{\text{th}} + E_{\text{Stark}}$ is given by the sum of the threshold frequency $\omega_{\text{th}} = E_{G}^f - E_{G}^i$ and a linear Stark shift, $E_{\text{Stark}} \propto \varepsilon$.

The threshold frequency $\omega_{\text{th}}$ is obtained numerically by simply calculating the ground state energies $E_{G}^f$ and $E_{G}^i$ of the two Anderson Hamiltonians $H^i$ and $H^f$ specified around Eq. (5.8), taking care to incorporate the gate-voltage dependence of the electron and hole levels, according to Eq. (5.7). When comparing experimental data with NRG-results, $E_{\text{Stark}}$ is treated as fit parameter, together with $\Gamma$, $D$ and $U_{ee}$.
Since only relative changes in gate voltage have physical relevance, the horizontal offset of the experimental data is chosen such that the value of $V_g$ for which the absorption contrast is strongest corresponds to the specific level-position $\varepsilon = \varepsilon_{\text{ee}}$ given by $\varepsilon = -U/2$.

From this procedure, we find that the following parameters for the EAM give excellent agreement with the experimental data: $U_{ee} = 7.5$ meV, $\Gamma = 0.7$ meV, and $D = 3.5$ meV.

### 5.8. Absorption lineshape

The energy renormalization of the transition energy allows us to determine the relevant parameters in our structure, but it does not let us draw conclusion about Kondo correlations in the initial state prior to absorption. Figures 5.9a-d show vertical cuts of the 2D plot from figure 5.8. In contrast to standard QD sample, the absorption all lineshapes are asymmetric with tails to blue laser detunings which becomes more prominent as one approaches the plateau edge. The peak contrast on the other hand deceases rapidly. Both are signatures of the strong exchange coupling to the FR as we will discuss on the following pages.

#### 5.8.1. Origin of the asymmetry in the absorption

As shown before, all the measured lineshapes are asymmetric with tails to blue laser detunings. The modification of the lineshape is related to the coupling of the QD electron and the FR. In fact the absorption at a detuning $\delta$ probes the structure of the FR as we will see.

We first give a intuitive picture of the origin of the tails. Consider the following experimental situation as depicted in figure 5.10a: The quantum dot is charged with a single electron and we apply a laser which is blue detuned with respect to the trion transition energy (the laser photon energy is larger than the trion transition energy). If the QD electron was completely decoupled from the Fermi reservoir, the absorption lineshape would be Lorentzian and suppressed at detuning $\delta$ by $1/\delta^2$. Since this is not the case in our sample, one can think of an absorption process, taking place in two-steps. First, the electron is excited to a virtual level, which is detuned by $\delta$ (figure 5.10b). Due to exchange interaction with the FR, this “excess” energy can be dumped into the FR by the generation of a particle-hole pair with an energy $\delta$(figure 5.10c). For simplicity, we assume for the moment that the
temperature $T$ of the FR is zero. For a two-dimensional density of states of the FR, the bandwidth of FR electrons which can contribute to excitation is proportional to the laser detuning $\delta$: electrons with an energy below $E_F - \delta$ cannot be excited since there are no available empty states below $E_f$. Therefore only electrons, which have an energy within a band $E_F - \delta$ and $E_F$ contribute. In a perturbative description this leads to a detuning dependent dephasing rate, where the exchange coupling contributes an additional phase-space factor $\propto \delta$ to the blue tail of the absorption lineshape.

In the case of a red detuned laser, the situation is different: for an absorption process to take place the missing energy $\delta$ has to be provided by the annihilation of a particle-hole pair in the FR. Since we assumed $T = 0$ this process does not exist. However, the simple explanation of a blue tail with a $1/\delta$ dependence in the presence of exchange interaction is not completely true. Even if the FR temperature is larger than the Kondo temperature, there is a small but finite probability to find the system in a strongly correlated Kondo state. In fact one can derive an analytical formula for a perturbative ($T_K \gg T$) lineshape for the local moment regime (when the QD contains exactly one electron) which is valid even at finite temperature. This lineshape is given by the following expression\cite{75}:

$$A(\delta) = \frac{3}{4} \frac{\delta/T}{1 - e^{-\delta/T}} \frac{\gamma_{\text{Korr}}/2\pi}{\delta^2 + \gamma_{\text{Korr}}^2/4}$$

(5.17)

with the Korringa relaxation rate $\gamma_{\text{Korr}} = 2\pi T/\ln^2(\max(|\delta|, T)/T_K)$. As one can see, Eqn. 5.17 is a lineshape which is Lorentzian when $|\delta| < T$. In this regime, the number of particle hole pairs which can be annihilated is equal to the number of particle-hole pairs which can be generated: This is a simple coupling to a thermal reservoir. For blue detunings $\delta > T$, the prefactor of the Lorentzian accounts for the absorption of the excess laser energy by the FR. However this will not lead to an exact $1/\delta$ tail, due to the logarithmic correction originating from the Korringa relaxation rate.

For a red detuned laser $\delta < T$ the number of particle-hole excitations which could provide energy for the absorption process decreases exponentially with increasing
|δ|. As can be seen from Eqn. 5.17, the red tails have an exponential dependence on the laser detuning an are sensitive to the temperature. In fact, we use the red tails to determine the temperature of the FR as explained in section 5.8.4.

We now consider the detailed form of the absorption lineshape \( A(\delta) \) as function of the detuning \( \delta \) between the applied laser frequency and the transition threshold: Figure 5.11a shows, on a log-log scale, the blue (\( \delta > 0 \)) tail of \( A(\delta) \) for dot 1, for the four values of gate voltage indicated by arrows in the inset of Fig. 5.8a. The inset to Fig. 5.11a compares the full unnormalized absorption lineshapes for the identical gate voltages in linear scale; the red absorption tail allows us to determine the temperature of the FR as \( T_{FR} = 180 \text{ mK} = 15.6 \mu\text{eV} \) (see section 5.8.2). The strong variation of the peak absorption strength and width in Fig. 5.11a inset is a consequence of the exponential dependence of the Kondo temperature 

\[
T_K(\varepsilon) = \sqrt{\Gamma D e^{-[1-(2\varepsilon/U_{ee}+1)^2][\pi U_{ee}/8\Gamma]}}
\]

(5.18)
on the gate voltage \( \varepsilon \). For this QD, \( T_K \) varies between 24\mu eV and 464\mu eV. All lineshapes carry the signatures of an optical interference effect induced by the sample structure (causing some lineshapes to become negative for small red detunings), and of independently measured fluctuations in gate voltage; both effects have been taken into account in the calculated lineshapes. Calculating the lineshapes by NRG (solid lines) without any further fit parameters, we find remarkable agreement with experiment for all four lineshapes depicted in Fig. 5.11a, demonstrating the validity of the EAM \[75\] for the coupled QD-FR system.

5.8.2. Influence of optical interference on measured lineshapes

In order to compare the results of numerical calculations with experimental data, we calculated interference-modified versions of the former, by proceeding as follows: state-of-the-art full density matrix (FDM) numerical renormalization group (NRG) techniques for calculating (non-equilibrium) spectral functions \( A_{NRG}(\delta) \) give us the absorptive part of the susceptibility, \( \chi_{NRG}'(\delta) \equiv A_{NRG}(\delta) \). From this we calculate the dispersive part using the Kramers-Kröning relation,

\[
\chi_{NRG}(\delta) = -P \int \frac{d\delta'}{\pi} \frac{\chi_{NRG}''(\delta')}{\delta - \delta'}.
\]

(5.19)

Inserting \( \chi_{NRG}(\delta) = [\chi_{NRG}'(\delta) + i\chi_{NRG}''(\delta)] \) into the expression for the detected signal \( A(\delta) = \text{Re}(i\chi(\delta)(1 + r e^{i\phi})) \) we obtain the interference-corrected prediction for the absorption lineshape, \( A_{\text{int}}(\delta) \).

In order to clarify the role of optical interference on the power-law exponents associated with the Kondo correlations, we compare the NRG results of figure 5.11b from the main text (dashed lines), which take into account interference, to their counterparts in the absence of optical interference (solid lines).

5.8.3. Gate voltage fluctuations

In contrast to conventional quantum dot samples, even small fluctuations \( \delta\varepsilon \) in the effective applied gate voltage \( \varepsilon \), originating from voltage fluctuations at the output

\(^2T_K\) looses significance for the black curve, for which the QD-FR system is in the mixed-valence regime.
Figure 5.11.: The absorption lineshape $A(\delta)$ for dot 1.  

- **a.** The blue tail of $A(\delta)/A(\delta_{\text{max}})$, plotted versus the laser detuning $\delta$ on a log-log scale. The experimental data were taken at an electron temperature of $T_{\text{FR}} = 180$ mK for the four values of gate voltage ($\varepsilon$) indicated by arrows in Fig. 2a; the corresponding Kondo temperatures $T_K(\varepsilon)$ are indicated by vertical lines in matching colors. NRG results (solid lines), obtained using the parameters from the fit in Fig. 5.8a, are in remarkable agreement with experiment. Inset: the measured full (unnormalized) absorption lineshape in linear scale for the identical $\varepsilon$ values.

- **b.** NRG results for $T = T_{\text{FR}}$ (solid lines) and $T = 0$ (dashed lines); the latter show the $\delta^{-0.5}$ behaviour expected in the strong-coupling regime, $T \ll \delta \ll T_K$.

- **c.** The rescaled lineshape $A(\delta)/A(T_K)$ versus $\delta/T_K$ shows a universal scaling collapse characteristic of Kondo physics.

The absorption lineshape $A(\delta)$ for dot 1, plotted versus the laser detuning $\delta$ on a log-log scale. The experimental data were taken at an electron temperature of $T_{\text{FR}} = 180$ mK for the four values of gate voltage ($\varepsilon$) indicated by arrows in Fig. 2a; the corresponding Kondo temperatures $T_K(\varepsilon)$ are indicated by vertical lines in matching colors. NRG results (solid lines), obtained using the parameters from the fit in Fig. 5.8a, are in remarkable agreement with experiment. Inset: the measured full (unnormalized) absorption lineshape in linear scale for the identical $\varepsilon$ values.

- **b.** NRG results for $T = T_{\text{FR}}$ (solid lines) and $T = 0$ (dashed lines); the latter show the $\delta^{-0.5}$ behaviour expected in the strong-coupling regime, $T \ll \delta \ll T_K$.

- **c.** The rescaled lineshape $A(\delta)/A(T_K)$ versus $\delta/T_K$ shows a universal scaling collapse characteristic of Kondo physics.

of the function generator and charge fluctuations in the QD environment, have an impact on the absorption lineshapes due to the strong non-linear dependence of the transition energy on the gate voltage. In the plateau center, where a small change $\delta \varepsilon$ in gate voltage corresponds to a negligible change in transition energy, fluctuations do not affect the lineshape. However, at the plateau edges, even small fluctuations $\delta \varepsilon$ cause measurable energy shifts of the threshold frequency $\omega_{\text{th}}(\varepsilon)$, and hence of the detuning $\delta = \omega - \omega_{\text{th}}(\varepsilon)$ of the laser (with frequency $\omega$) from the QD resonance. After having determined the temperature from the red tail of the lineshape in the plateau center (see section 5.8.4), we find that the lineshapes at the plateau edges are modified by gate voltage fluctuations. We take these fluctuations into account phenomenologically by convoluting the interference-corrected lineshape $A_{\text{int}}(\delta)$ with a Gaussian,

$$A_{\text{predicted}}(\delta) = \int dx A_{\text{int}}(\delta + x)p(x, \varepsilon), \quad p(x, \varepsilon) = \frac{e^{-(x/\sigma(\varepsilon))^2/2}}{\sqrt{2\pi}\sigma(\varepsilon)},$$  \hspace{1cm} (5.20)
with a gate-voltage dependent width $\sigma(\varepsilon)$. Here $\sigma(\varepsilon) = \langle [\omega_{th}(\varepsilon + \delta \varepsilon) - \omega_{th}(\varepsilon)]^2 \rangle^{1/2}$ represents the standard deviation of the fluctuations around the mean threshold frequency $\omega_{th}(\varepsilon)$, under fluctuations $\delta \varepsilon$ arising from fluctuations in the gate voltage, $\delta \varepsilon \sim \delta V_g$. The fluctuations $\delta V_g$ are assumed to Gaussian distributed, with a standard deviation of 10$\mu$V. These fluctuations predominantly alter the red tail of the absorption resonance away from the plateau center. We emphasize that our approach accounts only for fluctuations in the threshold frequency; it neglects the changes in the actual line shape induced by the fluctuations in $\varepsilon$. $A_{\text{predicted}}$ is the quantity plotted in the main text when comparing “NRG results for lineshape $A(\delta)$” to data.

### 5.8.4. Electron Temperature

A difficulty in the experiments is to determine the temperature of the electron gas during the measurements, which can differ substantially from the temperature of the sample holder. We extract the electron gas temperature from the optical lineshapes. For red detunings, such that $\delta < -T$, the number of electron-hole pairs that could provide the energy necessary for FR-assisted laser absorption scales exponentially with $\delta$ due to Fermi statistics: as a consequence, the absorption lineshape shows an exponential tail (see Eq. (7) of [75]), whose slope gives us the electron temperature $T$. For very large red detunings, the dominant line broadening is due to spontaneous emission and we recover the associated Lorentzian tail. To determine $T$, we actually fit the NRG lineshape for red detunings, taking into account optical interference (as described in Section 2.5.2 below).

### 5.8.5. Scaling collapse

For blue detunings satisfying $\delta > \max(T_{FR}, T_K)$, a perturbative description for $A(\delta)$ is possible. Both the frequency scale for which the perturbative $\sim \delta^{-1}$ dependence
Signatures of Kondo correlations in optical absorption

Figure 5.11a sets in and the peak absorption contrast itself, strongly depend on gate voltage. Remarkably, for gate voltages such that the initial ground state is a Kondo singlet, this dependence is such that it permits a scaling collapse: Figure 5.11c shows the normalized absorption lineshape $A(\delta)/A(T_K(\varepsilon))$ as a function of $\delta/T_K$, for the red, green and blue curves of Figure 5.11a (but omitting the black curve, which is in the mixed valence regime). We obtain a remarkable collapse of all three curves onto a universal scaling function of $\delta/T_K$, as expected [75] for the regime $T_{FR} \ll \delta \ll U_{ee}$. Thus, the $\varepsilon$-dependence of the crossover scale is captured by Eq. (5.18) for $T_K$; this observation is unequivocal proof that the Kondo effect is indeed present in our system.

5.8.6. Strong-coupling Kondo regime.

In the limit $T_{FR} < \delta < T_K$, a perturbative description of the lineshape is no longer valid. In the initial configuration, the exchange interaction between QD and FR induces a Kondo screening cloud that forms a singlet with the QD spin. This acts as a scattering potential that induces strong phase shifts for those low-energy Fermionic excitations whose energies are within $T_K$ from the Fermi level. In the final configuration after photon absorption, the QD has two electrons in a local singlet state. Therefore the Kondo screening cloud, and the scattering potential for FR electrons constituted by it, disappear in the long time limit: the corresponding ground state wave-function is a tensor product of the local singlet and free electronic states, with only weak phase shifts. Since the initial and final FR phase shifts differ (as depicted schematically in figure 5.5d), the FR does not remain a spectator during the $X^-$ transition: instead, the transition matrix element between the ground states of the initial and final configurations is vanishingly small. This leads to an AOC which manifests itself by transforming a delta-function resonance (of an uncoupled QD) into a power-law singularity [85] of the form $\delta^{-\eta}$, where the exponent $\eta$ characterizes the extent of AOC. For $T_{FR} \ll \delta \ll T_K$, the absorption lineshape of the $X^-$ transition is expected to show an analogous power-law singularity. The exponent $\eta$ is predicted [75, 91] to range between 0 and 0.5 (assuming no magnetic field), with $\eta \approx 0.5$ being characteristic for a Kondo-correlated initial state and an uncorrelated final state. This lineshape modification is a consequence of a redistribution of the optical oscillator strength, associated with the fact that the FR wave-function in the Kondo-correlated initial state has finite overlap with a range of final states consisting of electron-hole pair excitations out of a non-interacting FR.

If $T_{FR} \ll T_K$ and the optical detuning is reduced below $T_K$, the lineshape is predicted to smoothly cross over from the perturbative $1/\delta$ tail to the strong-coupling $1/\delta^{0.5}$ power law just discussed. This crossover is illustrated in Figure 5.11b (dashed lines) by NRG calculations, performed at $T_{FR} = 0$ for the three $\varepsilon$-values of Figure 5.11c: Remarkably, despite drastic differences in the $\delta > T_K$ tails due to different values of $T_K(\varepsilon)$, all three lineshapes show similar power-law exponents around $\eta \approx 0.5$ for $\delta \ll T_K$. For non-zero temperature, however, the $1/\delta^{0.5}$ power law is cut

While the actual frequency dependence in the perturbative regime has logarithmic corrections, these are masked by the optical interference effect, see Section 5.8.2.  

Deviations from scaling for $\delta < T_{FR}$ are expected, but masked by an insufficiently small signal-to-noise ratio of the experimental data.
It is instructive to gauge the effect of Kondo correlations in the measured lineshapes by comparing them to cases for which the Kondo effect is absent, so that the absorption lineshape lends itself to a perturbative description (based on the Anderson model). To show how the lineshapes differ in the absence of Kondo correlations, we here present a comparison of the $X^-$ line of dot 2 (used to obtain the lineshapes depicted in Fig. 4 of the main text), and another dot (referred to as dot 3) from the same sample. Dot 3 has a trion emission wavelength of 955 nm (as opposed to ~ 900 nm for dots 1 and 2); we observe that the dots emitting at this wavelength have much sharper lines, indicating weak coupling to the FR; this is a consequence of lower conduction band electron energy, which in turn increases the effective tunnel barrier to the FR.

Figure 5.13c shows that the lineshape of this weakly coupled dot 3 can be perfectly fit using modified version of Eqn. 5.17:

$$A_{\sigma}^{LM}(\delta) = \frac{3\pi}{4} \frac{\delta/T}{1 - e^{-\delta/T}} \frac{\gamma/(2\pi)}{\delta^2 + \gamma^2/4},$$

(5.21)

where $\gamma < T$ is a phenomenological relaxation rate. This line shape is a simplified version of the one derived in Ref. [90], where $\gamma$ was given by the scale-dependent Korringa relaxation rate $\gamma_{Kor}(\delta, T) = 2\pi T/\ln^2[\max(|\delta|, T)/T_K]$. As was argued in [75], this lineshape is strictly valid only in the limit $\max(|\delta|, T) \gg T_K$. Instead of the scale-dependent Korringa relaxation rate, we choose to use a constant $\gamma$ in our fits. We do however, take into account that the relaxation stems from the Anderson model, which restricts $\gamma < T$ (for $B = 0$) and introduces the $\frac{\delta/T}{1 - e^{-\delta/T}}$ factor, that accounts for the asymmetry between the FR electron-hole pair generation and annihilation processes.

In contrast to dot 3, an attempt to use Eqn. (5.21) with $\gamma \leq T$ to fit the lineshape of the strongly coupled dot 2 at $B_{\text{ext}} = 0$ fails dramatically, as show in figure. 5.13a.
Interestingly, the situation is different for $B_{\text{ext}} = 2$ Tesla, where the absorption lineshape of dot 2 can be fitted reasonably well with Eqn. (5.21), as shown in figure 5.13b. The reason for this striking change is related to the onset of the oscillations in peak contrast observed for the blue transition in the inset of figure 5.14a: as mentioned in the main text, a magnetic field exceeding 1 Tesla causes oscillations in the FR density of states, $\rho_{\text{FR}}$. The Kondo temperature, which depends exponentially on $\rho_{\text{FR}}$, will thus experience strong oscillations as well, which explains the observed strong oscillations in the peak contrast. Thus, the maximum in peak contrast observed at $B_{\text{ext}} = 2$ Tesla corresponds to a minimum for $T_K$, i.e. at $B_{\text{ext}} = 2$ Tesla we have, in effect, a weakly coupled dot, explaining why its lineshape can we fit reasonably well with the perturbative Eq. (5.21).

In summary, the striking difference seen in figure 5.13a and 5.13b between the $B_{\text{ext}} = 0$ and 2 Tesla lineshapes of dot 2 confirms our interpretation that for $B_{\text{ext}} = 0$, Kondo correlations are at the heart of the observed lineshapes.

### 5.8.7. Magnetic field-tuning of Kondo correlations

A direct extraction of the $1/\delta^{0.5}$ power law from the measured data is difficult due to the small accessible experimental window $T_{\text{FR}} < \delta < T_K$. Nevertheless, we are able to determine the power-law exponent corresponding to our data accurately by using the fact that the detailed form of the lineshape sensitively depends on the exponent $\eta$ which can be tuned by an external magnetic field[75]. Figure 5.14 shows the magnetic field dependence of the absorption lineshape for a second QD, dot 2, with parameters $U_{ee} = 7.5$ meV, $\Gamma = 1$ meV, $D = 6.5$ meV and $U_{eh} = (3/2)U_{ee}$, measured at $\varepsilon/U_{ee} = -0.43$ (where $T_K = 140 \mu$eV) and $T_{\text{FR}} = 15.6 \mu$eV. Measurements were carried out in Faraday geometry, where QD optical selection rules [96] ensure that if we choose right (left) hand circular polarization of the laser field, we can selectively probe the blue (red) trion transition that couples exclusively to the spin-up (spindown) initial state.

In comparison to the $B_{\text{ext}} = 0$ Tesla (black squares) results, the absorption lineshapes for the blue (blue dots) and red (red triangles) trion transitions at $B_{\text{ext}} = 1$ Tesla exhibit two striking features (Figure 5.14a): (i) The peak contrast increases (decreases) by a factor $\simeq 2$. (ii) The area under the blue (red) absorption curve increases (decreases) by less than 20%. These observations indicate that the change in the $B_{\text{ext}} \leq 1.5$ Tesla lineshapes is predominantly due to a line narrowing (broadening), associated with an increase (decrease) of the AOC power-law exponent $\eta$ of the blue (red) trion transition. To quantify the field-induced change in $\eta$, we plot in figure 5.14b the normalized lineshapes shown in figure 5.14a in a log-log plot, together with the corresponding NRG results (solid lines): the latter yield $\eta$ values of 0.5 at zero field, and 0.31 (red trion) and 0.66 (blue trion) at $B_{\text{ext}} = 1$ Tesla, proving the remarkable sensitivity of the measured lineshapes to the AOC determined power-law exponents. We emphasize that qualitatively similar features are observed for all field values $B \leq 1.5$ Tesla; for $B_{\text{ext}} > 1.5$ Tesla, the blue trion contrast exhibits oscillations (inset of figure 5.14a), most likely stemming from the modification of the FR density of states at high fields in Faraday geometry.
5.8. Absorption lineshape

Figure 5.14.: Magnetic field dependence of the absorption. a, The absorption line-shapes for a second QD (dot 2) with similar parameters (see text), for $\varepsilon/U_{ee} = -0.43$ at $B_{\text{ext}} = 0$ Tesla and $B_{\text{ext}} = 1$ Tesla for the blue/red trion transition. The magnetic field changes the strength of the AOC and the lineshape. The small peak appearing at $\delta \sim 80 \mu eV$ in the red trion absorption is due to the incomplete suppression of the laser polarization that couples to the blue trion transition. Inset: the peak absorption contrast showing good agreement with the NRG calculations for $B \leq 1.5$ Tesla. b, The normalized absorption lineshape for dot 2 in a log-log plot. These measurements pin the value of $\eta(B_{\text{ext}} = 0)$ to $\sim 0.5$, which is a direct signature of a Kondo singlet in the absorption lineshape. In addition they demonstrate the tunability of an orthogonality exponent.

Magnetization and lineshapes at finite magnetic field

The area under the (unnormalized) absorption lineshape is proportional to the initial occupancy $n_\uparrow (n_\downarrow)$ of the spin-up (spin-down) state. The small ($\lesssim 20\%$) field-induced change in the measured areas of Figure 5.14a, implies a small magnetization $m = \frac{1}{2}(n_\uparrow - n_\downarrow) \simeq 0.16$. In contrast, the corresponding magnetization for a free spin would have been $m = 0.40$. This measurement confirms that the static spin susceptibility of the initial configuration is substantially reduced relative to that of a free spin, providing yet another optical signature of the Kondo screening.

One of the most striking ways to identify the presence of Kondo correlations is to study how they are reduced by applying an external magnetic field. In this section, we outline what is expected to happen in some detail, based on NRG calculations and analytical arguments. We also show data that nicely illustrates the difference between dots with or without Kondo correlations.

In the presence of an external magnetic field $B_{\text{ext}}$ applied along the growth axis of the heterostructure (Faraday configuration), the electron and hole level energies will be Zeeman-shifted according to ($a = i, f$):

$$
\varepsilon_e^a = \varepsilon_e^a + \frac{1}{2} \sigma g_e \mu_B B_{\text{ext}} = \varepsilon_e^a - \frac{1}{2} \sigma B ,
$$

$$
\varepsilon_h^a = \varepsilon_h + \frac{1}{2} \sigma g_h \mu_B B_{\text{ext}} = \varepsilon_h + \frac{1}{2g_e} \sigma B ,
$$

where $g_e \simeq -0.6$ and $g_h \simeq 1.1$ for our dots, and we have defined $B = \mu_B |g_e| B_{\text{ext}}$. (5.22) (5.23)
Signatures of Kondo correlations in optical absorption

**Figure 5.15.** a. The magnetization \( m(B, T) \) of a localized spin coupled to a FR, with Zeeman energy \( H_{\text{Zeeman}} = -BS_z \), plotted as a function of \( B/T \) for several values of \( T_K \). All curves were obtained by NRG for the excitonic Anderson model of section 5.1.3. To mimic the initial configuration of dot 2 of the main text, we used \( n_i = 0, \Gamma = 1 \text{ meV}, D = 6.5 \text{ meV}, U_{ee} = 7.5 \text{ meV}, \varepsilon = -0.43U_{ee} \) (such that \( n^i_e \simeq 1 \) and \( T_K^{\text{exp}} = 140 \mu\text{eV} \)), and \( T = T_{\text{FR}} = 15.6 \mu\text{eV} \). Thus, the black curve with \( T_K = T_K^{\text{exp}} \) corresponds to the experimental situation of dot 2. For the other curves, we kept all parameters fixed except \( \Gamma \), which was decreased to yield smaller values of \( T_K \). b. The experimental magnetization of dot 2 at \( B = 1 \text{ Tesla} \), determined by directly integrating the area under the measured blue and red trion absorption lineshapes. For comparison, the predictions of NRG for the ”full area” as well as ”limited area” (i.e. integration of the area up to \( \delta_{\text{max}} = 3T_K \), as was done for the experimental points) cases are shown, along with a case where \( T_K \) is vanishingly small.

Creation of a trion, described by \( H_L \propto (\epsilon^e_+ h^e_+ e^{-i\omega_L t} + \text{h.c.}) \), can be induced in a spin-selective way [96]: by choosing right (left) circularly polarized laser field, one can exclusively couple to the state with initial electron spin-up \( \sigma = + \) (spin-down \( \sigma = - \)). For \( B > 0 \) (assumed henceforth), the spin-up (spin-down) electron has lower (higher) energy and the corresponding optical transitions are conventionally refereed to as blue (red) trion, with lineshapes \( A_+(\delta) \) (\( A_- (\delta) \)), since the \( B \)-induced shift in threshold frequency \( \omega_{\text{th}}^e (B) = E^e_G - E^f_G \) is positive (negative), as elaborated below.

We will discuss the effect of \( B \neq 0 \) on (i) the initial e-level magnetization \( m_i^e (B, T) = \frac{1}{2}(n_{e+,i} - n_{e-,i}) \), (ii) the absorption lineshape \( A_{\pm} (\delta) \), and (iii) the threshold frequency \( \omega_{\text{th}}^e (B) \), all for the \( X^- \) transition. Our discussion is very similar to that of Ref. [75], which analyzes analogous questions for the \( X^0 \) transition.

The magnetization \( m(B, T) \) of a free, localized spin, at finite temperature \( T \), evolves in a magnetic field with Zeeman energy \( H_{\text{Zeeman}} = -BS_z \) according to \( m_{\text{free}} = \frac{1}{2} \tanh(B/2T) \). Thus, \( m(B, T) \) crosses over from 0 to \( \frac{1}{2} \) on the scale \( B \gtrsim T \). In contrast, if the spin is exchanged-coupled to a FR, and the temperature is low enough that Kondo-screening occurs \( (T \ll T_K) \), the crossover scale increases to \( |B| \simeq T_K \). Thus, the magnetization for a Kondo-screened spin evolves much slower
with applied field than it does for a free spin, as illustrated in Fig. 5.15a. The reason is that Kondo screening of the local spin by the FR causes a strong reduction in the static spin susceptibility.

Since the area under the absorption curve $A_\sigma(\delta)$,

$$W_\sigma = \int_{-\infty}^{\infty} d\delta A_\sigma(\delta),$$  \hfill (5.24)

is proportional to $(1 - n_{ie})$ [by equation 5.12], the initial magnetization can be extracted from the normalized difference in areas under the blue and red shapes:

$$m^i_{ie}(B) = \frac{1}{2}(n^{i+}_{e} - n^{i-}_{e}) = \frac{W_+(B) - W_-(B)}{2(W_+(0) + W_-(0))}. \hfill (5.25)$$

The second equality assumes that the total initial occupancy is one, $n^{i+}_{e} + n^{i-}_{e} = 1$, which, to very good approximation, is the case for dot 2 of figure 5.14. We have confirmed numerically that $W_+(B) + W_-(B) = W_+(0) + W_-(0)$, even in the presence of optical interference induced modification of the absorption lineshape.

In practice, the determination of $m^i_{ie}$ from experimental lineshapes using Eq. (5.25) is complicated by (a) the difficulty in ensuring perfect circular polarization of the incident laser field, (b) the low signal-to-noise ratio for the red trion transition due to its enhanced broadening (arising from a magnetic-field induced reduction in its power-law exponent), (c) the low signal-to-noise ratio in the tails of the absorption line-shape which have a sizable contribution to the area, and (d) the modification of the FR density of states for $B > 1$ Tesla. Despite these complications, the results shown in figure 5.15 demonstrate unambiguously that the crossover scale for the initial magnetization to change significantly from 0 is not $B \simeq T$ but $B \simeq T_K$, implying that the initial state of dot 2 is a screened Kondo singlet. To determine the magnetization from the area of the experimental line shapes, we have integrated the lineshapes in the interval $-3T_K \leq \delta \leq 3T_K$ and focused on $B = 1$ Tesla where the modification of the FR density of states is still negligible. The dominant contributions to the error bar comes from the fact that with the limited integral, we find that the sum of the area under the blue and red trion transitions is 1.8 (instead of 2.0) times the area under the $B = 0$ lineshape. In addition, the additional peak visible at high frequency tail of the red trion transition depicted in figure 5.14a suggests that the suppression of the orthogonal polarization is incomplete: these two factors give rise to the error bars indicated in Fig. 5.15.
6. Summary and perspectives

Within the framework of this thesis, many-body interactions of an electron spin confined in self-assembled, charge tunable, InGaAs QDs with the solid-state environment was studied by optical means. In particular the coupling to two reservoirs, the mesoscopic ensemble of nuclear spins of the quantum dot host material and the nearby Fermionic reservoir was investigated.

Our experiments establish the potential of single optically active QDs in investigating many-body physics. In addition, they pave the way for a new class of quantum optics experiments where the simultaneous presence of non-perturbative coherent cavity/laser coupling and Kondo correlations on laser field and photon correlations could be investigated.

In the following we will discuss results in details, raise open questions and propose experiments which could answer these questions.

6.1. Nuclear spins

Origin of dragging

As discussed in chapter 3 an effective $\hat{s}\hat{I}_x$ coupling between the electron spin and the nuclear spins describes the observed dragging effect well. However the origin of this coupling is not known yet. There are three possible explanations: first, the QD electron couples to the nuclear spins by non-collinear dipolar hyperfine interaction. This interaction is usually neglected due to the s-type symmetry of the QD electron wavefunction. However, this contribution could still have a finite contribution to the hyperfine interaction. Second, the inhomogeneous strain field in self-assembled QDs leads to quadrupolar interaction (QI) between the electric quadrupole moment of the nuclear spins and the electric field gradient in the crystal lattice. In combination with the secular term of the Fermi contact hyperfine interaction, QI also leads to an effective $\hat{s}\hat{I}_x$ interaction. And lastly, non-collinear dipolar hyperfine interaction between the QD hole spin and the nuclear spin [52, 99] leads to a similar model, which describes the physics as well. So far, in single QD, no experiment has been carried out to identify whether the electron or the hole is responsible for optically induced dragging of excitonic transition in QDs.

We propose an experiment which can be performed in vertically stacked coupled quantum dots and in principle reveal the nature of dragging. The idea is the following: the applied gate voltage is adjusted such that each QD contain a single electron(figure 6.1a). Now a laser scan across the right (red) QD transition is done to build up a nuclear spin polarization by optical dragging. Since the two electrons in the ground state are in a singlet state, dragging occurs in the final state either by the electron in the left QD or by the hole in the right QD. For an electron-mediated dragging, the polarization would build up in the left QD. If the hole was responsible,
Summary and perspectives

Figure 6.1: a) Schematic for an experiment which could reveal, whether the electron-nuclear or the hole-nuclear spin coupling is the origin of dragging. b) Level diagram of an ESR experiment which reveals if nuclear spin fluctuations are suppressed in the presence of dragging.

the polarization would build up in the right QD. A second fast laser scan across the left (blue) QD transition measures the magnitude of the OH field. If no OH field is present in the left QD after dragging, this would be a strong indication for a hole-mediated dragging process as proposed in reference [52].

Suppression of nuclear spin fluctuations

Having demonstrated that the locking of the QD resonance to the incident laser frequency via selective DNSP strongly damps out the fluctuations in electronic transition energy, we address the possibility of suppressing the time-dependent fluctuations in the nuclear Overhauser field [43, 101]. Given that the effective Zeeman shift associated with the rms Overhauser field of the QD nuclei $B_{\text{nuc}}$ is comparable to the spontaneous emission rate, we would expect that the fluctuations in the Overhauser field would lead to sizable fluctuations of the resonant absorption signal on time-scales that are characteristic for the Overhauser field (few seconds). Since we lack controlled experiments exclusively demonstrating the role of the Overhauser field fluctuations on the absorption signal, we could only claim that the experiments presented in chapter 3 provide an indirect evidence for a suppression of nuclear Overhauser field fluctuations.

We emphasize that a narrowing of the Overhauser field variance would have remarkable consequences for quantum information processing based on spins. In particular, a major limitation for experiments detecting spin coherence in QDs is the random, quasi-static Overhauser field that leads to an inhomogeneous broadening of the spin transition with a short $T_2^*$ time [7]. Suppression of the long-timescale Overhauser field fluctuations by laser dragging may be used to ensure that the time/ensemble averaged spin coherence measurements yield a dephasing time that is limited only by the fundamental spin decoherence processes [63, 102, 103]. While replacing the more traditional spin-echo techniques with laser dragging would rep-
resent a practical advantage for optical experiments, a more intriguing possibility would be the slowing down of nuclear spin dynamics by a combination of large inhomogeneous quadrupolar shifts \[47\] and dragging, which may in turn prolong the inherent electron spin coherence time.

An experiments which would reveal the extent of the suppressions of the OH fluctuations could be carried out in an electron spin resonance (ESR) experiment as performed by Kroner et al. \[57\]. A schematic is shown in figure 6.1b. After the nuclear spins have been prepared by dragging any QD resonance, the gate voltage is set to the \(X\)- plateau. A laser applied on the blue Zeeman transition leads to spin-pumping and a shelving of the population in the \(|\downarrow\rangle\)-state. Now a radio-frequency (rf) field applied on the \(|\downarrow\rangle - |\uparrow\rangle\)-transition shuffles population back in to the \(|\uparrow\rangle\)-state, measurable as a recovery of the absorption signal. The linewidth of ESR signal would indicate a narrowing of the OH field for a prepared nuclear spin system.

### 6.2. Decay of nuclear spin polarization

Our results on hyperfine-dominated nuclear spin interaction demonstrate that the nuclear spin dynamics is solely determined by the coupling of each nucleus to the central electron spin. At ultra-low temperatures, the OH field decay for a QD well isolated from an electron reservoir is predominantly due to intra-dot spin diffusion. By saturating the diffusion process using multiple polarization cycles and reducing the extrinsic (gate voltage) fluctuations that enhance the diffusion rate, it should be possible to prolong the spin-echo \(T_2\) time of the electron spin \[102\].

The nuclear spins could be treated classically for experiments in this work. As was shown in chapter 4, a pure dephasing process interrupts the coherent evolution of the electron-nuclear spin system. Elimination of this dephasing, which we believe is largely extrinsic, would open up the possibility for observation of the coherent quantum dynamics taking place on sub-milliseconds timescales. An abrupt turn-on of the Fermi-contact hyperfine interaction could lead to a quantum quench similar to the one discussed in chapter 5.

### 6.3. Optical signatures of the Kondo effect

The remarkable agreement between our experimental data and the NRG calculations in chapter 5 clearly demonstrates Kondo correlations between a QD electron and the electrons in a FR. The optical probe of these correlations unequivocally shows the signatures of Anderson orthogonality physics associated with the quantum quench of Kondo correlations, with field-tunable power law exponents. The area under the absorption curve in a magnetic is proportional to the occupation with a certain spins state. The two different Zeeman lines can be separately addressed with circularly polarized light, revealing the magnetization of the QD electron. We have shown that the extracted magnetization is lower (0.16) than expected for a electron in thermal equilibrium in the absence of Kondo correlations.
Circumventing sample interference effects

Since all the measured lineshapes carry the signatures of optical interference induced by the sample structure, which leads to mixing of the absorptive and the dispersive part of the QD response, a different sample would be desirable. The optimum distance of the QD-sample surface distance should be 130 nm for a QD with a transition energy at 900 nm. However as discussed, even smallest deviations from this distance or transition energy will cause an admixture of the dispersive part of the susceptibility to de detected signal for large laser detunings $\delta$. Also it would be difficult to find a QD exactly matching this distance.

A possibility to circumvent the problem of interference effects is to use another detection scheme of the QD response. In this work the resonance fluorescence (RF) was neglected since the laser intensity was much larger than the QD RF signal. Recent experiments have shown that the laser photons can be suppressed up to 7 orders of magnitude using cross-polarized detection scheme [19, 27, 96], allowing for a detection of the QD fluorescence as low as $10^2$ counts per second. A Bragg mirror, processed on the bottom side of the sample leads to an increase of the RF counts by a factor of $20^1$. A further increase could be achieved by a solid-immersion lens [29].

Such a scheme could also be adopted in our measurements. One problem in our setup could be the fact, that the system is fiber-based, which means that the polarization has to be adjusted by applying strain to the fiber core with “paddles”. Although such paddles are specified with a $10^8$ suppression, in our laboratories only a suppression of $1/20000$ could be achieved $^2$.

Using a 2DEG

Surprisingly, the agreement between the experimental lineshapes and the NRG calculation is extremely good, given that our electron gas is a bulk-doped, 40 nm thick, GaAs layer with a non-constant density of states (DOS). Such a DOS would, for example, lead to a different bandwidth of states in the perturbative regime, which in turn leads to a modification of the power-law exponent.

It would be desirable to incorporate a two-dimensional electron (2DEG) that the QD electron couples to into the sample structure. Attempts have made during this work, so far without success. Simulations of a structure containing a remote-doped 2DEG at an AlGaAs/GaAs interface below the QD layer show that the necessary gate voltage to align the lowest QD level with the Fermi energy would in fact deplete the 2DEG. An alternative approach would be to grow a remotely doped InAs quantum well, just below the QDs.

In-plane magnetic field

As plotted in figure 5.14 the absorption contrast shows oscillations as a function a applied magnetic field above 1.5 T. We attribute these oscillations to the formation of Landau-levels in the FR. In order to avoid this issue, the external magnetic

$^1$private communication with Parisa Fallahi
$^2$private communication with Andreas Reinhard
field could be applied parallel to the electron gas, as usually done in transport experiments.

**Kondo effect and other excitonic states**

All resonant absorption experiments in this work on optical signatures of the Kondo effect have been carried out on the negatively charged exciton where the exchange interaction is strongest. As can be seen in the gate-voltage sweep in figure 5.7 other excitonic transitions also show signatures of strong exchange coupling of the electron to the FR.

**The neutral exciton**

For example the neutral exciton $X^0$ (one electron and one hole in the excited state). The problem in investigating Kondo correlations here arise from the exchange interaction between the electron and hole in the QD, which is with a magnitude of $200 - 300 \mu eV$ [16] larger than the Kondo temperature. Neglecting the anisotropic exchange interaction, at zero magnetic field, an electron spin flip process from the $|\uparrow\downarrow\rangle$ to the $|\uparrow\downarrow\rangle$ needs to overcome the energy difference of the exchange interaction. However, by applying a magnetic field of a few Tesla, both states can be brought into resonance, which would then allow to observe the Kondo exciton [90]. We point out again, that the gate-voltage dependent tunnel coupling leads to a lower Kondo temperature.

**The positively charged exciton**

It has been difficult to find a highly coherent system in self-assembled quantum dots so far. The main reason is the decoherence of the electron spin due to hyperfine interaction with the nuclear spins. In p-doped samples a coherent hole spin has been demonstrated recently [104]. Here we propose an alternative realization of a coherent $\lambda$-system in a n-doped structure, making use of the Kondo effect. In
n-doped samples, like the one we use, a single hole can be optically generated. A resonant laser can excite a positively charged trion state $X^+$ consisting of two holes in a local singlet and an electron. At temperatures $T \gg T_K$ the system consists of two weakly coupled two-level systems, where coupling occurs in the excited state via hyperfine interaction or co-tunneling (figure 6.2a). When the temperature is below $T_K$, the electron in the excited state forms a Kondo-singlet with the electrons in the FR, leading to only one effective excited state for both ground states. Such a system is expected to highly coherent.

**Kondo effect under strong laser excitation**

In this work, we used low laser excitation to quench the Kondo correlations allowing a perturbative treatment of the laser field by Fermi golden rule. A key interest in quantum optics is the coherence properties of photons emitted by the QD, which are modified under strong laser excitation, i.e. when the laser becomes non-perturbative. In the case of a simple two-level system, a strong laser field leads to a Rabi-splitting of the states, measurable in a resonance fluorescence spectrum as a Mollow-triplet [105] or as damped Rabi-oscillations in the second order correlation function. The interesting question here is whether or not the coherence properties are modified in the presence of Kondo correlations. In the case of the two-level system, the Rabi-oscillations are damped exponentially. Since timescales associated with the Kondo effect are determined by power-law decays [75], it would be interesting to see if the Rabi-oscillations show the same behavior.

We demonstrated that the absorption spectrum shows different regimes which can be associated with the perturbative (local-moment) and the non-perturbative (strong-coupling) coupling of the electron to the FR. As explained, in the perturbative regime showing a $1/\delta$ tail, the excess energy of the laser is carried away by a particle-hole excitation in the FR. Therefore, we expect that the emitted photons have an energy corresponding to the trion transition energy. In the case of the strong-coupling regime ($1/\delta^{1/2}$-tail), the situation is not clear and requires an intense experimental and theoretical investigations.

**Spin-dependent threshold frequency**

The shift of the (zero temperature) absorption threshold frequency $\omega_{\text{th}}^e(B) = E_G^e - E_G^i$ with magnetic field can be written as

$$\omega_{\text{th}}^e(B) - \omega_{\text{th}}(0) = \frac{1}{2} g_e B + \delta \omega_{\text{th}}^e(B).$$  \hfill (6.1)

The first term reflects the Zeeman energy of the photo-excited hole, the second the $B$-dependence of the ground-state energy of the electron system. In our situation, $\tilde{n}_e \simeq 1$ and $\tilde{n}_f \simeq 2$. The asymptotic behavior of the initial magnetization is $m_e^f = \chi_0 B$ for small fields, where the linear static susceptibility $\chi_0$ is of order $1/T_K$, and $|m_e^f| = \frac{1}{2}$ for large fields. This implies

$$\delta \omega_{\text{th}}^e = \begin{cases} \frac{1}{2} \chi_0 B^2 & (|B| \ll T_K), \\ \frac{1}{2} |B| & (T_K \ll |B| \ll |\varepsilon_e^i|) \end{cases}.$$  \hfill (6.2)
6.3. Optical signatures of the Kondo effect

\[ T_{FR} = 15.6 \mu eV, \quad B = \frac{1}{4}T_{K}^{exp} = 1.1T_{FR} \]

<table>
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<th>( B_{exp} = 0.5 ) Tesla</th>
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<td>( n_{e} = 0.70 )</td>
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<td>( n_{\bar{e}} = 0.29 )</td>
<td>( n_{\bar{e}} = 0.16 )</td>
<td>( n_{\bar{e}} = 0.09 )</td>
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Figure 6.3.: NRG calculations for the lineshapes \( A_{\pm}(\nu) \) for the excitonic Anderson model of section 5.1.3. For the upper panels, all parameters were chosen as for QD 2 in chapter 5. The three magnetic field values (from left to right: \( B_{ext} = 0.5, 1 \) and 1.5 Tesla) correspond to the first three non-zero field values shown in the inset of figure 5.14A of the main text. Lower panels differ from upper ones only in a 90 times smaller choice of \( T_{K} \), namely \( T_{K} = 0.1T_{FR} = 1.56 \mu eV \) instead of \( T_{K} = T_{exp}^{K} = 140 \mu eV \). The data in panel b correspond to the NRG lines shown in figure 5.14a of the main text.

In our experiment for dot 2, \( |B| \lesssim T_{K} \), hence the dominant contribution to the threshold shift will be the \( \frac{1}{2}\sigma \hbar |B| \) term from the hole. This shift is evident in the calculated NRG absorption spectra shown in figure 6.3. For the \( B \)-dependent spectra displayed in figure 5.14, the \( B \)-dependent shift in the position \( \omega_{\sigma}^{th}(B) \) of the peak maximum is subtracted out, i.e. the detuning refers to the \( \nu = \omega - \omega_{\sigma}^{th}(B) \).

Figure 6.3 illustrates the difference between the presence or absence of Kondo correlations in the initial configuration of the \( X^- \) transition on the lineshape. The lineshapes in the top (bottom) panels were calculated for \( T \ll T_{K} \) (or \( T_{K} \ll T \)), so that Kondo correlations are strong (or weak). Evidently, Kondo correlations cause the peaks to be much broader and less high, in effect redistributing oscillator strength over a larger frequency range.

In both upper and lower panels, a magnetic field splits the peak into two separate ones, whose areas \( W_{\pm} \) reflect the spin-dependent occupations \( 1 - n_{e}^{\pm} \approx n_{\bar{e}}^{\pm} \) of the Zeeman-split initial electron levels. The difference in areas between the blue and red transitions, \( W_{+} - W_{-} \), which is a measure of the initial magnetization \( m_{e}^{i} \) [see Eq. (5.25)], changes much more slowly with increasing field for the upper than the lower panels, in accord with the behavior shown in figure 6.3a. We emphasize that the splitting of the trion transitions with the applied magnetic field is primarily due...
to the hole-Zeeman effect as we discuss below.

**Fermi edge singularities**

The orthogonality catastrophe in our measurements arises from Kondo correlations which alter the structure of the FR. An interesting question is to what extent Coulomb interactions give rise to a change of the FR as an electron is excited on top of it. In conventional QD samples with a large tunnel barrier, the FR electron have no overlap with the QD valence band states. In a short tunnel barrier sample however the conduction band states are mixed with FR states, which allows for direct transition from a valence-band state to the FR. Such transition have already been observed in PL measurements [87], but not been studied resonantly.
A. Bibliography


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2008          QSIT Junior Meeting, Ausserferrera (Switzerland), Signatures of resonant nuclear spin polarization in self-assembled quantum dots
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2010 SNF Fellowship for prospective researchers

August 9, 2011
2.1. Schematic illustration of the QD growth process. InAs is deposited on top of a GaAs substrate by molecular beam epitaxy. After a critical thickness of $\sim 1.7$ ML strain due to the different lattice constants leads to the formation of QD on the wetting layer. A further overgrowth of the QDs with GaAs completes the growth process.

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