Doctoral Thesis

Anomalous and Critical Dynamics in a Dissipative Central Spin System

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Anomalous and Critical Dynamics in a Dissipative Central Spin System

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Abstract

The phenomena of anomalous and critical dynamics occur across a wide range of systems. Anomalous dynamics is a diffusion process that is characterized by rare events, termed Lévy flights. Critical dynamics is associated with diverging system properties in the vicinity of a critical point at which the system undergoes a phase transition. These effects can be studied not only in classical systems but also in quantum systems. A quantum system of particular interest is the central spin system, since several candidate systems for the building blocks (qubits) of quantum information processing applications can be mapped onto it. Here, a single central spin interacts with a large spin environment. Understanding the intricate coupled dynamics is key in overcoming a major roadblock to quantum information applications, which is rooted in the dissipation and decoherence of a spin qubit. Quantum dots, which are zero-dimensional nanostructures that confine electrons in all three spatial dimensions, constitute a qubit candidate. The confined electron spin interacts with the nuclear spin environment of the host material, realizing a driven-dissipative central spin system. Since many parameters of a quantum dot can be tuned, it is an extremely versatile system to study both experimentally and theoretically.

In this context, we identify open challenges, which include the preparation of the spin environment of a spin qubit in an ultra-narrow state to reduce the spin decoherence, the direct observation of Lévy flights in a single quantum system, as well as the investigation of critical dynamics of a dissipative solid-state system in an experimentally accessible regime. We address these questions theoretically in a driven-dissipative central spin system with a particular focus on an optically driven self-assembled quantum dot.

When the central (electron) spin is driven by two lasers in a coherent population trapping configuration, the coupling to the (nuclear) spin environment forces the system into an electron–nuclei dark state. As a consequence, the optical absorption is drastically reduced, which in turn suppresses the dominant nuclear spin dynamics. The standard deviation of the resulting ultra-narrow nuclear spin distribution is close to the single-spin regime. This greatly prolongs the spin coherence time of the electron, which is an important result in light of potential applications in quantum information processing. Remarkably, the mechanism that leads to the cooling of the nuclear spin environment can be described in terms of an anomalous diffusion process of the nuclear spins. We then show that anomalous nuclear diffusion can also be observed in a time-dependent scheme where the electron is driven by periodic laser pulses. Strikingly, these nuclear Lévy flights can be monitored in real-time by tracking the photon emission events, making this scheme a promising path towards the experimental observation of Lévy flights in a single quantum system. Finally, we find signatures reminiscent of critical phenomena in a spin environment that is controlled by the central spin. The latter is assumed to be driven by two lasers in the coherent population trapping configuration. For self-assembled quantum dots, the nuclear spins are forced to transition continuously from an unpolarized to a polarized state when the relative strength of the two lasers is tuned accordingly. The transition is accompanied by increased nuclear fluctuations. However, in the thermodynamic limit the feedback from the electron
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to the nuclear spins diminishes as a result of the asymmetric coupling between
the single electron and the large nuclear environment. Nevertheless, assuming a
constant coupling strength, we observe that the spectral gap at the critical point
closes in the thermodynamic limit, providing evidence of a non-analytic change of
the steady state at the critical point.
Zusammenfassung


Zusammenfassung

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Anomalous and critical dynamics are phenomena that are connected to the unusual and striking behavior of a system when certain system properties diverge. Both exhibit a universal character that allows them to be observed in various fields, from physics and biology to sociology and finance. We illustrate these phenomena with examples from different disciplines and provide a historic context. Then we motivate the choice of the quantum mechanical system in which we study anomalous and critical dynamics. We consider a quantum mechanical system that can be partitioned into a central (small) system, consisting of a single spin, and a large environment of spins. Additionally, the central spin is externally driven and experiences dissipation. We thus have a driven and open (dissipative) central spin system. From a fundamental perspective, the central spin system is one of the generic models for the interaction of a small quantum mechanical system with a larger environment. Such system–environment interactions are at the heart of the dissipation and decoherence that are a result of the intricate evolution of a coupled system. For quantum information processing applications, which aim at harnessing the power of a quantum system, dissipation and decoherence often represent an obstacle. Several promising candidate systems for the building blocks of such applications can be mapped onto the central spin system. Studying the rich dynamics of the central spin model is thus central to the understanding of the dominant decoherence mechanism of the central spin. Finally, we introduce the self-assembled quantum dot (QD) which is one such candidate system. It can be considered as a specific realization of a driven and open central spin system and therefore constitutes an interesting example to illustrate anomalous and critical dynamics in a open central spin system, which are the subject of this thesis.

1.1 Anomalous dynamics

Anomalous dynamics and its description is historically closely linked to the advances in the understanding of transport and ordinary diffusion phenomena. We start by considering the prototype example of diffusion, Brownian motion. Then we illustrate how the path of a Brownian particle can be viewed as a random walk in space. This description also applies to anomalous diffusion and thus allows us to highlight a major difference between ordinary and anomalous diffusion. Then we turn to examples of anomalous diffusion, one in a sociological context on a global scale, the other in quantum systems.
Ordinary versus anomalous diffusion

Brownian motion is a prototype example of ordinary diffusion. In 1785, Ingenhousz was the first to experimentally observe the random motion of coal particles in alcohol and in 1827, Brown observed the same random jittery motion of pollen in water [1]. In 1905, Einstein (and independently von Smoluchowski in 1906 [2]) provided a complete description of this Brownian motion and explained the random movement to be caused by the constant bombardment of small particles by the atoms and molecules in the liquid [3]. Since the particles are hit from different directions and the force at a given time is not completely balanced, the jitter of the particle results. Furthermore, Einstein in his work unified two coexisting approaches, i.e. the description as a stochastic random walk and as diffusion governed by a deterministic diffusion equation for the probability distribution of the position of the particle as a function of time [4]. The formulation in terms of an observable random walk, which connects the microscopic dynamics to macroscopic observables, is illustrated next.

Random walks are a mathematical description for the path of a particle, which is regarded as the addition of random steps that are distributed according to some probability function. In the case of Brownian motion, the increment of the particle’s position is governed by a Gaussian (bell-shaped) probability distribution [5]. The concept of a random walk can be extended to include a waiting (also trapping or dwell) time in between successive jumps. This is known as a continuous–time random walk, which is characterized by a jump length and waiting time distribution. We note that the description of diffusion processes in terms of random walks
1.1 Anomalous dynamics

![Graph of Gaussian and Lévy probability density functions](image)

Figure 1.2: The Gaussian (yellow) and a Lévy (blue) probability density functions are sketched. The Gaussian distribution decays quickly while the Lévy distribution shows a ‘heavy tail’.

can account for the memory effects in the dynamics of a system. Finally, random walks are an appropriate tool to study anomalous diffusion [4].

A hallmark of anomalous diffusion is the asymptotic power-law behavior of the probability distribution that governs the position or waiting time increment. These distributions, also known as Lévy distributions, decay much more slowly than the Gaussian distribution of Brownian motion and are sometimes said to have ”heavy-tails”. Therefore, extreme events, stemming from the tails of the distribution, are decisive for the evolution of the system. Early examples of anomalous diffusion appeared in 1926 in turbulent diffusion [6] and later in the transport of charge carriers in amorphous semiconductors [7, 8]. Since then, the concept of anomalous diffusion has been applied to the description of processes in various fields including rotating flows [9, 10], the propagation of light waves through a specially engineered system [11], as well as biological systems to describe certain foraging behavior of animals [12, 13, 14].

**Example of anomalous diffusion**

The first illustrative example of anomalous diffusion we consider is the circulation pattern of banknotes [15]. This example is also of practical value as it hints at the laws of human travel and the ensuing spread of diseases on a global scale. It was found that normal diffusion (Brownian-motion) is insufficient for describing the dynamic spatial redistribution of the banknotes. On the one hand, the probability distribution of the displacement length decays according to a power-law, making long-distance travel (due to e.g. air-travel) extremely important for the overall dynamics which is also known as superdiffusion. On the other hand, the waiting
time between two displacements is also shown to behave according to a power-law for the asymptotically long trapping times, a regime known as subdiffusion. Often both the super- and subdiffusive dynamics are termed Lévy flights.

### Anomalous diffusion in quantum systems

Anomalous diffusion was also used to explain several observations in quantum systems, of which laser cooling below the recoil limit by velocity selective coherent population (VSCPT) is a prominent example and of great significance for this thesis [16, 17]. Laser cooling of atoms, which was recognized with the 1997 Nobel prize in physics, is achieved by employing the mechanical effects of light to reduce the spread of the velocity $\vec{v}$ (and simultaneously of the momentum $\vec{p} = m\vec{v}$) distribution. This narrowing corresponds to lowering the temperature of the system. To study quantum mechanical effects in neutral atoms, it is clearly desirable to slow them from an average speed of $\sim 4000 \text{ km/h}$ at room temperature [18, 19, 20].

In a simplified picture, the mechanism that leads to the cooling of the atoms can be understood as the interplay of the momentum kicks the moving atom receives from absorbing laser photons (quanta of light) and from the photons it spontaneously emits in random directions, which results in a slowing of the atoms. An example of how such momentum kicks can modify the velocity of a classical object is observed e.g. in a game of billiards. However, even stationary atoms generally interact with the laser photons and subsequently recoil when they spontaneously emit a photon. One might therefore expect that this recoil, i.e. the recoil induced by a single photon, fundamentally limits the cooling.

Fortunately, VSCPT provides a route to circumvent the recoil limit by choosing a laser configuration for which there is a non-absorbing (dark) state for the slowest atoms at $p \simeq 0$ [21, 22]. This dark state is a result of a quantum interference effect [23, 24]. Since these slow atoms do not absorb laser photons and thus are not excited, they also do not spontaneously emit photons. Therefore, at $p \simeq 0$ the dominant mechanism for the redistribution of momentum ceases to work. Conversely, for $p \neq 0$ the atoms absorb and emit photons, and their momentum $p$ changes randomly. If an atom happens to experience a change in momentum such that after emission $p \simeq 0$, it will be trapped in this state. This mechanism allows the momenta to accumulate around $p \simeq 0$ and thus narrows the momentum distribution (and lowers the temperature). This example can be modeled as a continuous–time random walk in momentum space which shows subdiffusive behavior since the long trapping times around $p \simeq 0$ are governed by a slowly decaying Lévy distribution. It is worth noting that while the ensemble properties connected to the momentum distribution of the atoms can be extracted experimentally, there is to date no direct experimental observation of the power-law distribution of such an anomalous diffusion process in a single quantum system.

### 1.2 Critical dynamics at phase transitions

We now turn to the second topic of this thesis, which is the critical dynamics observed in connection with certain phase transitions. Phase transitions have traditionally been studied in classical thermodynamic systems, which are also the starting point of our discussion. Many of the concepts introduced in the classical
1.2 Critical dynamics at phase transitions

theory are carried over to the description of phase transitions in disciplines outside physics, to quantum phase transitions and dissipative phase transitions, which we will illustrate below.

Classical phase transitions

The boiling of water is an everyday example of a phase transition. When the temperature (or pressure) is changed beyond a certain value, the state of matter changes from a liquid to vapor. However, classical phase transitions occur in a wide range of systems and to give an additional example, we mention the loss of magnetization of a permanent magnet as the temperature exceeds a critical value. Phase transitions are often classified as abrupt (i.e. first order) or continuous (e.g. second order) [25]. The boiling of water is an example of an abrupt transition since certain system properties (in this case the density) of water change discontinuously. However, transitions which are smooth also exist, e.g. in a permanent magnet the magnetization decreases smoothly and vanishes at the critical point [26]. These transitions are classified as continuous phase transitions and attracted a lot of attention due to striking accompanying phenomena, which are also known as critical phenomena.

The first theoretical treatment of phase transitions that could be applied to a wide range of systems was developed by Landau in 1937 and corresponds to a mean field approach [27, 28, 25]. He introduced the important concept of an order parameter which describes the degree of order in the system. For a permanent magnet, the order parameter is its magnetization. It assumes a non-zero value below the critical point since the electron spins that give rise to the macroscopic magnetic effects are aligned in a given direction. As it is heated, the extra thermal energy allows some of the electron spins to flip and thereby destroys the ordering and the magnetization, i.e. the order parameter, vanishes [26, 25]. However, the predictions of Landau’s theory failed in the vicinity of the critical point of the phase transition in the 2D-Ising model, which serves as a prototype model to study phase transitions such as ferromagnetism [27]. This became apparent with Onsager’s solution of this model in 1944 [29]. Wilson realized in 1971 that this is connected to the importance of fluctuations over different length scales in the vicinity of the critical point which were not captured by Landau’s theory. He remedied this problem by modifying the renormalization group theory to deal with these fluctuations at the critical point [30, 31]. Furthermore, he showed that at the phase transition most system parameters become insignificant for the behavior of the system, and only the dimension and the order parameter are important. It is thus not surprising that vastly different physical systems show similar properties close to a continuous phase transition. For this seminal contribution, Wilson was awarded the Nobel prize in physics in 1982 [27].

In the last few decades, the classical theory of phase transitions has been expanded to encompass not only classical phase transitions in quantum systems and quantum phase transitions, but also dissipative phase transitions. Furthermore, the paradigm of phase transitions has more recently been applied to fields outside physics [25]. For example, whether certain properties of economic markets close to a financial crash can be understood as resulting from an analogue of a critical point is currently being investigated [32]. We will now highlight a few examples
of different types of phase transitions in various physical systems.

**Classical phase transitions in quantum systems and quantum phase transitions**

As experimental techniques made the study of quantum mechanical systems possible, researchers began investigating phase transition in these quantum mechanical systems. A prominent example of a classical phase transition in a quantum system is Bose-Einstein condensation (BEC): as the temperature of certain dilute gases is lowered below a critical value, the collection of atoms falls (or condenses) into the lowest energy state, giving rise to a single macroscopic quantum state with a narrow velocity distribution [33, 34, 35]. Thus the state of matter of this quantum system has changed.

In contrast to the BEC transition which occurs when the temperature is changed, quantum phase transitions occur at $T = 0$ K [36, 37]. At absolute zero, thermal fluctuations become irrelevant and quantum fluctuations drive the phase transition as a function of a parameter other than temperature [38]. As the control parameter is tuned across the critical point, the properties of the lowest-energy (ground) state change and the system transitions from one quantum phase to another. To illustrate this, we consider a ferromagnet for which we recall that thermal fluctuations flip spins that are initially aligned and once the temperature reaches a critical value, the magnetization is completely lost. If instead the temperature is kept constant at $T = 0$ K and a transverse magnetic field is turned on and gradually increased beyond a quantum critical point, the ferromagnetic ordering is lost. Here, quantum fluctuations rather than thermal fluctuations destroy the ferromagnetic ordering [36]. A realization of the described transition has been observed in different crystals [39, 40]. Even though quantum phase transitions occur at absolute zero temperature, investigating them is not just an academic endeavor since the existence of a quantum critical point can strongly influence the system properties for a wide range of parameters [41, 42]. This could be crucial to the understanding of certain phenomena at $T \neq 0$ K, e.g. high-temperature superconductivity [43, 44, 45, 46, 41].

**Dissipative phase transitions**

Dissipative phase transitions occur in non-equilibrium situations when the coupling of the system to an environment leads to dissipation, of which friction is an everyday example. The interplay between dissipation and external driving can lead to a phase transition as a function of the system–environment parameters. An interesting example of such a dissipative phase transition is the onset of lasing when the radiation emitted from a laser (light amplification by stimulated emission of radiation) changes from incoherent to coherent light as the pump power parameter is increased [47, 48, 49, 50]. A second dissipative phase transition that has been intensively studied theoretically occurs in the Dicke model, where an ensemble of atoms enters a superradiant phase once the light-matter interaction becomes sufficiently strong [51, 52, 53, 54, 55, 56]. Recently, the Dicke phase transition has been realized experimentally in a BEC [57, 58, 59]. Since then, the number of theoretical proposals for dissipative phase transitions burgeoned [60, 61, 62, 63, 64, 65, 66, 67, 68, 69], predominantly in atomic systems. However,
The dissipative central spin model

Since the emergence of quantum mechanics, much effort was directed towards the challenging task of isolating a quantum system from its environment. This was motivated by the fact that the environment can cause dynamics in the system that destroy the delicate quantum effects. However, if a quantum system is completely decoupled from its environment, no information can be extracted about the quantum properties of the system. In light of potential quantum information processing applications that are aimed at harnessing the quantum properties of a system, it is not only of fundamental interest, but also of practical importance to understand the coupled system–environment dynamics. Fortunately, many physical systems can be mapped onto a small number of general (canonical) system–environment models, which we will discuss next. Then we consider the role dissipation plays, not just as an obstacle that prevents quantum effects from being observed but also as a tool, e.g. to engineer interesting quantum mechanical states. Finally, we briefly introduce quantum dots which realize an open (dissipative) central spin model.

Canonical system–environment models

While a broad variety of quantum mechanical systems and environments have been studied, most of them can be mapped onto a small number of canonical models that are studied to gain insight into the dissipative effect of the environment. We consider the system and the environment to be composed of either one of two prototypes of quantum systems: spins and harmonic oscillators [70]. The system is described either by a continuous coordinate in a harmonic oscillator potential or by a spin with a discrete state space. If the environment is well described by extended modes, a mapping to an oscillator environment is often justified [71, 72]. In contrast, the environment is mapped to a spin environment if its modes are localized which is particularly relevant at low temperatures [73]. Often, the mutual coupling of the spins within the spin environment is much weaker than the coupling of an environmental spin to the central spin. The latter is thus often the dominant interaction for the spin environment and strongly modifies the spin environment. This gives rise to intricate feedback mechanisms between the system and environment (non-Markovian dynamics)[70, 74]. Combining the mapping of the system and environment to oscillators and spins, we obtain four canonical models for system–environment interactions shown in Fig. 1.3 [75, 76, 70]. In the following, we will be concerned with the central spin model, which is schematically depicted in the lower right graph of Fig. 1.3.

Dissipative dynamics of the central spin

To understand the motivation for studying a dissipative central spin model, we note that one of the intriguing properties of quantum systems is their ability...
Figure 1.3: Canonical system–environment models. The central system is either represented by a harmonic oscillator (top) or a spin (bottom). The environment is mapped onto a collection of harmonic oscillators (left) or spins (right). The central spin model is sketched on the bottom right.
to exist in a superposition of states, for which there is no classical counterpart. However, these superpositions are often fragile and prone to decay as a result of the interaction of the system with a large environment. In this situation, decoherence induced by the environment is an obstacle and motivated the scientific community to devise elaborate techniques that allowed for a sufficient isolation of the system so that quantum effects could be observed. Coherence, however, can also be observed in situations where a system experiences dissipation. In fact, dissipation recently came to the attention of physicists as a versatile tool rather than an obstacle. To give just one example, dissipation can be employed to prepare interesting quantum states [77, 78, 79, 80]. In this thesis, we are interested in the evolution of a central spin system as a result of not only the interplay between coherent driving and spin system–environment coupling but also the dissipative dynamics the central spin experiences.

1.4 Outline of this thesis

Chapter 2 introduces the QD model system. After reviewing relevant characteristics of self-assembled QDs, we detail how a single electron charged QD realizes a driven and open central spin system. We first consider the central electron spin and its coherent and dissipative couplings, which arise due to the interaction with laser fields and spontaneous emission. Then we highlight the most important interactions for the nuclear spins which constitute the spin environment in (strained) self-assembled QDs. Then we provide a detailed derivation of the hyperfine interaction between the central electron spin and the nuclear spin environment, which is at the heart of the schemes studied in this thesis. Finally, we highlight some
Introduction

of the effects, studied both theoretically and experimentally, that result from the feedback in the coupled electron-nuclear dynamics.

In chapter 3 we analyze coherent population trapping (CPT) in the central spin system. We show that the spin environment is drastically modified and its distribution can be narrowed to the single spin flip level. Concurrently, the central spin evolves into a dark (non-absorbing) state for a wide range of bare two-photon laser detunings, thereby broadening the CPT interference dip in the absorption curve. These results can be explained by an anomalous diffusion process reminiscent of the Lévy flights connected to VSCPT. This analysis can also be used to estimate the achievable narrowing of the nuclear distribution in the presence of imperfections. In self-assembled QDs, the large difference between electronic and nuclear time scales allows for a measurement-based verification of the desired electron–nuclei state. The treatment in this chapter is based on [89].

Chapter 4 is based on [90] and covers a scheme that allows the direct observation of Lévy flights in a single quantum system which has so far remained elusive. We investigate a periodically driven open central spin system and find that the dynamics of the spin environment is captured by Lévy flights. For the particular realization in a single-electron charged quantum dot, existing ensemble measurements [91] are consistent with Lévy dynamics. We employ Monte Carlo simulations to extract the probability distribution that governs the long waiting times between successive nuclear spin flip events and find a power law distribution with exponent \( \eta \approx -3/2 \). Remarkably, this power-law exponent is imprinted on the waiting time distribution of successive photon emission events. Due to the favorable ratio of electron-nuclear time scales in QDs, the nuclear Lévy flights can thus be tracked in real-time. In contrast to chapter 3, we pay particular attention to strain-induced nuclear quadrupolar interactions, which have been neglected in chapter 3. We also discuss the modifications induced by imperfections. We further show that the dominant intrinsic limitation of the scheme arising from nuclear quadrupole coupling can be minimized by adjusting the magnetic field or by implementing a spin echo sequence.

In chapter 5 we discuss the signatures of criticality in a dissipative central spin model where the central spin is driven by two lasers in a CPT setting. We develop a tractable description of the spin environment by deriving a reduced master equation. Applying this method to self-assembled QDs, we find that the spin environment transitions from an unpolarized to a polarized state when the two laser Rabi frequencies are equal. In contrast to the previous two chapters, directional rather than diffusive nuclear spin polarization processes are at the heart of this observation. Furthermore, we show that the nuclear probability distribution broadens as the transition point is approached before it develops a double-peaked structure. We thus find that by tuning the relative Rabi frequencies, the nuclear spin distribution can be narrowed. Then we proceed to the computation of the spectral gap of the Liouvillian in the thermodynamic limit, which when it closes at the transition point is a strong evidence that the system undergoes a dissipative phase transition. In the case of QDs, the spectral gap tends to zero in the thermodynamic limit at any value of the external control parameter. This is due to the fact that the feedback loop between the central spin and the nuclear environment is broken in the thermodynamic limit. We use mean field equations to investigate the origin of this decoupling. We find that the combination of the
particular scaling of the spin-environment coupling with the number of spins and the fact that the magnitude of the electronic parameter that feeds back to the nuclear spins is not directly proportional to the strength of the laser driving.
The quantum dot central spin model

The topic of this chapter is the realization of a driven–dissipative central spin model by self–assembled quantum dots (QD). The central spin is the electron spin, trapped in the QD (Fig. 2.1). The environment considered here is composed of the nuclear spins of the QD host material [76, 92, 93]. First, we review some important properties of self–assembled QDs. Then we introduce the central electron spin and its interaction with the external magnetic field, the laser fields, and the open radiation field reservoir. Next, we focus on the nuclear spins of the QD under an external magnetic field as well as the strain that naturally occurs in the strain–driven growth, which gives rise to quadrupolar interactions. After introducing the elementary interactions between the electron and nuclear spins, which are dominated by the Fermi–contact hyperfine interaction, we review some of the intriguing phenomena of the coupled electron–nuclei dynamics that have been observed in self–assembled QDs.

2.1 Semiconductor quantum dots

Semiconductor QDs are heterostructures that extend only a few nanometers in each spatial direction. This strong confinement leads to the quantization of the energy states of the charge carriers in the semiconductor material, similar to a particle–in–a–box [94]. Hence, they are effectively zero–dimensional objects. Since the discrete QD states resemble atomic states, QDs are often referred to as “artificial atoms” [95]. A range of fabrication methods allow to synthesize different types of QDs. Colloidal QDs are nanocrystals that form due to crystallization in a colloidal solution [96] and can be chemically post–processed. This led to a range of biological [97] and optoelectronical [98] applications. Electron confinement can also be realized by electrically defined QDs. The electrical gates are fabricated

Figure 2.1: QD central spin model
on top of a two-dimensional electron gas, which allows to locally isolate a small number of electrons [99]. Self-assembled QDs are grown using molecular beam epitaxy and form due to strain, as described in more detail below. In contrast to electrically defined QDs, self-assembled QDs confine both electrons and holes, leading to strong optical interaction. A strain-free alternative are QDs that are synthesized by molecular droplet epitaxy [100].

The optically active self-assembled QDs studied in this thesis are grown by molecular beam epitaxy [101]. First, the wafer is covered with a buffer layer of GaAs. Next, InAs is deposited on the GaAs buffer layer. The strain induced by the 7% lattice constant mismatch of the two materials leads to the formation of InAs islands at random locations, once 1.7 monolayers are deposited. The average spatial extent of these islands is about 10 nm along the growth direction and the in-plane diameter is 20–30 nm. The size of the QD determines the quantized energy levels and hence the emission wavelength \( \lambda \approx 1.1 \mu \text{m} \). To shift the wave length to \( \sim 950 \text{nm} \) such that silicon photodetectors can be used, a further layer of GaAs is grown on top of the QD layer at a higher temperature. This procedure is known as the partially covered island technique [102]. The diffusion of In leads to smaller QD heights of about 5 nm, resulting in the desired emission wave length. Even though the self-assembled QDs grown according to the described technique show atomic properties such as discrete states, they typically consist of \( N \approx 10^4 – 10^6 \) atoms [103, 104, 105].

To deterministically charge the QD with individual electrons, it is embedded in a field effect structure [106, 107]. An n-doped back gate, which acts as an electron reservoir, is separated from the QD layer by a tunnel barrier. An AlGaAs blocking layer separates the QDs from the semitransparent metallic top gate. The applied voltage between the gates as well as the voltage induced by the semiconductor–metal interface determine the electrostatic energy of the electron confined in the QD with respect to the Fermi energy. Depending on the applied voltage, electrons from the back contact can tunnel into the QD. Once an electron has tunneled into the dot, adding a second electron requires additional energy to overcome the Coulomb repulsion. This effect is known as the Coulomb blockade and is utilized to deterministically tune the charge state of the QD.

If the potential difference varies on a length scale that is comparable to a particle’s de Broglie wave length \( \lambda = \hbar/p \), quantum mechanical effects are expected. In the nanometer-sized self-assembled QDs, the variation of the potential experienced by the electron and holes arises due to the bandgap difference of the QD material to the surrounding material. Since the electron de Broglie wave length is on a nanometer scale, fully quantized states for the charge carriers result. At low temperatures it is assumed that the wave function of the charge carriers remains in the lowest energy state along the growth direction. This is justified since the confinement along the growth direction is much stronger than in-plane. The in-plane potential can be assumed to be approximately harmonic [108]. The quantized energy states are labeled as s, p, d... in accordance with atomic physics. The Bloch function of the lowest conduction band state has s-type orbital symmetry, leading to total angular momentum \( J = 1/2 \). The valance band has p-like orbital symmetry, yielding total angular moment \( J = 3/2 \). The highest energy state of the valence band is the heavy–hole band, while the light–hole and spin–orbit split–off band are detuned energetically by more than 10 meV and will therefore
be neglected in this thesis. The band structure gives rise to ’almost hard’ optical selection rules \[109\], which will be discussed later.

Combining deterministic charging of the QD with a single electron with the selection rules under optical excitation, we can examine the electronic states that will be of importance in this thesis. The ground state corresponds to a single-electron charged QD, where the electron resides in the s-shell of the conduction band. Upon optical excitation an electron–hole pair is created. The optically excited electron is promoted from the valence band to the conduction band and forms a singlet state together with the resident electron. The electrons and the hole are subject to Coulomb interactions and form a negatively charged exciton \((X^-)\), called trion.

\section{2.2 The central electron spin}

Self-assembled QDs can be deterministically charged with a single electron. The spin of this resident QD electron constitutes the central spin of the model. In this section, we first discuss the Hamiltonians that describe the coupling of the electron spin to an external magnetic field and to optical laser fields. Throughout this thesis, the magnetic field points along an axis perpendicular to the growth direction. We denote the direction of the magnetic field by \(z\) and the growth direction by \(x\). Furthermore, we consider optical illumination by laser fields, which propagate along the sample growth direction \(x\). Hence, the laser excitation is perpendicular to the magnetic field, a configuration known as Voigt geometry. For linearly polarized continuous wave (cw) excitation we describe the Hamiltonians in the \(z\)-basis, but we switch to the optical \(x\) basis for circularly polarized excitation by a periodic laser pulse train. Then we consider the coupling of the electron to the open radiation field reservoir which introduces dissipative dynamics. Finally, we discuss the Markovian master equation that captures both the coherent and dissipative evolution of the central electron spin.

\subsection{2.2.1 Electron Zeeman interaction}

The external magnetic field splits the energy levels of the electron spin via the Zeeman effect, yielding the two states \(|\uparrow\rangle, |\downarrow\rangle\). The excited states considered throughout the thesis are negatively charged exciton states, \(|\uparrow\downarrow\uparrow\rangle, |\uparrow\downarrow\downarrow\rangle\), where \(\uparrow\) and \(\downarrow\) denote the projection of the pseudospin of the hole. Since the two electrons form a singlet, the hole determines the Zeeman splitting of the excited states. The Zeeman Hamiltonian reads (\(\hbar = 1\) throughout this thesis)

\[
H_e = -\frac{\omega_e}{2} (|\uparrow\rangle \langle \uparrow| - |\downarrow\rangle \langle \downarrow|) + \frac{\omega_t}{2} (|\uparrow\downarrow\uparrow\rangle \langle \uparrow\downarrow\uparrow| - |\uparrow\downarrow\downarrow\rangle \langle \uparrow\downarrow\downarrow|) + \omega_t (|\uparrow\downarrow\rangle \langle \uparrow\downarrow| + |\uparrow\downarrow\rangle \langle \downarrow\downarrow|),
\]

where \(\omega_t = g_e \mu_e B\) and \(\omega_e = g_h \mu_e B\) are the electron (trion) Zeeman energy, \(g_e \ (g_h)\) is the effective electron (hole) g-factor, \(\mu_B\) is the Bohr magneton, and \(\omega_t\) is the optical transition frequency.
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Figure 2.2: The two electron spin states are coupled to a common excited state under cw excitation. In this configuration, CPT can be observed.

2.2.2 Continuous wave laser excitation

We consider linearly polarized cw lasers that couple the two Zeeman-split electron ground states to a common excited state. For large magnetic fields we can neglect one of the excited states and consider a three-level system only. The two orthogonally linearly polarized laser beams, called probe and coupling lasers, couple the ground states to the excited state \(|↑↓⇑⟩\). The Rabi frequency of the probe (coupling) laser is denoted by \(Ω_p\) (\(Ω_c\)) and the corresponding laser frequency is \(ω_p\) (\(ω_c\)) (Fig. 2.2). The Hamiltonian, describing the laser interaction in the rotating wave approximation [110], is then given by

\[
H_{\text{laser}} = Ω_p(|↑⟩⟨↑↓⇑| e^{iω_p t} + |↑↓⇑⟩⟨↑| e^{-iω_p t}) + Ω_c(|↓⟩⟨↑↓⇑| e^{iω_c t} + |↑↓⇑⟩⟨↓| e^{-iω_c t}).
\] (2.2)

After transforming into a rotating frame, we analyze the consequences of the laser Hamiltonian and introduce coherent population trapping (CPT) which is a quantum interference phenomenon. It can be understood by employing the concept of a dark state, which is a coherent superposition of the two ground states that does not couple to the laser fields. Many effects investigated in this thesis are a consequence of the existence of such a dark state.

Rotating frame

To eliminate the time dependence in the laser Hamiltonian \(H_{\text{laser}}\), a unitary transformation \(U\) to the rotating frame is applied. The transformed wave function describing the electronic system is given by [111]

\[
|\tilde{ψ}\rangle = U^\dagger |ψ\rangle.
\]
While the Schrödinger equation
\[ H |\psi\rangle = (H_0 + H_{\text{laser}}) |\psi\rangle = i \frac{\partial}{\partial t} |\psi\rangle \]
governs the evolution of $|\psi\rangle$, the dynamics of $|\tilde{\psi}\rangle$ is determined by
\[ H_{\text{rot}} |\tilde{\psi}\rangle = i \frac{\partial}{\partial t} |\tilde{\psi}\rangle \]
with
\[ H_{\text{rot}} = U^\dagger H U - i U^\dagger \frac{\partial U}{\partial t}. \]

We continue with the ansatz
\[ U = e^{-i A t}, \quad (2.3) \]
where
\[ A = \sum_j a_{jj} |j\rangle \langle j|. \quad (2.4) \]

The diagonal matrix elements of $A$ will be determined such that the fast time dependence of $H_{\text{laser}}$ is canceled. In the following calculation it is convenient to use the following notation for $H_{\text{laser}}$:
\[ H_{\text{laser}} = \sum_{k,l} h_{kl} |k\rangle \langle l|, \]
where $j,k,l$ run over $\uparrow, \downarrow, \uparrow \downarrow \uparrow \downarrow$, and $h_{\uparrow, \uparrow \downarrow \uparrow \downarrow} = \Omega_p e^{+i \omega_p t}$, $h_{\downarrow, \uparrow \downarrow \uparrow \downarrow} = \Omega_c e^{+i \omega_c t}$, $h_{kj} = h_{jk}^*$ with all other $h_{jk} = 0$. To proceed we rewrite the effective Hamiltonian in the rotating frame $H_{\text{rot}}$ using
\[ [H_0, A] = 0, \]
and
\[ U^\dagger \frac{\partial U}{\partial t} = U^\dagger (-i A U) = -i A, \]
which yields
\[ H_{\text{rot}} = U^\dagger H U - A = (H_0 - A) + U^\dagger H_{\text{laser}} U. \]

Expanding $U^\dagger H_{\text{laser}} U$ and using the ansatz (2.3) we find
\[
U^\dagger H_{\text{laser}} U = H_{\text{laser}} + it[A, H_{\text{laser}}] + \ldots
\]
\[
= \sum_{k,l} h_{kl} |k\rangle \langle l| + it \sum_j a_{jj} \sum_{k,l} h_{kl} |k\rangle \langle l| + \ldots
\]
\[
= \sum_{k,l} h_{kl} |k\rangle \langle l| + \sum_{k,l} it (a_{kk} - a_{ll}) h_{kl} |k\rangle \langle l| + \ldots
\]
\[
= \sum_{k,l} (1 + it (a_{kk} - a_{ll})) h_{kl} |k\rangle \langle l| + \ldots
\]
\[
= \sum_{k,l} e^{+it(a_{kk} - a_{ll})} h_{kl} |k\rangle \langle l|
\]
\[
= \Omega_p e^{+i \omega_p t} \Omega_p e^{+it(\tilde{\omega}_p - \tilde{\omega}_p)} |\uparrow\rangle \langle \uparrow| + \text{h.c.}
\]
\[
+ \Omega_c e^{+i \omega_c t} \Omega_c e^{+it(\tilde{\omega}_c - \tilde{\omega}_c)} |\downarrow\rangle \langle \downarrow| + \text{h.c.}
\]
2 The quantum dot central spin model

To eliminate the fast time dependence induced by the laser frequencies $\omega_p$ and $\omega_c$, we need to choose the parameters $a_{jj}$ such that they satisfy the following system of equations:

\[
\begin{align*}
    a_{\uparrow \uparrow} - a_{\uparrow \downarrow \uparrow \downarrow} &= -\omega_p, \\
    a_{\downarrow \downarrow} - a_{\uparrow \downarrow \uparrow \downarrow} &= -\omega_c.
\end{align*}
\]

The choice

\[
\begin{align*}
    a_{\uparrow \uparrow} &= -\frac{1}{2}(\omega_p - \omega_c), \\
    a_{\downarrow \downarrow} &= +\frac{1}{2}(\omega_p - \omega_c), \\
    a_{\uparrow \downarrow \uparrow \downarrow} &= +\frac{1}{2}(\omega_p + \omega_c)
\end{align*}
\]

solves the required system of equations and yields the time independent effective Hamiltonian

\[
H_{\text{rot}} = \begin{pmatrix}
    -\frac{\delta}{2} & 0 & \Omega_p \\
    0 & \frac{\delta}{2} & \Omega_c \\
    \Omega_p & \Omega_c & \frac{\Delta \omega_p + \Delta \omega_c}{2}
\end{pmatrix},
\]

where the one–photon laser detunings are defined as

\[
\begin{align*}
    \Delta \omega_p &= (\omega_{te} + \frac{\omega_h}{2} + \frac{\omega_e}{2}) - \omega_p, \\
    \Delta \omega_c &= (\omega_{te} + \frac{\omega_h}{2} - \frac{\omega_e}{2}) - \omega_c
\end{align*}
\]

and the two–photon detuning is given by

\[
\delta = \Delta \omega_p - \Delta \omega_c = \omega_e - (\omega_p - \omega_c).
\]

Coherent population trapping

The optically driven electronic system introduced above shows the phenomenon of CPT [23, 24, 112], which occurs in three–level systems where the two ground states are coupled to a common excited state. Central to the understanding of CPT is the dark state

\[
|D\rangle = \frac{1}{\sqrt{\Omega_p^2 + \Omega_c^2}}(\Omega_c |\uparrow\rangle - \Omega_p |\downarrow\rangle).
\]

The quantum amplitudes to couple this coherent superposition of the ground states to the excited state interfere destructively when the two lasers are detuned equally from the excited state. More precisely, when the effective laser Hamiltonian $H_{\text{rot}}$ is applied on the dark state we find

\[
H_{\text{rot}} |D\rangle = \begin{pmatrix}
    -\frac{\delta}{2} & 0 & \Omega_p \\
    0 & \frac{\delta}{2} & \Omega_c \\
    \Omega_p & \Omega_c & \frac{\Delta \omega_p + \Delta \omega_c}{2}
\end{pmatrix}
\begin{pmatrix}
    \frac{\Omega_p}{\sqrt{\Omega_p^2 + \Omega_c^2}} \\
    \frac{\Omega_c}{\sqrt{\Omega_p^2 + \Omega_c^2}} \\
    0
\end{pmatrix} = \begin{pmatrix}
    -\frac{\delta}{2} & 0 & \Omega_p \\
    0 & \frac{\delta}{2} & \Omega_c \\
    \Omega_p & \Omega_c & \frac{\Delta \omega_p + \Delta \omega_c}{2}
\end{pmatrix}
\begin{pmatrix}
    \frac{\Omega_p}{\sqrt{\Omega_p^2 + \Omega_c^2}} \\
    \frac{\Omega_c}{\sqrt{\Omega_p^2 + \Omega_c^2}} \\
    0
\end{pmatrix}
\]

\[
= \begin{pmatrix}
    -\frac{\delta}{2} & 0 & \Omega_p \\
    0 & \frac{\delta}{2} & \Omega_c \\
    \Omega_p & \Omega_c & \frac{\Delta \omega_p + \Delta \omega_c}{2}
\end{pmatrix} = 0.
\]
2.2 The central electron spin

Figure 2.3: Electronic levels in the rotating frame

Hence, at two–photon resonance ($\delta = 0$), the dark state $|D\rangle$ is unaffected by the interaction with the laser fields. This results in a dip in the absorption spectrum and the excited state population at $\delta = 0$ (Fig. 2.4 and Fig. 2.5). The width of the resulting CPT–dip can be shown to be characterized by $\sqrt{\Omega_p^2 + \Omega_c^2}$ in the limit where $\Omega_p, \Omega_c \ll \Gamma$ [112], where $\Gamma$ is the rate of spontaneous emission. Ideally, the system is completely transparent at $\delta = 0$. However, the coherent superposition of the electron spin states that constitutes the dark state is sensitive to dephasing of the electron ground states. Thus, pure dephasing results in a residual absorption rate at $\delta = 0$.

A prominent application of CPT is subrecoil laser cooling through velocity selective coherent population trapping (VSCPT) [21, 22, 16, 17, 113]. Furthermore, CPT is closely related to a plethora of intriguing phenomena including lasing without inversion [114], stimulated Raman adiabatic passage (STIRAP) [115, 116], electromagnetically induced transparency (EIT) [117, 118] and slow–light propagation [119, 120]. CPT has been observed not only in atomic systems but also in solid state emitters. In particular, CPT has been confirmed in self–assembled QDs for Zeeman split ground states of a single resident electron [121] or hole [122, 123]. Additionally, CPT has been reported in Nitrogen Vacancy centers in diamond [124].

2.2.3 Periodic pulsed laser excitation

The evolution of the central electron spin under cw excitation differs from the dynamics that results under periodic illumination by a laser pulse. We first introduce the optical basis which allows for a convenient description in the case of a circularly polarized laser pulse train. Then we derive the action of a pulse on an electron that is initially in the ground state. Finally, we briefly outline how certain electron Larmor precession frequencies are synchronized with the periodic
The excited state population vanishes when the two lasers satisfy the two–photon resonance condition. Here, the coupling laser detuning is kept constant at $\Delta \omega_c = 0$ (top) and $\Delta \omega_c = -0.75\Gamma$ (bottom), respectively, while the probe laser frequency is scanned.

The population excited state $|\uparrow \downarrow \uparrow\rangle$ is strongly reduced whenever $\Delta \omega_p = \Delta \omega_c$, i.e. $\delta = 0$. 

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Figure 2.6: Larmor precession under periodic pulsed excitation in the optical basis.

In the pulsed regime, we consider a resonant laser pulse train that is spectrally broad enough to couple all four transitions. Its pulse repetition period is denoted by $\tau$. To be specific, we assume the laser to be $\sigma_+$-polarized, thereby coupling $|\uparrow_x\rangle$ to $|\uparrow\downarrow\uparrow_x\rangle$ (Fig. 2.6), where the optical ($x$) basis is given by

$$
|\uparrow_x\rangle = \frac{1}{\sqrt{2}} (|\uparrow_z\rangle + |\downarrow_z\rangle),
$$

$$
|\downarrow_x\rangle = \frac{1}{\sqrt{2}} (|\uparrow_z\rangle - |\downarrow_z\rangle),
$$

$$
|\uparrow\downarrow\uparrow_x\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\uparrow_z\rangle + |\uparrow\downarrow\downarrow_z\rangle),
$$

$$
|\uparrow\downarrow\downarrow_x\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\uparrow_z\rangle - |\uparrow\downarrow\downarrow_z\rangle).
$$

The external magnetic field then couples $|\uparrow_x\rangle$ to $|\downarrow_x\rangle$ and $|\uparrow\downarrow\uparrow_x\rangle$ to $|\uparrow\downarrow\downarrow_x\rangle$. We note that not only the Larmor precession of the electron in self-assembled QD has been measured, but also the corresponding Larmor precession of the hole state [125].

In the following we will derive the wave function that describes the system after the action of a short, resonant, square laser pulse which satisfies

$$
t_p \ll \omega_c, \omega_t
$$

$$
\Delta = \omega - \omega_{te} \approx 0
$$

$$
g(t) = g[\Theta(t) - \Theta(t_p)],
$$

where $t_p$ is the pulse duration, $\omega$ is the laser frequency, $\omega_{te}$ is the optical transition frequency, $g$ describes the coupling strength and $\theta$ is the Heaviside step function. We thus neglect the Larmor precession during the action of the pulse. To avoid unphysical phase factors, we consider a quantum description of the laser field for
the moment and write the Hamiltonians as
\[
H_0 = \omega a^\dagger a + \omega_{\text{te}} (|\uparrow\downarrow\uparrow_x\rangle \langle \uparrow\downarrow\uparrow_x| + |\uparrow\downarrow\downarrow_x\rangle \langle \uparrow\downarrow\downarrow_x|)
\]
\[
H_{\text{int}} = g(a + a^\dagger) (|\uparrow\rangle \langle \uparrow \uparrow\uparrow_x| + |\uparrow\downarrow\downarrow_x\rangle \langle \uparrow_x|),
\]
where \(a\) and \(a^\dagger\) are the bosonic annihilation and creation operators of the quantized laser mode.

Going to the interaction picture, we find the corresponding interaction Hamiltonian
\[
H_I = e^{+iH_0 t} H_{\text{int}} e^{-iH_0 t}
\]
\[
\text{RWA, } \Delta = 0 = g(a^\dagger |\uparrow\rangle \langle \uparrow \uparrow\uparrow_x| + a |\uparrow\downarrow\uparrow_x\rangle \langle \uparrow_x|),
\]
where we applied the rotating wave approximation (RWA) and \(\Delta = 0\). We apply this Hamiltonian \(H_I\) on the initial wave function
\[
|\psi_I(t = 0)\rangle = \psi_{\uparrow}(0) |\uparrow_x, n\rangle + \psi_{\downarrow}(0) |\downarrow_x, n\rangle,
\]
in the Fock–basis where \(n\) is the photon number in the laser mode. Then we find the wave function at the end of the pulse duration \(t_p\) to be
\[
|\psi_I(t = t_p)\rangle = e^{-iH_0 t_p} |\psi_I(t = 0)\rangle
\]
\[
= \psi_{\uparrow}(0) \cos(g\sqrt{n t_p}) |\uparrow_x, n\rangle
+ \psi_{\downarrow}(0) \sin(g\sqrt{n t_p}) |\downarrow\uparrow\uparrow_x, n - 1\rangle.
\]
Transforming back to the Schrödinger picture, the final wave function reads
\[
|\psi(t = t_p)\rangle = e^{-iH_0 t_p} |\psi_I(t = t_p)\rangle
\]
\[
= \psi_{\uparrow}(0) \cos(g\sqrt{n t_p}) |\uparrow_x, n\rangle + \psi_{\downarrow}(0) |\downarrow_x, n\rangle
- i\psi_{\uparrow}(0) \sin(g\sqrt{n t_p}) |\downarrow\uparrow\uparrow_x, n - 1\rangle.
\]
after neglecting a global phase factor and setting \(e^{-i\Delta t_p \Delta = 0} \rightarrow 1\). In the limit of large photon numbers \(n\), where \(n - 1 \approx n\), we can replace \(g\sqrt{n}\) by the classical Rabi frequency \(\Omega\).

**Electron synchronization**

We now investigate the case of a periodic laser pulse train with the time–dependent Rabi frequency
\[
\Omega(t) = \sum_{k \in \mathbb{N}} \Omega [\Theta(t_k) - \Theta(t_k + t_p)],
\]
where \(t_k = (k - 1)\tau\) is the time at which the \(k\)th pulse arrives. An important characteristic of the final wave function (2.6) is that the amplitude in \(|\downarrow_x\rangle\) remains unaffected by the laser pulse. In particular, we now consider the case where the amplitude in \(|\downarrow_x\rangle\) at the arrival of the \(k\)th pulse is unity. Then, this is also true for the wave function at the end of the action of the laser pulse. In the time window that separates the pulses, the electron undergoes Larmor precession at the frequency \(\omega_e\). If the precession frequency is commensurable with the pulse repetition period, i.e.
\[
\omega_e \in \frac{2\pi}{\tau} \mathbb{Z}
\]
the amplitude in \(|\downarrow_x\rangle\) at the arrival of the \((k + 1)\)th pulse is unity and the optical absorption vanishes. Hence, the electron is synchronized with the laser pulse train.
2.2.4 Dissipation by spontaneous emission

The central electron spin experiences not only coherent dynamics, but also dissipative dynamics by coupling to the radiation field reservoir, resulting in spontaneous emission [126, 127]. In general, dissipative processes are incorporated into the density matrix equation of motion by the additional Lindblad term $\mathcal{L}_C$ of the form

$$\mathcal{L}_C(\rho) = \sum_i (C_i \rho C_i^\dagger - \frac{1}{2} \{C_i^\dagger C_i, \rho\}).$$

(2.8)

The Lindblad collapse operators $C_i$ describe the dissipative process and $\{A, B\} = AB + BA$ is the anti-commutator.

In Voigt geometry, the collapse operators in the $z$-basis are given by

$$C_{V,1} = \sqrt{\frac{\Gamma}{4}} \left| \uparrow \right\rangle \left\langle \downarrow \uparrow \uparrow \right|$$

$$C_{V,2} = \sqrt{\frac{\Gamma}{4}} \left| \downarrow \right\rangle \left\langle \downarrow \downarrow \right|$$

$$C_{H,1} = -i \sqrt{\frac{\Gamma}{4}} \left| \downarrow \right\rangle \left\langle \uparrow \uparrow \right|$$

$$C_{H,2} = -i \sqrt{\frac{\Gamma}{4}} \left| \uparrow \right\rangle \left\langle \downarrow \downarrow \right| .$$

If we consider a three-level system, then only $C_{V,1} = C_V$ and $C_{H,1} = C_H$ are included and the total decay rate of $\left| \uparrow \uparrow \uparrow \right\rangle_z$ is given by $\Gamma$. This is equivalent to the replacement of $\Gamma/2 \rightarrow \Gamma$ in $C_V$ and $C_H$.

Under periodic pulsed excitation, all four states are relevant. Since all four transitions have different transition energies, this would imply that we can track the decay channel by measuring the polarization state and the energy of the emitted photon. However, for fast measurements the uncertainty principle imposes a large energy uncertainty. This hinders the characterization of the decay channel through the energy measurement such that only the polarization can be extracted faithfully in a fast measurement. Two distinguishable collapse operators are then given by

$$C_V = \sqrt{\frac{\Gamma}{4}} \left( \left| \uparrow \right\rangle \left\langle \downarrow \uparrow \uparrow \right| + \left| \downarrow \right\rangle \left\langle \uparrow \downarrow \right| \right)$$

$$= \sqrt{\frac{\Gamma}{4}} \left( \left| \uparrow \right\rangle \left\langle \downarrow \uparrow \uparrow \right| + \left| \downarrow \right\rangle \left\langle \uparrow \downarrow \right| \right)$$

$$C_H = -i \sqrt{\frac{\Gamma}{4}} \left( \left| \downarrow \right\rangle \left\langle \uparrow \uparrow \right| + \left| \uparrow \right\rangle \left\langle \downarrow \uparrow \uparrow \right| \right)$$

$$= -i \sqrt{\frac{\Gamma}{4}} \left( \left| \downarrow \right\rangle \left\langle \uparrow \uparrow \right| + \left| \downarrow \right\rangle \left\langle \downarrow \uparrow \uparrow \right| \right),$$

where each operator is first written in the $z$- and then in the $x$-basis. If the polarization of the emitted light is measured in the circular basis, the collapse
operators are given by

\[ C_+ = \frac{1}{\sqrt{2}}(C_V + i C_H) = \sqrt{\frac{\Gamma}{2}} |\uparrow_x\rangle \langle \downarrow\downarrow x| \]

\[ C_- = \frac{1}{\sqrt{2}}(C_V - i C_H) = \sqrt{\frac{\Gamma}{2}} |\downarrow_x\rangle \langle \uparrow\downarrow x|. \]

The corresponding Lindblad superoperator \( \mathcal{L}_C \) does not depend on the polarization basis that is chosen for the measurement, as we can easily check

\[
\mathcal{L}_C(\rho) = (C_+ \rho C_+^\dagger - \frac{1}{2}\{C_+^\dagger C_+, \rho\}) + (C_- \rho C_-^\dagger - \frac{1}{2}\{C_-^\dagger C_-, \rho\})_+ \tag{2.9}
\]

\[
= \frac{1}{2}((C_V + i C_H)\rho(C_V + i C_H)^\dagger - \frac{1}{2}\{(C_V + i C_H)^\dagger(C_V + i C_H), \rho\}) \tag{2.10}
\]

\[
+ \frac{1}{2}((C_V - i C_H)\rho(C_V - i C_H)^\dagger - \frac{1}{2}\{(C_V - i C_H)^\dagger(C_V - i C_H), \rho\}) \tag{2.11}
\]

\[
= (C_V \rho C_V^\dagger - \frac{1}{2}\{C_V^\dagger C_V, \rho\}) + (C_H \rho C_H^\dagger - \frac{1}{2}\{C_H^\dagger C_H, \rho\}). \tag{2.12}
\]

For concreteness, we choose to work with the collapse operators in the circular basis.

### 2.2.5 Electron master equation in the absence of nuclear spins

To capture the evolution of the central electron spin under the combined influence of coherent and non-unitary (dissipative) processes, we introduce the Markovian master equation both for illumination by cw lasers and periodic laser pulses. Subsequently, we introduce the vectorization of the density matrix which is a useful tool for the numeric computation of the time evolution of the electron density matrix.

Under cw excitation, the dynamics is given by the Master equation [127, 126, 128]

\[
\dot{\rho} = \mathcal{L}(\rho) = -i[H_{\text{rot}}, \rho] + \mathcal{L}_C(\rho) \tag{2.13}
\]

\[
= -i[H_{\text{rot}}, \rho] + C_V \rho C_V^\dagger - \frac{1}{2}\{C_V^\dagger C_V, \rho\} + C_H \rho C_H^\dagger - \frac{1}{2}\{C_H^\dagger C_H, \rho\},
\]
2.2 The central electron spin

With $H_{\text{rot}}$ given by (2.5) and $L_C$ by (2.8).

To describe the evolution between two subsequent pulses of a laser pulse train, we replace $H_{\text{rot}}$ in (2.13) by $H_e$ given in (2.1), i.e.

$$\dot{\rho} = -i[H_e, \rho] + C_+ \rho C_+^\dagger - \frac{1}{2} \{C_+^\dagger C_+, \rho\} + C_- \rho C_-^\dagger - \frac{1}{2} \{C_-^\dagger C_-, \rho\}, \quad (2.14)$$

To compute the time evolution over many pulse periods, the electron density matrix is repeatedly evolved under the master equation and the action of the pulse. We note that equivalently, one may also use the Monte Carlo wave function approach to include the effect of dissipation [127].

**Vectorization**

For solving the equation of motion, it is often desirable to vectorize the density matrix. This results in a system of linear differential equations of the form $\dot{\vec{\rho}} = L \vec{\rho}$ [129]. Assuming that the density matrix is vectorized as

$$
\begin{pmatrix}
\rho_{11} & \rho_{12} & \cdots \\
\rho_{21} & \rho_{22} & \cdots \\
\vdots & \vdots & \ddots \\
\end{pmatrix}
\rightarrow
\begin{pmatrix}
\rho_{11} \\
\rho_{12} \\
\vdots \\
\rho_{21} \\
\rho_{22} \\
\vdots \\
\end{pmatrix},
$$

the Lindblad operator $L$ can be written as a matrix with enlarged dimensions, using the Kronecker product $\otimes$ [130], i.e.

$$
L = -i(H_{\text{rot}} \otimes 1^T - 1 \otimes H_{\text{rot}}^T) + C_V \otimes (C_V^T)^T - \frac{1}{2} (C_V^T C_V \otimes 1^T + 1 \otimes (C_V^T C_V)^T) + C_H \otimes (C_H^T)^T - \frac{1}{2} (C_H^T C_H \otimes 1^T + 1 \otimes (C_H^T C_H)^T).
$$

The equation of motion for the vectorized density matrix $\vec{\rho}$ is then given by

$$
\frac{\partial}{\partial t} \vec{\rho} = L \vec{\rho},
$$

where $\vec{\rho}$ is now multiplied from the left by the matrix $L$. Therefore, the time evolution of $\vec{\rho}(t)$ can be easily calculated numerically, using

$$
\vec{\rho}(t) = e^{Lt} \vec{\rho}(0).
$$

Alternatively, if only the steady state is of interest, one can solve the linear system of equations given by

$$
L \vec{\rho} = 0
$$

under the assumption of $\text{Tr}\rho = 1$. 


2 The quantum dot central spin model

2.3 The nuclear environment

The number of nuclei that constitute the self-assembled QDs under investigation is on the order of $N \approx 10^4 - 10^6$, but can vary considerably from dot to dot. The QDs are composed of an In$_{1-x}$Ga$_x$As alloy. The natural abundances are given in table 2.1.

In this thesis, the spin of the QD nuclei plays an important role. While the nuclear spin of As and both Ga isotopes is $I_{\text{As}} = I_{\text{Ga}} = \frac{3}{2}$, the spin of the two In isotopes is $I_{\text{In}} = \frac{9}{2}$ [131]. For simplicity, most calculations in this thesis are performed for nuclear spins $I = \frac{1}{2}$. In the case of the hyperfine interaction described in section 2.4, the nature of the coupling remains unaltered by this replacement. We therefore capture the essential features of the nuclear dynamics while the description and computation are tractable. This replacement is more delicate in the case of quadrupolar (see section 2.3.3) and non-collinear interactions (see section 2.4.2) since they require a non-vanishing quadrupole moment of the nuclei. This is only possible for $I > \frac{1}{2}$. However, for $I = \frac{3}{2}$ and the quadrupolar interaction we consider only couples $m = \pm\frac{3}{2}$ to $m = \mp\frac{1}{2}$, where $m$ is the spin projection along $z$. Hence, there are two independent effective two-level systems, i.e. $(m = \frac{3}{2}, m = -\frac{1}{2})$ and $(m = -\frac{3}{2}, m = +\frac{1}{2})$ (see section 2.3.3 and [132]). Then, the reduction to a spin-$\frac{1}{2}$ system corresponds to proper rescaling of the coupling constants. For $I = \frac{9}{2}$ this argument fails; we provide a justification for the specific case considered in chapter 4.

We start by describing the interaction of the nuclear spin environment with the external magnetic field, which induces a Zeeman splitting of the different nuclear spin states. Then we introduce the mutual dipolar interactions of the nuclear spins. However, in self-assembled QDs, the strain-driven growth leads to quadrupolar interactions, which strongly suppress the effect of the dipolar coupling. Finally, we introduce collective nuclear spin states which frequently form a convenient basis for the states of the nuclear environment. Furthermore, they allow us to illustrate certain properties of the spin environment that are experimentally relevant.

2.3.1 Nuclear Zeeman interaction

Similar to the central electron spin, the nuclear spin environment interacts with the external magnetic field. In the absence of any other interactions, in particular the quadrupolar interactions discussed below, the magnetic field defines the quantization axis for the nuclear spins. The Zeeman Hamiltonian for the nuclear spins is thus given by

$$H_n = -\sum_j \omega_n^j I_z^j,$$  \hspace{1cm} (2.15)

where the Zeeman frequency is given by $\omega_n = \gamma_n B$. The gyromagnetic ratio $\gamma_n$ of the different isotopes are given in table 2.1.
2.3 The nuclear environment

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$^{69}$Ga</th>
<th>$^{71}$Ga</th>
<th>$^{75}$As</th>
<th>$^{113}$In</th>
<th>$^{115}$In</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural abundance (%)</td>
<td>60.1</td>
<td>39.9</td>
<td>100</td>
<td>4.3</td>
<td>95.7</td>
</tr>
<tr>
<td>Gyromagnetic ratio ($10^7 \text{ rad} \text{T s}$)</td>
<td>6.44</td>
<td>8.18</td>
<td>4.60</td>
<td>5.90</td>
<td>5.88</td>
</tr>
</tbody>
</table>

Table 2.1: Natural abundances and gyromagnetic ratios of typical isotopes in QDs [131]

2.3.2 Dipolar interactions

Since magnetic dipoles interact with each other, it is expected that the nuclear spin magnetic moments give rise to a dipolar coupling of the form [133]

$$H_{\text{dip}} = \frac{\mu_0}{4\pi} \sum_{i \neq j} \frac{\gamma_i \gamma_j}{2r_{ij}^3} \left( \vec{I}_i \cdot \vec{I}_j - \frac{3(\vec{I}_i \cdot \vec{r}_{ij})(\vec{I}_j \cdot \vec{r}_{ij})}{r_{ij}^2} \right).$$

The dipolar interaction is strongly dependent on the distance between two nuclear spins. The dipole Hamiltonian $H_{\text{dip}}$ can be split into a spin–conserving (secular) and spin–non–conserving (nonsecular) part. On the one hand at low magnetic fields the nonsecular part leads to nuclear spin depolarization by transfer of spin angular momentum to the crystal lattice. On the other hand the secular terms allow for nuclear spin flip–flops between nuclear spins of the same species but at different sites. Thus it redistributes the nuclear spin polarization in the QD [134]. However, if the quadrupolar interaction, which is discussed below, induces large enough inhomogeneous shifts, the dipolar coupling is suppressed [135]. We will therefore neglect the dipolar coupling among nuclear spins in this thesis.

2.3.3 Quadrupolar interactions

In this section we consider the Coulomb interaction between the charge distribution $\rho(\vec{r})$ of a QD nucleus and the electric potential $V(\vec{r})$ created by the lattice. We are particularly interested in the gradients of the electric field that arise from breaking the cubic symmetry in QDs due to strain or interdiffusion of Ga and In [135].

First we consider the classical electrostatic energy $U$, given by

$$U = \int \rho(\vec{r})V(\vec{r}) \, dx \, dy \, dz.$$  

Performing a multipole expansion of the potential $V$ yields

$$V(\vec{r}) = V(0) + \sum_\alpha x_\alpha \left( \frac{\partial V}{\partial x} \right)_{r=0} + \frac{1}{2} \sum_{\alpha,\beta} x_\alpha x_\beta \left( \frac{\partial^2 V}{\partial x_\alpha \partial x_\beta} \right)_{r=0} + \ldots,$$

where $\alpha, \beta = x, y, z$. The resulting energy $U$ can be cast into a convenient form by defining $q$ to be the total charge of the nucleus, $\vec{E}(0)$ the electric field created by the lattice at the position of the nucleus, and $\vec{p}(\vec{r}) = \int \rho(\vec{r})(\vec{r} - \vec{r}') \, dV'$ the electric dipole moment [136]:

$$U = qV(0) - \vec{p} \cdot \vec{E}(0) + \frac{1}{6} \sum_{\alpha,\beta} Q_{\alpha\beta} \left( \frac{\partial^2 V}{\partial x_\alpha \partial x_\beta} \right)_{r=0},$$
where $Q_{\alpha\beta}$ are the elements of the quadrupole tensor of the nucleus and are given by

$$Q_{\alpha\beta} = \int (3x_\alpha x_\beta - r^2 \delta_{\alpha\beta}) \, dV'.$$

Note that we have assumed the density of the charges that create $V$ to vanish at the site of the nucleus. This leads to the Laplace equation $\sum_\alpha V_\alpha(0) = 0$ that we have directly employed to rewrite the quadrupolar term.

The nuclei have zero electric dipole moment, but have a finite quadrupole moment for nuclear spins $I > \frac{1}{2}$. Hence, in the quantum mechanical description, the lowest order Hamiltonian of interest is the quadrupolar Hamiltonian. For an axially symmetric potential, this quadrupolar Hamiltonian can be written as [132, 137]

$$H_Q = \frac{\omega_Q}{2} \sum_j ((I^j_z)^2 - \frac{I^j(I^j + 1)}{3} \mathbb{I}^j),$$

where $\omega_Q$ gives the quadrupolar energy and $z'$ is the principal axis.

In the following we assume that the principal axis $z'$ is along the growth direction $x$ [132]. The quadrupolar Hamiltonian can then be split into a diagonal and off–diagonal part, where the latter couples spin states with different total spin projection along $z$ (Fig. 2.8). This yields

$$H_Q = \frac{\omega_Q}{2} \sum_j ((I^j_z)^2 - \frac{I^j(I^j + 1)}{3} \mathbb{I}^j)$$

\[=\]

$$= \frac{\omega_Q}{2} \sum_j \left( \frac{1}{2} ((I^j)^2 - (I^j_z)^2) - \frac{I^j(I^j + 1)}{3} \mathbb{I}^j \right) + \frac{\omega_Q}{8} \sum_j ((I^j_+)^2 + (I^j_-)^2).$$

The diagonal part $H_Q^D$ introduces energy shifts that give rise to inhomogeneous level spacings. This leads to strong suppression of nuclear spin diffusion via the nuclear dipolar interaction [135]. The off–diagonal term $H_Q^O$ induces coherent rotation of the nuclear spin projection, which is however reduced at high magnetic fields through the nuclear Zeeman splitting $\omega_n$. These off–diagonal terms play an important role in the nuclear spin dynamics in a single electron charged self–assembled QD and will be discussed in more detail in section 2.4.2 and chapter 4.

### 2.3.4 Collective nuclear spin states

Since the collective properties of $N$ nuclear spins are of interest, it can be useful to work with the following collective nuclear spin operators, satisfying

$$I_{x,y,z} = \sum_j I^j_{x,y,z},$$

$$I_\pm = I_x \pm iI_y,$$

$$I^2 = I_x^2 + I_y^2 + I_z^2.$$
2.3 The nuclear environment

Figure 2.8: Inhomogeneous shifts of the nuclear Zeeman levels and couplings of different total angular momentum projection states.

In analogy to atomic Dicke states \[138\] we introduce nuclear Dicke states \( |J, m\rangle \), which are simultaneous eigenstates of \( I_z \) and \( I^2 \). They satisfy

\[
I_z |J, m\rangle = m |J, m\rangle ,
I^2 |J, m\rangle = J(J + 1) |J, m\rangle ,
I_\pm |J, m\rangle = \sqrt{J(J + 1) - m(m \pm 1)} |J, m \pm 1\rangle ,
\]

where \( m \) is the projection of the nuclear spin polarization along the \( z \)-direction. \( J \) runs from 0 to \( N I \) where \( I \) is the spin of a single nucleus. For a given \( J \), it holds that \( |m| \leq J \). Evidently, the effect of the collective raising and lowering operators \( I_\pm \) is to raise or lower the collective spin projection \( m \) by one unit, while leaving \( J \) fixed. Thus, if the Hamiltonian of interest can be written as a function of \( I_\pm, I_z \), the nuclear spin state will remain in the initial \( J \)-manifold and in each manifold \( m_J \) evolves independently. An example for this is the hyperfine interaction in the case of homogeneous coupling constant, which is the subject of section 2.4.

Even though the following discussion can be generalized to particles with \( I > \frac{1}{2} \) \[139\], the expressions become more cumbersome and offer no further insight. We therefore limit the following analysis to spin-\( \frac{1}{2} \) particles. For \( N \) nuclei with spin \( I = \frac{1}{2} \), there are a total of \( 2^N \) states. Hence, the nuclear states \( |J, m\rangle \) are in general degenerate. This is also clear if one considers the criterion of adding an angular momenta \( I_1 \) to \( I_2 \): The total \( J \) has to satisfy \( I_1 + I_2 \geq J \geq |I_1 - I_2| \). To illustrate this, we consider the simple example of consecutively adding the angular momenta of four spin-\( \frac{1}{2} \) particles. For \( I_1 = I_2 = \frac{1}{2} \) we find

\[
J = \begin{cases} 
I_1 + I_2 = 1, \\
I_1 - I_2 = 0.
\end{cases}
\]
Adding a third spin $I_3 = \frac{1}{2}$ yields

$$J = \begin{cases} \frac{1}{2}(I_1 + I_2) + I_3 = 3, \\ \frac{1}{2}(I_1 + I_2) - I_3 = \frac{1}{2}, \\ \frac{1}{2}(I_1 - I_2) + I_3 = \frac{1}{2}. \end{cases}$$

And finally, for the fourth spin $I_4 = \frac{1}{2}$ we find the angular momenta

$$J = \begin{cases} \frac{1}{2}(I_1 + I_2 + I_3) + I_4 = 2, \\ \frac{1}{2}(I_1 + I_2 + I_3) - I_4 = 1, \\ \frac{1}{2}(I_1 + I_2 - I_3) + I_4 = 1, \\ \frac{1}{2}(I_1 + I_2 - I_3) - I_4 = 0, \\ \frac{1}{2}(I_1 - I_2 + I_3) + I_4 = 1, \\ \frac{1}{2}(I_1 - I_2 + I_3) - I_4 = 0. \end{cases}$$

Hence, there are different ways of obtaining $J = 1, 0$, making these subspaces degenerate. The different $J$-subspaces of this example are depicted in Fig. 2.9. More generally, the degeneracy $D_J$ of each $J$ manifold can be shown to be [138]

$$D_J = \frac{(2J + 1)N!}{(N/2 + J + 1)!(N/2 - J)!}.$$ 

For a fixed $J$, all $2J + 1$ states $|J, m\rangle$ with the same degeneracy $D_J$ can be generated by repeatedly applying $I_{\pm}$. Hence the total number of nuclear spin states in a given $J$-manifold is

$$N_J = (2J + 1)D_J = \frac{(2J + 1)^2N!}{(N/2 + J + 1)!(N/2 - J)!}.$$ 

If every state is occurring with the same probability $2^{-N}$, the probability of being
2.4 The electron–nuclei interactions

in a certain $J$-manifold is given by

$$p(J) = \frac{N_J}{2^N} = \frac{1}{2^N} \frac{(2J + 1)^2 N!}{(N/2 + J + 1)!(N/2 - J)!}$$

$$\overset{N \to \infty}{\to} \frac{8J^2/N}{\sqrt{2\pi \sigma^2}} e^{-\frac{J^2}{2\sigma^2}},$$

where $\sigma^2 = \frac{N}{4}$. Here we have used the central limit theorem for the binomial distribution $\mathcal{B}$

$$\mathcal{B}(k; n, p) = \binom{n}{k} p^k (1-p)^{n-k}$$

$$\overset{n \to \infty}{\to} \frac{1}{\sqrt{2\pi np(1-p)}} \exp\left(-\frac{(k-np)^2}{2np(1-p)}\right) = \mathcal{N}(np, np(1-p)),$$

where $\mathcal{N}(\mu, \sigma^2)$ denotes the normal distribution with mean $\mu$ and variance $\sigma^2$.

Furthermore we note that the number of states with a given nuclear spin polarization $m$ is

$$B_m = \frac{N!}{(N/2 + m)!(N/2 - m)!}.$$ 

Under the assumption that any nuclear spin configuration is equally likely to occur, we find the probability for a given nuclear spin polarization $m$ to be

$$p(m) = \frac{B_m}{2^N} = \frac{1}{2^N} \frac{N!}{(N/2 + m)!(N/2 - m)!}$$

$$\overset{N \to \infty}{\to} \frac{1}{\sqrt{2\pi \sigma^2}} e^{-\frac{m^2}{2\sigma^2}},$$

where we have applied (2.19). This means that in a statistical ensemble, the nuclear polarization $m$ is distributed according to a Gaussian with mean $m = 0$ and standard deviation $\sigma = \sqrt{N/4}$.

It is also instructive to compute the conditional probability of $p(J|m)$. Assuming that the system is in a state with total spin projection $m$, $p(J|m)$ is a measure of how likely this state is realized in a certain $J$ subspace (Fig. 2.10). In particular, for relatively large nuclear spin projections, the relevant $J$ subspaces for a given polarization $m$ are those that satisfy $J \approx m$.

2.4 The electron–nuclei interactions

In this section we describe the dominant interactions between the central electron spin and the surrounding nuclear spins that constitute the spin environment. The most important interaction is the isotropic Fermi–contact hyperfine interaction, which is chiefly responsible for many of the striking experiments connected to nuclear spin effects in QDs. Due to its significance for the phenomena investigated in this thesis, we not only discuss the effect of the hyperfine interaction on the dynamics of the QD system but also provide a detailed derivation starting from the Dirac equation for the QD electron. Then we discuss the non–collinear hyperfine Hamiltonian which arises in self–assembled QDs with a large off–axis strain due to the interplay of the nuclear quadrupolar interactions with the hyperfine interaction.
2 The quantum dot central spin model

Figure 2.10: The conditional probability $p(J|m)$ shows that for large polarizations $m$, only subspaces that satisfy $J \approx m$ contribute.

2.4.1 Hyperfine interaction

In self–assembled QDs we are mainly concerned with interactions of the central electron with the environmental nuclei via the spin. However, spin is a concept that has to be introduced by hand into the Schrödinger equation in order to obtain agreement between theory and experiments such as the Zeeman effect. Therefore, the starting point of the discussion is the Dirac equation, which reconciles quantum mechanics and special relativity [140]. In the non–relativistic limit the Dirac equation reduces to the Pauli equation and spin comes out naturally. In what follows we start with the Dirac equation in the presence of electromagnetic fields. In particular, we will show the non–relativistic limits in which the electron and positron decouple. Then we specify the electromagnetic fields entering the Dirac equation to be the electric and magnetic field induced by a QD nucleus. This allows us to derive the coupling terms of the central electron in a QD with a nucleus. In a next step, we focus on the most relevant spin interaction terms and explicitly derive the effective Hamiltonian for the dominant term, the Fermi–contact hyperfine interaction. Lastly, we generalize the effective Hamiltonian to an ensemble of QD nuclear spins. The resulting Hamiltonian is simply referred to as the hyperfine Hamiltonian $H_{\text{hyp}} = \sum_j A_j \vec{S} \cdot \vec{I}_j$ and is essential for the phenomena discussed in this thesis.
2.4 The electron–nuclei interactions

Dirac equation

The Dirac equation for the electron is given by

$$i \frac{\partial \psi(\vec{r}, t)}{\partial t} = H_D \psi(\vec{r}, t) = (c\vec{\alpha} \cdot \vec{p} + \beta mc^2)\psi(\vec{r}, t), \quad (2.19)$$

where

$$\vec{\alpha} = \left( \begin{array}{ccc} 0 & \sigma_x & 0 \\ \sigma_x & 0 & \sigma_y \\ 0 & \sigma_y & 0 \end{array} \right), \quad \beta = \left( \begin{array}{cc} 1/2 & 0 \\ 0 & -1/2 \end{array} \right) \quad \text{(2.20)}$$

and $\mathbb{1}_2$ being the two–dimensional identity matrix and $\sigma_x, \sigma_y, \sigma_z$ are the Pauli matrices.

The $\vec{\alpha}$ and $\beta$ matrices act on the four–dimensional spinor

$$\psi(\vec{r}, t) = \left( \begin{array}{c} \psi_1^1(\vec{r}, t) \\ \psi_1^2(\vec{r}, t) \\ \psi_2^1(\vec{r}, t) \\ \psi_2^2(\vec{r}, t) \end{array} \right), \quad (2.22)$$

where $\psi_1$ is a two–component spinor representing the electron (particle) and $\psi_2$ stands for the positron (antiparticle). Each of these wave functions has again two components for the two orientations of spin. Note that the Dirac equation (2.19) couples the particle and antiparticle through the $\vec{\alpha}$ matrices.

Similar to the Schrödinger equation, one can separate the time and space coordinates in the Dirac equation. We write

$$\psi(\vec{r}, t) = \psi(\vec{r}) e^{-iWt} = \left( \begin{array}{c} \psi_1^1(\vec{r}) \\ \psi_1^2(\vec{r}) \\ \psi_2^1(\vec{r}) \\ \psi_2^2(\vec{r}) \end{array} \right) e^{-iWt}, \quad (2.23)$$

where $W = \sqrt{p^2c^2 + m^2c^4} = c\vec{\alpha} \cdot \vec{p} + \beta mc^2$ and $\psi(\vec{r})$ now satisfies the time-independent Dirac equation

$$H_D \psi(\vec{r}) = (c\vec{\alpha} \cdot \vec{p} + \beta mc^2)\psi(\vec{r}) = W\psi(\vec{r}), \quad (2.24)$$

which we will use below.

Dirac equation with nuclear electric and magnetic fields

In the presence of an electromagnetic field we replace $\vec{p} \rightarrow \vec{\pi}$, where $\vec{\pi} = \vec{p} - e\vec{A}$ with the vector potential $\vec{A}$ such that $\vec{B} = \nabla \times \vec{A}$. The scalar potential energy $V(\vec{r}) = e\Phi(\vec{r})$ with $\vec{E}(\vec{r}) = -\nabla \Phi(\vec{r})$ is added to the Hamiltonian which now reads

$$H_D = c\vec{\alpha} \cdot \vec{\pi} + \beta mc^2 + V(\vec{r}) = c\vec{\alpha} \cdot (\vec{p} - e\vec{A}) + \beta mc^2 + V(\vec{r}). \quad (2.25)$$

We specify the electric field $\vec{E}$ and the vector potential $\vec{A}$ to be induced by the QD nuclei. Then, we separate the electron and positron equations, thereby
deriving the desired terms that describe the coupling of the electron spin $\vec{\sigma}$ to these nuclear electric and magnetic fields. The resulting time–independent equations are given by

$$W\psi_1 = c\vec{\pi} \cdot \vec{\sigma}\psi_2 + (V(\vec{r}) + mc^2)\psi_1$$

$$W\psi_2 = c\vec{\pi} \cdot \vec{\sigma}\psi_1 + (V(\vec{r}) - mc^2)\psi_2.$$  \hspace{1cm} (2.26)

We isolate the positron wave function $\psi_2$ in equation (2.27) and find

$$\psi_2 = \frac{c}{W + mc^2 - V(\vec{r})}\vec{\pi} \cdot \vec{\sigma}\psi_1.$$ \hspace{1cm} (2.28)

Next we substitute the expression (2.28) for $\psi_2$ in equation (2.26). This yields an effective Hamiltonian $H'_1$ for the electron wave function $\psi_1$ such that

$$H'_1\psi_1 = W\psi_1$$

with

$$H'_1 = \vec{\pi} \cdot \vec{\sigma} \frac{c^2}{W + mc^2 - V(\vec{r})}\vec{\pi} \cdot \vec{\sigma} + (V(\vec{r}) + mc^2). \hspace{2cm} (2.29)$$

It is convenient to introduce the energy $\epsilon$ on top of $mc^2$ such that

$$W = \epsilon + mc^2.$$ \hspace{1cm} (2.30)

For further manipulations we introduce

$$F = \frac{c^2}{\epsilon + 2mc^2 - V(\vec{r})}$$ \hspace{2cm} (2.31)

and hence

$$H_1 = \vec{\pi} \cdot \vec{\sigma} F\vec{\pi} \cdot \vec{\sigma} + V(\vec{r}). \hspace{2cm} (2.32)$$

Before we take a closer look at $H_1$ we note that $\vec{\pi} = \vec{p} - e\vec{A}$ and

$$\frac{\partial F}{\partial x} = \frac{\partial}{\partial x} \left( \frac{c^2}{\epsilon + 2mc^2 - V(\vec{r})} \right) = \frac{c^2}{(\epsilon + 2mc^2 - V(\vec{r}))^2} \frac{\partial V}{\partial x} = -\frac{c^2e}{(\epsilon + 2mc^2 - V(\vec{r}))^2} E_x,$$

since $\vec{E} = -\frac{1}{e} \nabla V$ and the potential energy $V$ is $V = e\Phi$. Generalizing, this yields

$$\nabla F = -\frac{c^2e}{(\epsilon + 2mc^2 - V(\vec{r}))^2} \vec{E}. \hspace{2cm} (2.33)$$

The two types of terms found in $H_1 = \vec{\pi} \cdot \vec{\sigma} F\vec{\pi} \cdot \vec{\sigma} + V(\vec{r})$ are of the form

1. $T_1 = \pi_x \sigma_x F\pi_x \sigma_x = \pi_x F\pi_x$

2. $T_2 = \pi_x \sigma_x F\pi_y \sigma_y + \pi_y \sigma_y F\pi_y \sigma_x = i\sigma_z(\pi_x F\pi_y - \pi_y F\pi_x).$
The goal is to rewrite $H_1$ in a form where the nuclear electric field $\vec{E}$ and nuclear vector potential $\vec{A}$ enter explicitly. This allows for a physical interpretation of the interaction terms between the central electron spin and nuclear spins. To this end, we write out the two types of terms of $H_1$ explicitly and formulate them as a function of $F$ and $\nabla F$. Using $p_x = -i \frac{\partial}{\partial x}$ this yields

1. $$T_1 = (p_x - eA_x)F(p_x - eA_x)$$
   $$= -\left(\frac{\partial F}{\partial x}\right)\frac{\partial}{\partial x} - F\frac{\partial^2}{\partial x^2} + ie(A_x F \frac{\partial}{\partial x} + F \frac{\partial A_x}{\partial x}) + e^2A_x^2F$$
   $$= -i(\nabla F \cdot \vec{p})_x + Fp_x^2 - eF(\vec{A} \cdot \vec{p})_x + ie(\nabla F \cdot \vec{A})_x + ieF(\nabla \cdot \vec{A})_x + e^2FA_x^2$$

2. $$T_2 = i\sigma_z((p_x - eA_x)F(p_y - eA_y) - (p_y - eA_y)F(p_x - eA_x))$$
   $$= i\sigma_z\left(-\left(\frac{\partial F}{\partial y}\right)\frac{\partial}{\partial y} - F\frac{\partial^2}{\partial y^2} + ie(A_y F \frac{\partial}{\partial y} + F \frac{\partial A_y}{\partial y}) + e^2A_y^2F\right)$$
   $$= i\sigma_z(-i(\nabla F \times \vec{p})_z - eF(\vec{A} \times \vec{p})_z + ieF(\nabla \times \vec{A})_z + ie(\nabla \times \vec{A})_z)$$

Now we sum all terms and group them depending on whether they contain $F$ or $\nabla F$.

$$H_1 = F \left(p^2 + e^2A^2 - e\vec{A} \cdot \vec{p} + ie\nabla \cdot \vec{A} - ie\bar{\sigma} \cdot (\vec{A} \times \vec{p}) - e\bar{\sigma} \cdot (\nabla \times \vec{A})\right)$$
   $$+ \left(-i\nabla F \cdot \vec{p} + ie\nabla \cdot \vec{A} + \bar{\sigma} \cdot (\nabla F \times \vec{p}) - e\bar{\sigma} \cdot (\nabla \times \vec{A})\right) + V(\vec{r})$$

Substituting $F$ and $\nabla F$ yields $H_1$ in a form that contains the electric field $\vec{E}$ and vector potential $\vec{A}$ created by a nucleus explicitly, i.e.

$$H_1 = \frac{c^2}{\epsilon + 2mc^2 - V(\vec{r})}(p^2 + e^2A^2 - e\vec{A} \cdot \vec{p} + ie\nabla \cdot \vec{A} - ie\bar{\sigma} \cdot (\vec{A} \times \vec{p}) - e\bar{\sigma} \cdot (\nabla \times \vec{A}))$$
   $$- \frac{c^2e}{(\epsilon + 2mc^2 - V(\vec{r}))^2}(-i\vec{E} \cdot \vec{p} + ie\vec{E} \cdot \vec{A} + \bar{\sigma} \cdot (\vec{E} \times \vec{p}) - e\bar{\sigma} \cdot (\vec{E} \times \vec{A})) + V(\vec{r})$$

There are several terms in $H_1$ that couple the electron spin to the nuclei, including the spin–orbit $H_{so}$, isotropic $H_{ihf}$ and anisotropic $H_{ahf}$ hyperfine interactions [141]

$$H_{so} = -\frac{c^2e}{(\epsilon + 2mc^2 - V(\vec{r}))^2}\bar{\sigma} \cdot (\vec{E} \times \vec{p})$$
$$H_{ihf} = \frac{c^2e^2}{(\epsilon + 2mc^2 - V(\vec{r}))^2}\bar{\sigma} \cdot (\vec{E} \times \vec{A})$$
$$H_{ahf} = -\frac{e^2c^2}{(\epsilon + 2mc^2 - V(\vec{r}))^2}\bar{\sigma} \cdot (\nabla \times \vec{A})$$

While the spin–orbit interaction is suppressed in QDs due to the localization of the electron [99], the anisotropic hyperfine interaction is negligible compared to the isotropic hyperfine interaction for a s-type conduction electron [141]. Hence, from now on we focus on the isotropic hyperfine interaction $H_{ihf}$.
2 The quantum dot central spin model

Effective hyperfine Hamiltonian

Next we derive an effective Hamiltonian for the inhomogeneous hyperfine interaction between a nucleus and a QD electron with s–type wave function, where the starting point is $H_{\text{hf}}$. We substitute the expressions for $V(\vec{r})$, $\vec{E}$, $\vec{A}$ induced by the nucleus. Then we integrate over the spatial part of the electron wave function to obtain a Hamiltonian that contains only spin operators. The electric potential, electric field and vector potential created by the nucleus are given by

$$V(\vec{r}) = -\frac{1}{4\pi\epsilon_0} \frac{Ze^2}{r}$$ (2.34)

$$\vec{E} = -\frac{1}{4\pi\epsilon_0} \frac{Ze\vec{r}}{r^3}$$ (2.35)

$$\vec{A} = \frac{\mu_0}{4\pi} \mu_N \times \frac{\vec{r}}{r^3},$$ (2.36)

where $\mu_N = \gamma_n \vec{I}$ is the nuclear magnetic moment and $Z$ is the effective nuclear charge. Substituting these expressions, we obtain

$$H_{\text{hf}} = -\frac{\mu_0}{4\pi} \frac{1}{4\pi\epsilon_0} \left(\frac{c^2e^2}{\epsilon + 2mc^2} + \frac{1}{4\pi\epsilon_0} \frac{Ze^2}{r}\right) \frac{Z}{r^6} \vec{\sigma} \cdot (\vec{r} \times (\mu_N \times \vec{r}))$$

$$= -\frac{\mu_0}{4\pi} \frac{1}{4\pi\epsilon_0} \left(\frac{Ze^2}{r^2} \vec{\sigma} \cdot \vec{\mu}_N - (\vec{\sigma} \cdot \vec{r})(\vec{\mu}_N \cdot \vec{r})ight),$$

where we have used the vector triple product identity to rewrite

$$\vec{\sigma} \cdot (\vec{r} \times (\mu_N \times \vec{r})) = \vec{\sigma} \cdot (\vec{\mu}_N (\vec{r} \times \vec{r}) - \vec{r} (\vec{\mu}_N \cdot \vec{r})) = r^2 (\vec{\sigma} \cdot \vec{\mu}_N - (\vec{\sigma} \cdot \vec{r})(\vec{\mu}_N \cdot \vec{r})),$$

and $\vec{r} = \frac{\vec{r}}{|\vec{r}|}$.

Following [142], we introduce the parameter

$$d = \frac{1}{4\pi\epsilon_0} \frac{Ze^2}{2mc^2}$$ (2.37)

and rewrite $H_{\text{hf}}$

$$H_{\text{hf}} = \frac{\mu_0 \mu_B}{4\pi} \frac{d}{(1 + \frac{\epsilon}{2mc^2} + \frac{d}{2})^2} \frac{\vec{\sigma} \cdot \vec{\mu}_N - (\vec{\sigma} \cdot \vec{r})(\vec{\mu}_N \cdot \vec{r})}{r^4},$$ (2.38)

where we introduced the Bohr magneton $\mu_B = \frac{e}{2m}$ when $e < 0$ and $\hbar = 1$.

Now we integrate over the spatial part of the electron s–wave function $\psi_s$ to extract an effective Hamiltonian $H_{\text{hf}}^{\text{eff}}$ that only acts on the spin part of the electron wave function. Without loss of generality, we assume that $\vec{\mu}_N = \mu_N (0, 0, 1)$ defines the z-axis

$$H_{\text{hf}}^{\text{eff}} = \int \int \int \psi_s^* H_{\text{hf}} \psi_s dV$$

$$= \frac{\mu_0 \mu_B}{4\pi} \int_0^\infty \int_0^{2\pi} \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \psi_s^* \left[1 + \frac{\epsilon}{2mc^2} + \frac{d}{2}\right] \frac{d}{(1 + \frac{\epsilon}{2mc^2} + \frac{d}{2})^2} \frac{\vec{\sigma} \cdot \vec{\mu}_N - (\vec{\sigma} \cdot \vec{r})(\vec{\mu}_N \cdot \vec{r})}{r^4} \psi_s r^2 \cos \theta \, d\theta d\phi dr$$

$$= \frac{\mu_0 \mu_B}{4\pi} \int_0^\infty \int_0^{2\pi} \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \psi_s (r)^2 \frac{\vec{\sigma} \cdot \vec{\mu}_N - (\vec{\sigma} \cdot \vec{r})(\vec{\mu}_N \cdot \vec{r})}{r^4} \cos \theta \, d\theta d\phi dr$$

$$\approx \frac{\mu_0 \mu_B}{4\pi} f(\vec{r})|_{r=0}.$$
The last line follows because the radial integral is controlled by \( r < d \). Since \( d \) is a small quantity compared to the length scale over which the electron wave function changes considerably, \( f(\vec{r}) \) reduces to the modulus of the electron wave function at the site of the nucleus squared multiplied by the angular integral

\[
\int_0^{2\pi} \int_{-\pi/2}^{\pi/2} (\sigma \cdot \mu_N - (\hat{\sigma} \cdot \hat{r})(\mu_N \cdot \hat{r})) \cos \theta \, d\theta d\phi
\]

\[
= \int_0^{2\pi} \int_{-\pi/2}^{\pi/2} (\mu_N \sigma_z - (\sigma_x \cos \theta \cos \phi + \sigma_y \cos \theta \sin \phi + \sigma_z \sin \theta)(\mu_N \sin \theta)) \cos \theta \, d\theta d\phi
\]

\[
= 4\pi \mu_N \sigma_z - \int_0^{2\pi} \int_{-\pi/2}^{\pi/2} (\sigma_z \sin \theta)(\mu_N \sin \theta) \cos \theta \, d\theta d\phi
\]

\[
= \frac{8\pi}{3} \sigma_z \mu_N,
\]

where we have used \( \hat{r} = (\cos(\theta) \cos(\phi), \cos(\theta) \sin(\phi), \sin(\theta)) \) in spherical coordinates. Plugging in \( \vec{S} = \frac{1}{2} \sigma \) and \( \mu_N = \gamma_n \vec{I} \) yields the effective Hamiltonian for the isotropic hyperfine interaction

\[
H_{\text{eff}}^{\text{hf}} = \frac{4}{3} \mu_0 \mu_B \gamma_n |\psi_s(0)|^2 \vec{S} \cdot \vec{I}.
\]

To generalize to many nuclear spins we sum over the contribution of the individual nuclear spins

\[
H_{\text{hyp}} = \sum_j A_j \vec{S} \cdot \vec{I}_j,
\]

with

\[
A_j = \frac{4}{3} \mu_0 \mu_B \gamma_j |\psi_s(\vec{R}_j)|^2,
\]

where \( \vec{R}_j \) and \( \gamma_j \) are the position and gyromagnetic ratio of the \( j \)th nuclear spin, respectively. Evaluating \( |\psi_s(\vec{R}_j)|^2 \) for harmonic confinement of the electron in all three dimensions yields after normalization \([143, 108]\)

\[
|\psi(R_j)|^2 = \frac{1}{(\sqrt{\pi}l_0)^3} e^{-(R_j/l_0)^2},
\]

where \( l_0 \) is the confinement radius of the QD and \( R_j = |\vec{R}_j| \). It is instructive to introduce the total hyperfine interaction constant \( A_H \) of the material for a single species. To this end, we partition the QD into spherical shells and we assume a constant density \( \rho \) of nuclei in the QD. The shell at radius \( r \) contains \( N_r \) nuclei. We then integrate over the contributions from each shell and find

\[
A_H = \int dA = \int_0^\infty \left( \frac{4}{3} \mu_0 \mu_B \gamma \right) \left( \frac{1}{(\sqrt{\pi}l_0)^3} e^{-(r/l_0)^2} \right) \frac{(4\pi r^2 \, dr)}{N_r} |\psi(r)|^2
\]

\[
= \text{const.} \int_0^\infty \frac{1}{(\sqrt{\pi}l_0)^3} e^{-(r/l_0)^2} 4\pi r^2 \, dr = \text{const.}
\]

This implies, that the total hyperfine strength \( A_H \) is independent of the number of nuclear spins \( N \) in the QD. Therefore, in the homogeneous hyperfine coupling
constant approximation, the single nuclear spin coupling constant $A$ is the same for all nuclear spins and equal to

$$A = A_j = \frac{A_H}{N},$$

(2.40)

for all $1 \leq j \leq N$. Generally, however [144, 143, 145]

$$A_j = A_0 e^{-R_j^2/R_0^2},$$

(2.41)

where $A_0$ is the coupling constant of a nucleus at the center of the QD, which depends on $N$. In this thesis, we denote the hyperfine coupling constant of the $j^{th}$ nucleus by $A_j$. In the case of homogeneous coupling, we substitute $A_j = A = \frac{A_H}{N}$.

**Overhauser field and electron–nuclei flip–flops**

To analyze the effect of the Fermi-contact hyperfine interaction, it is convenient to partition the Hamiltonian into a diagonal and a flip-flop term, i.e.

$$H_{hyp} = \sum_j A_j(\mathbf{S} \cdot \mathbf{I}) = \sum_j A_j S_z I_j^z + \sum_j \frac{A_j}{2} (S_+ I_j^+ + S_- I_j^-).$$

The diagonal part gives rise to an effective magnetic field for the electron spin, called the Overhauser field

$$B_{Oh} = \frac{\sum_j A_j \langle I_j^z \rangle}{g_e \mu_B}.$$

Conversely, for each nuclear spin $1 \leq j \leq N$ the diagonal part of $H_{hyp}$ constitutes an effective magnetic field, called the Knight field,

$$B_{K}^j = \frac{A_j \langle S_z \rangle}{\gamma_n^j}.$$

The flip–flop terms $H_{ff} = \sum_j \frac{A_j}{2} (S_+ I_j^+ + S_- I_j^-)$ on the other hand allow for the transfer of angular moment from the electron to the nuclear spin system and therefore induce nuclear spin dynamics. However, at high magnetic fields, the flip–flop terms are suppressed due to the mismatch of the electron and nuclear Zeeman energies, i.e. $\omega_e \gg \omega_n$. The consequences of this will be discussed in more detail in chapters 3, 4, and 5.

**2.4.2 Non–collinear interaction**

The nuclear quadrupolar interaction introduced in section 2.3.3 couples states of different total angular momentum projections. These states are detuned by the nuclear Zeeman energy $\omega_n$. Since the energy is not conserved, we treat the off–diagonal quadrupolar terms $H_Q^O$ perturbatively via a Schrieffer–Wolff transformation [146, 89]. The goal of this section is to derive an effective Hamiltonian for Voigt geometry that describes the effect of $H_Q^O$ for effective spin-$\frac{1}{2}$ nuclei. We assume that the principle axis of the quadrupolar interactions points along the growth direction $x$ [132].
2.5 Coupled dynamics in the quantum dot spin system

The effective Hamiltonian arises due to the transformation

$$\hat{H} = e^{-S_Q} H e^{S_Q} = H + [H, S_Q] + \ldots$$

where we require

$$[H_n, S_Q] = -H_{\text{quad}}^O$$

such that $H_{\text{quad}}^O$ is canceled to first order. However, due to the interaction with the Overhauser field term

$$H_{\text{Oh}} = \sum_j A_j S_z I_j^z$$

there are second order off–diagonal terms of the form

$$[H_{\text{Oh}}, S_Q] = \sum_j A_j \frac{\omega_Q}{8 \omega_n} S_z ((I_j^z)^2 + (I_j^+)^2).$$

Taking advantage of the fact that the term above leads to two independent two–level systems (Fig. 2.8), the interaction for one of these effective two level systems can be written as a non–collinear Hamiltonian

$$H_{\text{nc}} = \sum_j A_{\text{nc}}^j S_z I_j^z,$$

where

$$A_{\text{nc}}^j = 2\sqrt{3} \frac{\omega_Q}{8 \omega_n} A_j = \frac{\sqrt{3} \omega_Q}{4 \omega_n} A_j.$$

2.5 Coupled dynamics in the quantum dot spin system

In the previous section we reviewed the interactions in the driven and dissipative QD spin system that are relevant for the topics in this thesis. These interactions lead to rich coupled dynamics of the central electron and the QD nuclear spins, which is the subject of this section. More specifically, we will first concentrate on the central electron spin and discuss the electron spin decoherence which is of great importance for quantum information processing applications. Then we focus on the nuclear spin dynamics and highlight some striking effects that arise due to dynamic nuclear spin polarization. Finally, we will summarize the external driving and dissipation mechanisms as well as the feedback pathways between the central electron and the nuclei that are considered in this thesis and are at the heart of the effects investigated in the subsequent chapters.

2.5.1 Electron spin decoherence

Self–assembled QDs have been proposed as candidates for quantum bits (qubit) that form the basis of quantum information schemes [81]. The projection of the electronic spin would form the two levels of the spin qubit. To harness the power of these schemes, it is crucial that the gate operation time is short compared to the time scale at which the quantum coherence of the qubit is lost. This spurred a large body of work concerned with the coherence of the electron spin in quantum dots [147, 148, 149].

Three different time scales characterize the loss of coherence of the electron spin [150, 151, 88], which we discuss following [88]:

$$\ldots$$
1. The relaxation time $T_1$ describes relaxation process that transfer some quantum amplitude from one spin state to the other, e.g. $|\uparrow\rangle \rightarrow |\downarrow\rangle$ due to the interaction of the electron spin with an environment. Such transitions leave their marks on the time evolution of the operator $\langle S_z \rangle(t)$. In the Bloch sphere picture, $T_1$ thus describes the decay of the longitudinal part of the Bloch vector. For self-assembled QDs at low temperatures and high magnetic fields is $T_1 \sim 20\ \text{ms}$ [152]. This long time scale can be intuitively understood, since the energy cost of flipping the electron spin is given by the large electron Zeeman energy.

2. The decoherence time $T_2$ on the other hand does not require energy exchange with the environment, but is instead related to decay of the off–diagonal density matrix elements that describe the coherence of the state. The coherence can decay without resulting in a transfer of population between the two state. If the initial wave function is given by $|\psi\rangle = \alpha |\uparrow\rangle + \beta |\downarrow\rangle$, then the effect of decoherence on the corresponding density matrix is

$$
\rho = \begin{pmatrix}
|\alpha|^2 & \alpha \beta^* \\
\alpha^* \beta & |\beta|^2
\end{pmatrix} \rightarrow \begin{pmatrix}
|\alpha|^2 & 0 \\
0 & |\beta|^2
\end{pmatrix}.
$$

Clearly, the superposition state $|\psi\rangle = \alpha |\uparrow\rangle + \beta |\downarrow\rangle$ has decayed to either $|\uparrow\rangle$ or $|\downarrow\rangle$ with probabilities that correspond to the initial quantum amplitudes of the two states. To quantify the decoherence of the initial superposition state, the temporal behavior of $\langle S_x \rangle(t)$ is studied. Unfortunately, in most experiments $T_2$ is not directly accessible since a measurement of $\langle S_x \rangle(t)$ has to be averaged over either a spatial ensemble of QDs or a temporal ensemble of the same QD in order to obtain good statistics. However, by employing the spin echo technique, one can partially circumvent this problem and obtain an echo time scale. Strictly, the echo time scale differs from $T_2$; nevertheless we will denote it by $T_2^*$ for simplicity. Alternatively, an electron mode–locking technique in a spatial ensemble of QDs can be used. Both approaches agree well and yield $T_2^* \sim 3\ \mu s$ [153, 85]. We note that similar time scales have been measured in gate–defined QDs [154, 155, 156].

3. In a spatial or temporal ensemble, the ensemble averaged quantity $\langle S_z \rangle_{\text{ensemble}}(t)$ is measured. However, the state of the environments of the electron varies spatially or temporally inducing a fast decay of $\langle S_z \rangle_{\text{ensemble}}(t)$, occurring on a time scale $T_2^* \sim 1 – 10\ \text{ns}$ in self–assembled QDs with an unprepared nuclear spin environment [86, 85, 157]. With proper engineering in a self–assembled QD molecule, it has been shown that $T_2^*$ can be prolonged by more than two orders of magnitude to $T_2^* \sim 0.2\ \mu s$ [158]. However, we stress that this fast decay time scale does not represent the time scale on which the coherence is lost in a single run of the experiment. We illustrate the origin of $T_2^*$ in more detail below.

The two most relevant environments of the confined QD electron that cause relaxation and decoherence are the lattice phonons and the host material nuclear spins. Phonons are particularly important for determining $T_1$ at high magnetic fields, whereas at low fields the hyperfine flip–flop terms are limiting $T_1$ [152]. The hyperfine interaction is also the dominant source for the spin decoherence. We will
2.5 Coupled dynamics in the quantum dot spin system

therefore restrict the following discussion to the effect of the hyperfine interaction on the electron coherence.

We begin this short introduction to hyperfine induced electron spin decoherence by classifying the different time scales of the coupled electron–nuclei dynamics in the absence of an external magnetic field and zero mean nuclear spin polarization according to the influential model due to Merkulov–Efros–Rosen [148]. The first time scale corresponds to the precession period of the electron in the initial Overhauser field. Due to the much slower nuclear spin dynamics the nuclear polarization can be taken to be constant at this stage, which is termed frozen fluctuation. In repeated measurements the initial nuclear spin polarization is different each time and hence when averaged the coherence of the electron is washed out. The second time scale is given by the evolution of the nuclear spin polarization under the hyperfine interaction with the electron spin. This can be visualized as the precession of each nuclear spin around the inhomogeneous Knight field induced by the central electron. The third stage leads to a decay of the total angular momentum in the electron–nuclei system via the dipole–dipole interaction among the nuclei in the host material. In the presence of large inhomogeneous quadrupolar interactions, the mechanisms leading to the second and third stage of the decay are strongly suppressed.

To illustrate the origin of the ensemble decoherence time $T_{2}^{*}$, we will perform a simple calculation at high magnetic fields and in the short time limit, i.e. the nuclei are assumed to be frozen, following [150, 159]. In this case, the flip–flop terms of the hyperfine interaction can be neglected and the Hamiltonian simplifies to

$$H = \omega_e S_z + \omega_{Oh} S_z,$$

where $\omega_e$ is the electron Zeeman splitting in the external magnetic field, and $\omega_{Oh} = \sum_{j=1}^{N} A_j \langle I_j^z \rangle$ is the precession frequency resulting from the Overhauser field $B_{Oh} = \omega_{Oh}/(g_e \mu_B)$. First, we consider the time evolution of the expectation value of the Heisenberg operator $S_x$, when the Overhauser field is characterized by its mean field alone, i.e. $B_{Oh}$ is a known value without fluctuations. Then we find

$$\langle S_x \rangle(t) = \frac{1}{2} \cos((\omega_e + \omega_{Oh})t),$$

for a spin which is initially oriented along the $x$-direction such that $\langle S_x \rangle(0) = 1/2$. However, in general the distribution $P(B_{Oh})$, which describes the probability with which a certain $B_{Oh}$ is realized, has a non–vanishing standard deviation $\sigma$. Therefore, the time evolution of $\langle S_x \rangle$ needs to be averaged over $P(B_{Oh})$. Before doing so, we note that in the limit of a large number of nuclear spins, the central limit theorem applies as described in section 2.3.4, and $P(B_{Oh})$ can be taken to be a Gaussian distribution of the form [148]

$$P(B_{Oh}) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{B_{Oh}^2}{2\sigma^2}\right),$$

where the standard deviation $\sigma$ is determined by

$$\sigma^2 = \frac{1}{4} \sum_j A_j^2,$$
for a single nuclear spin species with \( I = 1/2 \). Under the assumption of uniform coupling this further simplifies to

\[
\sigma = \frac{A_H}{2\sqrt{N}}, \quad (2.42)
\]

where \( A_j = A = \frac{A_H}{N} \) for all \( j \) and \( A_H \) is the total hyperfine coupling constant of the material. Having determined the probability distribution of \( B_{Oh} \), we can now calculate the time evolution of the ensemble averaged operator \( \langle S_x \rangle_{\text{ensemble}} \), yielding

\[
\langle S_x \rangle_{\text{ensemble}} = \int P(B_{Oh}) \frac{1}{2} \cos((\omega_e + g_e \mu_B B_{Oh})t) dB_{Oh}
\]

\[
= \frac{1}{4g_e \mu_B \sqrt{2\pi\sigma}} \left[ \int e^{-\frac{\omega_{Oh}^2}{2(\omega_e + g_e \mu_B B_{Oh})^2} + i\hbar \omega_{Oh} + i\hbar \omega_e} d\omega_{Oh} + \int e^{-\frac{\omega_{Oh}^2}{2(\omega_e + g_e \mu_B B_{Oh})^2} - i\hbar \omega_{Oh} - i\hbar \omega_e} d\omega_{Oh} \right]
\]

\[
= \frac{1}{2} \cos(\omega_e t) e^{-\left(\frac{T_2^*}{2}\right)^2},
\]

where

\[
T_2^* = \frac{\sqrt{2}}{g_e \mu_B \sigma} = \frac{\sqrt{8N}}{g_e \mu_B A_H}. \quad (2.43)
\]

The second expression holds for a single nuclear spin species with spin \( I = 1/2 \) and uniform hyperfine coupling. Above we used the identity for Gaussian integrals

\[
\int_{-\infty}^{+\infty} e^{-ax^2 + bx + c} dx = \sqrt{\frac{\pi}{a}} e^{b^2/a + c}.
\]

Physically, the decay of \( \langle S_x \rangle_{\text{ensemble}} \) arises due to the averaging over many different configurations of the nuclear spin environment. The fluctuations of the spin environment affect the electron spin via the Overhauser term, which modifies the energy splitting between the two electron spin states and thereby decoheres the electron. The decay follows a temporal Gaussian function rather than an exponential, which is a first signature of non–Markovian, or history–dependent, behavior [151]. An important conclusion from this simple calculation is the fact \( T_2^* \) is inversely proportional to the width of the nuclear distribution, i.e. \( T_2^* \propto \frac{1}{\sigma} \). Hence, the ensemble decoherence time \( T_2^* \) can be prolonged by narrowing the nuclear spin distribution.

While the simple calculation above neglected the flip–flop terms, several works analyzed their influence on the dephasing of an electron spin interacting with a narrowed nuclear spin environment and found various decay laws for the different time scales and experimental parameters [148, 143, 141, 134, 160, 161, 88]. At high magnetic fields, it has been shown that the dephasing starts with a quadratic decay at very short time scales, which transitions to a power–law decay followed by a Markovian regime at intermediate time scales. Later, there is an exponential decay before the decay occurs again according to a power–law in the long–time limit [88].

Clearly, engineering the nuclear spin environment is interesting not only from a fundamental perspective, but also for prolonging the coherence time of a QD spin qubit. One option to narrow the nuclear spin distribution that was pursued
was creating a large nuclear spin polarization. The improvement in $T_2^*$ is given by the standard deviation $\sigma$ of a polarized ensemble of nuclear spins. If the average polarization of the ensemble is given by $p = (\langle N_+ \rangle - \langle N_- \rangle)/N$, the probability to obtain a nuclear spin state with $N_+$ spins pointing up is described by the binomial distribution [143]

$$B(N_+, N, f_+) = \binom{N}{N_+} f_+^{N_+} (1 - f_+)^{N-N_+},$$

where $f_+ = \frac{\langle N_+ \rangle}{N} = (p + 1)/2$. Clearly, the mean of this distribution is $\langle N_+ \rangle$ as expected; the standard deviation is given by

$$\sigma = \sqrt{\frac{N}{4}(1 - p^2)}.$$

Hence, to obtain an improvement of a factor of 10 in $T_2^*$, which corresponds to a reduction in $\sigma$ by a factor of 10, a polarization $p \gtrapprox 99\%$ is required. We will review some experimental and theoretical studies that followed that route by employing dynamic nuclear polarization. An alternative to large nuclear spin polarization is to narrow the nuclear spin distribution by engineering an intrinsic feedback–loop that stabilizes the nuclear spin polarization and reduces its spread.

Experimental results of this approach are also discussed in section 2.5.2. In chapter 3 of this thesis we propose a scheme that ideally allows to cool the Overhauser field distribution to the single–spin flip level.

### 2.5.2 Dynamic nuclear polarization

In QDs, nuclear polarization can build up through dynamics that involves both external driving of the electron spin as well as dissipative dynamics. The resulting nuclear polarization is called dynamic nuclear polarization (DNP) in contrast to nuclear polarization that is generated by simple relaxation to a polarized state in equilibrium [162]. While the external coherent drive allows for the transfer of angular momentum from the laser fields to the nuclear spin environment via the hyperfine interaction with the electron, the non–unitary dynamics lowers the entropy by carrying away information about the system. The latter can enter through measurement or dissipative dynamics stemming from the coupling to the photon or phonon reservoir or the electrons in the Fermi sea. A commonly encountered and illustrative example are spontaneously emitted photons that lead to relaxation of the electron spin on the one hand and provide information about the state of the electron–nuclei system on the other hand [162]. The information carried by the photon not only lowers the entropy of the system but can also be utilized to read out the nuclear polarization because DNP shifts the electron energy levels (Overhauser shift, see section 2.4.1). Since QDs have been shown to have almost hard selection rules [163, 109], the feedback of DNP on the electron spin can strongly influence the interaction of the electron with the driving lasers. This property can be used to detect DNP through non–resonant (photoluminescence [164, 107, 165]) or resonant (differential transmission [166] and resonance fluorescence [167]) as well as pump–probe techniques [168]. In the case of an ensemble of QDs, the Overhauser field modifies the Larmor precession frequency which can be readout through a Faraday pump–probe experiment [103].
One of the goals aimed for was the creation of high DNP, motivated by improving the electron decoherence time. Overhauser fields on the order of $B_{\text{Oh}} \approx 3 \text{T}$ have been measured which corresponds to a nuclear spin polarization of approximately 60% [103]. While this is a remarkable achievement, the nuclear environment is far from a polarization exceeding $\sim 99\%$ that would increase $T_2^*$ by an order of magnitude. A recent theoretical study considered the achievable DNP in such an experimental setup [144, 169] and showed that nuclear dark states limit the production of DNP in the investigated experimental situation. At the same time inhomogeneities stemming from different hyperfine coupling to the central electron spin, the quadrupolar effects or different nuclear spin species could lead to a higher degree of nuclear spin polarization. These findings can be understood when considering that the relative polarization at which DNP saturates is found to vary as $\propto N^{-\frac{1}{2}}$, where $N$ is the group of nuclear spins for which the total spin $J$ is conserved. Inhomogeneities split up the nuclear spin environment into smaller groups of spins that conserve $J$. These smaller groups then show a larger relative saturation polarization resulting in a higher saturation polarization for the complete nuclear spin environment.

Various experiments investigated the typical time scales of buildup and relaxation of DNP in QDs. The DNP buildup time generally depends strongly on the specific experimental conditions, including the magnetic field and laser intensity and ranges from $1 \text{ms} - 10 \text{s}$ [103, 105, 170]. In contrast, the DNP relaxation is related to the structural properties and elementary interactions of the QD [135]. DNP in strained QDs was measured to survive up to $30 \text{h}$ in the dark [171], suggesting that nuclear spin diffusion to the bulk is strongly suppressed. While phonon–mediated decay can be neglected at temperatures of a few K, electron mediated decay due to fast co–tunneling to the Fermi reservoir can lead to nuclear depolarization on a millisecond time scale [105, 103].

Many experiments on DNP revealed rich physics that arises due to the feedback mechanisms between the central electron spin and the nuclear spin environment. For simplicity, in the following short discussion we focus on the electron–nuclei feedback mechanisms induced by the Overhauser field and the hyperfine flip–flop terms; we note that other nuclear interactions could play a similar role. The polarization of the nuclear environment collectively modifies the Zeeman levels / Larmor precession frequency through the Overhauser field $B_{\text{Oh}}$. At the same time, the transfer of angular momentum from the electron spin to the nuclear ensemble through the flip–flop terms $H_{\text{ff}} = \sum_j \frac{A_j}{2} S_+ I^+_j + S_- I^-_j$ is suppressed by the mismatch of the electron and nuclear Zeeman energies at high magnetic fields, i.e. $\omega_e \gg \omega_n$. In many settings, the energy mismatch between the initial and final states, denoted by $\Delta E$, is compensated by coupling to a reservoir. The rate of the corresponding second–order processes usually satisfies $R \propto \Delta E^{-2}$ [103]. Since the electron states involved in the process shift with the Overhauser field, $\Delta E$ depends not only on the external magnetic field, but also on the Overhauser field. This then leads to a feedback on the rate that causes the DNP in the first place. These types of feedback loops can be used to stabilize certain nuclear spin polarizations and to reduce the nuclear fluctuations, thereby increasing the electron coherence times.

Experimental studies on DNP revealed intricate feedback mechanisms that include hysteresis and bistability effects. Therefore DNP is strongly dependent on
2.5 Coupled dynamics in the quantum dot spin system

Figure 2.11: Summary of the interactions considered in the driven–dissipative QD central spin system.

the history and thus non–Markovian. A plethora of intriguing experiments have been reported [172, 173, 174, 170, 175, 176]. We would like to discuss three particular examples of these experiments, i.e. DNP in the presence of CPT [176], nuclei induced frequency focusing in an ensemble of QDs [91], as well as the dragging experiments [170, 175], and how they are connected to this thesis in more detail in section 2.5.3 below.

Clearly, DNP in the QD central spin system gives rise to a wealth of intriguing physical phenomena. At the same time it is relevant for potential quantum information applications as DNP provides a route to prolong the coherence time of the central electron spin.

2.5.3 Summary of electron–nuclei feedback discussed in this thesis

In this chapter we have reviewed the basic interactions that govern the coupled electron–nuclei dynamics in self–assembled QDs. We have discussed how the electron spin, and in particular its coherence, is affected by the nuclear environment and conversely, how this driven–dissipative central spin system allows the buildup of DNP. To conclude this chapter, we summarize the interactions that will be considered in this thesis and how they affect the coupled electron–nuclei spin dynamics (Fig. 2.11).

The central electron spin is coupled to the magnetic field $\vec{B}$ and it is optically excited by laser fields that propagate perpendicularly to the magnetic field. The laser excitation considered here either consists of two linearly polarized continuous wave lasers or a single circularly polarized pulsed laser that is periodic in time. The electron also interacts with the radiation field reservoir which introduces non–unitary dynamics; the optically excited electron can decay to the ground state via spontaneous emission.

The nuclear environment also interacts with the magnetic field. In self–assembled QDs, the nuclear spins experiences strain due to the lattice constant mismatch of the materials that constitute the QD as well as interdiffusion of In and Ga. This strain leads to nuclear quadrupolar interactions that can strongly modify the resulting coupled electron–nuclei dynamics. While the effect of strain is neglected in chapter 3, it is considered in chapters 4 and 5.

Optical excitation and the subsequent spontaneous emission provide a mechanism to provide a pathway to conserve energy in the buildup of DNP. In chapters 3
2 The quantum dot central spin model

Figure 2.12: Modification of electron Zeeman energy and Larmor frequency by the nuclear Overhauser field.

and 4, the dominant DNP mechanism stems from spontaneous emission assisted nuclear spin flips that result in diffusive dynamics. In contrast, in chapter 5 the directional DNP arising from absorption assisted nuclear spin flips plays an important role. The feedback of DNP on the central electron spin occurs through a modification of the electron Zeeman energy (or equivalently the Larmor precession frequency). It is important to note that these quantities are crucial in determining the coupling to the external optical driving and therefore strongly influence the evolution of the central electron spin (Fig. 2.12).

We would like to discuss three DNP experiments that are of particular importance for this thesis. The first experimental scheme we focus on here is CPT in solid state emitters. As stated earlier, CPT has been observed both in self–assembled QDs and NV centers [121, 122, 124]. Here, nuclear spin effects did not lead to a strong modification of the characteristic CPT spectrum. However, under different experimental conditions nuclear spin effects in CPT experiments have been reported both in QDs [176] and NV centers [177]. In QDs these include hysteresis, deviation from a Lorentzian shape of the CPT dip as well as a dependence of the CPT dip on the integration time [176]. In this context, chapter 3 summarizes the results that are expected in a similar setup, where it was assumed that the Fermi contact hyperfine interaction is the only nuclear spin interaction. One of the main results is that the nuclear spin evolution is described by an anomalous diffusion process that leads to a drastic cooling of the nuclear spin distribution. The fact that the corresponding theoretical results neither match the experimental observations of [176] nor could be observed in a different self–assembled QD sample can be attributed to the presence of strong strain–induced quadrupolar interactions that lead to directional nuclear spin dynamics and were neglected in the theoretical description in chapter 3. Nevertheless, the nuclear spin cooling by anomalous diffusion predicted in chapter 3 was observed in NV centers [177]. These experiments confirm the validity of the proposed cooling mechanism, provided the experimental system satisfies the assumptions on which the theoretical description is based on. While the basic assumptions of chapter 3 are met by NV centers, we would like to point out some of the important differences between the configuration used in these NV centers and QDs. First of all, the hyperfine interaction in NV center is stronger in the excited state compared to the ground state in contrast to single electron charged QDs. Furthermore, the single proximal $^{14}$N
spin dominates the hyperfine interaction. Additionally, the number of relevant nuclei that couple to the electron is much smaller in NV centers compared to QDs such that the time scale on which DNP become apparent is strongly reduced in NV centers, facilitating their observation.

The second striking example is the dragging effect [175, 170]. Here, the nuclear spins evolve such as to lock an incident laser to optical resonance. These experimental results can be described as resulting from strain–induced non–collinear interactions that lead to bidirectional DNP. Depending on the optical transition that is chosen, detuning the laser from resonance induces a nuclear spin polarization rate that favors or opposes the nuclear spins to polarize in such a way as to re–establish resonance through a Overhauser shift of the electronic levels [170, 175]. The two effects are termed dragging and anti–dragging, respectively. In this experimental scheme, the scan direction of the laser frequency is important and leads to hysteresis effects in the absorption spectra, which indicates the occurrence of bistability in the nuclear spin system. When the Overhauser field is locked to optical resonance, the nuclear spin fluctuations are substantially reduced even tough the DNP is relatively small. This can be understood as arising from the intrinsic feedback mechanism that stabilizes the Overhauser field that satisfies the optical resonance condition. Chapter 5 extends the mechanism that explains the dragging effects, i.e. the directional optically assisted nuclear spin flips due to the non–collinear interaction, to a two–color illumination scheme in the CPT configuration. By varying the strength of the two Rabi frequencies that drive the two transitions, one could expect a competition between dragging and anti–dragging effects.

Furthermore, combining the insight into directional DNP gained from dragging with the results from an experiment on an ensemble of QDs under periodic pulsed laser excitation, hints at a route to circumvent the detrimental effect of directional DNP for the nuclear spin cooling by anomalous diffusion. It was shown that the electron precession frequencies of an ensemble of singly charged QDs are focused into a discrete set of frequencies by DNP effects [91]. One might expect that for a single QD, shortening the interaction time with the laser fields by going to a pulsed excitation regime suppresses the directional, absorption assisted DNP terms that arise due to strain–induced nuclear interactions. In chapter 4, this situation is analyzed and the experimental observation of the anomalous diffusion effects is discussed.

To conclude, in this thesis we will analyze the rich dynamics that is a consequence of the aforementioned interactions. We discuss cooling mechanisms for the nuclear spin distribution as well as anomalous and critical dynamics in the nuclear spin system and derive the ensuing optical signatures that result from the feedback mechanism on the central electron spin.
Nuclear spin cooling using Overhauser field selective coherent population trapping

In the central spin system realized by solid–state emitters such as quantum dots (QD), the coupling of the central electron spin to the nuclear spin environment leads to decoherence. Here, we show that a quantum interference effect in the optical absorption from two electronic spin states of a solid–state emitter can be used to prepare the surrounding environment of nuclear spins in well–defined states, thereby suppressing electronic spin dephasing. The coupled electron–nuclei system evolves into a coherent population trapping state by optical–excitation–induced nuclear spin diffusion for a broad range of initial optical detunings. The spectroscopic signature of this evolution, where the single–electron strongly modifies its environment, is a drastic broadening of the dark resonance in optical absorption experiments. The dynamics that leads to nuclear spin cooling can be described as an anomalous diffusion process in close analogy to sub–recoil laser cooling. The large difference between electronic and nuclear time scales allows to verify the preparation of the desired nuclear spin state. This chapter is based on [89].

3.1 Motivation

Confined spins in solid–state emitters are subject to dephasing due to the coupling to various environments. Spin environments with long correlation times are especially important, because they lead to non–Markovian dephasing [149, 178]. In particular, hyperfine coupling to nuclear spins constitutes the dominant source of decoherence for spin qubits. It has been proposed that polarizing or measuring nuclear spins could alleviate the ($T_2^*$) decoherence process [149], which prompted theoretical [179, 180] as well as experimental efforts aimed at narrowing down the Overhauser field (OF) distribution [181, 182, 170]. These schemes could be considered a form of reservoir engineering; remarkably, recent experiments showed that a substantial manipulation of the nuclear spins (reservoir) could be achieved by using the electron spin (system) itself [183, 184, 172, 182, 170, 169].

In this chapter, we show that the $T_2^*$ decoherence time scale can be drastically prolonged by narrowing (cooling) the nuclear spin environment by applying coherent population trapping (CPT) to the spin states of a solid–state emitter. The phenomenon of CPT in three–level emitters [23] is at the heart of a number of key advances in quantum optics, such as sub–recoil cooling of atoms [21] and
slow–light propagation [119, 185]. In these experiments, optical excitation from two low energy (spin) states to a common excited state vanishes due to a quantum interference effect, leading to the formation of a dark resonance whenever the two driving laser fields satisfy the two–photon resonance condition as discussed in section 2.2.2. When the solid–state emitter is coupled to the spin environment via hyperfine interactions, we find that the spin environment (nuclei) modifies the (bare) laser detuning until two–photon resonance is established, forcing the central (electron) spin into a dark state. This holds for a wide range of initial detunings and results in a pronounced broadening of the dark resonance. Concomitantly, the dominant mechanism for the nuclear spin dynamics, which stems from spontaneous–emission assisted nuclear spin flips, is suppressed. Thereby, the prepared nuclear spin state is stabilized. The standard deviation of the ensuing ultra–narrow nuclear spin distribution is close to the single–spin limit. This, in turn, leads to a prolonged $T_2^*$ decoherence time for the electron spin.

An additional feature of the scheme is the possibility of using the measurement of resonant fluorescence to verify the preparation of a narrow nuclear spin distribution [186]; turning off the laser fields after determining the coupled system to be in the dark state can then be used to ensure that the Overhauser field distribution remains in the single–spin regime within the time scales determined by the (intrinsic) nuclear spin lifetime, reducing the standard deviation from $\sigma \propto \frac{1}{\sqrt{N}}$ (see (2.42)) to $\sigma \propto \frac{1}{N}$. The corresponding electron spin $T_2^* \propto \frac{1}{\sigma^2}$ time will then be prolonged by a factor $\sim \sqrt{N}$, where $N$ is the number of nuclear spins. Furthermore, nuclear spins in the prepared state remain unaffected by the electron–mediated indirect interactions, which eliminates a principal contribution to the electron spin $T_2$ time [149, 178].

Remarkably, the cooling of the nuclear spin environment can be considered the result of an anomalous diffusion process associated with optical excitation. This is in close analogy to sub–recoil laser cooling which arises in velocity selective coherent population trapping (VSCPT) as described in chapter 1 [21, 16]. Rather than narrowing the momentum distribution, here the nuclear spin (Overhauser field) distribution is narrowed. While in VSCPT the atoms experience a momentum kick in the process of optical absorption and emission, here the nuclear Overhauser field changes whenever a spontaneous–emission assisted nuclear spin flip occurs. As a consequence of the anomalous diffusion, the coupled electron–nuclei system dynamically switches back and forth between a trapped regime where nuclear spin diffusion slows down drastically due to the formation of an electronic dark state, and a non–trapped regime where optical excitation leads to fast nuclear diffusion [16]. The description in terms of anomalous diffusion not only offers deeper physical insight into the nuclear cooling mechanism, but also allows us to discuss the characteristic time scale on which substantial narrowing can be expected.

This chapter is organized as follows: We first specify the system and interactions under investigation in section 3.2. Then, we derive the equations that govern the dynamics of the coupled system and present an overview of the fundamental feedback mechanisms that are central to this scheme in section 3.3. In section 3.4, we first analyze the effect of the mutual coupling on the nuclear spin environment and discuss the observed cooling in various limits. Second, we review how the slow nuclear spin dynamics affects the central electron spin. In particular, we compute the optical signatures in CPT in the presence of the nuclear spin environment and
find that the optical absorption is drastically reduced for a wide range of laser detunings. Additionally, in section 3.5, we describe the nuclear dynamics that leads to the narrowing of the nuclear spin distribution in terms of an anomalous diffusion process. We close with a discussion of experimental implementations of this scheme in section 3.6.

### 3.2 Driven–dissipative electron–nuclei system

We consider a solid–state emitter where the two ground electronic spin states, denoted by $|\uparrow\rangle$ and $|\downarrow\rangle$, are coupled by two laser fields to a common optically excited state $|t\rangle$ (Fig. 3.1). The laser field with frequency $\omega_p$ ($\omega_c$) that couples the $|\uparrow\rangle \leftrightarrow |t\rangle$ ($|\downarrow\rangle \leftrightarrow |t\rangle$) transition with Rabi frequency $\Omega_p$ ($\Omega_c$) is referred to as the probe (coupling) field. The state $|t\rangle$ decays in turn via spontaneous emission back to the two ground spin states. For simplicity we assume for the rates $\Gamma_{t\uparrow} = \Gamma_{t\downarrow} = \Gamma/2$. Denoting the Zeeman energy of the electron spin due to the external field $B_z$ with $\omega_z$ and the energy of the optically excited state with $\omega_t$, we express the bare optical detunings relevant for the CPT system as

$$\Delta\omega_p = \omega_t + \omega_z/2 - \omega_p,$$

$$\Delta\omega_c = \omega_t - \omega_z/2 - \omega_c.$$  

In the absence of any spin interactions or decoherence, laser fields satisfying the two–photon resonance condition

$$\delta = \Delta\omega_p - \Delta\omega_c = 0$$

pump the electron spin into the dark state

$$|D\rangle = \frac{1}{\sqrt{\Omega_p^2 + \Omega_c^2}}(\Omega_c |\uparrow\rangle - \Omega_p |\downarrow\rangle),$$

which is decoupled from optical excitation. When $\Delta\omega_c = 0$ and $\Omega_p, \Omega_c \ll \Gamma$, the absorption lineshape of the emitter is a Lorentzian with a quantum interference induced transparency in the center, that has a width $\delta\nu_{\text{trans}} \sim (\Omega_p^2 + \Omega_c^2)/\Gamma \ll \Gamma$ (see 2.2.2).

In most solid–state emitters, the electronic spin states are mutually coupled via hyperfine interactions with a nuclear spin ensemble consisting of $N$ nuclei

$$H_{\text{hyp}} = A\sqrt{N} \sum_i \frac{A_i}{A\sqrt{N}} \left[I_z^i S_z + \frac{1}{2}(I_+^i S_- + I_-^i S_+)\right].$$

Here, $A_i$ is the hyperfine coupling constant between the emitter electron and the $i^{th}$ nucleus, with average coupling strength $A = \frac{A_H}{\sqrt{N}}$. As described in chapter 2, $A_H$ defines the total hyperfine coupling constant of the material. If the ensemble consists of different atomic species or isotopes, one can define an effective $A_H$ that corresponds to the maximal electron spin splitting when all nuclei are polarized. The electronic and nuclear spin operators are denoted by $S_\alpha$ and $I_\alpha^i$ ($\alpha = +, -, z$), respectively, where $S_\pm = S_x \pm iS_y$. It is convenient to introduce the nuclear quasi–spin operators

$$I_\alpha^{(n)} = \sum_{i=1}^N \left(\frac{A_i}{A\sqrt{N}}\right)^n I_\alpha^i,$$  

(3.1)
Figure 3.1: The energy level diagram of a solid–state emitter exhibiting CPT. In the absence of any spin environment (solid lines), the system evolves into the dark state if the (bare) two–photon detuning $\delta$ vanishes which leads to strongly reduced absorption. The direct flip–flop terms of the hyperfine coupling to the spin environment are strongly suppressed at high magnetic fields. However, they give rise to second–order optically–assisted flip–flops (dashed lines), e.g. upon excitation to the excited state, spontaneous emission occurs concurrent with an electron–nuclei flip–flop (red dashed lines).
where $\alpha = +, -, z$. This operator is unitless and of order one for typical nuclear spin states (see section 2.3.4). In general, these operators do not satisfy angular momentum commutation relations, i.e.

$$[I^{(n)}_\alpha, I^{(n)}_\beta] = \sum_{i,j=1}^N \left( \frac{A_i}{A\sqrt{N}} \right)^n \left( \frac{A_j}{A\sqrt{N}} \right)^n [I^i_\alpha, I^j_\beta]$$

$$= \sum_{i,j=1}^N \left( \frac{A_i}{A\sqrt{N}} \right)^n \left( \frac{A_j}{A\sqrt{N}} \right)^n i\epsilon_{\alpha\beta\gamma} I^i_\gamma \delta_{i,j}$$

$$= i\epsilon_{\alpha\beta\gamma} \sum_i \left( \frac{A_i}{A\sqrt{N}} \right)^{2n} I^i_\gamma$$

$$= i\epsilon_{\alpha\beta\gamma} I^{(2n)}_\gamma$$

$$\neq i\epsilon_{\alpha\beta\gamma} I^{(n)}_\gamma.$$ 

In the limit of a large external field ($\omega_z \gg A\sqrt{N}$), the direct electron–nuclei flip-flop processes $I_+ S_- + I_- S_+$ are strongly suppressed due to the large mismatch in the electronic and nuclear Zeeman splitting. In contrast, optical excitation does allow for energy conservation in an optically assisted electron–nuclei spin–flip process, as we will discuss in the next section.

### 3.3 Description of the coupled electron–nuclei dynamics

In this section, we start with the master equation for the coupled electron–nuclei system in the presence of spontaneous emission. To obtain a tractable description of this driven and dissipative central spin system, we apply the quasi–degenerate perturbation theory (Schrieffer–Wolff transformation) [146] to the master equation and thereby extract the rate at which optically assisted nuclear spin flips occur. We then use the separation of time scales in the system; the characteristic time scale at which the electron evolves to its quasi–steady state is much faster than the evolution of the nuclear spins and therefore the electronic degrees of freedom can be eliminated. This yields a purely nuclear master equation that provides additional insight. These calculations allow us to qualitatively discuss the implications of the electron–nuclei coupling on the central spin and the nuclear environment. We predict nuclear spin cooling and as a consequence striking optical signatures. These predictions will then be further investigated in the subsequent section.

#### 3.3.1 Derivations

The derivations start with the master equation that describes the evolution of the electron–nuclei density matrix in the presence of spontaneous emission. After transforming to a rotating frame to eliminate the time dependence of the laser Hamiltonian, we introduce the quasi degenerate perturbation theory that allows us to derive the second order terms that describe the optically assisted nuclear spin flips. Finally, we eliminate the electronic degrees of freedom in the limit of weak optical excitation and derive a nuclear master equation.
Master equation in the rotating frame

The starting point of the derivations is the master equation describing the coupled electron and nuclear system, obtained after eliminating the radiation field reservoir using a Born–Markov approximation \[187\]:

\[
\dot{\rho} = \frac{\Gamma}{2} (|\uparrow\rangle \langle \downarrow| \rho |\uparrow\rangle - \frac{1}{2} \{ |\uparrow\rangle \langle \uparrow| , \rho \} + ) \\
+ \frac{\Gamma}{2} (|\downarrow\rangle \langle \downarrow| \rho |\downarrow\rangle - \frac{1}{2} \{ |\downarrow\rangle \langle \downarrow| , \rho \} + ) - i[H, \rho] \\
= \frac{\Gamma}{2} (1_S \otimes \rho_{tt} - \{ |\uparrow\rangle \langle \uparrow| , \rho \} + ) - i[H, \rho],
\]

(3.2)

where \(\rho_{tt} = \langle \uparrow| \rho |\uparrow\rangle\) is an operator that acts on the Hilbert space of the nuclear spins, \(1_S = |\uparrow\rangle \langle \uparrow| + |\downarrow\rangle \langle \downarrow|\) and we defined \(\{A, B\}_+ = AB + BA\). The Hamiltonian of the system consists of the diagonal part \(H_0\) and the laser and hyperfine Hamiltonians,

\[
H = H_0 + H_{\text{laser}} + H_{\text{hyp}},
\]

(3.4)

\[
H_0 = \omega_t |\uparrow\rangle \langle \uparrow| - \omega_2S_z,
\]

(3.5)

\[
H_{\text{laser}} = \Omega_c e^{i\omega_c t} |\downarrow\rangle \langle \downarrow| + \Omega_p e^{i\omega_p t} |\uparrow\rangle \langle \uparrow| + h.c.,
\]

(3.6)

\[
H_{\text{hyp}} = A\sqrt{N} (\mathcal{I}(1)zS_z + \frac{1}{2} (\mathcal{I}(1) + S_+ + S_- + S_+ S_-))
\]

(3.7)

For the analysis below, we (initially) neglect the nuclear Zeeman energy. We assume that in the absence of optical excitation, the electron spin is well isolated from all reservoirs other than the nuclear spins \[83\], and spin–flip co–tunneling or phonon emission rates are negligible within the time scales of interest.

The basis transformation to a rotating frame (see 2.2.2)

\[
H' = e^{i\xi t}(H - \xi)e^{-i\xi t},
\]

(3.8)

with

\[
\xi = \left(\omega_t - \frac{1}{2}(\Delta\omega_c + \Delta\omega_p)\right) |\uparrow\rangle \langle \uparrow| - (\omega_2 + (\Delta\omega_c - \Delta\omega_p)) (\mathcal{I}(0)_z + S_z),
\]

(3.9)

renders the Hamiltonian time–independent,

\[
H' = \tilde{\omega}_z \mathcal{I}(0)_z + \frac{1}{2}(\Delta\omega_c + \Delta\omega_p) |\uparrow\rangle \langle \uparrow| - \delta S_z + H_{\text{laser}} + H_{\text{hyp}},
\]

(3.10)

where

\[
H_{\text{laser}} = \Omega_c |\downarrow\rangle \langle \downarrow| + \Omega_p |\uparrow\rangle \langle \uparrow| + h.c.
\]

and

\[
H_{\text{hyp}} = H_{\text{hyp}}.
\]

The effective nuclear energy in the rotating frame is \(\tilde{\omega}_z = \omega_z + (\Delta\omega_c - \Delta\omega_p) \approx \omega_z\). The dissipative part of (3.3) - containing secular terms exclusively - remains unchanged under the transformation, yielding the master equation

\[
\dot{\rho} = \frac{\Gamma}{2} (|\uparrow\rangle \langle \uparrow| \rho |\uparrow\rangle - \frac{1}{2} \{ |\uparrow\rangle \langle \uparrow| , \rho \} + ) \\
+ \frac{\Gamma}{2} (|\downarrow\rangle \langle \downarrow| \rho |\downarrow\rangle - \frac{1}{2} \{ |\downarrow\rangle \langle \downarrow| , \rho \} + ) - i[H', \rho] \\
= \frac{\Gamma}{2} (1_S \otimes \rho_{tt} - \{ |\uparrow\rangle \langle \uparrow| , \rho \} + ) - i[H', \rho].
\]

(3.11)
In the limit of a large external field \( \omega_z \gg A\sqrt{N} \), we derive the second order effects of the energetically suppressed hyperfine flip-flop interaction in a systematic approach using quasidegenerate perturbation theory [188, 146]. We calculate the rate at which optically assisted nuclear diffusion process (Fig. 3.1) occur and motivate the introduction of the novel concept of a generalized Overhauser field.

**Quasidegenerate perturbation theory**

The clear separation of energy scales in the Hamiltonian of (3.10) \( \omega_z \gg A\sqrt{N}, \delta, \Omega_p, \Omega_c, \Delta \omega_p, \Delta \omega_c \) allows us to partition the full Hamiltonian into a zero-order part \( H'_0 = \omega_z I_z(0) \) and a small perturbation \( V = H' - H'_0 \). The eigenvectors of \( H'_0 \) are grouped into well separated manifolds, labeled by the nuclear spin projection quantum number \( m \). According to this spectrum any operator \( O \) can be partitioned into a block diagonal \( O_D \) operator–containing terms that conserve \( m \)–and a block off–diagonal \( O_O \) operator–containing terms that drive transitions between different manifolds (\( m \) non–conserving terms). For the Hamiltonian this separation yields

\[
H'_D = H'_0 + V_D, \\
H'_O = V_O,
\]

where

\[
V_D = \frac{1}{2} (\Delta \omega_c + \Delta \omega_p) |t\rangle \langle t| - \delta S_z + (\Omega_c |\downarrow\rangle \langle \downarrow| + \Omega_p |\uparrow\rangle \langle \uparrow| + h.c.) + A\sqrt{N} I_z^{(1)} S_z/n,
\]

\[
V_O = \frac{A\sqrt{N}}{2} \left( I_+^{(1)} S_- + I_-^{(1)} S_+ \right).
\]

Note that the above choice of \( H'_0 \) is not unique. Other choices that for instance render \( V_O \) purely block–off–diagonal are equivalent, but complicate the fine-structure within the manifolds.

In the following we construct a similarity transformation, generated by an anti–hermitian operator \( S = -S^\dagger \)

\[
H = e^{-S} H' e^S
\]

that renders the transformed Hamiltonian block diagonal and thus decouples the different manifolds from each other. Since the above condition does not define \( S \) uniquely [189], we further demand \( S_D = 0 \) (canonical choice). Expanding the operators in orders of the perturbation

\[
H = \sum_{n=0}^{\infty} H^{[n]}, \quad S = \sum_{n=0}^{\infty} S^{[n]},
\]

one can derive conditional equations for \( S \) order by order. Exploiting the fact that \( V_O^2 \) is block diagonal (\( S^2_0 = S^2_+ = 0 \)) one finds the simple recursive equations:

\[
S^{[0]} = 0, \\
[ H'_0, S^{[1]} ] = -V_O, \\
[ H'_0, S^{[2]} ] = -[V_D, S^{[1]}], \\
[ H'_0, S^{[3]} ] = -[V_D, S^{[2]}] - \frac{1}{3} \left[ [V_O, S^{[1]}], S^{[1]} \right],
\]

...
3 Nuclear spin cooling

The expansion of the transformed Hamiltonian yields

\[ H^{[0]} = H_0', \]
\[ H^{[1]} = V_D, \]
\[ H^{[2]} = \frac{1}{2}[V_O, S^{[1]}], \]
\[ H^{[3]} = \frac{1}{2}[V_O, S^{[2]}], \]
\[ \vdots \]

(3.15)

Note that \( H^{[n]} \) only depends on lower orders of the transformation matrix. Higher order expressions and a detailed derivation are given in [146].

Second order corrections: Optically assisted nuclear spin flips

We now expand our system’s master equation (3.3) to second order in the perturbation in order to identify the dominant hyperfine processes in the electron–nuclei CPT setting. From (3.14) we derive the form of \( S^{[1]} \),

\[ S^{[1]} = \epsilon(S_+ I_+^{(1)} - S_- I_+^{(1)}), \]

(3.16)

where we defined the expansion parameter \( \epsilon = \frac{A\sqrt{N}}{2\omega_z} \). \( S^{[1]} \) generates the second order corrections to the Hamiltonian (see (3.15)):

\[ H^{[2]} = -\frac{1}{2}\epsilon A\sqrt{N}(S_+(I_+^{(1)} I_+^{(1)})_+ - I_+^{(2)}). \]

(3.17)

The full transformed Hamiltonian then reads,

\[ H = H'_0 + V_D + H^{[2]}, \]
\[ = \tilde{\omega}_z I_z^{(0)} + \frac{1}{2}(\Delta \omega_c + \Delta \omega_p) |t\rangle \langle t| + (\Omega_c |\downarrow\rangle \langle t| + \Omega_p |\uparrow\rangle \langle t| + h.c.) \]
\[ + A\sqrt{N}S_z(I_+^{(1)} - \frac{1}{2}\epsilon\{I_+^{(1)}, I_+^{(1)}\}_+ - \delta/(A\sqrt{N})) + \epsilon A\sqrt{N}\frac{1}{2}I_+^{(2)} \]
\[ = \tilde{\omega}_z I_z^{(0)} + \frac{1}{2}(\Delta \omega_c + \Delta \omega_p) |t\rangle \langle t| + H_{\text{laser}} \]
\[ + A\sqrt{N}S_z(I_+^{(1)} - \delta/(A\sqrt{N})) + \epsilon A\sqrt{N}\frac{1}{2}I_+^{(2)}, \]

(3.18)

(3.19)

where we have introduced the notion of the generalized Overhauser field

\[ \tilde{I}_+^{(1)} = I_z^{(1)} - \frac{1}{2}\epsilon\{I_+^{(1)}, I_+^{(1)}\}_+, \]

(3.20)

which contains all electron–nuclei interactions up to second order and can be interpreted in our context as an effective two–photon detuning, which adds (multiplied by \( A\sqrt{N} \)) to the external laser detuning \( \delta \).

In a similar procedure, the system Liouvillian

\[ \mathcal{L}'(\rho) = \frac{\Gamma}{2}(\mathbb{1} s \otimes \rho_t - \{|t\rangle \langle t|, \rho\}_+) \]
3.3 Description of the coupled electron–nuclei dynamics

is transformed under $S$ and expanded in orders of the perturbation:

$$
\mathcal{L}(\rho) = \sum_{n=0}^{\infty} \mathcal{L}^{[n]}(\rho).
$$

(3.21)

Realizing that the transformation leaves the excited electron state (and thus the
second term of the Liouvillian $\propto \{ |t\rangle \langle t|, \rho \}_+$ invariant, i.e. $e^{-S^{[1]}_t} |t\rangle = |t\rangle$, we
only have to transform the jump term $I_S \otimes \rho_I$. The Baker–Hausdorff formula yields

$$
e^{-S^{[1]}_t}(I_S \otimes \rho_I)e^{S^{[1]}_t} \approx I_S \otimes \rho_I + [I_S \otimes \rho_I, S^{[1]}] + \frac{1}{2} \left[ \left[ I_S \otimes \rho_I, S^{[1]}_+ \right], S^{[1]}_+ \right],
$$

(3.22)

which is particularly simple to calculate since $I_S S_\alpha = S_\alpha$. Grouping terms with
regard to orders of $\epsilon$, one finds

$$
\mathcal{L}^{[0]}(\rho) = \frac{\Gamma}{2}(I_S \otimes \rho_I - \{ |t\rangle \langle t|, \rho \}_+),
$$

$$
\mathcal{L}^{[1]}(\rho) = -\epsilon^2 \frac{\Gamma}{2} \left( S_- [\rho_I, I_+^{(1)}] + S_+ [I_-^{(1)}, \rho_I] \right),
$$

$$
\mathcal{L}^{[2]}(\rho) = \epsilon^4 \frac{\Gamma}{4} I_S \otimes D_1(\rho_I) + \epsilon^2 \frac{\Gamma}{2} S_+ \otimes D_2(\rho_I).
$$

(3.23)

Note that the above procedure is equivalent to a straightforward transformation of
the electronic jump operators $|\uparrow\rangle \langle t|$ and $|\downarrow\rangle \langle t|$ of the Liouvillian. The superoperators

$$
D_1(\rho) = I_+^{(1)} \rho I_-^{(1)} + I_-^{(1)} \rho I_+^{(1)} - \frac{1}{2} \{ I_+^{(1)} I_-^{(1)} + I_-^{(1)} I_+^{(1)} , \rho \}_+,
$$

$$
D_2(\rho) = I_-^{(1)} \rho I_+^{(1)} - I_+^{(1)} \rho I_-^{(1)} + \{ I_z^{(2)} , \rho \}_+,
$$

(3.24)

describe optically induced random nuclear spin diffusion processes caused by optically assisted electron–nuclei spin flip events. Note that second order terms in
the transformed Liouvillian arising from $S^{[2]}$ are non–secular and consistently neglected. $\mathcal{L}^{[1]}$ and the second summand in $\mathcal{L}^{[2]}$ do not affect the nuclear evolution
governed by $\hat{\rho}_I = \text{Tr}_S(\hat{\rho})$, since $\text{Tr}_S(S_\pm) = \text{Tr}_S(S_\mp) = 0$ (Tr$_S$ denotes the trace
over all electronic degrees of freedom). For the electron evolution, which occurs on
time scales $\propto \Gamma$ these terms merely represent a small $\epsilon^2$–correction ($\mathcal{L}^{[1]}$ acquires
an additional factor $\propto \epsilon$ since it is non–secular) and are consequently neglected in the following. If we further assume both lasers to be resonant with the bare
transitions ($\Delta \omega_c = \Delta \omega_p = 0$) and neglect the last term of (3.18), which accounts
for a small state independent nuclear diffusion–which will be discussed in section
3.4.1–we arrive at the master equation

$$
\hat{\rho} = \frac{\Gamma}{2}(I_S \otimes \rho_I - \{ |t\rangle \langle t|, \rho \}_+) - i[H_{\text{laser}} + \hat{H}_{\text{spin}}, \rho] + \epsilon^2 \frac{\Gamma}{4} I_S \otimes D_1(\rho_I)
$$

(3.25)

$$
\underbrace{\mathcal{L}_{0}(\rho)}_{\hat{\mathcal{L}}(\rho)} + \underbrace{\epsilon^2 \mathcal{L}(\rho_I)}_{\mathcal{L}(\rho_I)}
$$

where we further applied the trivial transformation into a frame rotating with
$\tilde{\omega}_I I_z^{(0)}$. $\hat{H}_{\text{spin}} = A \sqrt{N} S_z (I_z^{(1)} - \delta / (A \sqrt{N}))$ contains all electron–nuclei interactions
to leading order and can be interpreted as an effective two–photon detuning. The
3 Nuclear spin cooling

The nuclear spin cooling equation describes a nuclear diffusion process caused by the optically assisted hyperfine flip–flop processes that are lowest order in $\epsilon$. Note that in (3.25) we have neglected terms $\propto \epsilon^2$ that only affect the electron evolution as well as (intrinsic) nuclear spin evolution in the absence of hyperfine interaction. We will address the latter when we discuss the cooling time scales.

Before we analyze the qualitative features of the dynamics induced by (3.25), we present the derivation of a master equation that only acts on the nuclear spin degrees of freedom.

**Nuclear master equation**

In the weak excitation limit $\Omega_p, \Omega_c \ll \Gamma$, we can eliminate the state $|t\rangle$, yielding a master equation involving the nuclear and electronic spins only. For simplicity, we assume

$$\delta = 0 \quad (3.26)$$

$$\Omega_c = \Omega_p = \Omega, \quad (3.27)$$

which ensures that the relevant dark and bright electron spin states in the rotating frame are states polarized in the $x$-direction

$$|D\rangle = (|\uparrow\rangle - |\downarrow\rangle)/\sqrt{2},$$

$$|B\rangle = (|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2}. \quad (3.28)$$

A generalization to a finite detuning $\delta$ and arbitrary Rabi frequencies $\Omega_p, \Omega_c$ is straightforward, but offers no further insight. In the case of homogeneous nuclear coupling we then obtain from (3.25) the reduced master equation

$$\dot{\rho} = \Gamma_{\text{eff}} \left( S_x^+ \rho S_x^- - \frac{1}{2} \{ S_x^+ S_x^-, \rho \} \right)$$

$$+ \frac{\Gamma_{\text{eff}}}{2} [S_x, [S_x, \rho]] - iA\sqrt{N} I_z^{(1)}[S_z, \rho]$$

$$+ \epsilon^2 S \otimes D_1(\Gamma_{\text{eff}} \rho_{BB}), \quad (3.29)$$

where $S_x^\pm$ are the electron spin flip operators in $x$-basis ($S_x^- |B\rangle = |D\rangle, S_x^+ |D\rangle = |B\rangle$) and

$$\Gamma_{\text{eff}} = \frac{\Omega^2}{(\Gamma/2)^2 + (A\sqrt{N} I_z^{(1)}/2)^2} \frac{\Gamma}{2} \quad (3.30)$$

is an operator valued effective (electron) spin decay rate. The last line of (3.28) describes the nuclear spin diffusion determined by the nuclear operator proportional to the bright state population $\rho_{BB} = \langle B|\rho B \rangle$.

In order to eliminate the electronic degrees of freedom from (3.28) we use the fact that on the time scales of the electron evolution, the nuclear field can be considered as quasi–static and hence the electron settles quickly (on nuclear time scales) to its interim steady state. We find that on this coarse–grained time scale

$$\rho_{BB} = \frac{1}{2} \left[ 1 - \left( \frac{\Gamma_{\text{eff}}}{|\Delta_{\text{eff}}|} \right)^2 \right] \text{Tr}_S(\rho), \quad (3.30)$$

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3.3 Description of the coupled electron–nuclei dynamics

with $\text{Tr}_S$ denoting the trace over electron spin and

$$|\Delta_{\text{eff}}|^2 = \Gamma_{\text{eff}}^2 + \left(A\sqrt{\bar{N}}I_{z}^{(1)}\right)^2.$$  \hspace{1cm} (3.31)

Using this relation, the electron spin can be eliminated from (3.28), yielding a nuclear rate equation

$$\dot{\rho^n} = \text{Tr}_S(\dot{\rho}) = D_1(\Gamma_{\text{nuc}}\rho^n),$$  \hspace{1cm} (3.32)

where we defined the state-dependent nuclear diffusion rate

$$\Gamma_{\text{nuc}} = \epsilon^2 \left[1 - \left(\frac{\Gamma_{\text{eff}}}{|\Delta_{\text{eff}}|}\right)^2\right] \Gamma_{\text{eff}}.$$  \hspace{1cm} (3.33)

Clearly, $\Gamma_{\text{nuc}}$ vanishes for all states in the kernel of $\bar{I}_z^{(1)}$; these states fulfill the two-photon resonance condition imposed in (3.26) and are steady states of the dynamics.

3.3.2 Qualitative description of the coupled dynamics

We now analyze the obtained equations by first looking at the term $L_0(\rho)$ in (3.25), which determines the fast electronic dynamics on a time scale for which the nuclear spins can be assumed to remain unchanged. Then we consider the slow nuclear dynamics induced by $\bar{L}$.

The term $L_0(\rho)$ describes the evolution of the coupled system that leaves $\bar{I}_z$ a constant of motion; for a two-photon detuning $\delta$ each eigenvector $|\lambda\rangle$ of $\bar{I}_z$ (with eigenvalue $\lambda$) corresponds to a (quasi) steady state solution, i.e.

$$L_0(\rho_\lambda) = 0,$$

$$\rho_\lambda = \rho^e(\lambda) \otimes |\lambda\rangle \langle \lambda|.$$  \hspace{1cm} (3.34)

The eigenvalue $\lambda$ can be considered a generalization of the eigenvalue of $I_z^{(1)}$ which are proportional to $m$, the nuclear spin projection along $z$. Here, $\rho^e(\lambda) = \langle \lambda|\rho_\lambda|\lambda\rangle$ is the steady state solution of the optical Bloch equations for which

$$\delta_{\text{eff}} = \lambda - \delta$$

gives the effective two-photon detuning that determines the CPT condition. Hence, $L_0$ forces the electron into a (quasi) steady state, determined by the effective detuning $\delta_{\text{eff}}$ that depends not only on the bare optical detunings and external magnetic fields, but also the generalized Overhauser field.

The term $\bar{L}$ describes the evolution of $\bar{I}_z$ due to hyperfine assisted light scattering, taking place on time scales longer than that of the electron by a factor $\epsilon^{-2}$. The corresponding nuclear spin flip rates in positive ($R^+$) and negative direction ($R^-$) are:

$$R^{\pm} = \epsilon^2 \frac{\Gamma}{2} \rho^{e}_{tt}(\lambda) \left(I_{z}^{(1)}I_{\pm}^{(1)}\right),$$

where $\rho^{e}_{tt}(\lambda) = \langle t|\rho^e(\lambda)|t\rangle$ is the population in state $|t\rangle$. $R^{\pm}$ are directly obtained from (3.25) and are plotted in Fig. 3.2 (b) (dashed black lines). Each nuclear spin flip event changes $\langle \bar{I}_z \rangle$ by a value of order $A_i/(A\sqrt{\bar{N}})$. 

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The interplay of the dynamics due to $\hat{H}_{\text{spin}}$ and $\hat{L}$ is at the heart of the nuclear–spin cooling scheme we analyze in this chapter. For nuclear states with $\delta_{\text{eff}} = 0$ the system is in two–photon resonance and the electronic system is transparent such that $\text{Tr}\{\rho_{tt}\} = 0$: as a consequence, the nuclear spin diffusion vanishes since the rates directly depend on $\rho_{tt}$. Hence, the system is trapped in a dark state. Since the generalized Overhauser field in an electronic–nuclear dark state is locked to a fixed value, its variance will be strongly reduced (nuclear state narrowing) suppressing hyperfine–induced electron spin decoherence. Strikingly, by narrowing the generalized Overhauser field, the second order contribution to hyperfine–induced electron spin decoherence is eliminated as well. For all nuclear states satisfying $\delta_{\text{eff}} \approx 0$, the excited electronic state population will remain small ($\text{Tr}\{\rho_{tt}\} \propto \delta_{\text{eff}}^2$), ensuring that the spin diffusion rate will remain vanishingly low: we refer to this subspace as the “trapping region”. In contrast, nuclear states with $\delta_{\text{eff}} \neq 0$ render the electron optically active and the generalized Overhauser field experiences random diffusion (“recycling region”). Through successive spin–flip events, the nuclear reservoir probes different spin configurations with distinct $\langle \tilde{I}_z \rangle$. Eventually, the diffusion allows the nuclei to reach a configuration that yields $\delta_{\text{eff}} \approx 0$, thereby locking the system in the trapping region. Due to the quasi–continuous nature of the $\tilde{I}_z$ spectrum in the limit $N \gg 1$ (see section 3.4.4) and inhomogeneous hyperfine couplings, the dark–state condition $\delta_{\text{eff}} = 0$ can be satisfied for a wide range of initial detunings $\Delta \omega_p$.

### 3.4 Nuclear spin cooling and spectroscopic signatures

To confirm the predictions derived from (3.25), we consider, for simplicity, nuclear spin-1/2 systems. While our analysis applies to a broad class of solid–state emitters from various types of quantum dots (QD) to nitrogen–vacancy (NV) centers, we now focus on a single–electron–charged QD in Voigt geometry where the optically excited state has an electron–singlet and a hole [186, 176, 122], i.e. $|\uparrow \downarrow \uparrow \rangle$. A numerical analysis of the dynamics is then feasible in two limits. First, we consider the case of homogeneous hyperfine coupling for which we investigate the narrowing of the distribution starting from the exact master equation (3.11). This treatment intrinsically accounts for higher order hyperfine processes that are neglected in the master equation (3.25). To understand the limits of the narrowing, we discuss these higher order terms and their impact on the nuclear spin dynamics. Then, we focus on the inhomogeneous coupling regime for which we adapt a semiclassical description of the nuclear spin environment. The ensuing rate equations allow us to use Monte Carlo simulations to uncover the optical signatures that are expected in QDs and confirm the narrowing of the nuclear spin distribution.

#### 3.4.1 Homogeneous hyperfine coupling

For homogeneous coupling ($A_i = A = A_H/N$) we calculate the dependence of the steady state standard deviation of the Overhauser field $\sigma_{\text{OF}}$ directly from the full master equation (3.11), which contains all orders of the hyperfine interaction including processes that result in a (small) finite decay rate out of the dark state
3.4 Nuclear spin cooling and spectroscopic signatures

Figure 3.2: (a) Nuclear spin diffusion rates (black lines) depending on the nuclear spin projection $\lambda$ assuming homogeneous coupling. Parameters are $N = 4 \times 10^4$, $\Gamma = 1$ GHz, $A_H = \omega_z = 100$ $\mu$eV and $\Omega = \Omega_p = \Omega_c = 0.1$ GHz. The rates calculated using the quantum mechanical (for the subspace $J = \sqrt{N/2}$, $R_{\pm}^{\text{qm}}$ dashed line) and the semiclassical ($R_{\pm}^{\text{sc}}$ dotted line) descriptions show both qualitative and quantitative agreement. The red curve shows the nuclear spin distribution obtained when $\Omega = 0.02\Gamma$, assuming $T_2^{-1} = 100\text{s}^{-1}$. (b) The dependence of the steady state standard deviation of the Overhauser field $\sigma_{\text{OF}}$ as a function of $\Omega$ in the limit of homogeneous coupling; the solid (dashed) line is obtained by taking $T_2^{-1} = 100\text{s}^{-1}$ ($T_2^{-1} = 0\text{s}^{-1}$). Figure credit: Eric Kessler

as well as a finite electron spin decoherence rate $T_2^{-1}$. Figure 3.2 (b) shows $\sigma_{\text{OF}}$ for the case $\delta = 0$ and $T_2^{-1} = 100\text{s}^{-1}$ ($T_2^{-1} = 0\text{s}^{-1}$) as a function of $\Omega$, where we find that $\sigma_{\text{OF}}$ decreases with decreasing $\Omega$ until it reaches a minimum of $\sigma_{\text{OF}} \approx 2A_H/N$ ($\sigma_{\text{OF}} \approx 0.7A_H/N$) for $\Omega \approx 0.2\Gamma$. This result can be understood by recalling that the width of the transparency dip in CPT scales as $\Omega^2/\Gamma$, implying that the range of Overhauser field values yielding transparency can be narrowed simply by reducing $\Omega$. However, for $\Omega < 0.2\Gamma$, we find that $\sigma_{\text{OF}}$ increases rapidly; for such small values of $\Omega$, the coupled electron–nuclei system spends a substantial amount of time outside the transparency region due to finite decay out of the dark state, leading to the observed increase in steady state value of $\sigma_{\text{OF}}$. Nevertheless, we find that even for $\Omega_p = \Omega_c = 0.02\Gamma$ where $\sigma_{\text{OF}} \approx 9A_H/N$, the nuclear spin distribution is strongly peaked around states $|\lambda_0\rangle$ such that $\tilde{I}_z |\lambda_0\rangle = 0$ (Fig. 3.2 (a) solid red line).

To understand the physical origin of the limit in the cooling, we note that even though nuclear states in the kernel of $\tilde{I}_z^{(1)} - \delta/(A\sqrt{N})$ decouple completely from the electron degrees of freedom and the evolution of these states comes to rest as the system is trapped, higher order corrections - which we have neglected so far - can contribute to a finite, state–independent nuclear diffusion rate out of the trapping region. In the following we identify and discuss these corrections.

First, we consider the effect of the second order term $\frac{1}{2}\epsilon A\sqrt{N}\tilde{I}_z^{(2)}$ obtained in (3.18) and which we neglected in (3.25) and which does not commute with the generalized Overhauser field $\tilde{I}_z^{(1)}$. Consequently, the eigenstates of the generalized Overhauser field evolve under its action. To estimate the corresponding nuclear diffusion rate, we consider the equation of motion of the corresponding Heisenberg
operator
\[
\frac{d}{dt} \tilde{I}_z^{(1)} = -i \left[ \tilde{I}_z^{(1)}, \frac{1}{2} \epsilon A \sqrt{N} I_z^{(2)} \right],
\]  
(3.35)

Note that, since the perturbation commutes with the zeroth order (i.e. standard Overhauser field) part of the general Overhauser field \([\tilde{I}_z^{(1)}, \tilde{I}_z^{(2)}] = 0\) the effect is of higher order \(\propto \epsilon^2\). Furthermore, the spin operators are normalized such that typical matrix elements of \(I_+^{(n)} I_-^{(m)}\) are of order \(\sim \sqrt{N}^{-1/2}\), since
\[
I_+^{(n)} I_-^{(m)} = \sum_{i,j=1}^{N} \left( \frac{A_i}{A \sqrt{N}} \right)^n \left( \frac{A_j}{A \sqrt{N}} \right)^m I_+ I_-
\]
\[
= A_i = A_j = A \quad \sim N^{-1}(n+m)/2
\]
\[
\sim \sqrt{N}/2 \sim N^{1-(n+m)/2}.
\]

Here, we have evaluated the matrix elements for the collective Dicke states \(|J, m\rangle\) with a typical \(J \sim \sqrt{N}/2\) (see section 2.3.4). In fact, the number of bigger matrix elements (at most by a factor \(N\)) is exponentially small and can reduce the trapping region (which is defined as the set of eigenvectors with sufficiently small eigenvalues of \(\tilde{I}_z^{(1)} - \delta/(A \sqrt{N})\), and which is shown to be sizeable in section 3.4.4) by an insignificant fraction only. Therefore we can roughly estimate the rate of change
\[
\left| \frac{d}{dt} \tilde{I}_z^{(1)} \right| \sim \frac{\epsilon^2}{N} A \sqrt{N}.
\]  
(3.36)

This expression is by a factor \(N\) smaller than the optically induced nuclear diffusion rates
\[
R^\pm \sim \epsilon^2 \Gamma \tilde{I}_+^{(1)} \tilde{I}_-^{(1)} = \epsilon^2 \Gamma \sum_{i,j=1}^{N} \frac{A_i}{A \sqrt{N}} \frac{A_j}{A \sqrt{N}} I_+ I_- \frac{A_i}{A \sqrt{N}} \frac{A_j}{A \sqrt{N}} = 1/N \epsilon^2 \frac{1}{N} \Gamma N = \epsilon^2 \Gamma.
\]

Next we consider higher order corrections in the perturbation theory by expanding the Hamiltonian to third order. The generator of the third order correction \(S^{[2]}\) can be calculated using (3.14):
\[
S^{[2]} = \epsilon \delta \left( S_+ I_-^{(1)} - S_- I_+^{(1)} \right)
+ \epsilon \epsilon_c \left( I_-^{(1)} \langle \uparrow \rangle \langle \downarrow \rangle - h.c. \right) + \epsilon \epsilon_p \left( I_+^{(1)} \langle \downarrow \rangle \langle \uparrow \rangle - h.c. \right)
+ \epsilon^2 \left[ S_- I_+^{(2)} - S_+ I_-^{(2)} - 2 \left( S_+ I_-^{(1)} I_+^{(1)} - S_- I_+^{(1)} I_-^{(1)} \right) \right],
\]  
(3.37)

with the expansion parameters \(\epsilon_\delta = \frac{\delta}{\omega_z}, \epsilon_{p/c} = \frac{\Omega_{p/c}}{\omega_z} \ll 1\). The third order Hamil-
3.4 Nuclear spin cooling and spectroscopic signatures

The first term is of the exact same form as $H^{[2]}$ and can easily be incorporated in the above considerations of section 3.3.1 as a small ($\epsilon$) correction. The second and third term describe two different types of laser assisted nuclear diffusion. The first type supports the scheme, since $H_{\text{laser}}$ only couples to the electronic bright state $|B\rangle$. Thus this diffusion comes to rest whenever the electron is in the dark state $|D\rangle$. The second type of laser assisted nuclear diffusion couples to the dark state and thus, in principle, represents a possible escape mechanism from trapping states. However, since - as discussed above - the diffusion operator $I^{(2)}_z$ commutes with the zero order part of the generalized Overhauser field the contribution of the third term is yet of one order $\epsilon$ smaller than the process of (3.36) and thus safely negligible. The fourth and fifth term of (3.38) originate in third order contributions of the hyperfine interaction. While the fourth term can be incorporated in the definition of the generalized Overhauser field, the fifth term represents a state independent nuclear diffusion, effectively of the same order as the one of (3.36).

All these processes are taken into account exactly in the homogeneous simulations since they are based on (3.3) and account for the small but finite standard deviation in the steady state.

3.4.2 Inhomogeneous hyperfine coupling

To analyze the inhomogeneous coupling limit with a quasi–continuous Overhauser field, we adopt a semiclassical approach where the nuclear spins are described at all times by a product of $I^z_i$-eigenstates. Furthermore, for simplicity we replace the generalized Overhauser field $\hat{I}_z$ by the Overhauser field $I^{(1)}_z$ in the simulations since the spectrum, the density of states, and the nuclear spin dynamics $\dot{I}^{(1)}_z$ resulting from $\hat{I}_z$ and $I^{(1)}_z$ are equivalent. The proofs that justify this replacement will be given below in section 3.4.4. In this case, the master equation (3.25) reduces to rate equations which can be simulated using Monte Carlo techniques, and allowing us to uncover the spectroscopic signatures of the Overhauser field selective CPT cooling scheme.

The semiclassical limit can be derived from the master equation (3.25) by replacing the collective spin decay by independent decay of individual spins. This

\begin{equation}
H^{[3]} = -\epsilon^2 \delta \left( S_z \{ I^{(1)}_{+}, I^{(1)}_{-}\} + I^{(2)}_z \right) \\
+ \frac{1}{4} \epsilon^2 \left\{ I^{(1)}_{+}, I^{(1)}_{-}\right\}_+ H_{\text{laser}} \\
+ \frac{1}{2} \epsilon^2 I^{(2)}_z \left( \Omega_c \langle \downarrow | t \rangle - \Omega_p \langle \uparrow | t \rangle + h.c. \right) \\
+ \epsilon^2 A \sqrt{N} S_z \left( I^{(1)}_{+} I^{(1)}_{-} + h.c. \right) \\
+ \frac{1}{8} \epsilon^2 A \sqrt{N} \left( \{ I^{(2)}_{+}, I^{(2)}_{-}\}_+ + \{ I^{(1)}_{+}, I^{(1)}_{-}\}_+ - I^{(1)}_{+} I^{(2)}_z \right),
\end{equation}

(3.38)
3 Nuclear spin cooling

is accomplished by making the following substitutions in the master equation:

\[
I_+^{(1)} \rho I_-^{(1)} = \sum_{ij} \frac{A_i}{A\sqrt{N}} \frac{A_j}{A\sqrt{N}} I_+^{(1)} I_-^{(1)} \rho I_+^{(1)} I_-^{(1)} \rightarrow \sum_i \frac{A_i^2}{A^2 N} I_+^{(1)} I_-^{(1)} \rho I_+^{(1)} I_-^{(1)},
\]

\[
I_-^{(1)} \rho I_+^{(1)} = \sum_{ij} \frac{A_i}{A\sqrt{N}} \frac{A_j}{A\sqrt{N}} I_-^{(1)} I_+^{(1)} \rho I_-^{(1)} I_+^{(1)} \rightarrow \sum_i \frac{A_i^2}{A^2 N} I_-^{(1)} I_+^{(1)} \rho I_-^{(1)} I_+^{(1)},
\]

\[
\left\{ I_+^{(1)} I_-^{(1)} + I_-^{(1)} I_+^{(1)}, \rho \right\}_+ = \left\{ \sum_{ij} \frac{A_i}{A\sqrt{N}} \frac{A_j}{A\sqrt{N}} (I_+ I_- + I_- I_+), \rho \right\}_+ \rightarrow \left\{ \sum_i \frac{A_i^2}{A^2 N} (I_+ + I_-), \rho \right\}_+.
\]

The replacement of the collective spin operators by single spin operators is justified in the limit where coherences between nuclear spin product states vanish on time scales short compared to their lifetime; this condition would be satisfied in systems with large inhomogeneities - either in the nuclear spin splitting or in hyperfine coupling. We now coarse grain the nuclear motion with respect to the electron dynamics and from the resulting master equation we obtain rate equations that describe the nuclear spin evolution.

In order to mimic the inhomogeneous character of the hyperfine coupling we introduce a shell model of the QD with \( M \) different classes of nuclear spins; the nuclei in class \( \nu \) have identical \( a_\nu \) and their net spin polarization is \( m_\nu = \frac{1}{2}(N_\nu^+ - N_\nu^-) = \sum_{i=\nu} I_i^z \), where \( N_\nu^+ (N_\nu^-) \) denote the total number of up (down) spins in class \( \nu \). The derived rate equation for the joint probabilities \( \mathcal{P}(\{m_\mu\}) \) associated with the nuclear spin configuration \( \{m_\mu\} \) is given by

\[
\frac{\partial \mathcal{P}(\{m_\mu\})}{\partial t} = \sum_{\nu = 1}^{M} \mathcal{P}(\{\tilde{m}_\mu\}) N^-_\nu(\{\tilde{m}_\mu\}) \Gamma^+_\nu(\{\tilde{m}_\mu\})
\]

\[
+ \sum_{\nu = 1}^{M} \mathcal{P}(\{\tilde{m}_\mu\}) N^+_\nu(\{\tilde{m}_\mu\}) \Gamma^-_\nu(\{\tilde{m}_\mu\})
\]

\[
- \sum_{\nu = 1}^{M} \mathcal{P}(\{m_\mu\}) [N^-_\nu \Gamma^+_\nu(\{m_\mu\}) + N^+_\nu \Gamma^-_\nu(\{m_\mu\})],
\]

where \( \Gamma^\pm_\nu(\{m_\mu\}) = \left( \frac{A_\nu}{\sqrt{A_\nu}} \right)^2 \rho_\text{int}(\{m_\mu\}) \) are the rates at which nuclear spins of the \( \nu^{th} \) class are flipped if the nuclear spin polarizations in each class are given by \( \{m_\mu\} \) and where \( A_\nu \) is the average hyperfine coupling constant in the \( \nu^{th} \) class. \( \{\tilde{m}_\mu\} \) denotes the nuclear spin configuration that differs from the configuration \( \{m_\mu\} \) only in the \( \nu^{th} \) class, with polarization \( m_\nu - 1 \ (m_\nu + 1) \). The factors \( N^-_\nu (N^+_\nu) \) account for the number of nuclear spins in the configuration \( \{\tilde{m}_\mu\} \) \( \{\tilde{m}_\mu\} \) that could be flipped to reach \( \{m_\mu\} \).

We simulate the evolution of the nuclear spins with a Monte Carlo method. We assume in our numerical simulations that the QD contains 100 nuclear spins, grouped into five concentric shells \( (M = 5) \) with different hyperfine coupling constants that are determined by the 3D Gaussian electronic envelope function. The coupling constants for these shells are taken to be 0.0934\( \Gamma \), 0.0828\( \Gamma \), 0.0678\( \Gamma \), 0.0513\( \Gamma \), 0.0358\( \Gamma \) and the corresponding total numbers of nuclear spins in each

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Figure 3.3: Shells of nuclear spins and their hyperfine coupling constants to model inhomogeneous coupling (in units of $\Gamma$).

Shell are chosen to be 2, 8, 16, 28, 46 (see Fig. 3.3). The coupling constants are chosen to ensure that the standard deviation of the Overhauser field seen by the QD electron for nuclei in a completely mixed state satisfies $\sigma_{OF}(\rho) = \frac{\Gamma}{4}$. We do not keep track of the exact configuration within each class ($\nu$) of nuclear spins and assume that any configuration of spins leading to the same $m_\mu$ is equally likely and that the nuclear spin distribution in each shell is independent of the other shells.

Figure 3.4 (a) shows the result of such simulations as the probe field is scanned across the (bare) resonance while keeping $\Delta \omega_c = 0$, in the limit of a vanishing decay rate out of the dark state. To obtain the probe field absorption lineshape as well as the Overhauser field variance, we assume that for each probe field detuning, we start out from a completely mixed nuclear state $\rho^n$ and evolve the coupled system until $t_{fin} = 10^{12} \Gamma^{-1}$. We find that instead of exhibiting a narrow transparency dip at (bare) two–photon–resonance (as in the absence of hyperfine coupling, Fig. 3.4 (a), dashed curve), the coupled electron–nuclei system displays a drastically broadened transparency window with width significantly larger than $\Gamma$ (Fig.3.4 (a), green ($\Omega = 0.4\Gamma$) and blue ($\Omega = 0.2\Gamma$) lines). The two–photon resonance condition $\delta_{eff} = \lambda - \delta = 0$ can be satisfied for a large range of initial detunings because the set of Overhauser fields resulting from all possible nuclear spin configurations of the toy model QD described above is quasi–continuous. However, the density of states in the toy model is not a smooth function of the Overhauser field, which explains the observed variations in the absorption strength (and $\sigma_{OF}$) within the extended transparency region: the Overhauser field required to fulfill $\delta_{eff} = 0$ for some detunings $\Delta \omega_p$ is composed of a nuclear spin configuration that requires a large polarization in one of the nuclear spin classes. It will take many nuclear spin flips to reach such a state, since the initial state is taken to be Gaussian in all classes. For our finite simulation time, these configurations are not captured and lead to a finite absorption strength for some $\delta$.

Concurrent with the broadening of the dark resonance, the Overhauser field
distribution is narrowed dramatically from its value in the absence of optical excitation (Fig. 3.4 (b), black line) such that $\sigma_{\text{OF}}$ is smaller than the change induced by flipping one nuclear spin of the most weakly coupled class (Fig. 3.4 (b), green and blue lines). This narrowing of the distribution is a consequence of the feedback between the electron and nuclear dynamics discussed in section 2.5.2: Since optical absorption is strongly reduced for Overhauser fields satisfying $\delta_{\text{eff}} = 0$, so are the dominant nuclear spin flip processes. This leads to a stabilization of this specific Overhauser field and hence reduces the spread of the nuclear spin distribution.

To summarize, the semiclassical simulations show the striking features that are consequences of the optically induced nuclear spin diffusion which allows the coupled system to evolve into the electron-nuclei dark state $\rho_D = |D\rangle\langle D| \otimes \rho^D_n$, where $\rho^D_n$ is a nuclear spin density operator that yields $\delta_{\text{eff}} = 0$.

### 3.4.3 Suppression of $T_2$ decoherence

We note that when the system is in the prepared state, the effect of the indirect interaction, which is one of the major contributions that limit $T_2$ [190, 191], is strongly suppressed. The indirect interaction describes the electron–mediated coupling of two nuclear spins via the hyperfine interaction. This interaction is given by $H^{[2]}$ in equation (3.17). We now consider the evolution of $I^{(1)}_z$ under $H^{[2]}$, where we neglect the smaller second term that is discussed in section 3.4.1. Then, the dominant contribution is

$$\frac{dI^{(1)}_z}{dt} = i \left[ -\frac{1}{2} \epsilon A \sqrt{N} S_z \{ I^{(1)}_+ I^{(1)}_+ + I^{(1)}_- I^{(1)}_- \} \right]$$

$$= -\frac{i}{2} \epsilon A \sqrt{N} S_z \left( [I^{(1)}_+ I^{(1)}_+ + I^{(1)}_- I^{(1)}_-] \right)$$

$$= -\frac{i}{2} \epsilon A \sqrt{N} S_z \left( \sum_{i,j} \frac{A_j - A_i}{A \sqrt{N}} \frac{A_i}{A \sqrt{N}} \frac{A_j}{A \sqrt{N}} I^+_i I^+_j + \sum_{i,j} \frac{A_i - A_j}{A \sqrt{N}} \frac{A_i}{A \sqrt{N}} \frac{A_j}{A \sqrt{N}} I^-_i I^-_j \right)$$

$$= 0.$$  

The last line follows trivially for $i = j \Rightarrow A_i = A_j$. For $i \neq j$ the operators $I^+_i$ and $I^-_j$ commute. Hence, we conclude that the indirect interaction does not contribute to $T_2$ processes except for small contributions which are discussed in section 3.4.1.

### 3.4.4 Spectrum and density of states of the generalized Overhauser field

The semiclassical simulations above were performed for the Overhauser field rather than the generalized Overhauser field for simplicity. To justify this replacement, we show here that the nuclear spin flip rates for the two cases agree up to a correction of the order $\epsilon$. Furthermore, it is important that the spectrum and density of states of the generalized Overhauser field do not differ significantly from the spectrum and density of states of the Overhauser field; these quantities play a significant role in determining the evolution of the coupled electron–nuclei system and the ensuing optical signatures.
3.4 Nuclear spin cooling and spectroscopic signatures

Figure 3.4: (d) The absorption lineshape for Rabi frequencies $\Omega = 0.2 \Gamma$ (blue) and $\Omega = 0.4 \Gamma$ (green): in stark contrast to the standard CPT profile (dotted lines), the dark resonance is drastically broadened (solid lines). (e) $\sigma_{OF}$ for $\Omega_p = \Omega_c = 0.2 \Gamma$ (blue line) and $\Omega_p = \Omega_c = 0.4 \Gamma$ (green line) is reduced to the level below that of a single nuclear spin flip (red dashed line). The red line shows $\sigma_{OF}$ in the absence of lasers.
We first note that for the main part of the nuclear Hilbert space – namely the domain where the operator \( \tilde{\mathcal{I}}^{(1)}_z - \delta/(A\sqrt{N}) \) is large (recycling region) – the \( \epsilon \)-correction in the generalized Overhauser field represents a negligible perturbation to the hyperfine interaction. However, in the domain of small eigenvalues of \( \tilde{\mathcal{I}}^{(1)}_z - \delta/(A\sqrt{N}) \) which also referred to as trapping region, this perturbative picture is not trivially justified. To be more quantitative, we define the trapping region to be within the small interval

\[
L(\delta, \eta) = \left( \frac{\delta - \eta}{A\sqrt{N}} \right), \quad (3.40)
\]

centered around \( \delta/g \). This is illustrated in Fig. 3.5.

Before considering the modification of the density of states brought about by the generalized Overhauser field, we investigate the range of \( \mathcal{I}_z \) eigenvalues that make up a substantial part of the Hilbert space for homogeneous and inhomogeneous coupling. Translated to Overhauser field values, we find that for Overhauser fields exceeding \( \sim A\sqrt{N} = A_H/\sqrt{N} \) are not easily reached. Correspondingly, at bare two-photon detuning exceeding \( \delta \sim A\sqrt{N} \) the reduction of the optical absorption is not expected on a reasonable time scale (see Fig. 3.4 (a)). To this end, we first use Monte Carlo simulations to show that the density of eigenstates of the homogeneous (\( A_i = A_j = A \)) and inhomogeneous operator \( \mathcal{I}_z^{(1)} \), respectively, is identical (up to small corrections) for our system parameters (\( N = 10^4 \), Gaussian distribution of coupling strengths), see Fig. 3.6 (a). Using this equivalence, the following argument for the homogeneous case extends to inhomogeneous coupling. The collective Dicke states \( |J, m\rangle \) are eigenstates of the collective nuclear operator \( \mathcal{I}_z^{(0)} = \sum_i \mathcal{I}_z^i \), where \( J \) denotes the total spin and \( m \) the spin projection in \( z \)-direction (see chapter 2). They satisfy

\[
\mathcal{I}_z^{(0)} |J, m\rangle = m |J, m\rangle.
\]

The analytic expression for the degeneracy of each eigenvalue \( m/\sqrt{N} \) is

\[
B_m = \left( \frac{N}{N/2 + m} \right), \quad (3.41)
\]

and hence the number of eigenstates of the homogeneous and inhomogeneous operator \( \mathcal{I}_z^{(1)} \) with small eigenvalues is exponentially large in the number of spins. Still neglecting the \( \epsilon \)-correction of the generalized Overhauser field, this implies that the trapping regions close to the center of the spectrum \( |\delta/(A\sqrt{N})| \lesssim 1 \) constitute a substantial part of the Hilbert space and can be reached in reasonable times by the nuclear random diffusion. For trapping regions \( |\delta/(A\sqrt{N})| \gtrsim 1 \), i.e., very large laser detunings, the size of the trapping region drops exponentially (Fig.3.6(a)) and cannot be explored by the nuclear diffusion (see Fig. 3.4 (a)).

Next we show that the generalized Overhauser field fulfills the same property by deriving a relation between the spectrum of the \( \tilde{\mathcal{I}}^{(1)}_z \) and \( \mathcal{I}_z^{(1)} \) operators. In the homogeneous case the \( \epsilon \)-correction commutes with the unperturbed part

\[
[I_z^{(1)}, \{I_z^{(1)}, I_z^{(1)} \}_+] = 0,
\]
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Figure 3.5: For homogeneous hyperfine coupling we plot the eigenvalues \( \lambda_{I_z^{(1)}} \) of \( I_z^{(1)} \) with their corresponding degeneracy \( B_m \). The small interval \( L \) is shown in red. For homogeneous hyperfine coupling the separation of the eigenvalues is given by \( 1/\sqrt{N} \). For inhomogeneous hyperfine coupling the degeneracy is lifted (yellow). Trapping regions up to \( |I_z^{(1)}| \sim 1 \leftrightarrow \delta/(A\sqrt{N}) \sim 1 \) can be explored on reasonable time scales. The generalized Overhauser field shifts most states in energy by a small correction \( \epsilon \) (blue).

Figure 3.6: (a) Comparison of the density of eigenstates \( d(|\alpha|) \) of the homogeneous and inhomogeneous nuclear operator \( I_z^{(1)} \). The density of states is averaged over segments of size \( 1/\sqrt{N} \). While in the homogeneous case the density of states can be calculated exactly, in the inhomogeneous case it is evaluated using Monte Carlo simulations. (b) The number of eigenvalues in small intervals of size \( 1/\sqrt{N} \) of the generalized Overhauser field approximately equals the number of states for the standard Overhauser field. In the simulations we assumed \( N = 10^4 \) and unitless hyperfine coupling coefficients \( \sum a_i^2 = 1 \) arising from a Gaussian electron wave function. Figure credit: Eric Kessler
3 Nuclear spin cooling

and a common eigenbasis is again given by the Dicke states $|J, m\rangle$. The vast majority of states within the kernel of $I_z^{(1)}$, i.e. $|J, m\rangle$ with $m = 0$, lie in $J$ subspaces around $J = \sqrt{N}/2$ since the degeneracy of each subspace $J$ is given by

$$D_J = \left( \frac{N}{N/2 - J} \right) - \left( \frac{N}{N/2 - J - 1} \right).$$

Thus, up to a negligible fraction of states that are in a larger $J$ subspace, most states will be shifted in energy by a small amount of order $\epsilon$ since

$$\frac{1}{2} \epsilon \{ I_z^{(1)}, I_z^{(1)} \} + |J, m\rangle = \frac{1}{2} \epsilon \sum_i \sum_j \frac{A_i}{A \sqrt{N}} \frac{A_j}{A \sqrt{N}} (I^+_i I^-_j + I^-_i I^+_j) |J, m\rangle$$

$$A_{ij} = \frac{1}{2} \epsilon \left( I^+_i I^-_j + I^-_i I^+_j \right) |J, m\rangle$$

$$= \frac{1}{2} \epsilon \frac{1}{N} \left( J(J + 1) - m(m - 1) \right)$$

$$+ J(J + 1) - m(m + 1))$$

$$= \frac{1}{N} \left( J(J + 1) - m^2 \right)$$

$$\approx \epsilon \frac{1}{4}$$

The same can be shown for states with finite eigenvalue of $I_z^{(1)}$ in the range $\sim (-1, 1)$ which corresponds to $|m| \sim 1/\sqrt{N}$ and hence Overhauser fields up to $\sim A \sqrt{N}$. To summarize, the positive operator of the correction $\frac{1}{2} \epsilon \{ I_z^{(1)}, I_z^{(1)} \} +$ shifts the whole spectrum in the region of interest by a small amount $\propto \epsilon$ and thus preserves the required property of large density of states in any trapping region $L(\delta, \epsilon)$ close to the center of the spectrum.

In the inhomogeneous case this simple argument fails, since one cannot easily construct an eigenbasis of the generalized Overhauser field. We estimate the density of states in the following. For the inhomogeneous generalized Overhauser field operator $\tilde{I}_z^{(1)}$, the number of states within the trapping region $L(\delta, \eta)$ is given by

$$N^\epsilon (L(\delta, \eta)) = \int_{L(\delta, \eta)} dE \text{ Tr} \left( \delta \left( E - \tilde{I}_z^{(1)} + \frac{1}{2} \epsilon \{ I_z^{(1)}, I_z^{(1)} \} + \right) \right).$$

(3.42)

Approximating the $\delta$-functions by Lorentzians of width $\gamma \ll \eta$, i.e.

$$\delta(E - \tilde{I}_z^{(1)}) \approx \frac{\gamma}{(E - \tilde{I}_z^{(1)})^2 + \gamma^2},$$

and using the expansion

$$\frac{1}{A - \epsilon B} = \frac{1}{A} \sum_{n=0}^\infty \left( \epsilon \frac{B}{A} \right)^n$$

(3.43)

with the definitions

$$\tilde{A} = \gamma^2 + (E - I_z^{(1)})^2,$$

$$\tilde{B} = -\frac{1}{2} \{ (E - I_z^{(1)}), \{ I_+^{(1)}, I_-^{(1)} \} + \} - \frac{1}{4} \epsilon \left( \{ I_+^{(1)}, I_-^{(1)} \} + \right)^2 =: Q + \epsilon P,$$
we find
\[ N'(L(\delta, \eta)) = \sum_{n=0}^{\infty} U^{(n)} = N^0(L(\delta, \eta)) + \sum_{n=1}^{\infty} U^{(n)}, \] (3.46)
\[ U^{(n)} = \gamma \epsilon^n \int_{L(\delta, \eta)} dE \text{Tr} \left( \frac{1}{A} \left[ \frac{1}{A} \right]^n \right), \] (3.47)

where \( N^0(L(\delta, \eta)) \) denotes the number of eigenstates of the standard Overhauser field operator \( I_z \) within the trapping region \( L(\delta, \eta) \). A pessimistic approximation shows that the sum \( \sum_{n=1}^{\infty} U^{(n)} \) can be upper bounded by \( \sim N^0(L(\delta, \eta)) \) for large trapping regions \( \eta \gg \epsilon \), i.e. the number of eigenstates changes at most by a factor of order 1.

To get further insight into how the \( \epsilon \)-correction of the generalized Overhauser field modifies the number of states in a trapping region for inhomogeneous coupling, we turn to numerical methods. Using the above definitions the quotient of the number of states in the perturbed and unperturbed case can be written to first order in \( \epsilon \) as
\[ \frac{N'(L(\delta, \eta))}{N^0(L(\delta, \eta))} = \frac{\int_{L(\delta, \eta)} dE \text{Tr}(\frac{1}{A}(1 + \epsilon Q \frac{1}{A}))}{\int_{L(\delta, \eta)} dE \text{Tr}(\frac{1}{A})} = \frac{\sum \bar{n} \int_{L(\delta, \eta)} dE \, g(\bar{n}, E) f(\bar{n}, E)}{\sum \bar{n} \int_{L(\delta, \eta)} dE \, g(\bar{n}, E)}, \] (3.48)

where we defined
\[ g(\bar{n}, E) = \langle \bar{n} | \frac{1}{A} | \bar{n} \rangle, \] (3.49)
\[ f(\bar{n}, E) = 1 + \epsilon \langle \bar{n} | Q \frac{1}{A} | \bar{n} \rangle, \] (3.50)

for a nuclear product state \( |\bar{n}\rangle \). Expressions like the one of (3.48) can be efficiently evaluated using Monte Carlo simulation with importance sampling [192]. This method uses a more efficient sampling according to the probability distribution \( g(\bar{n}, E) \) (i.e. strongly weighted regions are favored in the sampling) instead of the sampling of random configurations \((\bar{n}, E)\) within the entire state space. In particular, it can be shown that
\[ \frac{\sum \bar{n} \int_{L(\delta, \eta)} dE \, g(\bar{n}, E) f(\bar{n}, E)}{\sum \bar{n} \int_{L(\delta, \eta)} dE \, g(\bar{n}, E)} = \lim_{t \to \infty} \frac{1}{t} \sum_{i=1}^{t} f(\bar{x}_i), \] (3.51)

where the vectors \( \bar{x}_i \) stand for particular configurations of the random variables \((\bar{n}, E)\) which are distributed according to \( g(\bar{x}_i) \). The algorithm realizing the above scheme contains the following steps:

1. We start from a random sample \( \bar{x} := (\bar{n}, E) \) \( (E \in L(\delta, \eta)) \).

2. Then we create a new sample \( \bar{x}' \) by randomly changing one coordinate \( x_i \) (the ratio of spin flips and change of \( E \) is defined a priori).

3. If \( g(\bar{x}')/g(\bar{x}) > s \)–where (in each step) \( s \) is a randomly created number \( \in (0, 1) \)–we add \( f(\bar{x}') \) to a variable \( F \), if not we discard the new state, return to \( \bar{x} \) and add \( f(\bar{x}) \) to \( F \).
4. We successively repeat steps (2) and (3) which lets the quantity $F/t$ ($t$ denotes the number of steps) converge to the desired quotient in (3.48)

This method ensures that regions of higher importance are explored more frequently than others (according to $g(\mathbf{\bar{n}}, E)$), increasing the performance of the algorithm. The simulations for $N = 10^4$ inhomogeneously coupled spins suggest that even for $\eta \sim \epsilon$ the number of states in both the perturbed and unperturbed case differ by less than a few percent (see Fig. 3.6 (b)).

Furthermore, since $[\vec{I}_z^{(1)}, \vec{I}_-^{(1)}] = [\vec{I}_z^{(1)}, \vec{I}_-^{(1)}] + \mathcal{O}(\epsilon)$ the diffusion rate of the generalized Overhauser field equals the one in the unperturbed case up to an $\epsilon$ correction. This justifies the conclusion that the diffusive dynamics of the generalized Overhauser field is well reproduced by that of the standard Overhauser field.

### 3.4.5 Generalization to multiple nuclear spin species

We have so far neglected the Zeeman energy of the nuclei, which is typically three orders of magnitude smaller than $\omega_z$. A homogeneous nuclear Zeeman term $\propto I_z^{(0)}$ has no effect on the analysis carried out above since it commutes with the generalized Overhauser field $\vec{I}_z^{(1)}$. However, if different nuclear species with different gyromagnetic ratios are involved, this is no longer the case since the correction $\propto I_+^{(1)} I_-^{(1)}$ in $\vec{I}_z^{(1)}$ includes the exchange of nuclear spin excitations between different nuclear species. Different Larmor frequencies associated with different species/isotopes will in general lead to a modulation of the generalized Overhauser field. Here, we will show that in the limit of large differences in Larmor frequencies, the dominant contribution to generalized Overhauser field stems from intra–species flip–flop terms. The fast time dependence of the inter–species flip–flop terms in this limit ensures that their contribution averages out and we can assume that each species/isotope couples to the electron independently. For the relevant nuclei the energy differences (between different species) are often so large (up to 10 MHz/T) that they cannot be neglected on the time scales of nuclear spin diffusion (see chapter 2, Table 2.1).

We consider here the case that $\mathcal{H}$ also contains an inhomogeneous nuclear Zeeman term

$$H_{nz} = \sum_j \omega_{nz,j} I_j z.$$

We assume a number of different nuclear species labeled by $s$ and define nuclear operators referring to species $s$ by $I_{\alpha}^{(n,s)} \equiv \sum_{j \in s} \left( \frac{A_j}{A_N} \right)^n I_{\alpha j}$ (where the sum runs only over the indices $j$ of nuclei belonging to species $s$). Then the correction term in the generalized Overhauser field splits into an intra–species part which commutes with $H_{nz}$ and a second (inter–species) part describing the exchange of spin excitations between different species:

$$\{I_+^{(1)}, I_-^{(1)}\}_+ = \sum_s \{I_+^{(1,s)}, I_-^{(1,s)}\}_+ + 2 \sum_{s>s'} \left( I_+^{(1,s)} I_-^{(1,s')} + I_-^{(1,s)} I_+^{(1,s')} \right).$$

We show in the following that for sufficiently large magnetic fields the latter terms are off–resonant and thus suppressed to leading order. Only the intra-species terms $\sum_s \{I_+^{(1,s)}, I_-^{(1,s)}\}_+$ survives in the generalized Overhauser field. To higher orders,
the inter–species terms provide small additional state–independent generalized Overhauser field–diffusion terms similar to the one generated by \( I_z^{(2)} \) (see equation (3.35)).

Generalizing the considerations of section 3.3.1 we consider \( H_{nz} \) to be part of \( V_D \) and modify the generator of the Schrieffer–Wolff transformation such that the terms connecting different species in (3.53) are canceled. This is achieved by adding

\[
T = S_z \sum_{s > s'} \frac{\epsilon A \sqrt{N}}{2(\omega_{nz,s} - \omega_{nz,s'})} \left( I_+^{(1,s)} I_+^{(1,s')} - I_-^{(1,s)} I_+^{(1,s')} \right) \equiv S_z \otimes X \tag{3.54}
\]

to \( S \), where \( X \) is an operator that only acts on the nuclear spin degrees of freedom. This modification has the following effects:

\[
H = e^{-S^{-T}(H' + H_{nz})} e^{S + T} = H' + H_{nz} - [S, H'] - [T, H'] - [S, H_{nz}] - [T, H_{nz}] + \frac{1}{2} [S + T, [S + T, H' + H_{nz}]] + \ldots,
\]

which leads to several new first and second order terms like \([S^{[1]}, H_{nz}], [T, H_0 + V_N + V_D], [T + S^{[1]}, [T + S^{[1]}, H_{nz}]]\) etc. Most of these terms are off–resonant either by \( \omega_z \) or \( \omega_{nz,s} - \omega_{nz,s'} \) and the secular terms lead to \( I_z^{(0,s)} \)–conserving second order corrections, which either modify the generalized Overhauser field or induce a small state–independent generalized Overhauser field–diffusion (similar to \( I_z^{(2)} \) in (3.35)).

Since they are similar to and smaller (by \( \epsilon \omega_{nz,s}/(A\sqrt{N}) \) or \( \epsilon A\sqrt{N}/(\omega_{nz,s} - \omega_{nz,s'}) \), respectively) than terms already considered, we do not discuss them in detail.

To determine the conditions under which it is allowed to neglect all non–secular terms, denote by \( \Delta_{nz} = \min\{|\omega_{nz,s} - \omega_{nz,s'}| : s \neq s'\} \) the nuclear Zeeman inhomogeneity and introduce \( \epsilon_{nz} = A\sqrt{N}\epsilon/\Delta_{nz} \). Exemplarily, we consider the laser term arising from \([T, V_D]\), which represents one of the major perturbations. It reads

\[
\frac{\Omega_p}{2} \left| \uparrow \right\rangle \left\langle X \right| - \frac{\Omega_c}{2} \left| \downarrow \right\rangle \left\langle X \right| + \text{h.c.} \propto |D\rangle \left\langle t \right| + \text{h.c.} \tag{3.55}
\]

It describes laser–assisted nuclear spin dynamics that changes the generalized Overhauser field and is only detuned by \( \Delta_{nz} \). Thus we need

\[
\frac{\Omega \epsilon A\sqrt{N}}{\Delta_{nz}} \ll \Delta_{nz} \iff \Delta_{nz}^2 \gg \frac{\Omega (A\sqrt{N})^2}{2\omega_z}. \tag{3.56}
\]

With typically \( \Delta_{nz} \sim 10^{-3}\omega_z \), the inequality (3.56) becomes \( 10^{-6} \gg \frac{\Omega \epsilon (A\sqrt{N})^2}{2\omega_z^2} \). In terms of \( \epsilon_{nz} \) (3.56) yields \( \Omega \epsilon_{nz} \ll \Delta_{nz} \). For typical values (all energies in \( \mu eV \)) of \( A\sqrt{N} \sim 1 \) and \( \Delta_{nz} \sim 10^{-2}\omega_z \) we could take \( \omega_z \sim 10^{-3} \), which yields \( \epsilon_{nz} \sim 10^{-1} \) and thus would require \( \Omega \sim 0.1 \). Thus if \( \Delta_{nz} \) is sufficiently large \( (A\sqrt{N}\epsilon, \Omega \epsilon_{nz} \ll \Delta_{nz}) \) these contributions are small and the non-secular terms can be neglected. This can always be ensured by sufficiently strong magnetic field. Using the same arguments one can show that under condition (3.56) also the other terms arising from \( T \) can safely be neglected.

Similarly, we have to transform the jump operators in the Liouvillian. Here, we consider only the first–order correction to \( \mathcal{L}^{[1]} \) arising from \( |\downarrow\rangle \left\langle t \right| \rightarrow |\downarrow\rangle \left\langle t \right| - \ldots \)
3 Nuclear spin cooling

\[ [S^{[1]} + T, \downarrow] \downarrow(t) \pm \ldots \]

The new terms such as \([T, \downarrow] \downarrow(t) = -\frac{1}{2} X \downarrow \downarrow(t)\) describe an additional slow nuclear spin dynamics that occurs only in the optically excited state, thus enhancing nuclear diffusion outside the trapping region and improving the scheme.

To conclude, if \(M > 1\) nuclear species (isotopes) are present, a simple extension of (3.25), where each isotope couples to the electron independently, will be valid provided that \(eA \sqrt{N}/|\omega^s - \omega^r| \ll 1\) \(\forall s \neq r\); here \(\omega^s\) is the Larmor frequency of the \(s^{th}\) nuclear species. Generalized Overhauser field terms describing flip-flops between different species will then also be off-resonant and can be neglected provided \(|\omega^s - \omega^r| \gg \Omega^2/\Gamma, eA \sqrt{N}\). In this limit, we obtain an intra-species generalized Overhauser field:

\[ \tilde{I}_z = I_z - \frac{\epsilon}{2} \sum_{s} \{I_{s+}, I_{s-}\} \]

3.4.6 Nuclear spin decoherence

In realistic systems, there may be other processes affecting the nuclear spins which have to be taken into account for a full description, in particular the dipolar interaction between nuclear spins and on-site quadrupolar terms; these terms would lead to a non-zero \(T_{2,nuc}^*\) time of the nuclear spin ensemble. In addition, there may be \(T_{2,nuc}\) processes affecting the nuclei, arising from fluctuating local magnetic fields. Here, we provide some simple estimates of how these processes relate quantitatively to other corrections considered in previous sections.

We consider pure dephasing of nuclear spins with rate \(\kappa = T_{2,nuc}^{-1}\)

\[ \dot{\rho} = \kappa \sum_j (I_j^z \rho I_j^z - \rho) \]

affecting all nuclei. Computing the contribution to \(\frac{d}{dt} \tilde{I}_z\) arising from this process we find

\[ \left| \frac{d}{dt} \tilde{I}_z \right| \sim \epsilon \kappa. \] (3.57)

as the effective \(T_{2,nuc}\)-induced diffusion rate of \(\tilde{I}_z\). At the end of the next section we discuss the conditions under which \(\kappa\) is small enough to allow for nuclear cooling to the single-spin level.

3.5 Anomalous nuclear spin diffusion

In this section, we describe the time evolution of the nuclear Overhauser field as a continuous-time random walk in order to gain further insight into the dynamics of the nuclear cooling mechanism. We find that this random walk shows anomalous diffusion that leads to asymptotic waiting times between successive nuclear spin flips that are governed by heavy-tailed Lévy distributions. Furthermore, this treatment allows us to extract not only the time needed for the coupled electron-nuclei system to reach the dark state but also the effective minimum width of the trapping region \(\hat{\delta}\) in the presence of intrinsic nuclear spin diffusion.

Before we describe the random walk analysis in detail, we sketch the motivation for deriving an expression for the time \(\langle \hat{t} \rangle\) to reach the dark state. The optically-assisted nuclear spin flips take place at a rate that is smaller by a factor \(\epsilon^2\) than the rate of photon scattering events without nuclear spin flips. This enables us to
monitor the sharp decrease in QD resonance fluorescence and to verify that the coupled system is in the trapping region on time scales short compared to average nuclear spin flip time. Once the system is found to be in the trapping region, a feedback mechanism can be used to turn the laser excitation off, ensuring that the desired/attained $\sigma_{OF}$ is preserved. This will be discussed at the end of this section.

Continuous–time random walks are used in a wide range of fields to describe stochastic processes that are characterized by two probability distributions: one for the spatial jump length and another for the waiting time between two consecutive jumps. If one allows the jump length distribution to assume a Lévy–type distribution that is marked by so–called “fat tails”, extremely long spatial jumps will occur. This is due to the fact that, asymptotically, Lévy distributions decay as power laws rather than exponentially, which gives rise to larger probabilities for extreme events that dominate the evolution of the system. These “fat tails” are also responsible for diverging variance and possibly infinite mean. Compared to Brownian motion, the described random walk will show superdiffusive behavior. On the other hand, if the waiting time distribution obeys Lévy distribution, the system can become trapped for long times between jumps, which leads to sub-diffusion [5]. Lévy distributions have been used to describe a very wide variety of phenomena as detailed in chapter 1. Of particular importance is the problem of subrecoil laser cooling [16], which shares many features with the anomalous diffusion process that appears in the cooling of nuclear spins in Overhauser field selective coherent population trapping.

Here, we analyze the anomalous diffusion of the nuclear Overhauser field due to optical excitation. The jump length distribution of the continuous–time random walk that describes this physical process is not of Lévy type, but is given by the distribution of the hyperfine coupling strength of the nuclei. The hyperfine interaction depends on the electronic envelope wave function, which is assumed to be Gaussian in the QD. A typical jump will thus induce a change in the Overhauser field by $A_{H}/N$. However, the waiting time distribution $P(t)$ between consecutive nuclear spin flips shows signatures of Lévy statistics.

For fixed laser detunings $\Delta \omega_p$ and $\Delta \omega_c$ the absorption depends on the value that the nuclear Overhauser field assumes. In particular, for $\delta_{eff} = \lambda - \delta = 0$ with $\delta$ fixed, absorption vanishes due to the formation of a dark state. We define the region around this dark state in the CPT dip to be the trapping region, while the remaining part is called the recycling region. For all practical purposes, the waiting time distribution in the recycling region does not exhibit Lévy statistics (see below). On the other hand, in the trapping region one finds an infinite average trapping time if no optical–excitation–independent nuclear diffusion processes are present. The fact that $P(t)$ in the trapping region is a Lévy distribution is responsible for the overall subdiffusive behavior of the random walk of the nuclear spins.

The temporal evolution of the system shows switching between two regimes: diffusion in the recycling region and locking in the trapping region. To describe these dynamics, we introduce the recycling time $\hat{t}$ and the associated probability distribution function $\hat{P}(\hat{t})$. The recycling time is the time an initially trapped Overhauser field would diffuse in the recycling region before returning to the trap. In other words, it is a measure for the time scales of switching between the
diffusive and trapping regimes.

First, we will neglect optical–excitation–independent nuclear spin diffusion and derive an expression for the time required to reach the trapping region. Here we focus on the case where $\Delta \omega_p = \Delta \omega_c = 0$ and $\Omega_p = \Omega_c$. To simplify the estimation of the recycling and trapping (waiting) time, we start by considering a limiting case where the width for the CPT transparency dip fulfills

$$\Omega^2 / \Gamma \ll A_H / N,$$  

(3.58)

i.e., a typical single nuclear spin flip will take the system out of the transparency window. We note that the condition $\Omega^2 / \Gamma \ll A_H / N$ is not necessarily optimal for nuclear spin cooling since it requires very small $\Omega$, which in turn leads to a small nuclear spin flip rate in the recycling region (see below) and longer than optimal return times. On the other hand, in this limit a single nuclear spin flip takes the system out of the trapping region, simplifying the analysis. Subsequently, we will provide an alternative description for the case $\Omega^2 / \Gamma \approx A_H / N$.

We assume that the width of the recycling region is determined by $A_H / \sqrt{N} \approx \Gamma / 4$; since the density of states of the Overhauser field quickly drops for large polarizations, the Overhauser field cannot explore extreme polarizations. This observation is supported by numerical simulations, justifying the assumption of hard walls at $A_H / \sqrt{N} \approx \Gamma / 4$. For the assumed parameter range, the light scattering rate is nearly constant outside the transparency dip and up to the hard walls that define the recycling region. We therefore take the light scattering rate to be constant in the recycling region and equal to $\Omega^2 / \Gamma$ in this simplified model. As mentioned above, the nuclear spin flip rate is suppressed by a factor $\epsilon^2$ compared to the light scattering rate, which leads to the nuclear spin flip rate

$$\tau_0^{-1} \approx \epsilon^2 \Omega^2 / \Gamma$$

(3.59)

in the recycling region. In the limit of many nuclear spin flips, the number of steps in the recycling region required to return to the trapping region is thus given by the total range of Overhauser field values the system explores divided by the size of the trap:

$$\langle M \rangle = \frac{A_H / \sqrt{N}}{\Omega^2 / \Gamma}.$$  

(3.60)

This expression is valid provided

$$\langle M \rangle \gg \left( \frac{A_H / \sqrt{N}}{A_H / N} \right)^2 = N,$$  

(3.61)

where the expression on the right corresponds to the number of spin flips that allows the system to diffuse to the hard walls, starting from an arbitrary polarization within the recycling region [17]. The condition (3.61) is equivalent to

$$\langle M \rangle = \frac{A_H / \sqrt{N}}{\Omega^2 / \Gamma} \gg N \Leftrightarrow \frac{\Omega^2}{\Gamma} \ll \frac{A_H}{N^{3/2}}.$$  

(3.62)

Since the time for a single spin flip is taken to be independent of the Overhauser field, the average (recycling) time to return to the trapping region is given by

$$\langle \hat{t} \rangle = \langle M \rangle \tau_0 = \frac{A_H / \sqrt{N}}{\Omega^2 / \Gamma} \frac{\Gamma}{1 / \epsilon^2}.$$  

(3.63)
Strictly speaking, we are interested in the time for an Overhauser field that is initially in the recycling region to reach the trapping region. In contrast, \( \langle \hat{t} \rangle \) gives the average return time from the recycling to the trapping region, starting from an Overhauser field that is initially in the trap. Since the analysis for the recycling time we presented is valid in the limit of many nuclear spin flips in the recycling region before the system reaches the trap, it follows that the Overhauser field explores the whole recycling region uniformly. In this case, the starting point of the Overhauser field becomes irrelevant in the sense that if the Overhauser field initially was in the trap, events where the Overhauser field returned to the trap after only a few spin flips in the recycling region are excluded from the analysis. As a consequence, the recycling (first return) time and the time for an Overhauser field that initially was in the recycling region to reach the trap, are comparable.

Now we derive an expression of \( \langle \hat{t} \rangle \) in the more relevant limit

\[
\frac{\Omega^2}{\Gamma} \simeq \frac{A_H}{N}. \tag{3.64}
\]

Clearly, \( \frac{\Omega^2}{\Gamma} \) does not satisfy condition (3.62) and therefore the number of steps needed to reach the trapping region is no longer given by (3.60). Since the step size is now comparable to the width of the trap, reaching the middle of the recycling region (where the trapping region is) starting from an arbitrary point within the hard walls is sufficient for trapping. The number of steps is then given by the whole interval divided by the step size squared:

\[
\langle \tilde{M} \rangle = \left( \frac{A_H/\sqrt{N}}{\Omega^2/\Gamma} \right)^2 = \left( \frac{A_H/\sqrt{N}}{A_N/N} \right)^2 \simeq N. \tag{3.65}
\]

Consequently, the time required to find the trap is given by

\[
\langle \hat{t} \rangle = \langle \tilde{M} \rangle \tau_0 = N \frac{\Gamma}{\Omega^2 \epsilon^2} = N \frac{A_H}{A_H \epsilon^2} = N^2 \frac{A_H}{A_H \epsilon^2} \simeq N^3, \tag{3.66}
\]

where the last expression follows for \( \omega_z \approx A_H \).

Given the strong \( N \) dependence of \( \langle \hat{t} \rangle \) corresponding to the time scale needed to establish \( \sigma_{\text{OF}} \sim A_H/N \), it is important to consider nuclear spin dynamics arising from optical–excitation–independent nuclear spin diffusion or decay processes, as well as the electron \( T_2 \). The ultimate limit for the latter is due to spin–orbit mediated spin–flip phonon emission with a rate \( \sim 10^{-7} \Gamma \) for \( \omega_z \sim A_H \) [152]. Physical processes leading to nuclear spin diffusion in the dark state include (a) nuclear spin diffusion mediated by dipole–dipole interactions, (b) electric field fluctuations leading to spatial shifts in the QD electron wave–function, (c) quadrupolar interaction with axis \( \parallel B_z \). Such processes would lead to a non–vanishing rate of nuclear spin flips that take the system out of the dark state. In the long term limit, this would establish a steady state between diffusion in the recycling region and finite–time trapping in the transparency region. Clearly, the presence of optical–excitation–independent nuclear spin diffusion processes limits the reduction in the standard deviation of the Overhauser field.

We denote the optical–excitation–independent nuclear spin diffusion rate that can arise from any of the mechanisms discussed above with \( \gamma_n \) and assume that \( N \gamma_n \ll \epsilon^2 \Omega^2/\Gamma \), so that the picture of a CPT dip is still valid. Next we want to
find an expression for the steady–state standard deviation of the Overhauser field for $\gamma_n \neq 0$. In this case, the system spends a substantial fraction of the total time both in the recycling and trapping region where the standard deviations are given by $A_H/\sqrt{N}$ and $\tilde{\delta}$, respectively. The standard deviation of the trapping region $\tilde{\delta}$ is a function of $\gamma_n$ and will be determined below. To obtain the desired expression for $\sigma_{OF}$ the contribution of the two regions are weighted by the fraction of time the system spends in the respective regions, i.e.

$$
\sigma_{OF} \simeq \tilde{\delta} \frac{\langle t \rangle}{\langle t \rangle + \langle \hat{t} \rangle} + \frac{A_H}{\sqrt{N}} \frac{\langle \hat{t} \rangle}{\langle t \rangle + \langle \hat{t} \rangle}.
$$

(3.67)

We start specifying the parameters that enter the first term of this expression which stems from the trapping region. In the presence of optical–excitation–independent nuclear spin diffusion, the time spent in the trap is given by

$$
\langle t \rangle = (N\gamma_n)^{-1}.
$$

(3.68)

The standard deviation in the trapping region is given by its effective width $\tilde{\delta}$, which is determined by the residual scattering rate in the trap $N\gamma_n$ and the expected pure nuclear spin flip rate in the CPT dip given by $\tau^{-1}_{\text{CPT}} = \epsilon^2 \frac{\Gamma}{\Omega^2} \langle |\tilde{I}_z|^2 \rangle$; i.e. $\tilde{\delta}$ satisfies

$$
e^2 \frac{\Gamma}{\Omega^2} \tilde{\delta}^2 = N\gamma_n,
$$

(3.69)

yielding

$$
\tilde{\delta} = \epsilon^{-1} \Omega \sqrt{N\gamma_n^2/\Gamma}.
$$

(3.70)

For the second term, which originates in the recycling region, we found the expression (3.66) for $\langle \hat{t} \rangle$. The standard deviation in the recycling region corresponds to the one of an unprepared nuclear spin environment, i.e. $A_H/\sqrt{N}$. Combining these two expressions, leads to (3.67).

The smallest steady–state (measurement–free) $\sigma_{OF}$ is obtained when the contribution from the trapping region (first term of (3.67)) and from the recycling region (second term) to $\sigma_{OF}$ are comparable. However, substantial Overhauser field narrowing in this case is only possible provided $\langle \hat{t} \rangle \ll \langle t \rangle$. This condition is unlikely to be satisfied for self–assembled QDs, since $\langle \hat{t} \rangle \sim 10^4$ s for $N = 10^4$, according to (3.66) with $A_H^{-1} \approx 10^{-11}$ s. On the other hand, a more modest narrowing yielding $\sigma_{OF} = 10A_H/N$ would give $\langle \hat{t} \rangle \sim 1$ s. This estimate is obtained with (3.66) by setting $\Omega^2/\Gamma \approx 10A_H/N$ in (3.59) while $\langle \hat{M} \rangle$ in (3.65) remains unaffected.

We now turn to the use of feedback from the scattered light intensity mentioned at the beginning of this section. The steady–state $\sigma_{OF}$ for a small trapping region is dominated by the optical excitation induced fluctuations in the Overhauser field in the recycling region. A strategy to reduce the $\sigma_{OF}$ below its steady–state value is to monitor the light scattering rate $W_{\text{scat}}$ of the coupled system. Since $W_{\text{scat}} \propto \delta_{\text{eff}}^2$ in the neighborhood of the trapping region, a drastic reduction in $W_{\text{scat}}$ verifies that the nuclear spins are in the desired state; turning the laser fields off upon acquiring this information will ensure that $\sigma_{OF}$ will be given by the width $\tilde{\delta} \sim A_H/N$ of the trapping region (see (3.67)). To achieve Overhauser field narrowing in the single spin limit

$$
\sigma_{OF} \sim \tilde{\delta} \sim A_H/N,
$$
it is necessary to ensure
\[ \gamma_n \leq A_H/N^3. \] (3.71)

This expression results from (3.69) for \( \tilde{\delta} \approx \Omega^2/\Gamma \approx A_H/N \) and \( \epsilon \approx N^{-1/2} \). For nuclear dephasing we find
\[ 1/T_{2,\text{nuc}} \leq A_H/N^2. \] (3.72)

This feedback is possible due to the fact that the time scale for flipping a nuclear spin is longer by a factor \( \epsilon^{-2} \) than that for a photon scattering event.

### 3.6 Conclusion

The present discussion was focused on self-assembled QDs, but we expect our findings to be relevant for a range of solid-state emitters. While we have concentrated on nuclear spin diffusion associated with the ground-state hyperfine coupling, the conclusions of our work remain unchanged if the solid-state emitter has hyperfine coupling leading to nuclear spin diffusion in the optically excited state [176]; in fact, such processes shorten \( \langle \tilde{t} \rangle \). This would be the case for example in QDs with vanishing heavy–light hole mixing leading to near–resonant hole–mediated nuclear spin–flips in the excited state or nitrogen vacancy (NV) centers in diamond as discussed next.

The central mechanism of our proposal has been experimentally realized in NV centers [177]. There, the spin ground states are realized by the \( m_s = \pm 1 \) states of the electronic ground state triplet and are optically coupled to a common excited state. The two photon detuning is given by the splitting of the two ground states, which is determined by a combination of the external magnetic field and the nuclear Overhauser field. If the Overhauser field cancels the external field, the electron is forced into the dark state. Contrary to the assumptions in this chapter, the dominant contribution to the hyperfine interaction in NV centers, which is due to the proximal nuclear spins, is much stronger in the electronic excited state. Nevertheless, our predictions are relevant for NV centers: If the electron is in the dark state, the excited state is depopulated and hence the nuclear dynamics is slowed down drastically. While in self–assembled QDs the nuclei are assumed to have \( I \neq 0 \), this is not true for the most abundant carbon isotope \( ^{12}\text{C} \). The dominant contribution to the Overhauser field stems from the single nitrogen \( ^{14}\text{N} \) nucleus while the \( ^{13}\text{C} \) spin environment couples more weakly to the central spin. As a consequence, the effective two–photon resonance can be established on a reasonable time scale only for a discrete set of external magnetic fields. This is in marked contrast to QDs, where a substantial reduction in the absorption is expected for any bare laser detuning as long as it does not exceed \( A_H/\sqrt{N} \). However, for these particular external fields, it was found that the NV center nuclear spins seek out a configuration that satisfies two–photon resonance, in agreement with our predictions. Furthermore, the limited number of participating nuclear spins implies that the time scale on which effective two–photon resonance is established is favorable in NV centers compared to QDs. Additionally, the distribution of the \( ^{13}\text{C} \) spin environment could be narrowed and the extension to a measurement–based scheme allowed the preparation of the \( ^{13}\text{C} \) environment in a desired state. Clearly, these results indicate that the nuclear spin cooling mechanism predicted in this chapter is effective in NV centers in diamond.
Real–time monitoring of Lévy flights in a single quantum system

Lévy flights are random walks where the dynamics is dominated by rare events. Even though they have been studied in vastly different physical systems, their observation in a single quantum system has remained elusive. Here, we analyze a periodically driven open central spin system and demonstrate theoretically that the dynamics of the spin environment exhibits Lévy flights. For the particular realization in a single–electron charged quantum dot driven by periodic resonant laser pulses, we use Monte Carlo simulations to confirm that the long waiting times between successive nuclear spin flip events are governed by a power–law distribution; the corresponding exponent \( \eta = -3/2 \) can be directly measured in real–time by observing the waiting time distribution of successive photon emission events. Remarkably, the dominant intrinsic limitation of the scheme arising from nuclear quadrupole coupling can be minimized by adjusting the magnetic field or by implementing spin echo. This chapter is based on [90].

4.1 Introduction

In chapter 3, we described the cooling of the nuclear spin environment in terms of an anomalous diffusion process due to the presence of an optically dark state, in analogy to velocity selective coherent population trapping. The narrowing of the nuclear spin distribution is thus accompanied by Lévy flights in the trapping time between two successive nuclear spin flip events. Here, we show that extending the scheme to time–dependent dark states may allow to avoid the detrimental effects of strain–induced directional nuclear spin polarization that hindered the observation of Lévy flights under continuous wave excitation described in chapter 3. By shortening the interaction time with the laser field, the directional absorption–assisted nuclear spin flips are strongly suppressed. Then the diffusive spontaneous–emission–assisted nuclear spin flips lead to Lévy flights in the nuclear spin environment which can be directly extracted and tracked in real time. This scheme is motivated by striking experimental results on an ensemble of self–assembled QDs under periodic excitation which are compatible with Lévy flights of the Overhauser field [193, 91, 194, 162, 195]. Even though we consider a single QD under periodic optical excitation we expect our findings to be relevant for other solid state emitters.

In this chapter, we consider a scheme that is based on the existence of time–dependent electronic dark states, which are characterized by the synchronization of the electron with the periodic laser pulse (see section 2.2.3). To be more specific,
if the electron Larmor precession period is commensurable with the pulse repetition period, then the electron is asymptotically forced into a time-dependent dark state, i.e. whenever the pulse arrives, the electron is found in a state that does not couple to the laser field. Under this synchronization condition, optically-assisted nuclear spin flip events are drastically reduced. Therefore, those nuclear spin configurations that synchronize the electron constitute nuclear trapping configurations, in analogy to chapter 3. Whereas in chapter 3 only a single nuclear trap existed for a given set of external parameters, in the time-dependent scheme considered here, multiple nuclear traps are expected. In this chapter, we show that in the vicinity of the nuclear traps, the time between two successive nuclear spin flip events is governed by a Lévy distribution. Importantly, the optical waiting time distribution also shows a power-law tail due to the proportionality of the optically-assisted nuclear spin flip rate and the spontaneous emission rate. Furthermore, the nuclear Lévy flights can be tracked in real-time by monitoring the photon emission events.

The chapter is structured as follows: Section 4.2 summarizes the interactions in the system under investigation. Then we show detailed derivations of the dynamics of the periodically driven-dissipative electron in the absence of nuclear spins in section 4.3. In section 4.4, we discuss the dominant electron–nuclei interactions and derive the rate at which optically-assisted nuclear spin flips occur perturbatively. We continue by analyzing the consequences of the coupled dynamics, in particular the emergence of Lévy flights in the nuclear spin environment (section 4.5) and the possibility of real-time monitoring (section 4.6). Finally, in section 4.7 we discuss the limitations of the scheme and close with a summary and outlook.

4.2 System

We study an electron in a single self-assembled QD coupled to the radiation field reservoir and illuminated by a ($\sigma^+$) circularly polarized laser pulse train propagating along the QD growth direction $x$. The external magnetic field $\vec{B} = B\hat{z}$ is applied transversely to the light propagation direction (Voigt geometry) as shown in Fig. 4.1. The electronic ground and excited states are given in the optical basis in section 2.2.3; throughout this chapter we use the optical basis and hereafter suppress the subscript $x$ on the basis states, i.e. $|\uparrow_x\rangle = |\uparrow\rangle$. The Hamiltonian describing the energy of the electronic states is given by (with $\hbar = 1$)

$$H_0 = + \frac{\omega_e}{2} (|\uparrow\rangle \langle \downarrow| + |\downarrow\rangle \langle \uparrow|)$$
$$+ \frac{\omega_t}{2} (|\uparrow\downarrow\uparrow\rangle \langle \downarrow\uparrow\downarrow| + |\uparrow\downarrow\downarrow\rangle \langle \downarrow\uparrow\uparrow|)$$
$$+ \omega_{te} (|\uparrow\downarrow\uparrow\rangle \langle \uparrow\downarrow\downarrow| + |\uparrow\downarrow\downarrow\rangle \langle \uparrow\downarrow\uparrow|),$$

where $\omega_e$ and $\omega_t$ are the Larmor precession frequencies in the ground (e) and excited states (t), respectively. The last term corresponds to the optical transition energy.

A $\sigma^+$-polarized periodic laser pulse train couples the states $|\uparrow\rangle$ and $|\uparrow\downarrow\uparrow\rangle$ [135] while leaving $|\downarrow\rangle$ unaffected. Hence, $|\uparrow\rangle$ is called bright and $|\downarrow\rangle$ dark state. The
4.2 System

Figure 4.1: (a) The magnetic field $B$ is applied in $z$-direction and the laser pulse train propagates along the $x$-direction, which is the QD growth axis. (b) The electron and the trion precess in the external magnetic field (green) and the $\sigma_+$-polarized laser only couples to $|\uparrow_x\rangle$ (blue). Spontaneous emission is measured in the circularly polarized basis (gray). On one hand, the coupling to the nuclear spin environment modifies the electron ground state precession frequency, while on the other hand the slow nuclear spin dynamics is dominated by optically-assisted nuclear spin flips (red).
action of a single pulse can be described by (see section 2.2.3)

\[
\rho_{f,\uparrow\uparrow}^\prime = \frac{1}{2} \rho_{i,\uparrow\uparrow}^\prime (1 + \cos \theta)
\]

\[
\rho_{f,\downarrow\downarrow\uparrow\uparrow}^\prime = \frac{1}{2} \rho_{i,\downarrow\uparrow\uparrow\uparrow}^\prime (1 - \cos \theta),
\]

where the pulse area \( \theta = 2 \int \Omega(t) \, dt \) determines the excited electron population and \( \Omega \) is the laser Rabi frequency and the indices \( i \) and \( f \) refer to the initial and final states, i.e. before and after the pulse, respectively.

We consider the dominant nuclear spin dynamics in strained self-assembled QDs as described in chapter 2. The Hamiltonians of the nuclear Zeeman interaction, the Overhauser field term, as well as quadrupolar interaction are given by

\[
H_n = - \sum_{j=1}^{N} \omega_n^j I_j^z,
\]

\[
H_{Oh} = \sum_{j=1}^{N} A_j S_z I_j^z,
\]

\[
H_Q = \sum_{j=1}^{N} \omega_Q^j \left( (I_j^z)^2 - \frac{I_j^z (I_j^z + 1)}{3} \right),
\]

where \( \omega_n^j \) and \( A_j \) are the Zeeman splitting and hyperfine coupling constant of the \( j^{th} \) nucleus, and \( \omega_Q^j \) is the quadrupolar energy for \( 1 \leq j \leq N \).

### 4.3 Periodic optical excitation of the electron

In this section, we introduce the coherent and dissipative interactions of the central electron spin in the absence of coupling to the nuclear spin environment. We first highlight striking features connected to the periodic pulsed excitation before we present the detailed derivation of these results.

The combined coupling of the four-level system to a periodic laser pulse train and the radiation field reservoir asymptotically forces the electron into a periodic quasi-steady state. This occurs by sequential excitation of the bright state by a laser pulse, followed by spontaneous emission of a photon at a rate \( \Gamma \) and the accompanying decay into one of the two ground states. If the pulse separation \( \tau \) satisfies \( \tau \gg \Gamma^{-1} \), the trion population decays completely within the pulse separation \( \tau \), which justifies the assumption on which (4.1) is based. The asymptotic bright state population \( \rho_{i,\uparrow\uparrow}^* \) before the pulse is then given by

\[
\rho_{i,\uparrow\uparrow}^* = \frac{\sin^2(\omega_e \tau/2)}{\frac{\tau}{2} + \sin^2(\omega_e \tau/2)},
\]

provided \( \theta = \pi \). Similarly, the coarse-grained spontaneous emission rate is given by

\[
\Gamma_{\text{spon}} = \frac{1}{\tau} \int_0^\tau \left( \frac{\rho_{\uparrow\uparrow\uparrow\uparrow}(t) + \rho_{\uparrow\uparrow\uparrow\downarrow}(t)}{2} \right) \, dt
\]

\[
= \frac{1}{\tau} \rho_{i,\uparrow\uparrow}^* = \frac{1}{2} \frac{\sin^2(\omega_e \tau/2)}{\frac{\tau}{2} + \sin^2(\omega_e \tau/2)}.
\]
If the bright state population at the arrival of the pulse vanishes, so does the optical absorption and hence the coarse–grained spontaneous emission rate $\Gamma_{\text{spon}}$. This occurs exactly if the electron precession frequency satisfies the synchronization condition

$$\omega_e \in \frac{2\pi}{\tau} \mathbb{Z}.$$ (4.2)

The expansion of $\Gamma_{\text{spon}}$ close to a synchronization condition (4.2) with $m \in \mathbb{Z}$ gives

$$\Gamma_{\text{spon}} = \frac{\tau}{2} \left( \omega_e - \frac{2\pi}{\tau} m \right)^2 + \mathcal{O} \left( \left( \omega_e - \frac{2\pi}{\tau} m \right)^4 \right).$$

The fact that $\Gamma_{\text{spon}}$ depends quadratically on $\omega_e$ is essential for the emergence of Lévy flights.

### 4.3.1 Derivations

We now turn to the derivation of the aforementioned results. First, we derive the electron density matrix at the arrival of a pulse as a function of the density matrix at the previous pulse. These recursive relations can be solved for their asymptotic limit. Then we introduce the concept of the coarse–grained spontaneous emission rate. While these derivations are carried out for an arbitrary pulse area $\theta$, we finally simplify the equations in the case of a $\pi$-pulse.

**Asymptotic electron density matrix**

The action of a single laser pulse on the electron wave function as well as the master equation that governs the evolution between two laser pulses have been discussed in detail in sections 2.2.3 and 2.2.5, respectively. Using these results, we now take the electron density matrix at the arrival of the $k$th pulse as an initial condition to derive the electron density matrix at the $(k+1)$th pulse. The periodic optical illumination, dissipation and the coherent Larmor precession force the electron asymptotically into a periodic quasi–steady state $\rho^{i,*}$.

In the following we assume that periodic pulses separated by a time $\tau \gg \Gamma^{-1}$ excite the four–level system. We take $\tau \gg \Gamma^{-1}$, such that the populations and coherences associated with the excited state manifold decay in the time window between two pulses. Since we consider high magnetic fields, we can further assume $|\omega_e|, \omega_t > \Gamma$, which corresponds to fast Larmor precession before the excited states decay to either ground state via spontaneous emission.

Starting with the electron density matrix at the arrival of the $k$th pulse at time $t_k = (k-1)\tau$, denoted by $\rho^{i,k} = \rho(t_k)$, we find that after the pulse duration $t_p$ the density matrix $\rho^{f,k} = \rho(t_k + t_p)$ is given by (see section 2.2.3)

$$\rho^{f,k} = \begin{pmatrix}
\frac{1}{2} \rho^{i,k}_{1,1} (1 + \cos \theta) & \rho^{i,k}_{1,4} \cos \frac{\theta}{2} & \frac{i}{2} \rho^{i,k}_{1,4} \sin \theta & 0 \\
\rho^{i,k}_{4,1} \cos \frac{\theta}{2} & \rho^{i,k}_{4,4} & i \rho^{i,k}_{4,4} \sin \frac{\theta}{2} & 0 \\
-\frac{1}{2} \rho^{i,k}_{1,1} \sin \theta & -i \rho^{i,k}_{4,1} \sin \frac{\theta}{2} & \frac{1}{2} \rho^{i,k}_{4,1} (1 - \cos \theta) & 0 \\
0 & 0 & 0 & 0
\end{pmatrix},$$ (4.3)
where we have assumed the initial density matrix to have vanishing matrix elements connected to the excited states, i.e.

\[
\rho_{i,k} = \begin{pmatrix}
\rho_{i,k}^{\uparrow\uparrow} & \rho_{i,k}^{\uparrow\downarrow} & 0 & 0 \\
\rho_{i,k}^{\downarrow\uparrow} & \rho_{i,k}^{\downarrow\downarrow} & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix}.
\]

The final density matrix \(\rho_{f,k}\) is the starting point for the subsequent evolution between the \(k\)th and \((k+1)\)th pulse. The dynamics itself is given by the master equation (see section 2.2.5)

\[
\dot{\rho} = -i[H_e, \rho] + C_+ \rho C_+^\dagger - \frac{1}{2}\{C_+^\dagger C_+, \rho\} + C_- \rho C_-^\dagger - \frac{1}{2}\{C_-^\dagger C_-, \rho\},
\]

where

\[
C_+ = \sqrt{\frac{\Gamma}{2}} |\uparrow\rangle \langle \uparrow\downarrow|, \\
C_- = \sqrt{\frac{\Gamma}{2}} |\downarrow\rangle \langle \uparrow\downarrow|.
\]

Since we assume that the excited state populations and coherences decay after a time \(\Gamma^{-1} \ll \tau\), we only need to derive the ground state density matrix elements of \(\rho_{i,k+1}\). To this end, we evolve the density matrix \(\rho_{f,k}\) according to the master equation for a time \(\tau\). Even though the evolution of the different matrix elements is coupled, one can define convenient linear combinations so that the equations partly decouple. This allows us to consecutively find analytical solutions for systems of linear differential equations of a smaller dimension, as outlined below.

First, we consider the total population in the ground state manifold \(\rho_{\uparrow\uparrow} + \rho_{\uparrow\downarrow}\), which is connected to the excited state population \(\rho_{\uparrow\downarrow\uparrow\downarrow} + \rho_{\downarrow\uparrow\downarrow\downarrow}\). Writing the Markovian master equation (4.4) for these density matrix elements yields

\[
\frac{\partial}{\partial t} \begin{pmatrix}
\rho_{\uparrow\uparrow} + \rho_{\uparrow\downarrow} \\
\rho_{\uparrow\downarrow\uparrow\downarrow} + \rho_{\downarrow\uparrow\downarrow\downarrow}
\end{pmatrix} = \begin{pmatrix}
0 & \frac{\Gamma}{2} \\
\frac{\Gamma}{2} & 0
\end{pmatrix} \begin{pmatrix}
\rho_{\uparrow\uparrow} + \rho_{\uparrow\downarrow} \\
\rho_{\uparrow\downarrow\uparrow\downarrow} + \rho_{\downarrow\uparrow\downarrow\downarrow}
\end{pmatrix}.
\]

This solution of this system of linear differential equations is trivial in the limit \(\tau \gg \Gamma^{-1}\) since the excited states have completely decayed to the ground states.

Next we consider the population difference in the ground states

\[
\frac{\partial}{\partial t}(\rho_{\uparrow\uparrow} - \rho_{\downarrow\downarrow}) = i\omega_e (\rho_{\uparrow\downarrow} - \rho_{\downarrow\uparrow}) + \frac{\Gamma}{2} (\rho_{\uparrow\downarrow\uparrow\downarrow} - \rho_{\downarrow\uparrow\downarrow\downarrow}),
\]

which is coupled not only to the ground state coherences

\[
\frac{\partial}{\partial t}(\rho_{\uparrow\downarrow} - \rho_{\downarrow\uparrow}) = i\omega_e (\rho_{\uparrow\uparrow} - \rho_{\downarrow\downarrow}) \quad (4.7)
\]

where

\[
\frac{\partial}{\partial t}(\rho_{\uparrow\uparrow} + \rho_{\downarrow\downarrow}) = 0,
\]
but also to the population difference in the excited states. The time evolution of the population difference in the excited states is defined through the following system of equations:

\[
\frac{\partial}{\partial t} \begin{pmatrix}
\rho_{\uparrow\uparrow,\uparrow\uparrow} - \rho_{\downarrow\downarrow,\uparrow\uparrow} \\
\rho_{\uparrow\downarrow,\uparrow\uparrow} - \rho_{\uparrow\uparrow,\downarrow\downarrow}
\end{pmatrix} = \begin{pmatrix}
-\frac{\Gamma}{2} & i\omega_i \\
-i\omega_i & -\frac{\Gamma}{2}
\end{pmatrix} \begin{pmatrix}
\rho_{\uparrow\uparrow,\uparrow\uparrow} - \rho_{\downarrow\downarrow,\uparrow\uparrow} \\
\rho_{\uparrow\downarrow,\uparrow\uparrow} - \rho_{\uparrow\uparrow,\downarrow\downarrow}
\end{pmatrix}.
\]

The solution can readily be found to be

\[
\begin{pmatrix}
\rho_{\uparrow\uparrow,\uparrow\uparrow} - \rho_{\downarrow\downarrow,\uparrow\uparrow} \\
\rho_{\uparrow\downarrow,\uparrow\uparrow} - \rho_{\uparrow\uparrow,\downarrow\downarrow}
\end{pmatrix}(t) = c_3 e^{-\left(\frac{\Gamma}{2} + i\omega_i\right)t} \begin{pmatrix}
-1 \\
1
\end{pmatrix} + c_4 e^{-\left(\frac{\Gamma}{2} - i\omega_i\right)t} \begin{pmatrix}
1 \\
1
\end{pmatrix} = \frac{1}{2} \rho_{\uparrow,\uparrow}^{i,k} (1 - \cos \theta) e^{-\frac{\Gamma}{2}t} \frac{\cos(\omega_i t) + \omega_i \sin(\omega_i t)}{i\sin(\omega_i t)},
\]

(4.8)

where the imposed initial conditions are \(c_3 + c_4 = \frac{1}{2} \rho_{\uparrow,\uparrow}^{i,k} (1 - \cos \theta)\) and \(c_3 + c_4 = 0\). In the limit of \(\tau \gg \Gamma^{-1}\), these two quantities again vanish at the arrival of the \((k + 1)^{th}\) pulse. We differentiate equations (4.6), (4.7) and (4.8) and get

\[
\frac{\partial^2}{\partial t^2} (\rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow}) + \omega_e^2 (\rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow}) = -\frac{\Gamma}{2} \rho_{\uparrow,\uparrow}^{i,k} (1 - \cos \theta) e^{-\frac{\Gamma}{2}t} \left(\frac{\Gamma}{2} \cos(\omega_i t) + \omega_i \sin(\omega_i t)\right),
\]

which is an uncoupled equation that only contains the ground state population difference.

To solve this differential equation, we note that it has the form of a driven harmonic oscillator. The driving term describes how the ground-state population difference is modified by spontaneous emission from the excited state. It also contains information about the Larmor precession in the excited states with frequency \(\omega_i\) prior to the decay to the ground states. Before writing down the general solution, we observe that the driving term tends to zero on a time scale given by the spontaneous emission rate \(\Gamma\). This hints at the fact that in the long time limit, we expect a solution that describes the Larmor precession in the ground states at a frequency \(\omega_e\). In fact, for fast precession both in the ground and excited state manifolds, i.e. \(\omega_e, \omega_i \gg \Gamma\), the lowest order equation

\[
\frac{\partial^2}{\partial t^2} (\rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow}) + \omega_e^2 (\rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow}) = 0
\]

contains the basic features. Nevertheless, care must be taken to include the initial conditions properly, which requires solving the differential equation including the driving term.

The general solution can be calculated (by using e.g. the Laplace transform) to be

\[
(\rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow})(t) = A_{1,k} \cos(\omega_e t) + A_{2,k} \sin(\omega_e t)
\]

\[
-\frac{1}{2} \rho_{\uparrow,\uparrow}^{i,k} (1 - \cos \theta) e^{-\frac{\Gamma}{2}t} \left(\frac{\Gamma/2}{(\omega_e^2 - \omega_i^2 + (\Gamma/2)^2)^2 + (\Gamma \omega_i)^2} \times \right.
\]

\[
\times \left[ (\omega_e^2 + \omega_i^2 + (\Gamma/2)^2) \frac{\Gamma}{2} \cos(\omega_i t) + (\omega_e^2 - \omega_i^2 - (\Gamma/2)^2) \omega_i \sin(\omega_i t) \right].
\]
The last term decays on the spontaneous emission time scale and is therefore irrelevant for the population difference at the arrival of the next pulse. The first two terms describe the Larmor precession in the ground state manifold with prefactors that are determined by the initial conditions

\[
(\rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow})(0) = \frac{1}{2} \rho_{\uparrow,\uparrow}^{i,k}(3 + \cos \theta) - 1,
\]

\[
\frac{\partial}{\partial t}(\rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow})(0) = i\omega_e (\rho_{\uparrow,\downarrow} - \rho_{\downarrow,\uparrow})(0) + \frac{\Gamma}{2} (\rho_{\uparrow,\downarrow,\uparrow,\downarrow} - \rho_{\downarrow,\uparrow,\downarrow,\uparrow})(0),
\]

\[
= -2\omega_e \Im(\rho_{\uparrow,\downarrow}^{i,k}) \cos \theta + \frac{\Gamma}{2} \frac{1}{2} \rho_{\uparrow,\uparrow}^{i,k}(1 - \cos \theta).
\]

The prefactors are then given by

\[
A_{1,k} = \frac{1}{2} \rho_{\uparrow,\uparrow}^{i,k}(3 + \cos \theta) - 1 + \frac{a}{2} \rho_{\uparrow,\uparrow}^{i,k}(1 - \cos \theta),
\]

\[
A_{2,k} = -2\Im(\rho_{\uparrow,\downarrow}^{i,k}) \cos \frac{\theta}{2} + \frac{1}{2} \rho_{\uparrow,\uparrow}^{i,k}(1 - \cos \theta) \frac{(1 - a)\Gamma/2 + b\omega_e}{\omega_e},
\]

where the parameters

\[
a = \frac{\omega^2 + \omega_1^2 + (\Gamma/2)^2}{(\omega^2_\omega + \omega_1^2 + (\Gamma/2)^2 + (\Gamma\omega_1)^2)} \left( \frac{\Gamma}{2} \right)^2,
\]

\[
b = \frac{\omega^2 - \omega_1^2 - (\Gamma/2)^2}{(\omega^2_\omega - \omega_1^2 + (\Gamma/2)^2 + (\Gamma\omega_1)^2)} \frac{\Gamma}{2\omega_1}
\]

describe the deviation of the solution from the simple harmonic oscillator equation. Since we assume high magnetic fields, both parameters are small, i.e. \(a \ll b \ll 1\). Physically, this means that the probabilities to decay via the \(\sigma_+\) or \(\sigma_-\) channel are equalized due to fast Larmor precession. In the limit \(\tau \gg \Gamma^{-1}\), we find that the population difference of the ground states at the arrival of the \((k+1)\)th pulse is

\[
(\rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow})^{i,k+1} \approx A_{1,k} \cos(\omega_e \tau) + A_{2,k} \sin(\omega_e \tau).
\]

The difference of the ground state coherences in the limit \(\tau \gg \Gamma^{-1}\) is given to lowest order by

\[
(\rho_{\uparrow,\downarrow} - \rho_{\downarrow,\uparrow})^{i,k+1} \approx -i A_{2,k} \cos(\omega_e \tau) + i A_{1,k} \sin(\omega_e \tau).
\]

The sum of the coherences is only zero for \(\pi\)-pulses, in general however

\[
(\rho_{\uparrow,\downarrow} + \rho_{\downarrow,\uparrow})^{i,k+1} = 2\Re(\rho_{\uparrow,\downarrow}^{i,k}) \cos \frac{\theta}{2}.
\]
4.3 Periodic optical excitation of the electron

To summarize, we find the zeroth order solutions in the limit $\tau \gg \Gamma^{-1}$

\[
\rho^{i,k+1}_{\uparrow \downarrow} \approx \frac{1}{2} \left[ 1 + \left( \frac{1}{2} \rho^{i,k}_{\uparrow \downarrow}(3 + \cos \theta) - 1 + \frac{a}{2} \rho^{i,k}_{\downarrow \uparrow}(1 - \cos \theta) \right) \cos(\omega_e \tau) \right. \\
+ \left. \left( -2 \text{Im}(\rho^{i,k}_{\uparrow \downarrow}) \cos \left( \frac{\theta}{2} \right) + \frac{1}{2} \rho^{i,k}_{\downarrow \uparrow}(1 - \cos \theta) \frac{(1 - a)\Gamma/2 + b\omega_c}{\omega_c} \right) \sin(\omega_e \tau) \right], \\
\rho^{i,k+1}_{\downarrow \uparrow} \approx \frac{1}{2} \left[ 1 - \left( \frac{1}{2} \rho^{i,k}_{\downarrow \uparrow}(3 + \cos \theta) - 1 + \frac{a}{2} \rho^{i,k}_{\uparrow \downarrow}(1 - \cos \theta) \right) \cos(\omega_e \tau) \right. \\
- \left. \left( -2 \text{Im}(\rho^{i,k}_{\downarrow \uparrow}) \cos \left( \frac{\theta}{2} \right) + \frac{1}{2} \rho^{i,k}_{\uparrow \downarrow}(1 - \cos \theta) \frac{(1 - a)\Gamma/2 + b\omega_c}{\omega_c} \right) \sin(\omega_e \tau) \right], \\
\rho^{i,k+1}_{\uparrow \uparrow} \approx \text{Re}\left(\rho^{i,k}_{\uparrow \downarrow}\right) \cos \left( \frac{\theta}{2} \right) \\
+ \frac{i}{2} \left[ \left( 2 \text{Im}(\rho^{i,k}_{\uparrow \downarrow}) \cos \left( \frac{\theta}{2} \right) - \frac{1}{2} \rho^{i,k}_{\downarrow \uparrow}(1 - \cos \theta) \frac{(1 - a)\Gamma/2 + b\omega_c}{\omega_c} \right) \cos(\omega_e \tau) \right. \\
\left. \left. + \left( \frac{1}{2} \rho^{i,k}_{\downarrow \uparrow}(3 + \cos \theta) - 1 + \frac{a}{2} \rho^{i,k}_{\uparrow \downarrow}(1 - \cos \theta) \right) \sin(\omega_e \tau) \right]. \right.
\]

Given the recursive expressions (4.9)–(4.11), we can solve them for a quasi-steady state $\rho^{i,k}$ in the asymptotic limit $k \to \infty$. This yields the electron density matrix at the arrival of an arbitrary pulse after many pulses have passed. The convergence to this asymptotic state is discussed for the special case $\theta = \pi$ below.

We start analyzing the recursive relations by noting that we only need to consider one of the ground state populations, since we assume

\[
\rho^{i,k}_{\uparrow \downarrow} + \rho^{i,k}_{\downarrow \uparrow} = 1 
\] (4.12)

for all $k$. Furthermore, (4.11) yields $\text{Re}(\rho^{i,k+1}_{\uparrow \downarrow}) = \text{Re}(\rho^{i,k}_{\downarrow \uparrow}) \cos(\theta/2)$ for the real part of the coherence $\rho^{i,k+1}_{\downarrow \uparrow}$. We find that $\text{Re}(\rho^{i,k}_{\downarrow \uparrow}) = 0$ if $\theta \neq \pi$. However, in the case of a $\theta = \pi$ pulse, the real part of the ground state coherence is automatically zero after every pulse. Hence, we find for a general $\theta$

\[
\text{Re}(\rho^{i,k}_{\downarrow \uparrow}) = 0. 
\] (4.13)

This reduces the system to two linear, coupled equations for $\rho^{i,k}_{\uparrow \downarrow}$ and $\text{Im}(\rho^{i,k}_{\uparrow \downarrow})$. These equations can easily be solved analytically; we exclude the uninteresting case $\theta \in 2\pi\mathbb{Z}$ where no population is transferred to the excited states. Then we solve for the imaginary part of the ground state coherence in the asymptotic limit and find

\[
\text{Im}(\rho^{i,k}_{\uparrow \downarrow}) = \frac{(1 - \cos \frac{\theta}{2})(c - \sin(\omega_e \tau) - \sin(\omega_e \tau) + a \sin(\omega_e \tau))}{\text{Denominator}} \right) / 2. 
\] (4.14)

The bright state population, which determines the optical absorption of the system, is given in the asymptotic limit by

\[
\rho^{i,k}_{\uparrow \downarrow} = \frac{1 - \cos(\tau \omega_e)}{\text{Denominator}}, 
\] (4.15)

where we have introduced

\[
\text{Denominator} = 2 - \cos \left( \frac{\theta}{2} (1 - \cos \frac{\theta}{2} + \cos(\tau \omega_e) \right) (1 - a) \right) \\
- \cos(\tau \omega_e)(1 + a) - c \left( 1 - \cos \left( \frac{\theta}{2} \right) \sin(\tau \omega_e) \right) 
\]
for brevity, with $c = [(1 - a) \Gamma/2 + b\omega_t]/\omega_c$.

Analyzing formula (4.15) for the synchronization condition $\omega_c \in \frac{2\pi}{\tau} Z$, we find that $\rho_{\uparrow\uparrow}^{i,*} = 0$. Hence, the entire population is in the dark state $\rho_{\downarrow\downarrow}^{i,*} = 1$.

**Asymptotic coarse–grained spontaneous emission rate**

A measurable quantity in this scheme is the rate at which photons are emitted. We first calculate the time–dependent spontaneous emission rate between the $k$th and the $(k + 1)$th pulse. For any $t_k + t_p \leq t \leq t_{k+1}$ holds

$$\tilde{\Gamma}_{\text{spon},k}(t) = \frac{\Gamma}{2} (\rho_{\uparrow\downarrow,\uparrow\downarrow}^{i,k} + \rho_{\downarrow\uparrow,\downarrow\uparrow}^{i,k})(t)$$

$$(4.5) \quad \Rightarrow \quad \frac{1}{2} \rho_{\uparrow\uparrow}^{i,k}(1 - \cos \theta) e^{-\frac{\Gamma}{2} t}$$

$$k \to \infty \quad \Rightarrow \quad \frac{1}{2} \rho_{\uparrow\downarrow}^{i,k}(1 - \cos \theta) e^{-\frac{\Gamma}{2} t} = \tilde{\Gamma}_{\text{spon}}(t).$$

We used (4.5) to express the time–dependent population in the excited state by the population in the bright state at the arrival of the pulse. We are interested in the overall absorption and hence the spontaneous emission rate between two pulses, rather than the instant at which the decay happens. Therefore, we average over the time interval between two pulses and obtain the coarse–grained spontaneous emission rate

$$\Gamma_{\text{spon},k} = \frac{1}{\tau} \int_{t_k}^{t_{k+1}} \tilde{\Gamma}_{\text{spon},k}(t) dt$$

$$(4.16) \quad \approx \frac{1}{\tau} \rho_{\uparrow\uparrow}^{i,k}(1 - \cos \theta)$$

$$k \to \infty \quad \approx \frac{1}{\tau} \rho_{\uparrow\downarrow}^{i,k}(1 - \cos \theta) = \Gamma_{\text{spon}}.$$

Clearly, the highest spontaneous emission rate is obtained for $\theta = \pi$.

**Perfect $\pi$–pulses**

The derivation of the asymptotic electron density matrix and the coarse–grained spontaneous emission rate was performed for arbitrary pulse areas $\theta$ and we found the most favorable pulse area for the scheme to be $\theta = \pi$. Focusing therefore only on this special case, we find that at high magnetic fields $|\omega_e|, \omega_t \gg \Gamma$ the expressions simplify drastically. The evolution between the $k$th and $(k + 1)$th $\pi$–pulse is then described by

$$\rho_{\uparrow\uparrow}^{i,k+1} \approx \frac{1}{2} \left[ 1 - (1 - \rho_{\uparrow\uparrow}^{i,k}) \cos(\omega_e \tau) \right],$$

$$\rho_{\downarrow\downarrow}^{i,k+1} \approx \frac{1}{2} \left[ 1 + (1 - \rho_{\uparrow\uparrow}^{i,k}) \cos(\omega_e \tau) \right],$$

$$\rho_{\uparrow\downarrow}^{i,k+1} \approx -\frac{i}{2} \left( 1 - \rho_{\uparrow\uparrow}^{i,k} \right) \sin(\omega_e \tau).$$

(4.17) (4.18) (4.19)

The asymptotic bright state population for a $\pi$–pulse can be shown to be

$$\rho_{\uparrow\downarrow}^{\pi,*} \approx \frac{1 - \cos(\omega_e \tau)}{2 - \cos(\omega_e \tau)} = \frac{\sin^2(\omega_e \tau/2)}{1 + \sin^2(\omega_e \tau/2)}.$$
4.3 Periodic optical excitation of the electron

Next we demonstrate how the bright state population approaches its asymptotic limit. To this end, we write down the recursive relation for the bright state population

\[ \rho_{\uparrow, \uparrow}^{i,k} = \frac{1}{2} \cos(\omega_e \tau) \rho_{\uparrow, \uparrow}^{i,k-1} + \frac{1}{2} (1 - \cos(\omega_e \tau)) \]

\[ = \alpha \rho_{\uparrow, \uparrow}^{i,k-1} + \left( \frac{1}{2} - \alpha \right), \]

where \( \alpha = \frac{1}{2} \cos(\omega_e \tau) \). It can be shown by induction that this recursive relation is solved by

\[ \rho_{\uparrow, \uparrow}^{i,k} = \alpha^{k-1} \rho_{\uparrow, \uparrow}^{i,1} + \left( \frac{1}{2} - \alpha \right) \sum_{p=0}^{k-2} \alpha^p \]

\[ = \alpha^{k-1} \rho_{\uparrow, \uparrow}^{i,1} + \left( \frac{1}{2} - \alpha \right) \frac{1 - \alpha^{k-1}}{1 - \alpha} \]

\[ = \alpha^{k-1} \rho_{\uparrow, \uparrow}^{i,1} + \frac{\frac{1}{2} - \alpha}{1 - \alpha} (1 - \alpha^{k-1}) \]

\[ = \frac{1}{2} - \alpha \]

\[ + \alpha^{k-1} \left( \rho_{\uparrow, \uparrow}^{i,1} - \rho_{\uparrow, \uparrow}^{i,*} \right), \]

where \( \rho_{\uparrow, \uparrow}^{i,1} \) is the bright state population at the arrival of the first pulse. To obtain the second line the partial sum for the geometric series \( \sum_{p=0}^{k-2} \alpha^p = \frac{1 - \alpha^{k-1}}{1 - \alpha} \) was used. In the last step, the identity

\[ \frac{1}{2} - \alpha \]

\[ = \frac{1 - \cos(\omega_e \tau)}{2 - \cos(\omega_e \tau)} = \frac{\sin^2(\frac{\omega_e \tau}{2})}{\frac{1}{2} + \sin^2(\frac{\omega_e \tau}{2})} = \rho_{\uparrow, \uparrow}^{i,*} \]

(4.20)

was plugged in. Since \( |\alpha| = |\frac{1}{2} \cos(\omega_e \tau)| \leq \frac{1}{2} \), the absolute error introduced when approximating the bright state population at the arrival of the \( k \)th pulse by the asymptotic value is bounded by

\[ |\rho_{\uparrow, \uparrow}^{i,k} - \rho_{\uparrow, \uparrow}^{i,*}| = |\alpha^{k-1} (\rho_{\uparrow, \uparrow}^{i,1} - \rho_{\uparrow, \uparrow}^{i,*})| \leq |\alpha|^{k-1} \leq 2^{-(k-1)}. \]

(4.21)

Finally, the coarse–grained asymptotic spontaneous emission rate for a \( \pi \)-pulse is given by

\[ \Gamma_{\text{spon}} = \frac{1}{\tau} \frac{\sin^2(\frac{\omega_e \tau}{2})}{\frac{1}{2} + \sin^2(\frac{\omega_e \tau}{2})} + \mathcal{O}(2^{-(k-1)}) \]

(4.22)

\[ = \frac{\tau}{2} (\omega_e - 2\pi m/\tau)^2 + \mathcal{O}(2^{-(k-1)}, (\omega_e - 2\pi m/\tau)^4). \]

(4.23)

On a side note, it is worth emphasizing that the synchronization condition and the quadratic behavior remain when the small parameters \( a \) and \( b \) are kept as in (4.15). However, the prefactor of the parabola is modified, making it narrower; in the context of the nuclear spin random walk described below, this would correspond to an increased sensitivity of the spontaneous emission rate to a single nuclear spin flip. This would be beneficial for many aspects of the scheme.
To conclude this section, we want to emphasize the assumptions of \( \tau \gg \Gamma^{-1} \gg \omega \omega^{-1} \) and state the main results again. The general expression of the asymptotic electron density matrix (4.16) achieves its maximal value at \( \theta = \pi \). Hence, applying a \( \pi \)-pulse is advantageous to get higher count rates, but the scheme is not limited to \( \pi \)-pulses. The second important result is that the spontaneous emission rate is zero if the synchronization condition \( \omega \in \frac{2\pi}{\tau} \mathbb{Z} \) is fulfilled, confirming the interpretation that the system achieves the dark state \( |\downarrow\rangle \) after a short train of pulses. If the synchronization condition is nearly fulfilled, the coarse–grained spontaneous emission rate behaves quadratically in \( \omega_e \).

### 4.4 Coupled electron–nuclei dynamics

The effect of the electron–nuclei interaction on the coupled system is twofold. On the one hand, the electron dynamics is modified by the Overhauser field which changes \( \omega_e \) to the effective electron precession frequency \( \omega_{\text{eff}} = \omega_e + \omega_{\text{Oh}} \). The dark state is then only reached at the arrival of a pulse if synchronization (4.2) is fulfilled for the effective precession frequency

\[
\omega_{\text{eff}} \in \frac{2\pi}{\tau} \mathbb{Z}.
\]  

The modified electron dynamics results in a coarse–grained spontaneous emission rate \( \Gamma_{\text{spon}} \) that depends on \( \omega_{\text{eff}} \) and thus on the nuclear Overhauser field \( B_{\text{Oh}} \), and vanishes if the synchronization condition is satisfied.

On the other hand, the nuclear dynamics is modified by optical excitation of the electron, which results in spontaneous emission assisted nuclear spin flips and leads to a slow time evolution of the nuclear Overhauser field. To describe such processes, we apply a Schrieffer–Wolff transformation \[146, 89\] to eliminate the energy non–conserving terms of \( H_Q \), as we show in detail below. We find second–order nuclear spin flip processes where the additional energy is supplied by a spontaneously emitted photon (Fig. 4.1 (b)) occurring with a rate

\[
R_{\text{nuc}} = \epsilon^2 \Gamma_{\text{spon}} \\
= \epsilon^2 \frac{1}{\tau^2} \sin^2(\omega_{\text{eff}} \tau/2) \\
= \epsilon^2 \frac{\tau}{2} \left( \omega_{\text{eff}}^2 - 2m/\tau \right)^2 + \mathcal{O} \left( (\omega_{\text{eff}}^2 - 2m/\tau)^4 \right)
\]  

where \( \epsilon = \sqrt{N \frac{A_{\text{eff}}}{4A_e}} \) under the assumptions of homogeneous hyperfine and quadrupolar coupling, \( \omega_n \gg A \), and \( I_z = \pm \frac{1}{2} \) for simplicity. While the replacement has been justified for spin-\( \frac{3}{2} \) in chapter 2, we will consider the case of spin-\( \frac{1}{2} \) in more detail in this section. The rate of optically–assisted nuclear spin flips \( R_{\text{nuc}} \) is drastically reduced in the vicinity of a synchronization condition (4.24). The signatures of this reduction can be observed in a single realization of a random walk of \( \omega_{\text{eff}} (I_z) \): the random walk freezes near a synchronization condition, which therefore constitutes a trap (red lines in Fig. 4.2 (a)). An unsynchronized electron is optically excited and induces an unbiased random walk of the nuclear polarization \( I_z = \sum_{j=1}^{N} \langle I_z \rangle \) until \( \omega_{\text{eff}} \) is synchronized. In the absence of other interactions, the time spent near such a trap can be on the order of the measurement time. We also simulated the
4.4 Coupled electron–nuclei dynamics

Figure 4.2: (a) The random walk of the effective electron precession frequency (blue) spends most of the time close to traps (red), indicating a Lévy distribution for long waiting times. The inset resolves the random walk between the traps. (b) $\Gamma_{\text{sp}}$ gives the spontaneous emission rate of the corresponding random walk in (a). (c) Exposing an ensemble of 500 QDs with normally distributed initial nuclear polarization ($\langle I_z \rangle = 5$, $\sigma = \sqrt{N}$) (red) to the pulsed laser train for $10^9 \tau = 10^9 / \Gamma$ leads to a final (blue) distribution of the effective electron precession frequency that is heavily peaked at the different traps (inset). For all simulations we used the parameters $\tau = 10 / \Gamma$, $\omega_e = 280 \omega_n / \tau$, $\omega_n = 0.18 \Gamma$, $N = 2000$, $\omega_Q = 0.013 \Gamma$, $A_H = 100 \Gamma$, and assumed inhomogeneous hyperfine coupling constants. Figure credit: Judith Höller

The time evolution of the distribution of $\omega_{\text{eff}}^e$, where the initial polarizations $\{I_j\}_{j=1}^N$ are drawn from a Gaussian distribution (red histogram in Fig. 4.2 (c)). We find that the probability of finding $\omega_{\text{eff}}^e$ within a single nuclear spin flip from a trap (4.24) exceeds 99%. These simulations are in excellent qualitative agreement with experiments carried out on an ensemble of single negatively-charged self-assembled QDs [91].

4.4.1 Schrieffer–Wolff transformation

In this section we detail the calculations that lead to the rate of optically assisted nuclear spin flips, $R_{\text{nuc}}$. First, we will apply a Schrieffer–Wolff transformation to eliminate energy non-conserving terms of the quadrupole Hamiltonian $H_Q$. In the limit $\Gamma \gg \omega_n$ a more elaborate treatment might be needed. Finally, we derive the rate of these optically assisted nuclear spin flips under more stringent assumptions.

For the Hamiltonians under consideration, the quadrupolar interaction is responsible for nuclear spin flips. To see which terms of $H_Q$ cause these spin flips, we split $H_Q$ into diagonal (D) and off-diagonal (O) parts, where the latter couples nuclear product states with different net polarization $\sum_{j=1}^N \langle I_j^z \rangle$ as described in section 2.3.3.

We write the total Hamiltonian as a dominant contribution $H_D$ and a perturbation $V = V_D + V_O$, where the perturbation is partitioned into a diagonal and an
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off–diagonal part as above

\[ H_D = H_0 + H_n, \]
\[ V_D = H_{O h} + H_Q^D, \]
\[ V_O = H_Q^O. \]

The Overhauser field and the diagonal quadrupolar Hamiltonian are treated as perturbations, since the corresponding coupling strengths \( A_j \) and \( \omega_Q^j \) are assumed to be much smaller than the nuclear Zeeman energy \( \omega_n^j \). It is apparent that the off–diagonal elements couple energy–detuned nuclear spin states \([89]\).

We apply a Schrieffer–Wolff transformation to eliminate the off–diagonal terms up to second order \([146, 89]\), i.e.

\[ \hat{H} = e^{-S}He^S \]

with

\[ S = \sum_{j=1}^{N} e_j^{[1]} \left( (I_+^j)^2 - (I_-^j)^2 \right) + \sum_{j=1}^{N} e_j^{[2]} S_z \left( (I_+^j)^2 - (I_-^j)^2 \right) - \sum_{j=1}^{N} 4 \left( e_j^{[1]} \right)^2 \left\{ (I_+^j)^2 - (I_-^j)^2, I_z^j \right\} \]

with the small parameters \( e_j^{[1]} = \frac{\omega_n^j}{16 \omega_n^2} \) and \( e_j^{[2]} = \frac{A_j}{\omega_n^2} e_j^{[1]} = \frac{A_j \omega_Q^j}{16 \omega_n^2} \). Even though the transformation on the Hamiltonian includes energy shifts that slightly modify the synchronization condition, they are not relevant for the scheme. Furthermore, any non–trivial nuclear spin dynamics that is obtained is treated non–perturbatively in the framework of the intrinsic limitation (see section 4.7). We therefore turn to the expansion of the dissipative part of the Liouvillian under the Schrieffer–Wolff transformation \([89]\), i.e.

\[ \tilde{\mathcal{L}}(\rho) = e^{-S} \mathcal{L}(\rho)e^S. \]

We start with the zeroth order Liouvillian

\[ \tilde{\mathcal{L}}^{[0]}(\rho) = \frac{\Gamma}{2} \left( \rho_{\uparrow\uparrow,\uparrow\uparrow} |\uparrow\rangle \langle \uparrow| + \rho_{\downarrow\downarrow,\downarrow\downarrow} |\downarrow\rangle \langle \downarrow| - \frac{1}{2} \left\{ |\uparrow\downarrow\uparrow\rangle \langle \uparrow\downarrow\uparrow| + |\uparrow\uparrow\downarrow\rangle \langle \uparrow\uparrow\downarrow|, \rho \right\} \right) \]

where \( \rho_{\uparrow\downarrow,\uparrow\downarrow} = \langle \uparrow\downarrow | \rho | \uparrow\downarrow \rangle \) only acts on the nuclear spin manifolds and \( \rho = \rho_e \otimes \rho_n + \rho_{\text{correl}} \). Since \( S^{[1]} \) only consists of nuclear operators, it does not lead to nuclear spin dynamics, i.e., \( \text{Tr}_e (e^{-S^{[1]}} \tilde{\mathcal{L}}(\rho)e^{+S^{[1]}}) = 0 \). For the second order, we note that the electron spin operator contained in \( S^{[2]} \) only acts on the electron ground–state manifold. Hence the transformation leaves the second term of \( \mathcal{L} \) invariant. Transforming the first term yields

\[ e^{-S^{[2]}} \left( \rho_{\uparrow\downarrow,\uparrow\downarrow} |\uparrow\rangle \langle \uparrow| + \rho_{\downarrow\uparrow,\downarrow\uparrow} |\downarrow\rangle \langle \downarrow| \right) e^{+S^{[2]}} \]

\[ \approx \rho_{\uparrow\downarrow,\uparrow\downarrow} |\uparrow\rangle \langle \uparrow| + \rho_{\downarrow\uparrow,\downarrow\uparrow} |\downarrow\rangle \langle \downarrow| + \left[ \rho_{\uparrow\downarrow,\uparrow\downarrow} |\uparrow\rangle \langle \uparrow| + \rho_{\downarrow\uparrow,\downarrow\uparrow} |\downarrow\rangle \langle \downarrow|, S^{[2]} \right] + \frac{1}{2} \left[ \left[ \rho_{\uparrow\downarrow,\uparrow\downarrow} |\uparrow\rangle \langle \uparrow| + \rho_{\downarrow\uparrow,\downarrow\uparrow} |\downarrow\rangle \langle \downarrow|, S^{[2]} \right], S^{[2]} \right] + \ldots \]

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Before explicitly calculating these terms, we note that in the $x$-basis $S_z = \frac{1}{2}(|↑⟩⟨↓| + |↓⟩⟨↑|)$. For clarity of the following expressions, we introduce the nuclear spin operator $O_j = (I_j^+)^2 - (I_j^-)^2$ such that

$$S^{[2]} = \sum_{j=1}^{N} \epsilon_j^{[2]} S_j O_j - \sum_{j=1}^{N} 4 \left( \epsilon_j^{[1]} \right)^2 \{O_j, I_j^z\}.$$

Since the second term of $S^{[2]}$ does not couple to $L$, only the first term of $S^{[2]}$ is relevant for the expansions of $\tilde{L}$. Explicitly we find

$$\tilde{L}^{[0]}(\rho) = \frac{\Gamma}{2} (\rho_{↑↓⇑} |↑⟩⟨↑| + \rho_{↑↓⇓} |↓⟩⟨↓|),$$

$$\tilde{L}^{[1]}(\rho) = \sum_{j=1}^{N} \frac{\Gamma}{4} \epsilon_j^{[2]} \left[ (\rho_{↑↓⇑} O_j - O_j \rho_{↑↓⇓}) |↑⟩⟨↑| + (\rho_{↑↓⇓} O_j - O_j \rho_{↑↓⇑}) |↓⟩⟨↓| \right],$$

$$\tilde{L}^{[2]}(\rho) = -\sum_{j,k=1}^{N} \epsilon_j^{[2]} \epsilon_k^{[2]} \frac{\Gamma}{8} \left( O_j O_k - \frac{1}{2} \{O_j, O_k\} \right) |↑⟩⟨↑| + \frac{1}{2} \left( O_j O_k + \{O_j, O_k\} \right) |↓⟩⟨↓|.\$$

The complete zeroth order Liouvillian as well as the first order Liouvillian have no effect on the reduced nuclear density matrix $\rho_n = \text{Tr}_e(\rho)$ since when tracing over the electron, the terms cancel each other due to $\text{Tr}_e(|↑⟩⟨↓|) = \text{Tr}_e(|↓⟩⟨↑|) = 0$. The second order Liouvillian leads to the lowest–order optically–assisted nuclear
spin flips. Tracing over the electron degrees of freedom yields

\[
\text{Tr}_e(\mathcal{L}^{[2]}(\rho)) = \sum_{j,k=1}^{N} -\epsilon_j^2 \epsilon_k^2 \frac{\Gamma}{8} \left( O_j \rho_k O_k - \frac{1}{2} \{ O_j O_k, \rho_k \} \right)
\]

\[
= \sum_{j,k=1}^{N} \epsilon_j^2 \epsilon_k^2 \frac{\Gamma}{8} \left( (I^j_+)^2 \rho_k (I^-_k)^2 + (I^j_-)^2 \rho_k (I^+_k)^2 \right.
\]

\[
- \frac{1}{2} \left\{ (I^j_+)^2 (I^-_k)^2 + (I^j_-)^2 (I^+_k)^2, \rho_k \right\} \right)
\]

\[
- \sum_{j,k=1}^{N} \epsilon_j^2 \epsilon_k^2 \frac{\Gamma}{8} \left( (I^j_+)^2 \rho_k (I^+_k)^2 + (I^j_-)^2 \rho_k (I^-_k)^2 \right.
\]

\[
- \frac{1}{2} \left\{ (I^j_+)^2 (I^-_k)^2 + (I^j_-)^2 (I^+_k)^2, \rho_k \right\} \right),
\]

where \( \rho_k = \langle \uparrow\downarrow\uparrow \mid \rho \mid \uparrow\downarrow\uparrow \rangle + \langle \uparrow\downarrow\downarrow \mid \rho \mid \uparrow\downarrow\downarrow \rangle \) is an operator and \( I^\pm_k = I^\pm x \pm i I^\pm y \).

In the semiclassical limit, we assume that the correlations between the electron and nuclear spin systems are negligible at all times, i.e. \( \rho = \rho_e \otimes \rho_n \). This implies that \( \langle \uparrow\downarrow\uparrow \mid \rho(t) \mid \uparrow\downarrow\uparrow \rangle = \langle \uparrow\downarrow\downarrow \mid \rho_n(t) \otimes \rho_n(t) \mid \uparrow\downarrow\downarrow \rangle = \rho_{\uparrow\downarrow\uparrow,\uparrow\downarrow\downarrow}(t) \rho_{\uparrow\downarrow\uparrow,\uparrow\downarrow\uparrow}(t) \) is an operator that acts on the nuclear spins, where \( \rho_{\uparrow\downarrow\uparrow,\uparrow\downarrow\downarrow}(t) \) is the population in one of the excited electronic (trion) states.

We further simplify the computations by considering each nuclear spin to be an effective spin \( \frac{1}{2} \) particle such that we can replace \((I^\pm_k)^2 \rightarrow I^\pm_k\). The spin of a Ga or As nucleus satisfies \( I = 3/2 \), and therefore the quadrupolar interaction \( H_Q \) splits into two decoupled spin-1/2 systems, which further motivates this simplification (see chapter 2). In the case of In, \( I = 9/2 \), we note that even though \( I^2_\pm \) introduces extra factors in the rate that are bounded by \( \frac{9}{2} \cdot \frac{11}{2} \approx 25 \), there are many more nuclear spin configurations that realize each nuclear spin polarization state. How the latter effects enter the nuclear spin flip rate will be discussed below; importantly, they counteract the modification of the nuclear spin flip rate induced by replacing \( I_\pm \rightarrow I = 1/2 \) and we expect qualitatively similar nuclear spin dynamics.

We now introduce further assumptions on the nuclear density matrix \( \rho_n \). For a given total polarization \( \langle I_z \rangle \), \( \rho_n \) is assumed to correspond to an ensemble of products of \( I^2_z \)-eigenstates \( |n^{(I_z)} \rangle \) with total polarization \( \langle I_z \rangle \). There are \( N_{\langle I_z \rangle} \) such product states, where

\[
N_{\langle I_z \rangle} = \frac{N!}{(N - \langle I_z \rangle)! (\frac{N}{2} + \langle I_z \rangle)!}.
\]

The product states are given by

\[
|n^{(I_z)}_{\alpha} \rangle = \otimes_j |I^2_z = 1/2, m^\alpha_j \rangle \quad \forall \alpha = 1, ..., N_{\langle I_z \rangle},
\]

where \( m^\alpha_j \in \{-\frac{1}{2}, \frac{1}{2}\} \). Furthermore, they satisfy

\[
\sum_{j=1}^{N} I^z_j |n^{(I_z)}_{\alpha} \rangle = \sum_{j=1}^{N} m^\alpha_j |n^{(I_z)}_{\alpha} \rangle = \langle I_z \rangle |n^{(I_z)}_{\alpha} \rangle \quad \forall \alpha = 1, ..., N_{\langle I_z \rangle}.
\]
Then the nuclear density matrix is written as

\[ \rho_{(I_z)} = \sum_{\alpha=1}^{N(I_z)} \frac{1}{N(I_z)} |n^{(I_z)}_\alpha \rangle \langle n^{(I_z)}_\alpha |. \]

It was assumed that any product state with total projection \( \langle I_z \rangle \) is equally likely and occurs with a probability \( \frac{1}{N(I_z)} \). If the coherences between the product states of \( |n^{(I_z)}_\alpha \rangle \) can be taken to vanish at all times, the only relevant terms in \( \tilde{L}^{[2]}[\rho] \) are those for which \( j = k \). Terms with \( j \neq k \) lead to coherences between product states which decay quickly if the system experiences large inhomogeneities.

Finally, \( R_\pm (t) \approx \text{Tr}_n \left( \sum_{j=1}^{N} (\epsilon_j^{[2]} \frac{2}{\Gamma} I^j_{\pm} \rho^{(I_z)}_{\pm}(t) \rho^{(I_z)}_{\pm}) I^j_{\mp} \right) = N_\pm \epsilon^2 \frac{\Gamma}{8} \rho^{(I_z)}_{\pm}(t), \)

where \( \rho^{(I_z)}_{\pm} = \rho^{(I_z)}_{\uparrow \downarrow \uparrow \downarrow}(t) + \rho^{(I_z)}_{\uparrow \downarrow \downarrow \uparrow}(t) \) is the trion population, given that the nuclear spin polarization is \( \langle I_z \rangle \). In the last step, it was assumed that the hyperfine and quadrupolar couplings are homogeneous such that \( \bar{\epsilon} = \epsilon_j^{[2]} = \frac{A_{\omega_j^2}}{16(\omega_j)^2} \) for all \( 1 \leq j \leq N \). Coarse graining these rates yields

\[ R_\pm = \frac{1}{\tau} \int^{t_{k+1}}_{t_k} N_\pm \epsilon^2 \frac{\Gamma}{8} \rho^{(I_z)}_{\pm}(t) \, dt = \frac{N_\pm}{4} \epsilon^2 \Gamma_{\text{spon}}. \]

The total rate to flip a nuclear spin is given by \( R_{\text{nuc}} = R_+ + R_- \), whence that we find

\[ R_{\text{nuc}} = \epsilon^2 \Gamma_{\text{spon}}, \quad (4.25) \]

with \( \epsilon = \sqrt{\frac{N}{2}} \bar{\epsilon} \).

To conclude this section, the quadrupolar coupling together with the Overhauser term induces diffusion in the nuclear spin polarization at a rate \( R_{\text{nuc}} \). Due to the linear relation between \( R_{\text{nuc}} \) and \( \Gamma_{\text{spon}} \), the nuclear spin flip rate also depends quadratically on \( \omega^2_{\text{eff}} \) close to a synchronization condition.

### 4.5 Lévy flights in the nuclear environment

To quantify the freezing of a random walk of \( \omega^2_{\text{eff}} \) in the vicinity of a trap, we introduce the probability distribution of the waiting time between two successive nuclear spin flip events \( P_{\text{nuc}}(\Delta t_{\text{nuc}}) \) and show that in the asymptotic limit it behaves according to a power--law. A striking feature of the ensuing Lévy flight behavior of the nuclear trapping times is that the tails of the distribution dominate the dynamics. We perform a simple calculation that allows to trace the origin of the power--law tail to the quadratic dependence of the nuclear spin flip rate on \( \omega^2_{\text{eff}} \) in the vicinity of a trap. Then we present a more rigorous derivation of the asymptotic trapping time distribution.

Long waiting times occur for \( \omega^2_{\text{eff}} \) close to a trap since

\[ \Delta t_{\text{nuc}} \sim R_{-1}^{\text{nuc}} \propto \left( \omega^2_{\text{eff}} - \frac{2 \pi}{\tau_m} m \right)^{-2}. \]
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This relationship of $\Delta t_{\text{nuc}}$ to $\omega_{\text{eff}}$ allows us to write

$$P_{\text{nuc}}(\Delta t_{\text{nuc}})d\Delta t_{\text{nuc}} = \Pi_{\text{nuc}}(\omega_{\text{eff}})d\omega_{\text{eff}},$$

(4.26)

where the probability to reach an effective precession frequency $\omega_{\text{eff}}$ in the trap is denoted by $\Pi_{\text{nuc}}(\omega_{\text{eff}})$ and is assumed to be uniform [17]. Estimating the change in $\Delta t_{\text{nuc}}$ induced by an infinitesimal change in $\omega_{\text{eff}}$ via

$$\frac{d\Delta t_{\text{nuc}}}{d\omega_{\text{eff}}} \propto (\omega_{\text{eff}} - \frac{2\pi}{\tau} m)^{-3} \propto \Delta t_{\text{nuc}}^{3/2},$$

(4.27)

results in the power-law distribution

$$P_{\text{nuc}}(\Delta t_{\text{nuc}}) \propto \Delta t_{\text{nuc}}^{-3/2}.$$  

(4.28)

This expression is valid for $\Delta t_{\text{nuc}} \gg (R_{\text{nuc}}|_{\omega_{\text{eff}}=\frac{2\pi}{\tau} m, A})^{-1}$, as explained in more detail below. As a consequence of the power-law (or fat tail) of the distribution, the expectation value of $\Delta t_{\text{nuc}}$ diverges. This Lévy behavior of the nuclear spin flip waiting time is thus based on the quadratic dependence of $\Gamma_{\text{spon}}$ and $R_{\text{nuc}}$ on $\omega_{\text{eff}}$.

We adapt the more rigorous derivation of the Lévy distribution given for VSCPT [16, 17] to the nuclear spin environment. The nuclear spin flip rate is given by

$$R_{\text{nuc}}(\omega_{\text{eff}}) = \epsilon^2 \frac{\tau}{2} (\omega_{\text{eff}})^2,$$

where without loss of generality $\omega_{\text{eff}}$ is considered to be close to the trap $m = 0$. The conditional probability density to be trapped for a time $\Delta t_{\text{nuc}}$, given $\omega_{\text{eff}}$ is

$$P_{\text{nuc}}(\Delta t_{\text{nuc}} | \omega_{\text{eff}}) = R_{\text{nuc}}(\omega_{\text{eff}}) e^{-R_{\text{nuc}}(\omega_{\text{eff}}) \Delta t_{\text{nuc}}}
\left(\epsilon^2 \frac{\tau}{2} (\omega_{\text{eff}})^2\right) e^{-\Delta t_{\text{nuc}} \epsilon^2 \frac{\tau}{2} (\omega_{\text{eff}})^2}.
$$

The total probability density to be trapped for a time $\Delta t_{\text{nuc}}$ is obtained by integrating the joint probability distribution of $\Delta t_{\text{nuc}}$ and $\omega_{\text{eff}}$ over $\omega_{\text{eff}}$. Therefore we first need to specify the probability density of $\omega_{\text{eff}}$. Here, we assume inhomogeneous hyperfine couplings so that any $\omega_{\text{eff}}$ in the trap $[-\omega_{\text{trap}}, \omega_{\text{trap}}]$ can be reached with equal probability, i.e.

$$\Pi(\omega_{\text{eff}}) = \frac{1}{2\omega_{\text{trap}}}.$$

(4.29)

For this to be true, we choose $\omega_{\text{trap}}$ to be much smaller than the average Overhauser shift induced by a single nuclear spin flip, i.e.

$$\omega_{\text{trap}} \ll A.$$

(4.30)

The trapping time at $\omega_{\text{trap}}$ is equal to $\tau_1$ and satisfies

$$\tau_1 \gg \left(R_{\text{nuc}}|_{\omega_{\text{eff}}=A}\right)^{-1} \approx \frac{2}{\epsilon^2 A^2 \tau}.$$

(4.31)
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Then, the trapping time probability distribution is given by

\[ P_{\text{nucl}}(\Delta t_{\text{nucl}}) = \int_{-\omega_{\text{trap}}}^{\omega_{\text{trap}}} P_{\text{nucl}}(\Delta t_{\text{nucl}}|\omega_e^{\text{eff}})\Pi(\omega_e^{\text{eff}})d\omega_e^{\text{eff}}. \]

\[ = \frac{2}{2\omega_{\text{trap}}} \int_{0}^{\omega_{\text{trap}}} P_{\text{nucl}}(\Delta t_{\text{nucl}}|\omega_e^{\text{eff}})d\omega_e^{\text{eff}} \]

\[ = \frac{1}{\omega_{\text{trap}}} \int_{0}^{\omega_{\text{trap}}} \left( \epsilon^2 \frac{\tau}{2}(\omega_e^{\text{eff}})^2 \right) e^{-\Delta t_{\text{nucl}}\epsilon^2 \frac{\tau}{2}(\omega_e^{\text{eff}})^2}d\omega_e^{\text{eff}} \]

\[ = \frac{1}{\omega_{\text{trap}}} \left( \frac{\tau_1^{1/2}}{\Delta t_{\text{nucl}}^{3/2}} \gamma \left( \frac{3}{2}, \frac{\Delta t_{\text{nucl}}}{\tau_1} \right) \right) \]

where \( \Delta t_{\text{nucl}}/\tau_1 \to \infty \) \( \sim \frac{\sqrt{\pi} \tau_1}{4\Delta t_{\text{nucl}}^{3/2}} \)

(4.32)

where we have used the incomplete gamma function

\[ \gamma(s, x) = \int_{0}^{x} u^{s-1}e^{-u}du \]

and its limit

\[ \lim_{x \to \infty} \gamma(s, x) = \Gamma(s), \]

where \( \Gamma(s) \) is the Gamma function. In particular, we applied this to \( \gamma(\frac{3}{2}, x) \to \frac{1}{2}\Gamma(\frac{1}{2}) = \sqrt{\pi} \). This derivation confirms the results obtained with the short calculation presented above.

In the following sections, this derivation is adapted and generalized to include a constant offset in the nuclear and optical rates. Using this result, we show that the exponent of the nuclear trapping times is imprinted on the optical waiting times. We also discuss the modified trapping probability distribution in the presence of imperfections.

4.6 Real–time monitoring of the Lévy flights

The physical observable in an experiment is the waiting time \( \Delta t_{\text{spon}} \) between two successive photon emission events, irrespective of whether the photon emission is accompanied by a nuclear spin flip or not. The rate of photon emission events is given by

\[ \Gamma_{\text{spon}} = \frac{\tau}{2}(\omega_e^{\text{eff}})^2. \]

(4.33)

The derivation of the asymptotic waiting time distribution of successive photon emission events is analogous to the derivation for the nuclear trapping times since \( \Gamma_{\text{spon}} = \epsilon^{-2} R_{\text{nucl}} \) and where \( \Pi(\omega_e^{\text{eff}}) \) remains unchanged. However, the photon scattering rate is enhanced by a factor of \( \epsilon^{-2} \) compared to the nuclear spin flip rate at \( \omega_{\text{trap}} \). Equivalently, the time window between successive photon emission events is shortened by a factor of \( \epsilon^2 \) compared to the time between nuclear spin flip events. Therefore, the expression

\[ P_{\text{spon}}(\Delta t_{\text{spon}}) \sim \frac{\sqrt{\pi} \epsilon^2 \tau_1}{4\Delta t_{\text{spon}}^{3/2}} \]

(4.34)
Counts per unit time ($\Gamma$) \\
Sanpon ($1/\Gamma$) \\
$R_0=0$ \\
$R_0=10^{-8}\Gamma$ \\
-1.49 \\
-1.53 \\

Figure 4.3: The optical waiting time distribution for $R_0 = 0$ (red) is extracted from 100 time traces. The corresponding power–law exponent in this simulation is $-1.49$, consistent with the theoretical prediction of $-3/2$. For $R_0^{-1} = 10^8/\Gamma$ (blue) we find a regime where a power–law with exponent $-1.53$ is observed, followed by an exponential decay. All other parameters are the same as in Fig. 4.2. Figure credit: Judith Höller

is already valid for $\Delta t_{\text{spon}} \gg \epsilon^2 \left( R_{\text{nuc}} \right)_{\omega_{\text{eff}}=\frac{2\pi}{\tau_m\pm A}}^{-1}$.

As a result of the proportionality of the rate of nuclear spin flip events $R_{\text{nuc}}$ to spontaneous emission events $\Gamma_{\text{spon}}$, the power–law exponent of the nuclear trapping time distribution $P_{\text{nuc}}$ is imprinted on the optical waiting time distribution $P_{\text{spon}}$. Consequently, by finding the distribution of optical waiting times, the distribution of nuclear trapping times can be deduced. Furthermore, since the rate of photon detection events is enhanced by a factor $\epsilon^{-2}$ compared to the nuclear spin flip rate, it is possible to monitor the Lévy flight behavior of nuclear trapping times in real–time even with modest photon detection efficiencies.

To numerically verify the power–law behavior of the waiting time distribution, we averaged over 100 Monte–Carlo simulations (of the type shown in Fig. 4.2 (a)). We then plot a histogram of the waiting times and extracted a power–law exponent $\eta = -1.49$, shown in Fig. 4.3 (red dotted line), in excellent agreement with the predicted distribution of optical waiting times (4.34). To reduce the noise in the tail, one could use a logarithmic bin size [196], but even with this method the noise increases for the tail of the distribution. We further note that we employ the most common method to extract the power–law exponent by fitting the slope of the resulting straight line in a double–logarithmic plot. However, it was shown that under certain circumstances this introduces systematic errors [196].

### 4.7 Limitations of the scheme

Experimentally observable waiting times are limited by additional interactions that have been neglected so far. To investigate their impact on the probability distribution of the optical waiting and nuclear trapping times, we assume that these imperfections can be modeled as an additional constant spontaneous emis-
sion rate $\Gamma_0$ and nuclear spin flip rate $R_0$, respectively. We will first derive the modified waiting and trapping time distributions. Then, we discuss possible physical origins of these additional rates. Next, we focus on three specific limitations arising due to the dephasing of the electron spin and imperfect laser polarization (both yielding $\Gamma_0 \neq 0$) as well as the intrinsic evolution of the system under the quadrupolar interaction in the time window that separates the laser pulses (giving rise to $R_0 \neq 0$). Finally, we discuss the possibility of implementing a displaced echo pulse sequence. This adds flexibility to the scheme and thereby minimizes the contribution of the intrinsic evolution to the nuclear spin flip rate.

4.7.1 Modified trapping time distribution

We assume that the imperfections can be modeled as an additional constant nuclear spin flip rate $R_0$ and spontaneous emission rate $\Gamma_0$, respectively, such that the overall nuclear spin flip rate becomes

$$\tilde{R}_{\text{nuc}} = R_{\text{nuc}} + R_0,$$

and the coarse–grained spontaneous emission rate reads

$$\tilde{\Gamma}_{\text{spon}} = \Gamma_{\text{spon}} + \Gamma_0.$$

The constant rates $R_0$ and $\Gamma_0$ prevent diverging average trapping and waiting times, respectively, since they effectively limit the trapping and waiting times to $R_0^{-1}$ and $\Gamma_0^{-1}$. This scenario corresponds to a truncated Lévy flight [197].

We exemplarily derive the modified nuclear trapping time distribution $\tilde{P}_{\text{nuc}}$, by generalizing the treatment given by [16, 17] and section 4.5 to include a constant offset $R_0$ in the nuclear spin flip rate, i.e.

$$\tilde{R}_{\text{nuc}}(\omega_{\text{eff}}^e) = \frac{\epsilon^2}{2} \tau^2 (\omega_{\text{eff}}^e)^2 + R_0,$$

where without loss of generality $\omega_{\text{eff}}^e$ is considered to be close to the trap $m = 0$. The modified optical waiting time distribution $\tilde{P}_{\text{spon}}$ is obtained by replacing $R_0 \rightarrow \Gamma_0$ and $\frac{\epsilon^2}{2} \tau^2 (\omega_{\text{eff}}^e)^2 \rightarrow \frac{\tau}{2} (\omega_{\text{eff}}^e)^2$.

The conditional probability density to be trapped for a time $\Delta t_{\text{nuc}}$, given $\omega_{\text{eff}}^e$ is

$$\tilde{P}_{\text{nuc}}(\Delta t_{\text{nuc}} | \omega_{\text{eff}}^e) = \tilde{R}_{\text{nuc}}(\omega_{\text{eff}}^e) e^{-\tilde{R}_{\text{nuc}}(\omega_{\text{eff}}^e) \Delta t_{\text{nuc}}} = \left( \frac{\epsilon^2}{2} \tau^2 (\omega_{\text{eff}}^e)^2 + R_0 \right) e^{-\Delta t_{\text{nuc}} \frac{\epsilon^2}{2} \tau^2 (\omega_{\text{eff}}^e)^2} e^{-R_0 \Delta t_{\text{nuc}}}.$$

The total probability density to be trapped for a time $\Delta t_{\text{nuc}}$ is obtained by integrating the joint probability distribution of $\Delta t_{\text{nuc}}$ and $\omega_{\text{eff}}^e$ over $\omega_{\text{eff}}^e$. We again assume $\Pi(\omega_{\text{eff}}^e) = \frac{1}{2\omega_{\text{trap}}}$, which leads to the restrictions $\omega_{\text{trap}} \ll A$ and $\tau_1 \gg \left( R_{\text{nuc}} | \omega_{\text{eff}}^e = A \right)^{-1} \approx \frac{2}{\epsilon^2 A^2 \tau}$. Then, the trapping time probability distribution is
given by

\[
\hat{P}_{\text{nuc}}(\Delta t_{\text{nuc}}) = \int_{-\omega_{\text{trap}}}^{\omega_{\text{trap}}} \hat{P}_{\text{nuc}}(\Delta t_{\text{nuc}}|\omega_e) \Pi(\omega_e) d\omega_e
\]

\[
= \frac{2}{2\omega_{\text{trap}}} \int_{-\omega_{\text{trap}}}^{\omega_{\text{trap}}} \hat{P}_{\text{nuc}}(\Delta t_{\text{nuc}}|\omega_e) d\omega_e
\]

\[
= \frac{1}{\omega_{\text{trap}}} \int_{0}^{\omega_{\text{trap}}} \left( \frac{1}{2} \Delta t_{\text{nuc}}^{3/2} \gamma \left( \frac{3}{2}, \Delta t_{\text{nuc}} \right) + \frac{R_0}{2} \frac{\tau_1^{1/2}}{\Delta t_{\text{nuc}}^{1/2}} \gamma \left( \frac{1}{2}, \Delta t_{\text{nuc}} \frac{\tau_1}{\tau_1} \right) \right) e^{-R_0 \Delta t_{\text{nuc}}} d\omega_e
\]

\[
\Delta t_{\text{nuc}}/\tau_1 \rightarrow \infty \rightarrow \sqrt{\pi} \tau_1^{3/2} (1 + 2R_0 \Delta t_{\text{nuc}}) e^{-R_0 \Delta t_{\text{nuc}}}
\]

where we have used the limits \(\gamma(\frac{1}{2}, x) \rightarrow \Gamma(\frac{1}{2}) = \sqrt{\pi}\) and \(\gamma(\frac{3}{2}, x) \rightarrow \frac{1}{2} \Gamma(\frac{1}{2}) = \frac{\sqrt{\pi}}{2}\).

Clearly, in the limit \(R_0 = 0\) we recover

\[
P_{\text{nuc}}(\Delta t_{\text{nuc}}) \sim \frac{\sqrt{\pi} \tau_1}{4\Delta t_{\text{nuc}}^{3/2}}.
\]

For \(R_0 \neq 0\) and \(\Delta t_{\text{nuc}} \gg R_0^{-1}\) the exponential term dominates and leads to an exponential decay. However, in an intermediate regime where the background rate \(R_0\) is much smaller than \(R_{\text{nuc}}\) at a single spin flip away from synchronization (i.e. \(R_0 \ll R_{\text{nuc}}|\omega_{\text{eff}} = m \frac{2\hbar}{T} \pm \Lambda\)), a power–law regime is observed before the exponential cutoff.

The modified probability distribution of successive photon emission events \(\hat{P}_{\text{spon}}\) behaves analogously, i.e.

\[
\hat{P}_{\text{spon}}(\Delta t_{\text{spon}}) \sim \frac{\sqrt{\pi} \epsilon^2 \tau_1}{4\Delta t_{\text{spon}}^{3/2}} (1 + 2\Gamma_0 \Delta t_{\text{spon}}) e^{-\Gamma_0 \Delta t_{\text{spon}}}
\]

and is valid for \(\Delta t_{\text{spon}} \gg \epsilon^2 \tau_1\).

A model simulation including a constant offset \(R_0 \neq 0\) shows a power law distribution with the power–law exponent \(-1.53\) (blue dotted line in Fig. 4.3) for intermediate waiting times and an exponential decay for large waiting times.

### 4.7.2 Physical origin of the limitations

The physical origin of \(R_0\) can be classified as system–specific or intrinsic to the scheme. In the case of an electron localized in a self–assembled QD, the coupling of the electron spin to phonons [198] or to a Fermionic reservoir [199] could limit the trapping times. In addition, finite pulse duration and imperfect laser polarization lead to a finite decay rate out of the electronic dark state. These effects can be suppressed by proper engineering.

In contrast, the scheme is intrinsically limited by the interaction that causes the optically assisted random walk, i.e. the quadrupolar interaction \(H_Q\) in self–assembled QDs. In the time windows that separate the successive laser pulses, \(H_Q\) leads to a coherent precession between different \(I_z\) eigenstates; the precession
frequency is dominated by the energy detuning $\sim \omega_n$. In the absence of a measurement interaction interrupting the coherent evolution, no actual nuclear spin flip can occur due to the energy mismatch of the coherently coupled nuclear states. However, the laser pulse terminating the period of coherent evolution under $H_Q$ effects such a measurement and could lead to an actual nuclear spin flip. Consequently, the rate at which these intrinsic nuclear spin flips occur depends on the bright state population in a different nuclear spin manifold at the arrival of a pulse. Similar intrinsically limiting mechanisms appear for systems with dominant non-collinear or hyperfine flip-flop interactions, where the latter is discussed in [162].

Before concluding this section, we note that the relationship between $R_0$ and $\Gamma_0$ depends on the physical origin of the offsets. To illustrate this, we consider the examples of electron dephasing and the intrinsic limitation, both of which will be discussed in more detail below. In the case of electron dephasing an additional constant offset $\Gamma_0$ is added to the coarse-grained spontaneous emission rate. These photon emission events also contribute to optically-assisted nuclear spin flips and therefore give rise to $R_0 = \epsilon^2 \Gamma_0$. However, the situation is different for the intrinsic limitation. The intrinsic limitation arises due to a coherent rotation between nuclear spin states that are different $I^z$ eigenstates. The energy mismatch between these states prevents actual nuclear spin flips. However, the laser pulse terminating the period of coherent evolution under $H_Q$ can effect a measurement interaction and could lead to a nuclear spin flip. The rate at which these intrinsic nuclear spin flips occur is denoted by $R_0$. Since such an intrinsic nuclear spin flip has to be accompanied by spontaneous emission, the intrinsic spontaneous emission rate equals the intrinsic nuclear spin flip rate, i.e. $\Gamma_0 = R_0$. This is advantageous when extracting the power-law exponent from the optical waiting time distribution since the cut-off for $P_{\text{spon}}$ is not at $\epsilon^2 R_0^{-1}$ but rather the power-law is expected to extend up to times $R_0^{-1}$.

4.7.3 Electron dephasing

As a first application of the modified waiting time distribution derived above, we now include the effect of pure dephasing of the electron spin. Clearly, one expects that dephasing destroys the dark state and therefore allows for optical excitation even if the synchronization condition is fulfilled. Thereby, it induces nuclear spin flips out of the trapped state.

First, the asymptotic electron density matrix in the presence of pure dephasing is derived. To this end, pure dephasing is introduced in the master equation, using the following two collapse operators:

$$C_{\text{deph},1} = \sqrt{\gamma} |\uparrow_z\rangle \langle \uparrow_z| = \sqrt{\gamma} \left( |\uparrow\rangle \langle \uparrow| + |\uparrow\rangle \langle \downarrow| + |\downarrow\rangle \langle \uparrow| + |\downarrow\rangle \langle \downarrow| \right)$$

$$C_{\text{deph},2} = \sqrt{\gamma} |\downarrow_z\rangle \langle \downarrow_z| = \sqrt{\gamma} \left( |\uparrow\rangle \langle \uparrow| - |\uparrow\rangle \langle \downarrow| - |\downarrow\rangle \langle \uparrow| + |\downarrow\rangle \langle \downarrow| \right).$$

Following the procedure in section 4.3, the only equations that are modified by dephasing are the ground state population difference and ground state coherence difference. Assuming that the lowest order equation for the population difference
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suffices and $\theta = \pi$, we find the following system of equations

$$\frac{\partial}{\partial t} \begin{pmatrix} \rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow} \\ \rho_{\uparrow,\downarrow} - \rho_{\downarrow,\uparrow} \end{pmatrix} = \begin{pmatrix} -\gamma & +i\omega_e \\ +i\omega_e & -\gamma \end{pmatrix} \begin{pmatrix} \rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow} \\ \rho_{\uparrow,\downarrow} - \rho_{\downarrow,\uparrow} \end{pmatrix}.$$ 

With the initial conditions given in (4.3), we find the solution

$$\begin{pmatrix} \rho_{\uparrow,\uparrow} - \rho_{\downarrow,\downarrow} \\ \rho_{\uparrow,\downarrow} - \rho_{\downarrow,\uparrow} \end{pmatrix}(t_{k+1}) = \rho_{\uparrow,\downarrow}(t_k) e^{-\gamma \tau} \begin{pmatrix} -\cos(\omega_e \tau) \\ i\sin(\omega_e \tau) \end{pmatrix}.$$

The solution is the same as without dephasing, but scaled with a factor $e^{-\gamma \tau}$.

In the asymptotic limit $k \to \infty$ we find the density matrix at the arrival of the pulse to be

$$\rho^{\ast} = \begin{pmatrix} 1-e^{-\gamma \tau} \cos(\omega_e \tau) & 1-e^{-\gamma \tau} \sin(\omega_e \tau) \\ \frac{2-e^{-\gamma \tau} \cos(\omega_e \tau)}{2} & \frac{1}{2} e^{-\gamma \tau} \sin(\omega_e \tau) \end{pmatrix} \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix}.$$

Now the asymptotic coarse–grained spontaneous emission is given by

$$\Gamma_{\text{spon,deph}} = \frac{1}{\tau} \int_{t_k}^{t_{k+1}} \frac{\Gamma}{2} (\rho_{\uparrow,\uparrow,\uparrow,\uparrow}(t) + \rho_{\uparrow,\downarrow,\downarrow,\uparrow}(t)) \, dt$$

$$= \frac{1}{\tau} \left( \frac{1 - e^{-\gamma \tau} \cos(\omega_e \tau)}{2 - e^{-\gamma \tau} \cos(\omega_e \tau)} \right)$$

$$\approx \frac{1}{\tau} \left( \frac{1 - \cos(\omega_e \tau)}{2 - \cos(\omega_e \tau)} + \gamma \tau \frac{\cos(\omega_e \tau)}{(2 - \cos(\omega_e \tau))^2} \right),$$

after a Taylor expansion in the small parameter $\gamma \tau$.

Having derived $\Gamma_{\text{spon,deph}}$ the nuclear spin flip rate $R_{\text{nuc,deph}} = \epsilon^2 \Gamma_{\text{spon,deph}}$ can be computed. In the limit $\gamma \tau \ll 1$, it is given by

$$R_{\text{nuc,deph}} = \epsilon^2 \frac{1}{\tau} \left( \frac{1 - \cos(\omega_e \tau)}{2 - \cos(\omega_e \tau)} + \gamma \tau \frac{\cos(\omega_e \tau)}{(2 - \cos(\omega_e \tau))^2} \right)$$

$$\approx \epsilon^2 \left( \gamma + \frac{\tau}{2} \left( 1 - \frac{3}{2} \gamma \tau \right) \left( \omega_e - \frac{2\pi}{\tau} \right)^2 \right),$$

for electron precession frequencies close to the synchronization condition. Pure dephasing thus adds a constant offset first to the spontaneous emission rate as well as to the nuclear spin flip rate, which limits the observable optical waiting and nuclear trapping times to $\Gamma_0^{-1} = \gamma^{-1}$ and $R_0^{-1} = (\epsilon^2 \gamma)^{-1}$, respectively. However, except for this constant offset, the modified nuclear spin flip rate is still quadratic near the synchronization condition, such that in the regime where a power–law behavior of the nuclear trapping time is expected, the exponent remains unaltered.

4.7.4 Imperfect laser polarization

To estimate the effect of imperfect laser polarization, we go back to the treatment of the system without nuclear spins. We furthermore assume that the $\sigma_+$-polarized transition is driven with a Rabi frequency $\Omega$, while the $\sigma_-$-polarized transition,
4.7 Limitations of the scheme

Figure 4.4: The excited state population after the pulse as a function of $\omega_e$ shows that taking into account the imperfect laser polarization is most important in the vicinity of a synchronization condition. i.e. $|\downarrow\rangle \leftrightarrow |\uparrow\downarrow\rangle$ is coupled with a Rabi frequency $\epsilon_{\text{pol}} \Omega$. Clearly, the state $|\downarrow\rangle$ is not completely dark anymore. Then the populations in the excited states after the pulse are given by

$$\rho^{f,k}_{\uparrow\downarrow \uparrow\uparrow} = \rho^{i,k}_{\uparrow\uparrow} \sin^2(\theta/2),$$
$$\rho^{f,k}_{\uparrow\downarrow \downarrow\downarrow} = \rho^{i,k}_{\downarrow\downarrow} \sin^2(\epsilon_{\text{pol}} \theta/2).$$

Assuming $\theta = \pi$, we numerically compute the asymptotic population in the excited states directly after the action of the 100th pulse, i.e. $\rho_{l,t}^{f,*} = \rho_{l,t}^{i,*} + \rho_{l,t}^{i,*}$ for various $\omega_e$ (see Fig. 4.4). Clearly, precession frequencies $\omega_e$ in the vicinity of a synchronization condition are particularly sensitive to imperfect laser polarization. In Fig. 4.5 we therefore plot the excited state population at the end of the action of a pulse at the synchronization condition for different $\epsilon_{\text{pol}}$.

The imperfect laser polarization introduces a rate that can be modeled as a constant offset $\Gamma_0$. This is given by

$$\Gamma_0 = \frac{1}{\tau} \left( \rho^{f,*}_{\uparrow\uparrow \uparrow\uparrow} + \rho^{f,*}_{\uparrow\downarrow \downarrow\downarrow} \right)_{\omega_{\text{eff}} = m \frac{2\pi}{\tau}}.$$

For the constant offset in the nuclear rate, we find $R_0 = \epsilon^2 \Gamma_0$. In order to observe a power–law regime before the exponential cut–off the condition $R_0 \ll R_{\text{nucl}} |\omega_{\text{eff}} - m \frac{2\pi}{\tau}| = A \approx \frac{1}{2} \epsilon^2 A^2 \tau$ needs to be satisfied. This yields

$$\rho^{f,*}_{\uparrow\uparrow \uparrow\uparrow} + \rho^{f,*}_{\uparrow\downarrow \downarrow\downarrow} \ll \frac{1}{2} (A \tau)^2.$$
4 Real–time monitoring of Lévy flights

Figure 4.5: The excited state polarization at the synchronization condition rapidly goes to zero as $\epsilon_{\text{pol}}$ approaches zero. The log–log plot resolves the behavior at small $\epsilon_{\text{pol}}$. In particular, for $\epsilon_{\text{pol}} \leq 10^{-2}$, the excited state population is negligibly small $\rho_{\text{f},\text{t}} \leq 5 \cdot 10^{-4}$.

For the above simulations $\tau = 39\Gamma^{-1}$ was used, but for a conservative estimate $\tau = 10\Gamma^{-1}$, which leads to $\rho_{\text{f},\text{t},\text{t}} + \rho_{\text{f},\text{t},\text{t}} \ll 5 \cdot 10^{-3}$. The simulations shown in Fig. 4.5 indicate that this condition is satisfied for $\epsilon_{\text{pol}} \leq 10^{-2}$.

4.7.5 Intrinsic limitation

In the time window between two pulses, the quadrupolar interaction leads to a coherent rotation between nuclear spin states of different $\langle I_z \rangle$. The frequency of the coherent rotation is determined by the coupling strength $\omega_Q$ as well as the energy detuning $\sim \omega_n$. Since the nuclear Zeeman detuning $\omega_n$ dominates over the coupling strength, the frequency of the coherent rotation is predominantly given by $\omega_n$. The intrinsic rate $R_0$ is determined by the probability amplitude of being in a nuclear spin state different from the one at the $k$th pulse at the arrival of the $(k+1)$th pulse. Therefore, the goal of minimizing $R_0$ can be achieved if the nuclear spins rotate by an angle that is a multiple of $2\pi$ in the time $\tau$. Thus $R_0$ is minimized for synchronized nuclear spins, i.e.

$$\omega_n^k \in \frac{2\pi}{\tau} \mathbb{Z}.$$  \hspace{1cm} (4.37)

To achieve nuclear synchronization, one can tune the magnetic field $B$ which changes $\omega_n^k$. Additionally, the pulse repetition period $\tau$ can be adjusted to improve the synchronization of the nuclear spins.

4.7.6 Displaced echo sequence

In self–assembled QDs, it is difficult to synchronize multiple nuclear spin species with different Larmor precession frequencies for a given range of magnetic fields $B$ and pulse repetition periods $\tau$. Especially $\tau$ cannot be extended too far without affecting the electron spin dynamics: the average number of nuclear spin flips
4.7 Limitations of the scheme

The echo pulse arrives at $t_{\text{echo},k}$, i.e. a time $\tau_{\text{echo}}$ after $t_k$. It is displaced from $t_k + \tau/2$ by $\Delta t_{\text{echo}}$. Therefore, the evolution that is not reversed by the displaced echo pulse occurs on the time scale of $2\Delta t_{\text{echo}}$.

between two traps is inversely proportional to $\tau$. This constraint can be lifted by applying an echo pulse on the electron. It has to be displaced in time from $\tau/2$, such that the electron dynamics that gives rise to $R_{\text{nuc}}$ - and thus the Lévy flights - is not completely reversed. A displaced echo pulse at

$$t_{\text{echo},k} = t_k + \frac{\tau}{2} - \Delta t_{\text{echo}}$$

defines a new synchronization condition

$$\omega_e \in \frac{\pi}{\Delta t_{\text{echo}}} \mathbb{Z}$$

for the electron. Therefore, $\tau$ can now be extended to optimize the nuclear synchronization, thereby minimizing $R_0$, without affecting the spacing between different trapping configurations.

In the following we outline how the new electron synchronization condition (4.38) in the presence of a displaced echo pulse can be derived. We assume that the duration of the echo pulse is short enough to include only the coherent coupling to the laser and neglect all other interactions during the pulse. Furthermore, the population of the excited states is assumed to have decayed to the ground states by the time the echo pulse arrives, i.e. $\Gamma^{-1} \ll \tau_{\text{echo}}$. We will therefore restrict the discussion to the ground state manifold and further assume for brevity that the excitation pulse is a $\theta = \pi$-pulse.

The derivation contains three stages. First, we calculate the electron ground state density matrix just before the arrival of the displaced echo pulse at $t_{\text{echo},k} = t_k + \tau_{\text{echo}}$ as a function of $\rho^{i,k}$. Then we determine the effect of the echo pulse. Lastly, this electron density matrix gives the initial conditions for the evolution.

Figure 4.6: The echo pulse arrives at $t_{\text{echo},k}$, i.e. a time $\tau_{\text{echo}}$ after $t_k$. It is displaced from $t_k + \tau/2$ by $\Delta t_{\text{echo}}$. Therefore, the evolution that is not reversed by the displaced echo pulse occurs on the time scale of $2\Delta t_{\text{echo}}$. 

$$\omega_e \in \frac{\pi}{\Delta t_{\text{echo}}} \mathbb{Z}$$
4 Real–time monitoring of Lévy flights

up to $t_{k+1}$ when the next excitation pulse arrives. We denote the density matrix just before the arrival of the echo pulse by $\rho_{t_{\text{echo}},k}$ and just after the action of the echo pulse by $\rho_{t_{\text{echo}},k}$.

To find $\rho_{t_{\text{echo}},k}$, we replace $\tau \rightarrow \tau_{\text{echo}} = \frac{\tau}{2} - \Delta t_{\text{echo}}$ in (4.17)–(4.19):

$$
\rho_{t_{\text{echo}},k}^{\uparrow,\uparrow} \approx \frac{1}{2} \left[ 1 - (1 - \rho_{t_{\text{echo}},k}^{i,\uparrow}) \cos(\omega_e \tau_{\text{echo}}) \right],
$$

$$
\rho_{t_{\text{echo}},k}^{\downarrow,\downarrow} \approx \frac{1}{2} \left[ 1 + (1 - \rho_{t_{\text{echo}},k}^{i,\downarrow}) \cos(\omega_e \tau_{\text{echo}}) \right],
$$

$$
\rho_{t_{\text{echo}},k}^{\downarrow,\uparrow} \approx \frac{i}{2} (1 - \rho_{t_{\text{echo}},k}^{i,\downarrow}) \sin(\omega_e \tau_{\text{echo}}).
$$

Visually, the electron precesses in the plane of the equator of the Bloch sphere between $|\uparrow\rangle$ and $|\downarrow\rangle$. Then the echo pulse rotates the state around the $x$-axis by an angle $\pi$ [110]. We thus find

$$
\rho_{t_{\text{echo}},k}^{f} = e^{-i\frac{\pi}{2} \hat{x}} \rho_{t_{\text{echo}},k}^{i} e^{i\frac{\pi}{2} \hat{x}},
$$

where we have used

$$
\hat{x} = |\uparrow\rangle \langle \downarrow| + |\downarrow\rangle \langle \uparrow| = |\uparrow\rangle \langle \uparrow| - |\downarrow\rangle \langle \downarrow| \langle \downarrow| = |\uparrow\rangle \langle \uparrow| - |\downarrow\rangle \langle \downarrow|.
$$

The final step involves the evolution from $t_{\text{echo},k}$ to $t_{k+1}$ according to the coupled equations

$$
\frac{\partial}{\partial t} \begin{pmatrix}
\rho_{t_{\text{echo}},k}^{\uparrow,\uparrow} - \rho_{t_{\text{echo}},k}^{\downarrow,\downarrow} \\
\rho_{t_{\text{echo}},k}^{\downarrow,\uparrow} - \rho_{t_{\text{echo}},k}^{\uparrow,\downarrow}
\end{pmatrix} = \begin{pmatrix}
i\omega_e & 0 \\
0 & i\omega_e
\end{pmatrix} \begin{pmatrix}
\rho_{t_{\text{echo}},k}^{\uparrow,\uparrow} - \rho_{t_{\text{echo}},k}^{\downarrow,\downarrow} \\
\rho_{t_{\text{echo}},k}^{\downarrow,\uparrow} - \rho_{t_{\text{echo}},k}^{\uparrow,\downarrow}
\end{pmatrix},
$$

with the initial conditions given by $\rho_{t_{\text{echo}},k}$, i.e.

$$
\rho_{t_{\text{echo}},k}^{f} = \rho_{t_{\text{echo}},k}^{i,\uparrow} - \rho_{t_{\text{echo}},k}^{i,\downarrow} = -(1 - \rho_{t_{\text{echo}},k}^{i,\downarrow}) \cos(\omega_e \tau_{\text{echo}}),
$$

$$
\rho_{t_{\text{echo}},k}^{f} = \rho_{t_{\text{echo}},k}^{i,\downarrow} + \rho_{t_{\text{echo}},k}^{i,\uparrow} = +i(1 - \rho_{t_{\text{echo}},k}^{i,\downarrow}) \sin(\omega_e \tau_{\text{echo}}).
$$

This leads to

$$
\rho_{t_{\text{echo}},k}^{i,\uparrow} - \rho_{t_{\text{echo}},k}^{i,\downarrow} = \rho_{t_{\text{echo}},k}^{f,\downarrow} \cos(\omega_e 2\Delta t_{\text{echo}}).
$$

In the asymptotic limit this yields the bright state population

$$
\rho_{t_{\text{echo}},k}^{f,\uparrow} = \frac{\sin^2(\omega_e \Delta t_{\text{echo}})}{1 + \sin^2(\omega_e \Delta t_{\text{echo}})}.
$$

Clearly, the electron now obeys the modified synchronization condition

$$
\omega_e \in \frac{\pi}{\Delta t_{\text{echo}}}, \mathbb{Z},
$$

which is exactly (4.38). Hence, we showed that by adding a displaced echo pulse, the synchronization condition for the electron is now determined by $\Delta t_{\text{echo}}$ rather than $\tau$. If the intrinsic limitation can be reduced by synchronizing the nuclear precession frequencies with the pulse repetition period $\tau$, the displaced echo pulse lifts restrictions on $\tau$ that occur due to electron synchronization. This applies for example if quadrupolar interactions are dominant; therefore, the displaced echo pulse can be used to extend $\tau$ and improve the nuclear synchronization of multiple nuclear species. The displaced echo pulse thus allows for further reduction of $R_0$, which is beneficial for the observation of the power–law tail that characterizes the Lévy flights.
4.8 Conclusion and outlook

In summary, we have demonstrated that the polarization of the spin environment of a central spin system that is driven by a resonant periodic pulse train exhibits Lévy flights. Our calculations indicate that the signatures of these Lévy flights can be observed by measuring the waiting time distribution of the emitted photons. This results from the decoupling of the central spin and the environment, as well as the proportionality of the environmental–spin–flip and spontaneous emission rates. While we illustrated the scheme with self-assembled quantum dots, we expect our findings to be relevant for a broad class of solid-state emitters including nitrogen–vacancy centers. More generally, our results show that the previously established connection between Lévy flights and dark states [16, 89] can be extended to systems with explicit time–dependence.

The power–law of the waiting time distribution between successive photon emission events is remarkable from a further perspective: It usually obeys an exponential decay law. In particular, the waiting time between successive photon emission events was used to describe the expected quantum jumps of an atomic electron, when in an atomic three–level system one transition is strongly driven while the weakly driven transitions connects to a metastable state [200]. When the photon emission on the strongly driven transition is recorded, one finds dark periods that correspond to the time intervals during which the electron is shelved in the metastable state. It was shown that the waiting time distribution contains a fast and a slow exponential decay [201]. The fast decay is associated with the photon detection events when the electron cycles on the strong transition. On the other hand, the slow decay is associated with the shelving in the metastable state and is responsible for the dark periods [201, 202]. A recent experimental realization of this idea in a solid–state spin qubit found that the long waiting times were rooted in two consecutive spin flips between the photon detection events and are governed by a slow exponential decay [203]. However, here we found a striking departure from a (bi)exponential waiting time distribution: The long waiting times, associated with electron synchronization, are distributed according to a power–law.
Signatures of a dissipative phase transition controlled by a single quantum system

We theoretically investigate the dissipative dynamics of an optically illuminated single confined spin-$\frac{1}{2}$ particle in a coherent population trapping (CPT) configuration. This central spin interacts with a spin environment, a situation that frequently occurs in solid-state emitters. The dynamics of this driven and open central spin system forces the environment into a steady state that depends on the relative strength of the two driving laser Rabi frequencies. When the relative strength of the Rabi frequencies is tuned accordingly, the spin environment exhibits physical properties that are reminiscent of a system undergoing a continuous dissipative phase transition. Remarkably, the transition is controlled by the central spin, i.e. a single quantum system. For illustration, we apply the analysis to a single electron charged quantum dot (QD). We discuss not only how the central spin controls the phase of the spin environment, but also the feedback of the nuclear spin environment on the quantum dot electron and the ensuing optical signatures. However, the control of the electron over the nuclear spin environment diminishes when the spin environment is enlarged in the thermodynamic limit. This is a consequence of the asymmetric coupling between the single (central) electron spin and the large nuclear spin environment.

5.1 Introduction

Recently, several schemes have been proposed to observe dissipative phase transitions in various systems. A wealth of proposals have been put forward for atomic systems [60, 61, 62, 63, 64, 65] and the the Dicke phase transition has been experimentally realized in a dissipative setting, where a Bose-Einstein condensate was coupled to an optical cavity [57, 58, 59]. However, only a few schemes for dissipative phase transitions in solid-state systems have been proposed so far [66, 67, 68, 69], and their experimental realization remains challenging. This motivates us to study the dynamics of a driven and dissipative central spin embedded in a solid-state spin environment. In particular, we focus on a single QD, an interesting realization of a dissipative central spin model, as described in detail in chapter 2. Furthermore, a single QD is an experimentally relevant, accessible, tunable, and extensively studied system.

In self-assembled QDs, signatures of an abrupt phase transition in the nuclear spin environment were found as a function of the strength of the transverse mag-
Signatures of a dissipative phase transition

This raises the question whether features reminiscent of a continuous phase transition could be observed in this system. Moreover, experiments on dragging effects in these QDs showed that the nuclear spin environment can be modified when scanning the laser detuning of an optical transition [170]: Depending on the optical transition that is selected, the nuclear spins either polarize such as to maintain (dragging) or avoid (antidragging) the resonance [175]. The ensuing spectroscopic signatures could be explained as arising from optically-assisted nuclear spin flips due to the non-collinear hyperfine interaction [175, 135], which is introduced in section 2.4.2. For a single electron charged QD, the spin of the ground state determines whether dragging or antidragging effects occur: The Knight shift (see section 2.4) alters the energy of the two spin states differently. The resulting modification of the laser detuning of the two transition causes optical resonance to be either a stable or an unstable state of the coupled electron-nuclei system. If the two ground states are simultaneously optically coupled to a common excited state in a CPT setting, as described in section 2.2.2, one could expect a competition between dragging and antidragging effects. Moreover, tuning the relative strength of the two laser Rabi frequencies correspondingly, one might suspect a continuous transition from a dragging to an antidragging phase. Even though the configuration we discuss is strongly motivated by self-assembled QDs, the scheme can be applied to a more general system. Therefore, we formulate the problem for a general dissipative central spin interacting with a spin environment, but use the subscript “e” to denote the central (electron) spin and “n” for the (nuclear) spin environment. The description can be used to investigate the signatures of criticality in a spin environment that is controlled by a single driven and dissipative quantum system.

This chapter is organized as follows: Section 5.2 introduces the master equation that governs the coupled system-environment dynamics and constitutes the starting point for the derivation of a purely environmental master equation in section 5.3. This tractable master equation for the environment is obtained by employing a hierarchy of time scales in the system; in particular the distinction between the fast evolution of the central system and the slow dynamics of the spin environment. This allows us to capture the relevant dynamics with a reduced set of variables. Then, in section 5.4, we discuss the signatures that are reminiscent of a continuous dissipative phase transition in the spin environment when the analysis is applied to QDs. Finally, in section 5.5, we conclude by summarizing our results.

5.2 System

We consider the dynamics of a single confined spin-$\frac{1}{2}$ particle and the $N$ spins in the environment to be captured by the Markovian master equation for the system-environment density matrix $\rho$ [128, 187, 188], i.e.

$$\dot{\rho} = \mathcal{L}(\rho) = \mathcal{V}(\rho) + \mathcal{L}_c(\rho), \quad (5.1)$$

where $\mathcal{L}$ is the Liouvillian superoperator. The Lindblad superoperator $\mathcal{L}_c$ is defined by (2.13) and describes the effect of spontaneous emission on the density matrix as discussed in section 2.2.5. We have also defined a superoperator that
5.2 System

Figure 5.1: The probe and coupling lasers drive the transitions from the Zeeman-split ground states to a common excited state $|t\rangle$.

does not contain the coherent dynamics

$$\mathcal{V}_\rho = -i[H, \rho],$$

with the Hamiltonian

$$H = H_e + H_n + H_{int},$$

where

$$H_e = +r \sin \phi (|\uparrow\rangle \langle t| + |t\rangle \langle \uparrow|) + r \cos \phi (|\downarrow\rangle \langle t| + |t\rangle \langle \downarrow|)$$

$$H_n = -\omega_n I_z$$

$$H_{int} = A_z S_z I_z + A_x S_z I_x.$$ 

Here, we assumed (bare) one- and two-photon resonance, i.e. $\Delta \omega_p = \Delta \omega_c = \delta_0 = 0$ and introduced the convenient parameterization of the laser Rabi frequencies

$$\Omega_p = r \sin \phi \quad r = \sqrt{\Omega^2_p + \Omega^2_c}$$

$$\Omega_c = r \cos \phi \quad \phi = \tan^{-1} \left( \frac{\Omega_p}{\Omega_c} \right),$$

such that the relative strength is controlled by a single parameter ($\phi$). The configuration of the central spin is depicted in Fig. 5.1. Furthermore, we assumed homogeneous Zeeman splittings $\omega_n$ in the spin environment and homogeneous system-environment couplings $A_z$ and $A_x$. The interaction Hamiltonian is motivated by the dominant role the Overhauser term (section 2.4) and the non-collinear hyperfine interaction (section 2.4.2) play in QDs. We employ collective operators for the environmental spins (section 2.3.4).
5 Signatures of a dissipative phase transition

Since all interactions that enter (5.1) conserve the total angular momentum \( J \), it is convenient to describe the system in the Dicke eigenbasis \( |J,m\rangle \) (section 2.3.4), where \( m \) is the spin projection along the \( z \)-direction. Given that a certain \( J \)-subspaces occurs with a probability \( p(J) \), we can compute the dynamics of the system by analyzing the evolution in a fixed \( J \)-subspace. Finally, we sum over all \( J \)-subspaces. In the following, the total spin polarization of the environment is of interest, which is characterized by the corresponding probability distribution

\[
p(m) = \sum_J p(m|J)p(J),
\]

where

\[
p(m|J) = \langle m| \rho^e_J |m\rangle,
\]

and we recall from section 2.3.4 that

\[
p(J) \approx \frac{8J^2/N}{\sqrt{2\pi\sigma}} e^{-\frac{J^2}{2\sigma^2}},
\]

where \( \sigma = \sqrt{N/4} \).

5.3 Master equation for the spin environment

The master equation (5.1) describes the interplay between the coherent and dissipative evolution of the coupled system. To analyze the dynamics it causes, we make use of the separation of time scales in the system. To be more specific, the central system spin evolves on a much faster time scale compared to the spin environment. This allows us to simplify the master equation and finally arrive at a purely dissipative master equation for the spin environment only. This analysis is based on the fact that the highest rate in the system described by (5.1) is the spontaneous emission rate \( \Gamma \). The second largest energy scale is given by the Zeeman energy of the environment \( \omega_n \), followed by the smaller effective Rabi frequency \( r = \sqrt{\Omega^2_p + \Omega^2_c} \) and the system-environment coupling \( A_z \). Finally, \( A_x \) constitutes the smallest energy scale in this system and is only kept to second order in perturbation theory. To summarize, the separation of energy scales is

\[
\Gamma \gg \omega_n \gg r, A_z \gg A_x.
\]

This is utilized to first adiabatically eliminate the excited state \( |t\rangle \) of the central system and thereby reduce the central system from a three-level to an effective two-level system. Then, the large environmental Zeeman energy \( \omega_n \) favors Fock-like environmental spin states, which enables us to eliminate coherences between environmental spin states of different \( \langle I_z \rangle \). This drastically reduces the dimension of the Hilbert space of the relevant states. Finally, tracing over the degrees of freedom of the central spin yields a purely dissipative master equation for the spin environment.

5.3.1 Adiabatic elimination of the excited central system state

The separation of energy scales (5.6) and more specifically

\[
\Gamma \gg r
\]

This is utilized to first adiabatically eliminate the excited state \( |t\rangle \) of the central system and thereby reduce the central system from a three-level to an effective two-level system. Then, the large environmental Zeeman energy \( \omega_n \) favors Fock-like environmental spin states, which enables us to eliminate coherences between environmental spin states of different \( \langle I_z \rangle \). This drastically reduces the dimension of the Hilbert space of the relevant states. Finally, tracing over the degrees of freedom of the central spin yields a purely dissipative master equation for the spin environment.
implies that the excited state population and the coherences between the ground states and the excited state $|t\rangle$, which constitute the fast subspace, remain small because they decay on the fast time scale $\sim \Gamma^{-1}$. Therefore, it is justified to describe the dynamics by an effective two-level system consisting of the two central spin ground states $|↑\rangle$ and $|↓\rangle$, which we refer to as the slow central spin subspace. This is accomplished by projecting the master equation (5.1) onto these ground states using the projection superoperators $P_e$ and $Q_e$ which act on an arbitrary operator $O$ according to [188]

$$
P_e(O) = P_e O P_e \tag{5.8}
$$
$$
Q_e(O) = O - P_e(O), \tag{5.9}
$$

with the projection operator $P_e = |↑\rangle \langle ↑| + |↓\rangle \langle ↓|$ that projects onto the slow subspace. Applying the projection superoperators on (5.1) and using $P_e + Q_e = 1$ yields the exact equations

$$
\dot{P}_e(\rho) = P_e \mathcal{L} P_e(\rho) + P_e \mathcal{L} Q_e(\rho) \tag{5.10}
$$
$$
\dot{Q}_e(\rho) = Q_e \mathcal{L} P_e(\rho) + Q_e \mathcal{L} Q_e(\rho), \tag{5.11}
$$

where explicitly $P_e \mathcal{L} P_e(\rho) = P_e \left( \mathcal{L}(P_e(\rho)) \right)$. The first approximation that we introduce is based on the separation of time scales (5.7) and consists of assuming that the fast subspace $Q_e(\rho)$ reaches its steady state quickly compared to the slow subspace $P_e(\rho)$. Therefore, we solve (5.11) in steady state by setting $\dot{Q}_e(\rho) = 0$ and plug the solution

$$
Q_e^{ss}(\rho) \overset{Q_e}{=} Q_e^{ss} - (Q_e \mathcal{L} Q_e)^{-1} Q_e \mathcal{L} P_e(\rho) \tag{5.12}
$$

into (5.10). Note that (5.12) describes the steady state of the fast subspace density matrix elements as a function of the slow subspace $P_e(\rho)$. Using (5.12) we can now rewrite (5.10) as a function of the slow variables only:

$$
\dot{P}_e(\rho) = P_e \mathcal{L} P_e(\rho) - P_e \mathcal{L} Q_e(Q_e \mathcal{L} Q_e)^{-1} Q_e \mathcal{L} P_e(\rho). \tag{5.13}
$$

While (5.13) describes the effective dynamics in the slow subspace, it contains the term $(Q_e \mathcal{L} Q_e)^{-1}$, which can be difficult to compute. We therefore once again use the hierarchy of energy scales (5.7) to derive a meaningful expansion of this term. Since the Lindblad superoperator $\mathcal{L}_c$ is the only term that contains the spontaneous emission rate $\Gamma$, it dominates over $\mathcal{V}$. Therefore we can use the Taylor expansion

$$(Q_e \mathcal{L} Q_e)^{-1} = (Q_e \mathcal{L}_c Q_e)^{-1} - (Q_e \mathcal{L}_c Q_e)^{-1}(Q_e \mathcal{V} Q_e)(Q_e \mathcal{L}_c Q_e)^{-1} + (Q_e \mathcal{L}_c Q_e)^{-1}(Q_e \mathcal{V} Q_e)(Q_e \mathcal{L}_c Q_e)^{-1}(Q_e \mathcal{V} Q_e)(Q_e \mathcal{L}_c Q_e)^{-1} + \ldots$$

The advantage of this expansion is based on the simple structure of $(Q_e \mathcal{L}_c Q_e)$, which allows us to invert this term more easily compared to $(Q_e \mathcal{L} Q_e)$. Before using the expansion in (5.13), we note that $P_e \mathcal{L} Q_e = P_e \mathcal{V} Q_e + P_e \mathcal{L}_c Q_e$. However, $Q_e \mathcal{L} P_e = Q_e \mathcal{V} P_e$ since spontaneous emission only leads to a relaxation of the fast
variables to the slow variables and thus \( P_e \mathcal{L}_c Q_e \neq 0 \) while \( Q_e \mathcal{L}_c P_e = 0 \). For similar reasons \( P_e \mathcal{L}_c P_e = 0 \). Finally, we find the three lowest orders

\[
\dot{P}_e(\rho) = \dot{\mathcal{L}} \rho = P_e \mathcal{V} P_e(\rho) \\
+ \left( - (P_e \mathcal{V} Q_e)(Q_e \mathcal{L}_c Q_e)^{-1}(Q_e \mathcal{V} P_e(\rho)) \right) \\
+ (P_e \mathcal{L}_c Q_e)(Q_e \mathcal{L}_c Q_e)^{-1}(Q_e \mathcal{V} Q_e)(Q_e \mathcal{L}_c Q_e)^{-1}(Q_e \mathcal{V} P_e(\rho)) \\
+ \left( (P_e \mathcal{V} Q_e)(Q_e \mathcal{L}_c Q_e)^{-1}(Q_e \mathcal{V} Q_e)(Q_e \mathcal{L}_c Q_e)^{-1}(Q_e \mathcal{V} P_e(\rho)) \right) \\
+ \ldots
\]

(5.14)

Here, we have defined the new Liouvillian superoperator \( \dot{\mathcal{L}} \), which describes the effective dynamics in the slow subspace.

Applying (5.14) to the master equation (5.1) we find that adiabatic elimination of the excited state leads to new decoherence processes that are characterized by the operators

\[
C_D = \sqrt{\frac{\bar{\Gamma}}{2}} |D\rangle \langle B| \\
C_B = \sqrt{\frac{\bar{\Gamma}}{2}} |B\rangle \langle B|,
\]

where the decay rate \( \bar{\Gamma} = 4r^2/\Gamma \) and the dark and bright basis states are given by

\[
|D\rangle = \cos(\phi) |\uparrow\rangle - \sin(\phi) |\downarrow\rangle, \\
|B\rangle = \sin(\phi) |\uparrow\rangle + \cos(\phi) |\downarrow\rangle.
\]

5.3.2 Adiabatic elimination of environmental spin coherences

After having eliminated the excited state of the central system, we turn to the spin environment. Since \( J \) is conserved by all the interactions in (5.3), we consider a fixed \( J \)-subspace that occurs with probability \( p(J) \). Then we use the second largest energy scale \( \omega_n \) to eliminate the coherences between states \( |J,m\rangle \) and \( |J,m'\rangle \) with different total spin projection \( m \neq m' \). To this end, we define the superoperators

\[
\mathcal{P}_n^J(O) = \sum_{m=-J}^{+J} |J,m\rangle \langle J,m| O |J,m\rangle \langle J,m| \\
\mathcal{Q}_n^J(O) = \sum_{m=-J}^{+J} \sum_{m' \neq m'} |J,m\rangle \langle J,m| O |J,m'\rangle \langle J,m'|. 
\]

Then we apply the equivalent of (5.13) to Liouvillian operator \( \dot{\mathcal{L}} \) that describes the effective dynamics in the slow electronic subspace. This yields

\[
\dot{\mathcal{P}}_n^J(\rho) = \mathcal{P}_n^J \dot{\mathcal{L}} \mathcal{P}_n^J(\rho) - \mathcal{P}_n^J \dot{\mathcal{Q}}_n^J(\mathcal{Q}_n^J \dot{\mathcal{Q}}_n^J)^{-1} \mathcal{Q}_n^J \dot{\mathcal{Q}}_n^J(\rho). 
\]

(5.15)
For the expansion of \((Q^J_n \hat{L} Q^J_n)^{-1}\), we partition \(\hat{L}\rho\) into a large environmental Zeeman superoperator \(\mathcal{V}_n\) and the small remainder \(\hat{L}_r\), i.e.

\[
\hat{L}\rho = -i[H_n, \rho] + \left(\hat{L}\rho + i[H_n, \rho]\right). 
\]

The only interaction that takes the system from the slow to the fast environmental subspace and vice versa is the second term of the system-environment Hamiltonian, i.e. \(A_n S L_x\). Therefore, the nuclear Zeeman interaction yields \(P^J_n \mathcal{V}_n Q^J_n = Q^J_n \mathcal{V}_n P^J_n = 0\). This simplifies the correct grouping of the terms by order in the expansion compared to (5.14) and yields the perturbative equation in the slow nuclear subspace

\[
P^J_n(\rho) = P^J_n \hat{L} P^J_n(\rho) - (P^J_n \hat{L}_r Q^J_n)(Q^J_n \mathcal{V}_n Q^J_n)^{-1}(Q^J_n \hat{L}_r Q^J_n)^{-1}(Q^J_n \hat{L}_r P^J_n(\rho)) + (P^J_n \hat{L}_r Q^J_n)(Q^J_n \mathcal{V}_n Q^J_n)^{-1}(Q^J_n \hat{L}_r Q^J_n)(Q^J_n \mathcal{V}_n Q^J_n)^{-1}(Q^J_n \hat{L}_r P^J_n(\rho)) + \ldots
\]

### 5.3.3 Tracing over the degrees of freedom of the central spin

To extract the dynamics of the spin environment, we trace over the degrees of freedom of the central spin, which yields the purely dissipative environmental master equation for a fixed \(J\)-subspace

\[
\dot{\rho}_n^J = \mathcal{L}_n^J(\rho_n) = \sum_m C_{+}^{J,m} \rho_n^J(C_{+}^{J,m})^\dagger - \frac{1}{2}\{(C_{+}^{J,m})^\dagger C_{+}^{J,m}, \rho_n^J\} + \sum_m C_{-}^{J,m} \rho_n^J(C_{-}^{J,m})^\dagger - \frac{1}{2}\{(C_{-}^{J,m})^\dagger C_{-}^{J,m}, \rho_n^J\}.
\]

The nuclear collapse operators are given by

\[
C_{+}^{J,m} = \sqrt{\Gamma_{+}^{J,m}} |J, m+1\rangle \langle J, m|,
\]

where

\[
\Gamma_{+}^{J,m} = \frac{A^2 \Gamma^3 \sin(2\phi)^2 (\eta_{+}^{J,m})^2}{8r^2 \omega_n^2} \left(1 + m A_z r^2 (3 \Gamma^2 A_z - 4 \omega_n \cos(2\phi) \Gamma^2 + 8r^2 A_z) \right),
\]

(5.16) and

\[
\eta_{+}^{J,m} = \sqrt{J(J+1) - m(m+1)}.
\]

For the nuclear master equation to be a meaningful description of the dynamics, the correlations between the central spin and the spin environment should remain small. To gain further insight into the validity of tracing out the central spin, we consider the quantum mutual information \(I(\rho)\), which compares the correlated system-environment state \(\rho\) and the uncorrelated state \(\rho_c \otimes \rho_n\). The reduced density matrices are given by \(\rho_c = \text{Tr}_n(\rho)\) and \(\rho_n = \text{Tr}_c(\rho)\). The quantum mutual information is defined as \([205]\)

\[
I(\rho) = S(\rho_c) + S(\rho_n) - S(\rho),
\]

where

\[
S(\rho) = -\text{Tr}(\rho \log \rho).
\]
Figure 5.2: On the left, the von Neumann entropy of the correlated state $\rho$ (blue line) and the sum of the entropies of the electron and nuclear systems (red line) are shown. On the right, the quantum mutual information $I(\rho)$ is depicted. It corresponds to the difference of the quantities shown on the left and remains much smaller across a wide range of $\phi$. It reaches its maximum at the transition point $\phi_c$.

where $S = -\rho \ln \rho$ is the von Neumann entropy and for the uncorrelated state $S(\rho_e \otimes \rho_n) = S(\rho_e) + S(\rho_n)$ holds [187]. The quantum mutual information $I(\rho)$ can be expressed in terms of the quantum relative entropy between the states $\rho$ and $\rho_e \otimes \rho_n$, which quantifies the distinguishability of the two states. However, we note that it is not the relative entropy of entanglement that constitutes a measure of entanglement [205]. To compute the quantum mutual information $I(\rho)$, we start out with the Markovian master equation (5.1) for a single electron charged QD (see section 5.4), for which the steady state can be computed exactly in the case of very small $J$. We calculate the von Neumann entropies of the correlated, and reduced density matrices for $J = 9$ over a range of $\phi$ and plot $I(\rho)$ (Fig. 5.2). If $\rho$ is uncorrelated such that $\rho = \rho_e \otimes \rho_n$, the quantum mutual information $I(\rho)$ vanishes. We find that $I(\rho)$ and equivalently, the relative entropy, is much smaller than the von Neumann entropy of the uncorrelated state. This suggests that the correlations remain small and tracing over the central system degrees of freedom yields a meaningful master equation for the spin environment. Furthermore, we note that the quantum mutual information attains its maximum at $\phi = \phi_c = \pi/4 = 45^\circ$, where we expect the system to undergo a transition (Fig. 5.2)

5.4 Signatures of criticality

The environmental spin master equation derived in the last section is the starting point for the analysis of criticality in a spin environment which is controlled by an open and driven central spin. In this section, we illustrate the consequences of this equation for a single electron charged QD, where the electron couples to the nuclear environment of the QD host material [135]. We first establish the cor-
respondence between the parameters in our description and the QD parameters. Then, we investigate the nuclear steady state as a function of the relative strength of the two laser Rabi frequencies. The relative driving strength is quantified by the parameter $\phi$ (5.4), with the critical value $\phi_c = \pi/4 = 45^{\circ}$ or equivalently, $\Omega_p = \Omega_c$. We characterizing the different phases by the dynamic nuclear polarization (DNP) of the steady state, where the Overhauser shift $A \langle I_z \rangle$ acts as the order parameter. Then we show that the DNP changes continuously at the critical point $\phi_c$ and therefore hints at a continuous transition. We also evolve the nuclear spin distribution across $\phi_c$, where we find increased fluctuations around the phase boundary.

We briefly discuss how the presence of multiple nuclear species affects the nuclear probability distribution. Then, we compute the low-excitation spectrum of the Liouvillian $\mathcal{L}_{\text{nuc}}$, which aims at detecting non-analyticities of the nuclear steady state. Subsequently, we discuss the issues that arise when taking the thermodynamic limit in this system. Finally, we compute the low-excitation spectrum in the thermodynamic limit for a system with constant hyperfine coupling constants, i.e. where $A_z$ and $A_x$ are independent of $N$.

### 5.4.1 Realization in quantum dots

We consider a self-assembled QD charged with a single electron (central) spin and the optically excited trion state $|t\rangle = |\uparrow \downarrow \uparrow \rangle$. The spin environment is assumed to be composed of the $N \approx 10^6$ nuclei. The interaction between the electron and nuclear spins is taken to be dominated by the Overhauser term and the non-collinear interactions (see sections 2.4 and 2.4.2), such that the hyperfine flip-flop terms can be neglected [175]. We assume that all nuclear spins couple equally to the electron, yielding $A_z = A = A_H/N$ and $A_x = A_{ne}$, where $A_H$ is the total hyperfine coupling constant of the material (see chapter 2). We use the typical values $A_z = 10^{-4}\Gamma$, $A_x = 7 \cdot 10^{-2}A_z$, $\omega_n = 0.5\Gamma$, and $r = 0.1\Gamma$ unless stated otherwise. We recall from chapter 2 that on the one hand the state of the nuclear environment modifies the two-photon laser detuning, i.e. $\delta_{\text{eff}} = \delta_0 + AI_z + A_{ne}I_x$. On the other hand, the non-collinear interaction $A_{nc} S_z I_x$ induces slow nuclear spin dynamics through optically-assisted nuclear spin flips.

### 5.4.2 Phase characterization by dynamic nuclear polarization

We start by characterizing the two phases by the nuclear polarization of the steady state. For $\phi < \phi_c$ ($\Omega_p < \Omega_c$) the nuclear steady state is characterized by a vanish-
5 Signatures of a dissipative phase transition

...ing average nuclear spin polarization, where the most likely total spin projection of the system is zero. However, as \( \phi \) is increased to \( \phi > \phi_c \) \( (\Omega_p > \Omega_c) \) a vanishing nuclear spin polarization becomes unstable. The nuclear spins become polarized and can be aligned parallel or anti-parallel to the external magnetic field \( \vec{B} \).

These observations can be illustrated with the analogy to dragging and antidragging: Initially, the lasers satisfy bare two-photon resonance, i.e. \( \delta_0 = 0 \). One could therefore associate the phase for which the two-photon resonance is stable, i.e. \( \delta_{\text{eff}} = 0 \), with dragging effects. Then, \( \delta_{\text{eff}} = \delta_0 = 0 \) implies that the nuclear spins remain unpolarized and the electron populates the dark state \( |D\rangle \). Since dragging occurs due to the \( |\uparrow\rangle \) spin ground state, one could expected the nuclear spins to remain unpolarized when the population is predominantly in the state \( |\uparrow\rangle \). To verify this, we calculate the overlap of the two spin states with the dark state \( |D\rangle \) i.e. \( \langle \uparrow | D \rangle = \cos(\phi) \) and \( \langle \downarrow | D \rangle = \sin(\phi) \). Clearly, when \( \cos(\phi) > \sin(\phi) \) or equivalently \( \phi < \phi_c \), the population in \( |\uparrow\rangle \) exceeds the population in \( |\downarrow\rangle \). Therefore, our results are consistent with the dragging and antidragging effects.

Experimentally, a vanishing nuclear spin polarization and hence Overhauser field means that the laser detunings are not modified. These therefore continue to satisfy the two-photon resonance condition in the presence of the nuclear spins and the CPT dip remains at \( \delta_0 = 0 \). This in turn implies that the absorption of the system is strongly suppressed since the electron remains in the dark state \( |D\rangle \) for \( \phi < \phi_c \). For \( \phi > \phi_c \) in contrast, the nuclear spins are in a mixed state of parallel or anti-parallel nuclear spin states. The electron thus sees a modified magnetic field which detunes the lasers from two-photon resonance. Since the absorption curve is symmetric with respect to the effective detuning, both the parallel and anti-parallel configurations lead to dip in the absorption curve at \( \delta_0 = \pm |\omega_{\text{OH}}| \). Hence, the two phases can also be distinguished by the location of the dip(s) in the optical absorption spectrum.

5.4.3 Continuous change in the dynamic nuclear polarization

One of the defining traits of a continuous phase transition is the continuous change of the order parameter at the phase boundary. In the system under investigation this corresponds to a continuous change of the Overhauser field at \( \phi = \phi_c \). To characterize the transition we look at the behavior of the stable solution(s) of \( \langle I_z \rangle \) as \( \phi \) is tuned across \( \phi_c \). To this end, we compute the zeros of \( \text{sgn} \left( \frac{d\langle I_z \rangle}{dt} \right) \) as a function of \( m \) and \( \phi \) for a fixed \( J \) subspace (Fig. 5.4). The solutions occur when the sign changes as a function of \( m \) for a given \( \phi \). They are stable if \( \frac{d\langle I_z \rangle}{dt} \) changes from positive to negative as \( m \) is varied across the solution from below. We numerically find that for \( \phi < \phi_c \) there is one stable solution at \( \langle I_z \rangle = 0 \). As \( \phi \) increases there is a bifurcation into two stable branches and an unstable solution at \( \langle I_z \rangle = 0 \).

Furthermore, we note that at the phase boundary, the nuclear polarization in a fixed \( J \)-subspace increases by the smallest possible quantity, which corresponds to a single nuclear spin flip (Fig. 5.4). The accompanying change in the Overhauser field is given by \( A = A_{\text{H}}/N \). However, as the number of nuclear spin is increased in the thermodynamic limit, the jump in the stable solutions of the Overhauser...
Figure 5.4: The sign of $\langle \hat{I}_z \rangle$ is plotted for $J = \sqrt{N/3}$ as a function of $m$ and $\phi$ in the immediate vicinity of its critical value. The solutions of $\langle I_z \rangle$ occur when the sign changes from positive (yellow) to negative (blue). Clearly, $\langle I_z \rangle = 0$ is stable for small $\phi$ and becomes unstable beyond the critical value of $\phi$. For larger $\phi$ two new stable solution emerge which are symmetric with respect to $m = 0$, i.e. the polarization points parallel or anti-parallel to the applied magnetic field. At the critical value the stable solution of the nuclear polarization only jumps by a single nuclear spin flip.

field tends to zero and the change of the stable solutions of the Overhauser field become continuous.

### 5.4.4 Evolution of the nuclear distribution

An additional signature of continuous phase transitions is an increase in the fluctuations as the critical point is approached. We therefore compute the steady state nuclear density matrix and plot the probability of the nuclear polarization $m$ given by (5.5) as a function of $\phi$. We note that due to limited computational power, we truncate the probability distribution at $J_{\text{max}} = 3\sqrt{N}/2$ which captures more than 95% of the total probability. We do not normalize the probability distribution, but plot the truncated probability

\[
p(m) = \sum_{J=0}^{N/2} p(m|J)p(J) \approx \sum_{J=0}^{J_{\text{max}}} p(m|J)p(J).
\]

Below the critical point ($\phi < \phi_c$) the distribution is peaked around zero nuclear spin polarization, which is in accordance with the stable solution presented in Fig. 5.4. As $\phi \rightarrow \phi_c$ from below, the distribution becomes broader, indicating
increased fluctuations (Fig. 5.7). Above the phase boundary, the distribution develops two peaks which correspond to the two polarized nuclear spins states directed parallel and anti-parallel to the external magnetic field (Fig. 5.8).

The width of the distribution affects the $T_2^*$ time of the electron spin qubit [149, 148] (see section 2.5), which is imprinted on the depth of the CPT dip [158]. One could attempt to extract this time scale by applying the following procedure. First, the Rabi frequencies ($\phi$) are adjusted. After waiting for the system to find its steady state distribution, a fast CPT scan is performed. Repeating this procedure and averaging the results could allow for the extraction of $T_2^*$ as a function of $\phi$. We indeed find that the CPT dip becomes less pronounced as the critical point is approached from below (Fig. 5.7). Above the critical point there are two symmetric dips (Fig. 5.8). The dips for $\phi > \phi_c$ are smaller as a consequence of a broad double-peaked probability distribution (Fig. 5.6).

### 5.4.5 Multiple nuclear species

We briefly discuss the effect of multiple nuclear species on the scheme, in particular the modification of the maximum Overhauser shift. We assume that the hyperfine coupling constant of each species is homogeneous such that $J$ is conserved for each species separately [206]. We compute the probability distribution $p(\omega_{\text{Oh}})$ of the Overhauser shift $\omega_{\text{Oh}} = \sum_j A_j \langle I_j^z \rangle$. Since we are predominantly interested in the
Figure 5.6: The unnormalized probability distribution for $\phi > \phi_c$ bifurcates and the two peaks of the distribution correspond to a polarization parallel or anti-parallel to the external magnetic field. Here, $J_{\text{max}} = 1500$ was taken.
Figure 5.7: Averaging fast CPT scans over the nuclear spin distribution for $\phi < \phi_c$ from Fig. 5.5 reveals that the broadening of the distribution as $\phi \rightarrow \phi_c$ is reflected by an imperfect suppression of the absorption in the CPT dip.
5.4 Signatures of criticality

Figure 5.8: For $\phi > \phi_c$, averaging fast CPT scans over the nuclear spin distribution from Fig. 5.6 leads to two symmetric dips in the spectrum. The dips correspond to the configurations of polarized nuclear spin environment, where the polarization points parallel and anti-parallel to the external magnetic field, respectively. Since the accompanying nuclear spin distribution is much broader than for $\phi < \phi_c$, the suppression of the absorption in the dip is far from perfect.
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Figure 5.9: The unnormalized probability distribution \( p(\omega_{Oh}) \) for the two nuclear species In and As averaged over \( J_{\text{max}}^{1,2} = 1100 \) (blue line). The homogeneous hyperfine coupling constants per nuclear spin were taken to be \( A_{z}^{\text{As}} = 6.4 \cdot 10^{-5} \Gamma \) and \( A_{z}^{\text{In}} = 2.5 \cdot 10^{-4} \Gamma \). Since we treat spin-\( \frac{1}{2} \) nuclei, we have scaled the couplings with the spin \( I = \frac{3}{2}, \frac{9}{2} \) of the nuclei such that the maximum Overhauser shift is consistent. For comparison, the red line shows one species with the same average coupling constant, where \( J_{\text{max}} = 1500 \) such that in both simulations more than 95% of the probability is captured. In the case of two species, the system evolves into configurations showing slightly larger Overhauser shifts than in the equivalent homogeneous case.

Modification of the maximum obtainable Overhauser field, we focus on \( \phi > \phi_c \) for which the nuclear spins are polarized.

We consider the QD to be composed of As and In nuclei with \( N_{\text{As}} = N_{\text{In}} = 5 \cdot 10^5 \) and hyperfine couplings \( A_{z}^{\text{As}} = 6.4 \cdot 10^{-5} \Gamma \) and \( A_{z}^{\text{In}} = 2.5 \cdot 10^{-4} \Gamma \). We compare the resulting distribution of the Overhauser field to the probability distribution for a single species, i.e. \( N = N_{\text{As}} + N_{\text{In}} = 10^6 \) and \( A = (A_{z}^{\text{As}} + A_{z}^{\text{In}})/2 \). We find that in the presence of these two nuclear species, the nuclear probability distribution is broadened and the probability to obtain a large Overhauser shift \( \omega_{Oh} \) is slightly increased (Fig. 5.9, blue line) compared to a single species with the same average hyperfine coupling constant (Fig. 5.9, red line). This is consistent with earlier theoretical findings for the nuclear spin polarization in QDs when the hyperfine coupling is inhomogeneous, e.g. [144]: In each fixed \( J \) manifold the projection \( m \) is bounded by \( |m| \leq J \) and hence the total nuclear spin projection is limited by the fact that \( p(J) \) rapidly decreases for large \( J \).

5.4.6 Spectral gap

A non-analytic change in the steady state of the system at a continuous phase transition is accompanied by the closing of the spectral gap of the Liouvillian in the thermodynamic limit [66]. The gap \( \Delta \) is determined by the largest non-zero eigenvalue \( \lambda \) of the Liouvillian, i.e.

\[
\Delta = |\max\{\text{Re}(\lambda_i)|\lambda_i \neq 0\}|.
\] (5.17)

Due to the purely dissipative character of the nuclear Liouvillian the eigenvalues assume real negative values. We numerically compute the eigenvalues of the Liouvillian for typical \( J \) around \( J = \sqrt{N}/2 \) and plot the gap as a function of \( \phi \).
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Figure 5.10: The gap of $L_{\text{nuc}}^J$ for $J = \sqrt{N}/2 = 500$ and $J = \sqrt{N} = 1000$ as a function of $\phi$. The gap decreases with increasing $\phi$. (Fig. 5.6).

For larger $J$ the gap tends to zero more quickly than for smaller $J$ for $\phi > \phi_c$ (Fig. 5.10). For $\phi < \phi_c$ the vanishing eigenvalue represents the steady state of the system and the gap to the next eigenvalue is finite. For $\phi > \phi_c$ the second largest eigenvalue diminishes. The existence of two eigenvalues that tend to zero might be expected, given that the probability distribution was found to peak at two finite polarizations of the same magnitude but opposite sign.

To investigate the spectrum of the Liouvillian as the thermodynamic limit $N \to \infty$ is approached, we compute the gap for $J = \sqrt{N}/2$ and for different $\phi$ as a function of $N$ on a double logarithmic scale (Fig 5.11). We find that as $N$ increases, the gap tends to zero independent of whether $\phi < \phi_c$ or $\phi \geq \phi_c$. To understand the origin of this behavior we turn to semiclassical equations in the next section.

5.4.7 Thermodynamic limit

We analyze the semiclassical equations to shed some light on the vanishing of the gap observed in the thermodynamic limit $N \to \infty$. We assume that the correlated electron–nuclei state can be factorized. We introduce the normalized polarizations $x = \langle I_x \rangle /N$, $y = \langle I_y \rangle /N$, and $z = \langle I_z \rangle /N$ such that $|x|, |y|, |z| \leq \frac{1}{2}$ independent of $N$. This allows us to identify the root of the vanishing gap more easily. Furthermore, we assume that on the time scale on which $x$, $y$, and $z$ evolve, the electron can be taken to be in a steady state that depends on the current values of the nuclear spin expectation values. The evolution of the latter can thus be
Figure 5.11: In the thermodynamic limit $N \to \infty$, the gap tends to zero not only for $\phi \geq \phi_c$ but also for $\phi < \phi_c$. This indicates that the dynamics of the coupled electron-nuclei system is interrupted for $N \to \infty$ and that the thermodynamic limit is problematic. We therefore cannot use the closing of the gap as evidence of a non-analytic change of the steady state.
studied using the Heisenberg equations of motion, e.g. \( \dot{x} = i[H, I_x]/N \), with

\[
H = -\omega_n I_z + \frac{A_H}{N} S_z I_z + \frac{A_{Hc}}{N} S_z I_x, \tag{5.18}
\]

where we defined \( A_z = A = A_H/N \) and \( A_x = A_{nc} = A_{nc}^H/N \) to make the \( N \)-dependence of the single nuclear spin coupling constants explicit. In this notation \( A_H \) and \( A_{nc}^H \) are independent of \( N \). Then the system of differential equations becomes

\[
\begin{align*}
\dot{x} &= \left( \omega_n - \frac{A_H}{N} \langle S_z \rangle \right) y \tag{5.19} \\
\dot{y} &= -\left( \omega_n - \frac{A_H}{N} \langle S_z \rangle \right) x - \frac{A_{nc}^H}{N} \langle S_z \rangle z \tag{5.20} \\
\dot{z} &= \frac{A_{nc}}{N} \langle S_z \rangle y, \tag{5.21}
\end{align*}
\]

where \( \langle S_z \rangle \) is taken to be the steady state value, which depends on the effective two-photon detuning

\[
\delta_{\text{eff}} = \frac{A_H}{N} \langle I_z \rangle + \frac{A_{nc}^H}{N} \langle I_x \rangle = A_H z + A_{nc}^H x. \tag{5.22}
\]

Clearly, \( \delta_{\text{eff}} \) only depends on the relative polarization of the spin environment and is then independent of \( N \). We thus find that the feedback of the nuclear spin environment on the electron is the same for a given relative polarization of the spin environment. Computing \( \langle S_z \rangle \) in the steady state yields

\[
\langle S_z \rangle = \frac{\cos(2\phi)}{2} \frac{\Gamma^2 \delta_{\text{eff}}^2 + (\delta_{\text{eff}}^2 - 2r^2)^2}{\Gamma^2 \delta_{\text{eff}}^2 + (\delta_{\text{eff}}^2 - 2r^2)^2 + 6\delta_{\text{eff}}^2 r^2 \sin(2\phi)^2} \approx \frac{\cos(2\phi)}{2} \frac{1 - 6\delta_{\text{eff}}^2 r^2 \sin(2\phi)^2}{\Gamma^2 \delta_{\text{eff}}^2 + (\delta_{\text{eff}}^2 - 2r^2)^2}, \tag{5.23}
\]

where the last approximation holds for most cases of interest and \( \delta_{\text{eff}} \) is given by (5.22). Clearly \( |\langle S_z \rangle| \leq \frac{1}{2} \) and it is dominated by the term \( \frac{\cos(2\phi)}{2} \) which only depends on the relative Rabi frequencies but no other external parameter. We now consider the feedback of the electron to the nuclear spins and illustrate our arguments e.g. with

\[
\dot{z} = \frac{A_{nc}}{N} \langle S_z \rangle y. \tag{5.25}
\]

If the feedback strength is to remain independent of \( N \) as the thermodynamic limit is taken, we require

\[
\frac{A_{nc}}{N} \langle S_z \rangle \overset{\text{const.}}{=} \langle S_z \rangle \overset{\text{const.}}{=} \frac{1}{N} \Rightarrow \langle S_z \rangle \overset{\text{const.}}{=} N. \tag{5.24}
\]

This is clearly impossible to satisfy: First, \( |\langle S_z \rangle| \leq \frac{1}{2} \) and is thus bounded. Second, scaling the external drives, e.g. \( r \) with \( N \) does not provide a remedy, since the dominant contribution to \( \langle S_z \rangle \), i.e. \( \frac{\cos(2\phi)}{2} \), is not directly proportional to any external parameter other than \( \phi \). Therefore,

\[
\dot{z} = \frac{A_{nc}}{N} \langle S_z \rangle y \overset{N \to \infty}{\to} 0, \tag{5.25}
\]
Figure 5.12: The gap of $\mathcal{L}^J_{\text{nucl}}$ for $J = \sqrt{N}/2$ as a function of $N$ for hyperfine coupling constants that are independent of $N$. For large $\phi > \phi_c$, the gap tends to zero rapidly in the thermodynamic limit. For $\phi < \phi_c$ the gap does not vanish, contrary to Fig. 5.11.

which means that the feedback of the single electron spin on the nuclear environment vanishes in the thermodynamic limit. Hence, the nuclear dynamics is decoupled from the evolution of the electron that was responsible for controlling the dynamics of the nuclear spins for finite $N$. This decoupling of the nuclear spins then implies that the dissipative rates in the nuclear Liouvillian approach zero, which in turn results in a vanishing spectral gap.

### 5.4.8 Constant hyperfine coupling

Having identified the root of the vanishing gap, we now take the hyperfine coupling constants $A$ and $A_{\text{nc}}$ to be independent of $N$. This clearly does not describe the physical situation in QDs, but it is motivated by similar assumptions in the literature [66]. We then find that in the thermodynamic limit, the gap remains finite for $\phi < \phi_c$ and vanishes only for $\phi \geq \phi_c$ (Fig. 5.12 and Fig. 5.13). Thus, this result suggests that under the assumption of constant hyperfine couplings, non-analyticities in the nuclear steady state occur at $\phi_c$.

### 5.5 Conclusion and outlook

In summary, we have analyzed a central spin system, where the central spin is driven by two lasers in a CPT configuration. We derived a reduced master equa-
5.5 Conclusion and outlook

Figure 5.13: At the critical point $\phi_c = \pi/4$, the gap tends to zero in the thermodynamic limit under the assumption of hyperfine coupling constants that are independent of $N$.

...tion that describes the evolution of the spin environment. Applying this equation to a electron confined in a QD and interaction with the nuclear spin environment, we characterized the nuclear steady state as a function of an external parameter given by the relative strength of the two laser Rabi frequencies. We found that the nuclear steady state undergoes a qualitative change at the critical value that is defined by equal Rabi frequencies. The signatures in the nuclear spin system include a change in the DNP behavior, a quasi-continuous change of the stable solutions of the nuclear spin polarization, and increased nuclear fluctuations in the vicinity of the critical value. We also showed that the relative strength of the two Rabi frequencies can be utilized to narrow the nuclear spin distribution. Interestingly, many of these effects feed back onto the electron which therefore responds differently to optical excitation. This can readily be measured and makes this scheme attractive for experimental investigation. However, the spectral gap of the Liouvillian and the rates describing the nuclear evolution vanish in the thermodynamic limit. This can be traced back to the asymmetric hyperfine coupling between the electron and nuclear spins as well as the fact that the feedback from the electron to the nuclear spins is not directly proportional to any external driving that could be scaled appropriately. Nevertheless, under the assumption of hyperfine couplings that are independent of the number of nuclear spins, we find that the gap closes above the critical point, hinting at non-analyticities in the nuclear steady state.
Conclusion and outlook

In this thesis, the complex dynamics in a driven–dissipative central spin system was investigated and applied to self-assembled QDs. Depending on the interplay between continuous wave or pulsed laser excitation and electron–nuclei interactions, the resulting nuclear spin evolution was shown to be dominated by either diffusive or directional DNP. In the diffusive regime, we developed schemes that take advantage of the existence of an electronic dark state to induce anomalous nuclear spin diffusion. This allows to cool the nuclear spins and prepare an ultranarrow nuclear spin state which is desirable for quantum information processing applications. Furthermore, in self-assembled QDs the nuclear Lévy flights can be monitored in real time, which represents and important step towards the direct experimental observation of Lévy flights in a single quantum system. Exploiting directional DNP, we extended the mechanism that governs dragging effects to two-color continuous wave excitation in a CPT configuration [175, 170]. This allowed us to study the dependence of the stability of the electron–nuclei dark state on the relative strength of the driving laser Rabi frequencies. We found that the behavior of the system as a function of the relative Rabi frequencies shares certain features of a dissipative continuous phase transition, including increased fluctuations around the transition point as well as a continuous change of the order parameter.

Experimentally, the preparation of a ultranarrow nuclear spin state with the help of anomalous diffusion has been verified in Nitrogen Vacancy centers [177]. However, in self-assembled QDs the directional DNP effects that arise under continuous wave excitation mask the anomalous diffusive character. To circumvent this problem, pulsed laser excitation suppresses these directional terms and therefore represents a promising route towards the nuclear spin cooling by anomalous diffusion and the direct observation of Lévy flights in self-assembled QDs. To achieve this goal, it might nevertheless be necessary to determine the electron dephasing time and potentially optimize the QD sample structure such that the dephasing time is long enough. Furthermore, the implementation of the displaced echo pulse sequence might prove to be inevitable in order to suppress the nuclear spin dynamics arising from the intrinsic evolution. As an alternative for both the continuous wave and the pulsed laser excitation scheme, strainfree droplet QDs could be used [207, 100, 135]. In these QDs the directional diffusion due to strain-induced quadrupolar interactions are drastically reduced compared to self-assembled QDs. Then, the anomalous diffusion under continuous wave and pulsed excitation would arise due to the hyperfine flip-flop terms. In NV centers, one might be able to implement the pulsed excitation scheme within one of the three nitrogen spin states, where the anomalous diffusion takes place in the ensemble of $^{13}$C spins. Then the pulse repetition period should be chosen large enough such
that at least one trap is located within the $^{14}$N resonance.

A future direction could include investigating whether ergodicity breaking in anomalous diffusion in a central spin system could be directly observed. A system is ergodic if the time average of the system properties equals the ensemble average. In subrecoil laser cooling it has been proposed that anomalous diffusion should lead to ergodicity breaking since the trapping times diverge [16]. Intuitively, one can understand this as follows: For a total experimental interaction time $T_{\text{tot}}$, trapping events with precession frequencies satisfying $R_{\text{nuc}}(\omega_e)^{-1} \ll T_{\text{tot}}$ can occur many times in $T_{\text{tot}}$. One therefore expects that the time and ensemble average coincide. However, different $\omega_e$ satisfying $R_{\text{nuc}}(\omega_e)^{-1} > T_{\text{tot}}$ cannot be distinguished from one another. A realization of any such $\omega_e$ has a high probability to stay within a trap for the complete interaction time $T_{\text{tot}}$, which results in an ensemble average that is not directly related to the time average anymore. In subrecoil laser cooling, it was shown that these effects are expected to lead to a modified momentum distribution, which was also experimentally verified [208]. This constitutes an indirect confirmation of ergodicity breaking. However, in self-assembled QDs, both time and ensemble average of the time spent in a trap are experimentally accessible. This might open the path to a direct experimental comparison of time and ensemble averages in anomalous diffusion.


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