Electrical Control and Coherence of Spin Qubits in Indium Gallium Arsenide and Silicon Quantum Dots

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Abstract

Since the scientific community reached the consensus that computing obeying the laws of quantum mechanics offers advantages over computing obeying the laws of classical mechanics many researchers have tried to address challenges arising in implementation of quantum computers. The advantage of using quantum processors lies in the fact that they can allow faster computation compared to classical processors and can solve problems inaccessible to their classical counterparts.

Indium gallium arsenide ($\text{In}_x\text{Ga}_{1-x}\text{As}, 0 \leq x \leq 1$) and silicon (Si) are materials which are used to mass produce electronic components crucial for the functioning of modern life. In the last fifteen years the condensed matter community has tried to embed a quantum bit based on semiconductor quantum dots in these materials. One of the advantages of implementing a quantum bit in these materials is that the technology of producing nanostructures in these materials has been known and perfected for many years before researchers realized that these nanostructures show prospect of embedding a quantum bit.

This thesis focuses on the problem of control and coherence of electron spin qubits in InGaAs and Si quantum dots. The role of hyperfine interaction, spin-orbit interaction, the valley degree of freedom and charge noise are all appropriately addressed.

In Chapter 1 attention is payed to the properties of InGaAs and Si, which are relevant for the implementation of work in this thesis. The band structure of InGaAs and Si are discussed with a special emphasis to differences of their band spectra. Furthermore, the hyperfine, spin-orbit and exchange Hamiltonians in semiconductor quantum dots are derived.

Chapter 2 provides an introduction to spin-based quantum computing in quantum dots. The chapter starts with introducing some basic quantum mechanical methods. Three most commonly used semiconductor quantum bits are introduced. Finally, attention is given to the way in which the time evolution of a system is described in quantum mechanics.

Chapter 3 deals with obstacles in fully polarizing nuclear spins in a double semiconductor quantum dot by driving the system through the singlet-triplet plus $S - T_+$ anti-crossing. A fully polarized nuclear spin bath would significantly improve the coherence time of the singlet-triplet zero $S - T_0$ qubit. The presented findings suggest that spin-orbit interaction plays a crucial role in preventing any significant buildup of nuclear polarization.

Chapter 4 deals with spin qubits in Si quantum dots. Valley-orbit mixing due to the tilts in the Si/SiGe interface along with the presence of the stray field
of the micromagnet produces valley dependent \( g \)-factors and valley dependent Rabi frequencies. When valley relaxation is present the spin resonance condition changes with valley relaxation producing errors in controlling the electron spin qubit in Si quantum dots.

In the Chapter 5 a novel method to manipulate the single electron spin based on adiabatic Landau-Zener sweeps is introduced. The manipulation method relies on spin-orbit interaction and requires \( g \)-factor engineering. The advantages of this method is that it can be operated without the precise knowledge of the spin resonance condition, can be done all-electrically and is robust against the uncertainties in the nuclear magnetic field.

In Chapter 6 the form of mediated exchange (superexchange) is calculated. The system under study is a triple quantum dot loaded with two electrons, with one of the quantum dots serving as the mediator. A non-trivial first order double “sweet spot” is found. In this point the superexchange is insensitive to detuning noise. Furthermore, the superexchange in the presence of spin-orbit interaction is derived, before the conclusion in Chapter 7.
Zusammenfassung

Seit bei Wissenschaftlern bekannt ist, dass Computer auf der Basis von Quantenmechanik Vorteile haben gegenüber Computern, die nach den Gesetzen der klassischen Mechanik funktionieren, befassen sich viele Wissenschaftler mit der Realisierung von Quantencomputern. Der Vorteil der Quantenprozessoren liegt in der Tatsache, dass sie eine viel höhere Rechnergeschwindigkeit als klassische Rechner ermöglichen und somit Probleme lösen können, die den klassischen Rechnern nicht möglich sind.

Indium-Gallium-Arsenid (In$_x$Ga$_{1-x}$As, 0 ≤ x ≤ 1) und Silikon (Si) sind Werkstoffe, die im modernen Leben für die Massenproduktion von Elektronikbestandteilen verwendet werden, welche für das Funktionieren des modernen Leben entscheidend sind. In den letzten fünfzehn Jahren haben die Wissenschaftler im Bereich der kondensierten Materie versucht, ein Quantenbit, das aus Halbleiter-Quantenpunkten besteht, auf diese Werkstoffe zu implementieren. Ein Vorteil dieser Methode besteht darin, dass die Technologie, welche Nanostrukturen in diesen Materialien erzeugt, bereits bekannt und perfektioniert war, bevor Wissenschaftler erkannten, dass diese Nanostrukturen die Möglichkeit des Implementierens eines Quantenbits boten.

Diese Doktorarbeit konzentriert sich auf das Problem der Kontrolle und Kohärenz von Elektronenspin Qubits in InGaAs und Si Quantenpunkten. Es wird entsprechend eingegangen auf die Rolle der Hyperfeinstrukturwechselwirkung, Spin-Bahn-Wechselwirkung, Der valley-Freiheitsgrad und Ladungsrauschen.

In Kapitel 1 wird auf die Eigenschaften von InGaAs und Si eingegangen, die wichtig sind für die Grundlage der Berechnung in dieser Doktorarbeit sind. Es wird die Bandstruktur von InGaAs und Si diskutiert mit besonderer Betonung auf die Unterschiede ihrer Bandspektren. Darüberhinaus werden der Hyperfeinstruktur, Spin-Bahn und Austausch Hamiltonoperatoren in Halbleiter Quantendots abgeleitet.


Kapitel 3 befasst sich mit den Schwierigkeiten der kompletten Polarisation von Kernspins in einem doppelten Halbleiter Quantenpunkt indem man es durch ein Singlet-Triplet mit $S - T_+$ Anti-Crossing leitet. Eine komplett polarisierte Ansammlung von Spins könnte die Kohärenzzeit des $S - T_0$ Qubit wesentlich

v
verbessern. Die vorliegenden Forschungsergebnisse lassen darauf schließen, dass die Spin-Bahn Wechselwirkung eine Schlüsselrolle in der Vermeidung der Entstehung einer Kernspinpolarisation hat.


Acknowledgments

Some teenagers dream to be fighter pilots, some to be movie stars I dreamed to work on theoretical quantum physics since the age 15 or 16. Just a minor obstacle needed to be crossed on that way, I did not know a first thing about physics or math. I was also not too talented for neither. I almost failed seventh grade because of physics, obtaining a minimal passing grade of 2 in the winter semester. For me this thesis is a proof that desire wins over talent (although this seems like a surreal dream from which I don’t want to wake up).

I would like to thank my adviser Prof. Dr. Guido Burkard and Assoc. Prof. Dr. Dimitrije Stepanenko from the University of Belgrade for mentoring and supervision during the course of this thesis. I also feel obliged to mention two persons who helped a lot in the early days of my PhD, Dr. Niklas Rohling and Miguel Angel Rodriguez Moreno (I would have probably gone crazy without you guys). I would also like to thank Dr. Alexander Pearce, Dr. Adrian Auer, Dr. Andrey Moskalenko, Dr. Erik Welander, Matthew Brooks, Dr. Csaba Geza Peterfalvi, Maximilian Russ and Vladislav Shkolnikov for many discussions regarding physics during the later years of my PhD.

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1. Physics of semiconductors

1.1. Indirect and direct bandgap semiconductors

Electrons are fermions and therefore they cannot occupy identical states, thus obeying the Fermi-Dirac distribution \([1]\)

\[
f(E) = \frac{1}{e^{(E-\mu)/k_BT} + 1}. \tag{1.1}\]

Here, \(E\) is the energy, \(k_B\) is the Boltzmann constant, \(T\) is the temperature and \(\mu\) is the chemical potential. The Fermi level \(E_F\) is such a level where \(E = \mu\) at \(T = 0\). The position of the Fermi level with respect to the conduction and valence bands is crucial in determining the conductive properties of a material. Electrons in crystalline solids, like semiconductors occupy discrete energy levels called bands. At zero temperature the highest band filled with electrons is called the valence band and the lowest unoccupied band is called the conduction band \([2, 3]\).

Insulators are such materials in which the Fermi level lies between the valence and conduction band, with the energy difference of the valence and conduction bands (the band gap) being large. In case of semiconductors, the Fermi level lies between the valence and conduction bands but the band gap is smaller compared to insulators. In semiconductors, electrons occupy the valence band at absolute zero, however doping or heating induce electron occupation of the conduction band. Metals are such materials in which the valence and conduction bands overlap at zero temperature.

The dependence of energy on wavevector for a given material defines the band structure (or the \(E - k\) diagram, band diagram). Two types of semiconductors exist with respect to the position of the minimum of the conduction band in \(k\) space, indirect and direct semiconductors. In direct semiconductors like GaAs, InAs and monolayer transition-metal dichalcogenides, the minimum of the conduction band is located on top of the maximum of the valence band in \(k\) space. In indirect semiconductors, like Si, Ge and multilayer transition-metal dichalcogenides, the minimum of the conduction band is not located on top of the valence band in \(k\) space Fig. 1.1.
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![Diagram of direct and indirect semiconductors](image)

Figure 1.1. Direct and indirect semiconductors. The valence band states $E < 0$ and conduction band states $E > 0$

The coupling of the spin and orbital motion is going to influence the way the band diagram looks. In the most general form spin-orbit interaction has the following form \[1\]

$$H_{so} = \frac{\hbar}{4m_0^2c^2} (\sigma \times (\nabla V_0)) \cdot \mathbf{p},$$ \hspace{1cm} (1.2)

where $m_0$ is the free electron mass, $c$ is the speed of light, $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ is the vector of Pauli matrices, $\mathbf{p}$ the momentum operator and $V_0$ the potential. Another useful form of the spin-orbit Hamiltonian is obtained for spherical potentials $\nabla V_0 = \frac{r}{r^3} \frac{dV_0}{dr}$. After some vector algebra

$$H_{so} = \frac{1}{2m_0^2c^2} \frac{1}{r} \frac{dV_0}{dr} \mathbf{L} \cdot \mathbf{S},$$ \hspace{1cm} (1.3)

where the spin operator is defined as $\mathbf{S} = \frac{\hbar}{2} \sigma$.

In many cases, electrons and holes in solids can be described as free particles having an effective mass $m^*$ \[2, 3\]. Expanding the expression for the $E_{\nu}(\mathbf{k})$ in a Taylor series, the following expression is obtained around the extrema of parabolic dispersion relation

$$E_{\nu}(\mathbf{k}) = E_{\nu}(0) + \frac{k^2}{2} \frac{\partial E_{\nu}^2(\mathbf{k})}{\partial k^2},$$ \hspace{1cm} (1.4)

with the first derivative being zero because the point of interest is at an extrema of energy. The effective mass is therefore calculated from

$$\frac{\hbar^2}{m^*_\nu} = \frac{\partial E_{\nu}^2(\mathbf{k})}{\partial k^2},$$ \hspace{1cm} (1.5)
yielding the following dispersion relation

\[ E_\nu(k) = E_\nu(0) + \frac{k^2 \hbar^2}{2m^*_\nu}, \]  

where \( \nu = e \) for electrons and \( \nu = h \) for holes.

A powerful method to describe the band structure of direct bandgap semiconductors in more detail is the \( \mathbf{k} \cdot \mathbf{p} \) method [4]. The Bloch theorem states that the wavefunction of an electron in a periodic potential can be written as a product of a planewave part and the so-called Bloch wavefunction, having the same periodicity as the lattice \( \psi_\nu(r) = e^{i\mathbf{k} \cdot \mathbf{r}} u_k(\mathbf{r}) \), where \( \mathbf{r} \) is the position, \( \mathbf{k} \) the wave vector and \( \nu \) is the band index [1]. The starting point is the most general Schrödinger equation of an electron in a crystal being subjected to spin-orbit interaction

\[ \left( \frac{\mathbf{p}^2}{2m_0} + V_0 + \frac{\hbar}{4m_0 c^2} (\mathbf{\sigma} \times (\nabla V_0)) \cdot \mathbf{p} \right) e^{i\mathbf{k} \cdot \mathbf{r}} u_\nu(\mathbf{r}) = E_\nu(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{r}} u_\nu(\mathbf{r}). \]  

(1.7)

Here, \( \mathbf{p} \) is the momentum operator, \( m_0 \) is the free electron mass, \( V_0 \) is the potential at the lattice site and \( E_\nu(\mathbf{k}) \) is the energy corresponding to the band index \( \nu \) and the wavevector \( \mathbf{k} \). As the momentum operator is defined as \( \mathbf{p} = -i\hbar \nabla \) it will act on the wavefunction in the following way

\[ \mathbf{p} e^{i\mathbf{k} \cdot \mathbf{r}} u_\nu(\mathbf{r}) = \hbar \mathbf{e}^{i\mathbf{k} \cdot \mathbf{r}} u_\nu(\mathbf{r}) + e^{i\mathbf{k} \cdot \mathbf{r}} \mathbf{p} u_\nu(\mathbf{r}), \]

\[ \mathbf{p}^2 e^{i\mathbf{k} \cdot \mathbf{r}} u_\nu(\mathbf{r}) = \hbar^2 k^2 e^{i\mathbf{k} \cdot \mathbf{r}} u_\nu(\mathbf{r}) + 2\hbar \mathbf{e}^{i\mathbf{k} \cdot \mathbf{r}} \mathbf{p} u_\nu(\mathbf{r}) + e^{i\mathbf{k} \cdot \mathbf{r}} \mathbf{p}^2 u_\nu(\mathbf{r}). \]  

(1.8)

Inserting Eqs. (1.8) into Eq. (1.7) and after some rearranging the following Schrödinger equation for the Bloch wavefunctions is obtained

\[ \left( \mathbf{H} + \frac{\hbar}{m_0} \mathbf{k} \cdot \mathbf{\pi} \right) u_\nu(\mathbf{r}) = E_\nu(\mathbf{k}) u_\nu(\mathbf{r}), \]  

(1.9)

where

\[ \epsilon_\nu(\mathbf{k}) u_\nu(\mathbf{r}) = E_\nu(\mathbf{k}) u_\nu(\mathbf{r}) - \frac{\hbar^2 k^2}{2m_0} u_\nu(\mathbf{r}) \]  

(1.10)

and \( \mathbf{\pi} \) is given by

\[ \mathbf{\pi} = \mathbf{p} + \frac{\hbar}{4m_0 c^2} (\mathbf{\sigma} \times (\nabla V)). \]  

(1.11)

Eq. (1.7) can be solved for an adequately chosen model potential \( V_0 \) in the case when \( k = 0 \). A commonly used potential for \( V_0 \) is the spherical Coulomb potential. Then the Eq. (1.7) reduces to a hydrogen atom equation, with known solutions
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Figure 1.2. The spectrum of InGaAs. Here, $\Delta$ is the energy difference between the valence and conduction bands (band gap) and $\delta_{so}$ is the split-off band splitting. The valence band and conduction band are split off by a band gap of $\Delta_{GaAs} = 1.424$ eV for GaAs and $\Delta_{InAs} = 0.354$ eV at room temperature. The spin-orbit split-off is $\delta_{so} = 0.34$ eV for GaAs and $\delta_{so} = 0.31$ eV for InAs.

given by hydrogen wavefunctions $\phi_{\nu lm}$.

The central idea of the $k \cdot p$ method is that the part $H' = k \cdot \pi$ can be treated as a perturbation. The second order correction of energy is

$$E_{\nu}(\mathbf{k}) = E_{\nu}(0) + \frac{\hbar^2 k^2}{2m_0} + \frac{\hbar^2 k^2}{m_0^2} \sum_{m \neq n} \frac{\langle m, 0 | \pi_\alpha | n, 0 \rangle \langle m, 0 | \pi_\beta | n, 0 \rangle}{E_n(0) - E_m(0)}. \quad (1.12)$$

Eq. (1.12) is a starting point for many models calculating the energy spectrum of direct bandgap semiconductors (the two band model, the Kane model [5], the extend Kane model [6]). These models vary in the number of states included in the calculation. When the number of bands is large and a couple of lower energy bands are of interest a Schrieffer-Wolff transformation could be applied to decouple bands with higher energies states from bands with lower energy (for more details about the Schrieffer-Wolff transformation see Chapter 6). The $k \cdot p$ theory gives the following spectrum of InGaAs Fig. 1.2. The valence band and
conduction band are split off by a band gap of $\Delta_{\text{GaAs}} = 1.424$ eV for GaAs and $\Delta_{\text{InAs}} = 0.354$ eV at room temperature. The spin-orbit split-off is $\delta_{\text{so}} = 0.34$ eV for GaAs and $\delta_{\text{so}} = 0.31$ eV for InAs.

The Bloch part of the wavefunction of an electron in the conduction band is $s$-type. The Bloch part of hole wavefunction has a $p$-character. The fact that the Bloch part of the conduction band wavefunction is $s$-type will turn out to be crucial for the form of the hyperfine coupling in InGaAs semiconductor quantum dots.

### 1.2. Spin-orbit interaction in semiconductors

Before deriving the form of spin-orbit interaction in semiconductors two important symmetry properties will be defined. Hamiltonians which have time-reversal symmetry are invariant when the time-inversion operation is applied [4]

\begin{align}
t &\to -t, \\
p &\to -p, \\
S &\to -S,
\end{align}

(1.13)

where $t$ is time, $p$ is the momentum operator and $S$ is the spin operator. The Hamiltonian of a crystal lattice having inversion symmetry remains the same upon the operation

\begin{equation}
r \to -r,
\end{equation}

(1.14)

where $r$ is the position operator. The spin-orbit interaction does not break time-reversal symmetry $H_{\text{so}} \sim L \cdot S$ (see Eq. (1.3)). However, when the crystal has a center of inversion

\begin{equation}
L \cdot S = (r \times p) \cdot S = ((-r) \times (-p)) \cdot (-S) = -L \cdot S \Rightarrow L \cdot S = 0.
\end{equation}

(1.15)

Therefore, one concludes that two types of crystal lattices exist with respect to the type of spin-orbit that can be present. Lattices without a center of inversion in which an electron experiences a spin-orbit interaction even without macroscopic field gradients. This type of spin-orbit interaction is called the atomic or Dresselhaus spin-orbit interaction and it exists in materials like GaAs and InAs [7]. Another type of crystal lattices are those with a center of inversion, having a vanishing Dresselhaus spin-orbit interaction like Si Fig. 1.3.

The Dresselhaus spin-orbit interaction has the following form (see Eq. (1.3))

\begin{equation}
H_{\text{so}} = \hbar(k) \cdot S.
\end{equation}

(1.16)
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Here \( h(k) \) is the spin-orbit field

\[
h_x = \frac{\tilde{\beta} \hbar^3}{m^* \sqrt{2m^*\Delta}} k_x (k_y^2 - k_z^2),
\]

where \( \tilde{\beta} \) is a dimensionless parameter (\( \tilde{\beta} = 0.07 \) for GaAs), \( m^* \) is the effective electron mass, \( \Delta \) is the band gap. In the case of a quantum well with a structure axis in the [001] direction the wavevector is going to have a significant average value in the \( z \) direction. After averaging \( k_z^2 \) the following expressions are obtained for the spin-orbit field

\[
h_x = -\beta k_x; \quad h_y = \beta k_y; \quad h_z = 0,
\]

where \( \beta \) is the Dresselhaus spin-orbit constant \( \beta = \tilde{\beta} \hbar^3 \langle k_z^2 \rangle / (m^* \sqrt{2m^*\Delta}) \). Therefore, the Dresselhaus spin-orbit interaction has the form

\[
H_D = \frac{\beta}{k} (k_y S_y - k_x S_x).
\]

Another type of spin-orbit interaction in quantum wells and quantum dots is due to the presence of an electric field gradient perpendicular to the plane of the two dimensional electron gas and is called the Rashba spin-orbit interaction or the confining potential asymmetry spin-orbit contribution [8]. In quantum wells and quantum dots the gradient of the electric field in the \( z \) direction originates from the asymmetry due to the interface of two materials. In that case the Rashba

![Figure 1.3. GaAs (left) and Si (right) crystal lattices. Gallium (red), arsenic (blue) and silicon (dark green).](image)
Figure 1.4. The energy diagram of Si. The band gap at 4K is $\Delta = 1.17$ eV and $\Delta = 1.12$ eV at room temperature, while the spin-orbit split splitting between $j = 3/2$ and $j = 1/2$ holes is $\delta_{\text{so}} = 44$ meV.

type spin-orbit is described with the Hamiltonian

$$H_R = \frac{\alpha}{\hbar}(k_x S_y - k_y S_x),$$

(1.20)

where $\alpha$ is the Rashba spin-orbit constant. The spin-orbit Hamiltonian is the sum of the Rashba and Dresselhaus spin-orbit interactions.

$$H_{\text{so}} = H_D + H_R = \frac{\beta}{\hbar}(k_y S_y - k_x S_x) + \frac{\alpha}{\hbar}(k_x S_y - k_y S_x).$$

(1.21)

1.3. The energy diagram of bulk silicon

From a spin qubit perspective silicon (Si) has an advantage over InGaAs with only 4.7% of nuclear spin species having a non-zero nuclear spin. In contrast to InGaAs Si is an indirect band gap semiconductor so $k \cdot p$ theory cannot be used to obtain the band diagram of Si. A common method for obtaining the band
diagram of Si is phenomenological tight binding. Obtaining the band diagram of Si is beyond the scope of this thesis and the reader is referred to for a more detailed overview [9, 10].

Si has a diamond-like structure with a maximum of the valence band at the Γ point and a minimum of the conduction band at the \( X \) point located at \( 0.85k_0 \) where \( k_0 \) represents the edge of the Brillouin zone Fig. 1.4. In bulk Si the minimum of the conduction band is 6 fold degenerate. In the remaining part of the thesis these minima of the Brillouin zone will be referred to as valleys. The band gap at \( 4K \) is \( \Delta = 1.17 \) eV and \( \Delta = 1.12 \) eV at room temperature, while the spin-orbit split splitting between \( j = 3/2 \) and \( j = 1/2 \) holes is \( \delta_{so} = 44 \) meV.

1.4. Si quantum dots

A six-fold degenerate minimum of the conduction band in bulk Si gets split into pairs of four and two fold degenerate minima by tensile strain at the Si/SiGe or Si/SiO\(_2\) interface [11, 12]. This split-off is on the order of magnitude of \( 10 \sim 200 \) meV [11, 13]. Further asymmetries of the confining potential due to the presence of the Si/SiGe or Si/SiO\(_2\) interface or electric fields further split the lowest two energy minima (valleys) Fig. 1.5.

The fact that another degree of freedom exists presents an additional difficulty in implementing spin qubits based on electron spins in Si quantum dots. The valley degree of freedom represents another degree of freedom which lifts the Pauli spin blockade and allows occupancy of parallel spin states of two electron spins in a single quantum dot in the lowest orbital state. The magnitude of this splitting \( 0.1 \sim 1 \) meV is crucial for the implementation of spin-based quantum dot qubit in Si quantum dots. Authors in [14] showed that the magnitude of this splitting can be controlled by varying the confinement potential of the quantum dot [15]. If this splitting can be made larger than the thermal broadening of the lead, the electron spin could be initialized, controlled and readout without occupying higher energy valley states. Another phenomenon diminishing the efficiency of coherent control of the electron spin qubit in Si quantum dots is the valley-orbit mixing. When valley-orbit mixing is present due to tilts of the Si/SiGe interface, the \( g \)-factors and EDSR Rabi frequencies become valley dependent, further limiting the coherence and control efficiency of the electron spin qubit by valley relaxation (see Chapter 4 and [16, 17]).
1.5. Exchange interaction in double quantum dots

Here the form of exchange interaction in double quantum dot will be derived, with individual dots being denoted as left (L) and right (R) \[18\]. The starting point is the Hubbard Hamiltonian of two electrons in a double quantum dot in zero magnetic field

\[
H = \begin{pmatrix}
U - \epsilon & 0 & \sqrt{2}t & 0 & 0 & 0 \\
0 & U + \epsilon & \sqrt{2}t & 0 & 0 & 0 \\
\sqrt{2}t & \sqrt{2}t & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}, \text{ written in the basis of }
\begin{align*}
S(2, 0) \\
S(0, 2) \\
S(1, 1) \\
T_0(1, 1) \\
T_-(1, 1) \\
T_+(1, 1)
\end{align*}
\]

The numbers in the parentheses denote the charge state, (2, 0) is the doubly occupied (L) QD, (0, 2) is the doubly occupied (R) QD and (1, 1) singly occupied (L) and (R) QD’s. Furthermore, S denotes the singlet state while \( T_0, T_- \) and \( T_+ \) denote the \( s = 1, m_s = 0, -1, 1 \) triplet states. The terms in the Hamiltonian are the Coulomb energy of the doubly occupied dot \( U \), the detuning of the (L) QD with respect to the (R) QD (assumed negative) \( \epsilon \) and the overlap between
the dots \( t \). In typical electrostatically defined quantum dots \( U = 1 - 5 \) meV, and \( t \sim 0.01U - 0.1U \). It should be noted that the Coulomb energy of neighboring dots being occupied is \( V = 0.01 - 0.1U \) and is therefore neglected. The states \( S(2,0), S(0,2) \) will be detuned from the \( S(1,1), T_0(1,1) \) states when the detuning \( |\epsilon| \ll U \). Then, the states \( S(2,0), S(0,2) \) can be decoupled from the \( S(1,1), T_0(1,1) \) with a Schrieffer-Wolff transformation. The Hamiltonian is split into the diagonal part \( H_0 \) and the part containing interactions \( H_2 \). An anti-Hermitian \( S \) is found, such that satisfies the following equation

\[
[H_0, S] + H_2 = 0.
\]  

(1.23)

When such \( S \) is found, the Hamiltonian decoupling doubly occupied from singly occupied states has the following form

\[
\hat{H} = H_0 + \frac{1}{2}[H_2, S] = \begin{pmatrix}
\frac{2t^2}{U^2 - \epsilon^2} + U - \epsilon & \frac{2t^2U}{U^2 - \epsilon^2} & 0 & 0 & 0 & 0 \\
\frac{2t^2U}{U^2 - \epsilon^2} & \frac{2t^2}{U^2 + \epsilon} + U + \epsilon & 0 & 0 & 0 & 0 \\
0 & 0 & -\frac{4t^2U}{U^2 - \epsilon^2} & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}, \text{ in the basis } \begin{cases}
S(2,0) \\
S(0,2) \\
S(1,1) \\
T_0(1,1) \\
T_-(1,1) \\
T_+(1,1)
\end{cases}.
\]  

(1.24)

By definition of the exchange splitting \( J \) is the energy difference between the \( T_0(1,1) \) and \( S(1,1) \) states yielding the following expression for the exchange interaction in double quantum dots.

\[
J = \frac{4t^2U}{U^2 - \epsilon^2}.
\]  

(1.25)

Excluding the highly detuned doubly occupied states the following Heisenberg Hamiltonian is obtained in the \( \{(\uparrow, \uparrow), (\uparrow, \downarrow), (\downarrow, \uparrow), (\downarrow, \downarrow)\} \) basis

\[
H_s = JS_1 \cdot S_2
\]  

(1.26)

The Heisenberg interaction is a crucial building block in implementing the single electron spin qubit, the \( S - T_0 \) qubit and the exchange only qubit as it mediates single and two qubit operations. The Schieffer-Wolff transformation will be discussed in more detail in Chapter 6.
1.6. Contact hyperfine interaction in semiconductor quantum dots

In this section the hyperfine Hamiltonian between the single electron spin and many nuclear spins in a semiconductor quantum dot is derived [19]. Initially, the interaction of a spin 1/2 electron interacting with a magnetic field and the attractive potential of a single proton will be derived (the case of the hydrogen atom). In contrast to the hydrogen atom, the wavefunction of the electron is overlapping with as many as $10^6$ nuclear spins. This happens due to the fact that the confining potential of the quantum dot is usually confining the electron to $\sim 20 \cdot 10^{-9}$ m in contrast to the hydrogen atom where the first Bohr radius is $5.29 \cdot 10^{-11}$ m. By generalizing the hydrogen atom to the case of a single electron interacting with many nuclear spins, the form of the hyperfine interaction in semiconductor quantum dots will be derived.

The starting point of the derivation is the Schrödinger equation of the electron in the electromagnetic field

$$H = \frac{1}{2m_e} (p - q \mathbf{A}(r))^2 + qV_0(r) - \frac{1}{\hbar} g\mu_B \mathbf{S} \cdot \mathbf{B}. \quad (1.27)$$

Here, $p$ is the momentum operator of the electron, $\mathbf{A}(r)$ is the vector potential of the proton, $q$ is the electron charge, $V_0(r)$ is the Coulomb potential, $g$ is the electron $g$ factor, $\mu_B$ is the Bohr magneton, $\mathbf{S}$ is the operator of the electron spin. Furthermore, $\mathbf{B}$ is the magnetic field generated by the proton with the following relation holding

$$\mathbf{B} = (\nabla \times \mathbf{A}(r)). \quad (1.28)$$

Furthermore, the vector potential of the proton can be represented in the following way

$$\mathbf{A}(r) = \frac{\mu_0}{4\pi} \frac{\mathbf{M} \times \mathbf{r}}{r^3}, \quad (1.29)$$

where $\mathbf{M}$ is the magnetic dipole moment of the proton. Inserting Eq. (1.29) into Eq. (1.28) and inserting the result of that into Eq. (1.27) and neglecting terms proportional to $A^2(r)$ (because the vector potential generated by a proton
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is rather small)

\[
H = \frac{1}{2m} \left( p^2 - q \frac{\mu_0}{4\pi} \left( p \cdot \frac{(M \times r)}{r^3} + \frac{(M \times r) \cdot p}{r^3} \right) \right) + qV_0(r) - \frac{\mu_0 g \mu_B}{4\pi\hbar} S \cdot \left( \nabla \times \frac{M \times r}{r^3} \right). \tag{1.30}
\]

Using the algebra of the vector product with keeping track of the commutation properties the following expression is obtained

\[
\frac{1}{r^3} (M \times r) \cdot p = p \cdot (M \times r) \frac{1}{r^3} = \mathbf{L} \cdot \mathbf{M}, \tag{1.31}
\]

where \( \mathbf{L} \) is the angular momentum operator \( \mathbf{L} = \mathbf{r} \times \mathbf{p} \). By inserting Eq. (1.31) into Eq. (1.30), the following Hamiltonian is obtained

\[
H = \frac{1}{2m} \left( p^2 - q \frac{\mu_0}{2\pi} \mathbf{L} \cdot \mathbf{M} \right) + qV_0(r) - \frac{\mu_0 g \mu_B}{4\pi\hbar} S \cdot \left( \nabla \times \frac{M \times r}{r^3} \right). \tag{1.32}
\]

The second term in the parentheses can be modified as

\[
H_{\text{orb.}} = q \frac{\mu_0}{4\pi m} \mathbf{L} \cdot \mathbf{M} = -\mathbf{M} \cdot \mathbf{B}^e. \tag{1.33}
\]

where, \( \mathbf{B}^e = q \frac{\mu_0}{4\pi m r^3} \mathbf{L} \) is the magnetic field of the electron influencing the nuclear moment \( \mathbf{M} \). The magnetic field originates form the current loop created by a motion of a charge (electron) around the proton.

Now the last term in Eq. (1.30) will be transformed

\[
H_{\text{HF}} = -\frac{\mu_0 g \mu_B}{4\pi\hbar} S \cdot \left( \nabla \times \frac{M \times r}{r^3} \right). \tag{1.34}
\]

Finite dimensions of the proton \( \rho_0 \) are assumed. The magnetic field of the proton can be derived for two limiting cases, for \( \rho_0 \ll r \) \( H_{\text{HF}}^{\rho_0 \ll r} \) and for \( r \to 0 \) \( H_{\text{HF}}^{r \to 0} \). First the term when \( \rho_0 \ll r \) will be derived. Assuming a proton magnetic moment in \( z \) direction and estimating the curl of Eq. (1.28)

\[
B_x = \frac{\mu_0}{4\pi} 3M r^1, \\
B_y = \frac{\mu_0}{4\pi} 3M r^1, \\
B_z = \frac{\mu_0}{4\pi} 3M \left( \frac{2z^2 - r^2}{r^5} \right). \tag{1.35}
\]
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Substituting these expressions into Eq. (1.34)

$$H_{\text{HF}}^{0 \to r} = \frac{\mu_0 g \mu_B}{4\pi} \frac{1}{\hbar r^3} \left( \mathbf{S} \cdot \mathbf{M} - 3 \frac{(\mathbf{S} \cdot \mathbf{r})(\mathbf{M} \cdot \mathbf{r})}{r^2} \right).$$  \hspace{1cm} (1.36)

Now the term when $r \to 0$ will be calculated. The starting point in the derivation is Gauss’s magnetism law

$$\oint \mathbf{B} \cdot d\mathbf{A} = 0,$$  \hspace{1cm} (1.37)

stating that a magnetic flux integrated over any closed surface equals to zero. In the case under study, the closed surface comprises of a disk centered at the origin in the $x$ – $y$ plane and the upper part of a sphere having an infinite radius. The flux through the upper part of the sphere is 0 (as the sphere has an infinite radius and the magnetic field goes $1/r^3$). Therefore, only the flux through the plane remains. The flux through the plane comprises of the flux outside of the proton radius $\Phi_e(\rho_0)$ and the flux inside the proton radius $\Phi_i(\rho_0)$. This two contributions must be equal with opposite signs.

$$\Phi_e(\rho_0) = 2\pi \int_{\rho_0}^{\infty} r dr \left[ -\frac{\mu_0}{4\pi} M \frac{2\pi}{r^3} \right] = -\frac{\mu_0}{4\pi} M \frac{2\pi}{\rho_0},$$  \hspace{1cm} (1.38)

and

$$\Phi_i(\rho_0) = \pi \rho^2 B.$$  \hspace{1cm} (1.39)

This yields

$$B = \frac{\mu_0}{4\pi} M \frac{2\pi}{\rho_0^3}.$$  \hspace{1cm} (1.40)

Writing the expression of the matrix element

$$\langle \psi_{nlms} | H_{\text{HF}}^{r \to 0} | \psi_{n'm'm's'} \rangle = \frac{\mu_0 2\mu_B M}{4\pi} \langle s | S_z | s' \rangle \frac{8\pi}{3} \psi_{nlm}^* (\mathbf{r} = 0) \psi_{n'm'm'}^* (\mathbf{r} = 0),$$  \hspace{1cm} (1.41)

where $\psi_{nlms}$ are the hydrogen atom wavefunctions corresponding to the quantum numbers $n, l, m, s$ respectively. This matrix element corresponds to the so-called contact hyperfine Hamiltonian

$$H_{\text{HF}}^{r \to 0} = \frac{\mu_0}{4\pi} \frac{8\pi}{3} M \left( \frac{2\mu_B S}{\hbar} \right) \delta(\mathbf{r}).$$  \hspace{1cm} (1.42)

The full hyperfine Hamiltonian is given by summing Eq. (1.33), Eq. (1.36) and
1. Physics of semiconductors

Eq. (1.42)

\[ H_{HF} = H_{orb} + H_{HF}^{\rho_0 \ll r} + H_{HF}^{r \to 0} \]

\[ = -\frac{\mu_0 \mu_0 \mu_n g g_n}{4\pi \hbar^2} \left( \frac{\mathbf{I} \cdot \mathbf{L}}{r^3} + \frac{3}{r^5} \frac{(\mathbf{I} \cdot \mathbf{r})(\mathbf{S} \cdot \mathbf{r})}{r^5} - \frac{\mathbf{I} \cdot \mathbf{S}}{r^3} + \frac{8\pi}{3} \mathbf{I} \cdot \mathbf{S}\delta(\mathbf{r}) \right), \quad (1.43) \]

where \( \mu_B \) is the Bohr magneton, \( \mu_N \) is the nuclear magneton, \( g \) is the electron \( g \) factor and \( g_n \) is the nuclear \( g \) factor, and \( \mathbf{I} = h\mathbf{M}/g_n \mu_n \) the nuclear spin operator.

Similarly to the Hydrogen atom, the electron in the ground state of a quantum dot is going to have an \( s \)-type spatial wavefunction. In contrast to the hydrogen atom, the electron spin confined in semiconductor quantum dot interacts with as many as \( 10^6 \) nuclei. The hyperfine Hamiltonian derived here takes the following form

\[ H_{HF} = -\frac{\mu_0 \mu_B g}{4\pi \hbar^2} \sum_j g_j \mu_j \left( \frac{\mathbf{I}_j \cdot \mathbf{L}}{|\mathbf{r}_j|^3} + \frac{3}{|\mathbf{r}_j|^5} \frac{(\mathbf{I}_j \cdot \mathbf{r}_j)(\mathbf{S} \cdot \mathbf{r}_j)}{|\mathbf{r}_j|^5} - \frac{\mathbf{I}_j \cdot \mathbf{S}}{|\mathbf{r}_j|^3} + \frac{8\pi}{3} \mathbf{I}_j \cdot \mathbf{S}\delta(\mathbf{r}_j) \right), \quad (1.44) \]

where the sum over \( j \) runs over all possible nuclear spins.

An electron in a semiconductor quantum dot is going to have an \( s \)-type periodic part of the Bloch wavefunction. Due to this, the contribution of the first, second and third term in the upper equation is negligible and only the contact term remains for electrons. In contrast, the periodic part of the Bloch wavefunction is going to be \( p \)-type in case of holes and therefore, the first, the second and third terms are going to be dominating over the contact one. Therefore, the hyperfine interaction for an electron inside a quantum dot takes the following form

\[ H_{HF} = \sum_j A_j \mathbf{S} \cdot \mathbf{I}_j = \mathbf{S} \cdot \mathbf{h}, \quad (1.45) \]

where \( A_j \) is the strength of interaction between the electron spin and \( j \)th nuclear spin, and \( \mathbf{h} \) is the operator of the Overhauser field [20, 21].
2. Spin qubits in semiconductor quantum dots

2.1. The density matrix

Pure states in quantum mechanics can be described with a wavefunction $|\psi\rangle$ [1, 19]. However, the wave function description of the state is insufficient to describe mixed states. Therefore, a different mathematical object must be used to include the description of mixed states. Both mixed and pure states can be described with a density matrix $\rho$. The diagonal terms of the density matrix are interpreted as probabilities, and the off-diagonal terms are so-called coherence terms, describing superpositions of states. The density matrix is Hermitian ($\rho = \rho^\dagger$) and has unit trace $\text{Tr}(\rho) = 1$. The density matrix of a pure state satisfies the following equality $\text{Tr}(\rho^2) = 1$, while the density matrix of a mixed state satisfies $0.5 \leq \text{Tr}(\rho^2) < 1$ with $\text{Tr}(\rho^2) = 0.5$, corresponding to a totally mixed state.

The density matrix of any two level system can be represented in the following way [1]

$$\rho = \frac{\hat{I} + \sigma \cdot \mathbf{P}}{2}, \quad (2.1)$$

where the polarization vector is denoted as $\mathbf{P} = \langle \sigma \rangle$, $\hat{I}$ is the unit matrix and $\sigma$ is the vector of the Pauli matrices which are Hermitian and traceless

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \quad (2.2)$$

Using the definition of the Pauli matrices, the density matrix can be represented as

$$\rho = \frac{1}{2} \begin{pmatrix} 1 + P_z & P_x - iP_y \\ P_x + iP_y & 1 - P_z \end{pmatrix}. \quad (2.3)$$

The polarization vector of a pure state has unit length $|\mathbf{P}| = 1$ and corresponds to a point on the surface of the so-called Bloch sphere [22]. Pure states correspond to states on the surface of a Bloch sphere $|\psi\rangle = \cos(\theta/2)|0\rangle + \exp(i\phi)\sin(\theta/2)|1\rangle$, where $\theta$ is the azimuthal angle and $\phi$ is the polar angle Fig. 2.1.
2. Spin qubits in semiconductor quantum dots

![Image of the Bloch sphere](image)

Figure 2.1. The Bloch sphere. Pure states are on the surface of the Bloch sphere, while totally mixed state lie in the center of the Bloch sphere.

2.2. Time dependent problems

1. Time evolution of the density matrix and relaxation

A time-dependent Schrödinger equation describes the time evolution of a wavefunction. Similarly, the Liouville-von Neumann equation describes the time evolution of a density matrix,

\[
\frac{i\hbar}{\hbar} \frac{\partial \rho}{\partial t} = [H(t), \rho]
\]  

(2.4)

where, \( H(t) \) is the time-dependent Hamiltonian and \( \rho \) the density matrix.

In the case when an uncorrelated (Markovian) environment is present, the time evolution of the density matrix describing the \( N \) dimensional relevant system follows the Lindblad master equation, often referred to as the Gorini-Kossakowski-Sudarshan-Lindblad equation [23–25]

\[
\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [H(t), \rho] + \sum_{n,m=1}^{N^2-1} \Gamma_{n,m} \left( L_n \rho L_m^\dagger - \frac{1}{2} \left( \rho L_m^\dagger L_n + L_n^\dagger L_m \rho \right) \right).
\]

(2.5)

Here, \( \Gamma_{n,m} \) is the positive relaxation rates between the states \( n \) and \( m \) and \( L_n \) are Lindblad dissipation operators.
2. Spin qubits in semiconductor quantum dots

The time evolution of the wavefunction in quantum mechanics is described with the time-dependent Schrödinger equation

\[ i\hbar \frac{d}{dt} |\psi(t)\rangle = H(t) |\psi(t)\rangle. \tag{2.6} \]

When the Hamiltonian \( H(t) \) is time independent the time evolution of the state vector is calculated in a following manner

\[ |\psi(t)\rangle = e^{-\frac{iHt}{\hbar}} |\psi(0)\rangle. \tag{2.7} \]

In many problems of quantum mechanics, the Hamiltonian \( H(t) \) explicitly depends on time. For any Hamiltonian which commutes with itself at all time instances \( t_1 \) and \( t_2 \) \([H(t_1), H(t_2)] = 0\) the time evolution of the state vector can be described in the following way

\[ |\psi(t)\rangle = e^{-\frac{i}{\hbar} \int_0^t H(t') dt'} |\psi(0)\rangle. \tag{2.8} \]

In the case when the Hamiltonian is not commuting at different time instances \([H(t_1), H(t_2)] \neq 0\) the time evolution of the state can be calculated in the following way

\[ |\psi(t)\rangle = G(t) |\psi(0)\rangle, \tag{2.9} \]

where \( G(t) \) is the propagator. Common methods of computing the propagator in an approximate manner \( G(t) \) are the Dyson series expansion \([26]\) and the Magnus expansion \([27]\). Writing more about these methods goes beyond the scope of this thesis. In the remaining part of the section a list of exact and approximate methods will be given for solving time-dependent problems relevant to the work conducted in this thesis.

3. The rotating wave approximation

The rotating wave approximation (RWA) is commonly used to derive approximate time-independent Hamiltonians for periodically driven two level systems. The Hamiltonian of such of a process is given by

\[ H(t) = \begin{pmatrix} \frac{E}{2} & \Omega \cos \omega t \\ \Omega \cos \omega t & -\frac{E}{2} \end{pmatrix}. \tag{2.10} \]
Here the $\pm E/2$ are energies of the two level system, $\Omega$ is the Rabi frequency, proportional to the power of driving and $\omega$ is the driving Larmour frequency. The first step in obtaining the time independent RWA Hamiltonian $\tilde{H}$ is to apply the following unitary transformation

$$\tilde{H} = U(t)H(t)U(t)\dagger - i\hbar U(t)\dot{U}(t)\dagger, \quad (2.11)$$

where $U(t)$ is given by

$$U(t) = \begin{pmatrix} e^{i\omega t/2} & 0 \\ 0 & e^{-i\omega t/2} \end{pmatrix}. \quad (2.12)$$

Inserting Eq. (2.12) into Eq. (2.11), setting $\tilde{\omega} \to \omega$ and rewriting the cosine as a sum of two complex exponentials

$$U(t)H(t)U(t)\dagger - i\hbar U(t)\dot{U}(t)\dagger = \begin{pmatrix} E - \hbar\omega & \Omega \left(1 + e^{2i\omega t}\right) \\ \frac{\Omega}{2} \left(1 + e^{-2i\omega t}\right) & -E + \hbar\omega \end{pmatrix}. \quad (2.13)$$

By neglecting the fast oscillating term $e^{\pm2i\omega t}$ the time-independent RWA Hamiltonian is obtained

$$\tilde{H} = \frac{1}{2} \begin{pmatrix} E - \hbar\omega & \Omega \\ \Omega & -E + \hbar\omega \end{pmatrix}. \quad (2.14)$$

4. Two level Landau-Zener-Stückelberg-Majorana problems

The two Landau-Zener-Stückelberg-Majorana model (often refereed to as the Landau-Zener model) is used to describe a two level system, coupled with a constant interaction strength with an energy of the levels varying linearly in time, described with a Landau-Zener Hamiltonian [28–31]

$$H(t) = \begin{pmatrix} -vt & \tau \\ \tau & vt \end{pmatrix}. \quad (2.15)$$

Here, $v$ is the so-called Landau-Zener velocity, $t$ is time and $\tau$ is the strength of interaction which is assumed to be real. Writing the time dependent Schrödinger equation with the Landau-Zener Hamiltonian, the set of coupled equations for the components of the wavefunction $c_1$ and $c_2$ is obtained

$$-vtc_1 + \tau c_2 = i\hbar\dot{c}_1, \quad (2.16)$$
$$\tau c_1 + vt c_2 = i\hbar\dot{c}_2. \quad (2.17)$$
From here, the differential equation for calculating $c_1$ is obtained

$$\ddot{c}_1(t) = \left( \frac{i}{\hbar} - \frac{v^2 t^2}{\hbar^2} - \frac{\tau^2}{\hbar^2} \right) c_1(t),$$

(2.18)

with solution [32]

$$c_1(t) = k_1 D_{\frac{\tau^2}{2\hbar}} \left( \sqrt{2} e^{-i\frac{\pi}{4}} \sqrt{\frac{v}{\hbar}} t \right) + k_2 D_{\frac{\tau^2}{2\hbar}} \left( \sqrt{2} e^{i\frac{3\pi}{4}} \sqrt{\frac{v}{\hbar}} t \right).$$

(2.19)

Where $D_{\mu}(x)$ are the parabolic cylinder functions which are solutions of the Weber differential equation

$$\frac{d^2 f(x)}{dx^2} + \left( \mu + 1 - \frac{1}{4} x^2 \right) f(x) = 0.$$  

(2.20)

The other component ($c_2(t)$) of the wavefunction is obtained by inserting the Eq. (2.19) into Eq. (2.16) and by applying the following relation for parabolic cylinder functions

$$\frac{d}{dx} \left( e^{x^2} D_{\mu}(x) \right) = \mu e^{x^2} D_{\mu-x}(x).$$

(2.21)

This procedure yields the following expression for the other component of the wavefunction [32]

$$c_2(t) = \frac{\tau}{\sqrt{2} \nu \hbar} e^{-\frac{i\pi}{4}} \left( -k_1 D_{\frac{\tau^2}{2\hbar}} -1 \left( \sqrt{2} e^{-i\frac{\pi}{4}} \sqrt{\frac{v}{\hbar}} t \right) + k_2 D_{\frac{\tau^2}{2\hbar}} +1 \left( \sqrt{2} e^{i\frac{3\pi}{4}} \sqrt{\frac{v}{\hbar}} t \right) \right).$$

(2.22)

The constants $k_1$ and $k_2$ are found from initial conditions for $c_1$ and $c_2$

$$k_1 = \frac{\Gamma \left( 1 - i \frac{\tau^2}{2\hbar} \right)}{\sqrt{2\pi}} \left[ D_{\frac{\tau^2}{2\hbar}} -1 \left( \sqrt{2} e^{i\frac{3\pi}{4}} \sqrt{\frac{v}{\hbar}} t \right) c_1(t_i) \right.$$  

$$\left. - \sqrt{\frac{2\nu \hbar}{\tau}} e^{i\frac{\pi}{4}} D_{\frac{\tau^2}{2\hbar}} \left( \sqrt{2} e^{i\frac{3\pi}{4}} \sqrt{\frac{v}{\hbar}} t \right) c_2(t_i) \right],$$

$$k_2 = \frac{\Gamma \left( 1 - i \frac{\tau^2}{2\hbar} \right)}{\sqrt{2\pi}} \left[ D_{\frac{\tau^2}{2\hbar}} -1 \left( \sqrt{2} e^{-i\frac{\pi}{4}} \sqrt{\frac{v}{\hbar}} t \right) c_1(t_i) \right.$$  

$$\left. + \sqrt{\frac{2\nu \hbar}{\tau}} e^{i\frac{\pi}{4}} D_{\frac{\tau^2}{2\hbar}} \left( \sqrt{2} e^{-i\frac{\pi}{4}} \sqrt{\frac{v}{\hbar}} t \right) c_2(t_i) \right].$$

(2.23)
Assuming \( c_1(t \to -\infty) = 1 \) and \( c_2(t \to -\infty) = 0 \), and using the asymptotic expansion of parabolic cylinder functions \([33]\), gives the Landau-Zener formula

\[
|c_2(t \to \infty)|^2 = 1 - e^{-\frac{2\pi r^2}{\hbar \omega}}.
\] (2.24)

5. Multilevel Landau-Zener problems

Unlike the two level Landau-Zener problem, multilevel Landau-Zener problems do not not have a known exact analytic solution in a general case. The earliest works from Demkov and Osherov \([34, 35]\) discussed a single level with an energy linearly varying in time interacting with a set of parallel, non-degenerate levels, with time independent energies. It was shown that in this case the probabilities at \( t \to \infty \) are in accordance with a simple semi-classical model.

Furthermore, Carroll and Hioe \([36]\) showed that a three level Landau-Zener crossing is solvable up to an inverse Fourier transform for the components of the wavefunction. Damski and Zurek \([37]\) devised a treatment of the two level Landau-Zener problem, treating the evolution in an adiabatic manner everywhere except close to the anti-crossings where the evolution is non-adiabatic. Following on their work Rangelov, Pillo and Vitanov \([38]\) expanded the formalism to treat three level Landau-Zener processes in the case when a single level with an energy linearly varying in time intersects with two non-degenerate levels with constant energies.

6. Fock’s adiabatic theorem

A common approach of treating adiabatic time-dependent problems is approximating them within the Fock’s adiabatic theorem, stating that the system remains in the instantaneous eigenstate of the Hamiltonian if the Hamiltonian is slowly varying in time. In the case of Landau-Zener processes where \( n \) time dependent levels with an energy linearly varying in time \( v_n t \) intersect with \( m \) levels, with a constant interaction matrix element \( \tau_{nm} \), the adiabatic requirement is

\[
v_1\hbar, v_2\hbar, \ldots v_n\hbar \ll \tau_{1,1}, \ldots \tau_{1,m}, \ldots \tau_{n,1}, \ldots \tau_{n,m}.
\] (2.25)

2.3. Quantum computers

In classical computation all information is stored into classical bits having two distinct values 0 and 1. In early 1980s Benioff \([39]\), Manin \([40]\), Feynman \([41]\) and
Deutsch [42] concluded that computing abiding the laws of quantum mechanics would provide advantages over computing abiding the laws of classical mechanics. In quantum computing, the basic unit of information is a quantum bit (qubit). In contrast to a classical bit, other than the pure $|0\rangle$ and $|1\rangle$ states a qubit can also be in a quantum superposition of these states $(|0\rangle \pm |1\rangle)/\sqrt{2}$. Furthermore, two or more quantum particles can be correlated in such a way that one cannot distinguish between the quantum states of the particles. This phenomenon is called entanglement and it has been described as “spooky action at a distance” by Albert Einstein. Einstein, Podolski and Rosen devised a thought experiment proving that quantum mechanics violates local realism and is therefore incomplete [43].

In 1964 John S. Bell derived a theorem stating that quantum mechanics violates either locality or realism [44]. Locality means that distant objects cannot influence one another on timescales shorter than the time it takes a photon to propagate between them. The context of realism was defined a bit differently compared to modern philosophy. Realism in the context of quantum mechanics means that all experimental outcomes possible prior to the experiment could have occurred as an experimental outcome. In his seminal paper Bell also proposed an experiment that would prove his theorem was correct and that quantum mechanics indeed violates either locality or realism. Since then many researchers have conducted experiments proving that quantum mechanics indeed violates locality or realism [45–50]. Latest cutting edge experiments with spins in nitrogen vacancies [51] and entangled photons [52, 53] have simultaneously closed loopholes in the Bell experiments, proving the Bell theorem below the statistical error margin.

Quantum computing exploits superposition and entanglement to create quantum processors comprising of correlated qubits. One of the most common used protocols to encrypt data is the RSA protocol. The security of the RSA protocol is based on the fact that factoring products of prime numbers with classical computers is a difficult, time demanding process. On the other hand, Shor’s quantum algorithm would allow factoring of integers in polynomial times, significantly outperforming sub-exponential times of the best known classical algorithm [54]. Another quantum algorithm outperforming classical algorithms is the Grover’s search algorithm [55]. Although it provides only quadratic speedup it can still significantly outperform classical algorithms when the number of entries is large.

1. Single qubit gates

A quantum gate is a device performing operations on qubits. All quantum gates can be represented with unitary matrices and visualized as rotations on the Bloch sphere [22, 56].
The Haddamard gate is extremely important for quantum computing

\[
H = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}.
\]  

(2.26)

If the input state was \(|0\rangle\) the Haddamard gate yields \((|0\rangle + |1\rangle)/\sqrt{2}\). Furthermore, if the input state was \(|1\rangle\) the Haddamard gate \(H\) yields \((|0\rangle - |1\rangle)/\sqrt{2}\). The \(X\) gate rotates the state of the qubit for an angle of \(\pi\) around the \(x\) axis of the Bloch sphere

\[
X = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}.
\]  

(2.27)

Likewise, the \(Y\) gate rotates the state of the qubit for an angle of \(\pi\) around the \(y\) axis of the Bloch sphere

\[
Y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}.
\]  

(2.28)

The \(Z\) gate rotates the state of the qubit for an angle of \(\pi\) around the \(z\) axis of the Bloch sphere

\[
Z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.
\]  

(2.29)

The phase shift gate \(R_\phi\) adds a phase of local \(\phi\) to one of the qubit states while it leaves the other qubit state unchanged, for instance \(|0\rangle \rightarrow |0\rangle\) and \(|1\rangle \rightarrow \exp(i\phi)|1\rangle\). It corresponds to a rotation of the qubit state on the Bloch sphere for an azimuthal angle \(\phi\)

\[
R_\phi = \begin{pmatrix} 1 & 0 \\ 0 & e^{i\phi} \end{pmatrix}.
\]  

(2.30)

2. Two qubit gates

Two qubit gates simultaneously operates on two qubits [22, 56]. The SWAP gate swaps the state of two qubits.
2. Spin qubits in semiconductor quantum dots

\[ \text{SWAP} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \quad (2.31) \]

in the \{\ket{0}\ket{0}, \ket{1}\ket{0}, \ket{0}\ket{1}, \ket{1}\ket{1}\} basis. A universal gate is such a gate which, combined with single qubit gates, performs any two qubit operation. SWAP gate is not universal as it has too much symmetry, however performing square root of a SWAP operation is an universal operation when combined with single qubit gates.

\[ \sqrt{\text{SWAP}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \frac{1}{2}(1+i) & \frac{1}{2}(1-i) & 0 \\ 0 & \frac{1}{2}(1-i) & \frac{1}{2}(1+i) & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \quad (2.32) \]

in the \{\ket{0}\ket{0}, \ket{1}\ket{0}, \ket{0}\ket{1}, \ket{1}\ket{1}\} basis.

Another important gate for quantum computing is the controlled-NOT gate CNOT. To understand the operation of the CNOT gate one of the qubits will be addressed to as the control qubit and the other will be addressed as the target qubit. If the control qubit was in the \ket{0} state the state of the target qubit is unchanged. However, if the state of the control qubit is \ket{1} the state of the target qubit is changed. On operation like this is described with the following unitary matrix

\[ \text{CNOT} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix}. \quad (2.33) \]

Common alternatives to the CNOT and \sqrt{\text{SWAP}} gates are the CSIGN gate, also known as CPHASE or CZ \[56\]

\[ \text{CSIGN} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & -1 \end{pmatrix}, \quad (2.34) \]

the iSWAP gate, implemented commonly in superconducting qubits,
2. Spin qubits in semiconductor quantum dots

\[ \sqrt{i \text{SWAP}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & i & 0 \\ 0 & i & 0 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \]  

(2.35)

the SWAP\(^\alpha\), occurring commonly in spintronic quantum computation,

\[ \text{SWAP}^\alpha = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \frac{1}{2} (1 + e^{i \pi \alpha}) & \frac{1}{2} (1 - e^{i \pi \alpha}) & 0 \\ 0 & \frac{1}{2} (1 - e^{i \pi \alpha}) & \frac{1}{2} (1 + e^{i \pi \alpha}) & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \]  

(2.36)

and the Berkley B gate

\[ B = \begin{pmatrix} \cos \left(\frac{\pi}{8}\right) & 0 & 0 & i \sin \left(\frac{\pi}{8}\right) \\ 0 & \cos \left(\frac{3\pi}{8}\right) & i \sin \left(\frac{3\pi}{8}\right) & 0 \\ 0 & i \sin \left(\frac{3\pi}{8}\right) & \cos \left(\frac{3\pi}{8}\right) & 0 \\ i \sin \left(\frac{\pi}{8}\right) & 0 & 0 & \cos \left(\frac{\pi}{8}\right) \end{pmatrix}. \]  

(2.37)

It should be noted that any two qubit gate can be implemented by a combination of any other two qubit gate and single qubit gates.

3. The DiVincenzo criteria

In his groundbreaking paper, David DiVincenzo proposed a set of 5 criteria, necessary in order to achieve computing abiding the laws of quantum mechanics [57].

1) Qubits must be well characterized and a must be scalable. A subspace defining the qubit states must be well defined. Lets assume a Hilbert space containing \( N > 2 \) states exists. One must be able to identify the 2 states comprising the qubit subspace. Furthermore, the possibility to build a quantum processor having many interacting qubits must exist.

2) The state of the qubit must be reliably initialized.

3) All quantum bits must be isolated from corruptive couplings to their environments. In semiconductor system this requirement is quite difficult achieve due to the coupling of qubits to charge noise [58], nuclear spin noise [59] and interface defects [17]. A noisy environment coupled to the qubit causes decoherence i.e., the qubit starts behaving like a classical mixture and the quantum mechanical
2. Spin qubits in semiconductor quantum dots

nature of the qubit is irreversibly lost.

4) A universal set of single and two qubit gates, necessary for quantum computing must exist for a given qubit. As isolating the qubit fully from the environment completely is impossible this requirement is connected with the previous one. In the sense that the time in which single and two qubit gates are performed much be much shorter than the typical time in which the qubit decoheres.

5) The state of the qubit must be reliably measured after the manipulation of the qubit.

2.4. Inhomogeneous dephasing, transverse and longitudinal relaxation

Inhomogeneous dephasing is a reversible loss of qubit coherence. The loss of coherence occurs when the splitting between qubit eigenstates $|0\rangle$ and $|1\rangle$ or the interaction between the qubit eigenstates differ for different realizations of the qubit. Inhomogeneous dephasing time $T_2^*$ is the average time, after which a qubit loses quantum mechanical behavior, thus evolving into a classical mixture of states. For a single electron spin qubit in a single quantum dot and a two electron spin qubits in a double quantum dot a typical $T_2^* \sim 1 - 10$ ns, mainly limited by the uncertainty in the magnetic field originating from nuclear spins. Inhomogeneous dephasing can be reversed by a sequence of $\pi/2 - \pi - \pi/2$ pulses (the so-called Hahn echo [60]). Inhomogeneous dephasing can be understood on a model involving 2 states $|0\rangle$ and $|1\rangle$. A superposition of the $|0\rangle$ and $|1\rangle$ states is prepared, described by the following density matrix

$$\rho(0) = \begin{pmatrix} 0.5 & 0.5 \\ 0.5 & 0.5 \end{pmatrix}. \quad (2.38)$$

After waiting a sufficiently long time, the density matrix of the system becomes

$$\rho(t = \infty) = \begin{pmatrix} 0.5 & 0 \\ 0 & 0.5 \end{pmatrix}, \quad (2.39)$$
due to inhomogeneous dephasing.

Transverse relaxation is an irreversible loss of qubit coherence due to the fact that the qubit splitting or the interaction between the qubit eigenstates varies in time during the operation of the qubit. Unlike the inhomogeneous dephasing the transverse relaxation cannot be removed with a spin echo [60]. Furthermore, it is the factor limiting the coherence time of the qubit. Maximally achieved transverse relaxation times of $T_2 \sim 200 \mu$s for a CPMG sequence [61] for a $S - T_0$ qubit.
2. Spin qubits in semiconductor quantum dots

embedded in an electrostatically defined double quantum dot and $T_2 \sim 1.2 - 4.5 \text{ms}$ for a single electron spin embedded in a self assembled quantum dot. Nuclear dipole-dipole interactions are a mechanism producing a time dependent magnetic field limiting the coherence of the electron spin qubit in both of this realizations [62]. For a self assembled quantum dot in InGaAs the quadrupolar interactions slow down dipole-dipole mediated nuclear spin dynamics [63, 64], thus potentially allowing for long living electron spin qubits.

When longitudinal relaxation occurs, one of the qubit states (for instance $|1\rangle$) relaxes towards the state closest to the thermal equilibrium state (for instance $|0\rangle$). The longitudinal relaxation time is sometimes refereed to as the spin-lattice relaxation time and happens on $T_1 \sim \text{ms}$ timescales in semiconductor quantum dots. The excess energy between the qubit eigenstates in a qubit relaxation process is usually transmitted to a phonon. The following relation between the inhomogeneous dephasing time, transverse relaxation time and longitudinal relaxation time holds

$$T_2^* \leq T_2 \leq 2T_1.$$  \hspace{1cm} (2.40)

2.5. Single electron spin qubits in single quantum dots

Since the seminal proposal of Loss and DiVincenzo [18], many groups have tried to embed a spin qubit based on the single electron spin in a quantum dot [65–68]. The logical qubit states $|0\rangle$ and $|1\rangle$, are the projection of the spin to the direction of the external magnetic field $|\uparrow\rangle$ and $|\downarrow\rangle$. The $\uparrow$ and $\downarrow$ stand for the values of the spin projection $m_s = 1/2$, $m_s = -1/2$ respectively.

The initialization of single electron spin states can be achieved by coupling a quantum dot to a nearby lead [68]. Assuming that the width of the Fermi-Dirac distribution of the lead is smaller than the Zeeman splitting of the electron spin, the chemical potential of the lead is tuned between the electron spin eigenstates. If this is the case the electron spin can only tunnel to the lower eigenstate and this is how the electron spin can be initialized [68].

Two single qubit axes of control are given by the external magnetic field (taken along $z$) and the inhomogeneous in-plane magnetic field. The inhomogeneous in-plane magnetic field can originate from the spin-orbit field [66, 69], stray magnetic field of a micromagnet [70, 71] or random in-plane nuclear magnetic fields [72].

When the in-plane magnetic field originates from the spin-orbit field, stray magnetic field of a micromagnet or random in-plane nuclear magnetic field, the wavefunction of the electron needs to be periodically displaced inside the quantum dot [17, 69, 73]. As the wavefunction of the electron is oscillating back and fourth, the electron experiences a periodic time-dependent magnetic field and coherently
changes the spin. The periodic oscillations of the electron wavefunction require a source of microwaves. This technique of controlling the electron spin is called Electric Dipole Spin Resonance (EDSR).

However, the efficiency of EDSR is often limited by an unknown static component of the magnetic field, parallel with the \( z \)-axis, originating in the unknown static magnetic field of the nuclear spins. This is often the case in InGaAs as this material does not have stable isotopes with a non-zero value of the nuclear spin. On the other hand natural Si has \( \approx 95.3\% \) of stable isotopes with a zero value of the projection of the nuclear spin. The remaining \( \approx 4.7\% \) can be eliminated with isotopic purification techniques [74–76]. When the unknown magnetic field of the nuclear spins is present the electron spin is not rotating around the \( x \)-axis but around a sum axis of the nuclear spin magnetic field and the external magnetic field (parallel with \( z \)) and the \( x \)-axis of control.

However, the magnetic field of the nuclear spins is also varying in time. The nuclear spins are going to experience the magnetic field of the electron and are going to start to precess in this field at different rates. This precession of the nuclear spins in the magnetic field of the electron is called Knight shift and happens on \( \mu s \) timescales [62]. As a consequence the electron spin is going to experience a time dependent magnetic field causing the electron spin to decohere.

Furthermore, the nuclear spins are also exchanging angular momentum with distant dipole-dipole interactions [62]. This can be understood on a simple example comprising of two nuclear spins, one close to the electron spin and one further

![Figure 2.2. Quantum control of single electron qubit states on the Bloch sphere.](image)
away from the electron spin. The initial system is a $|\uparrow\rangle$ nuclear spin close to the electron spin and a nuclear spin further away from the electron spin initially in the $|\downarrow\rangle$ state. When the close and the distant nuclear spin exchange spin angular momentum the electron spin experiences a small change in the nuclear magnetic field, as the two nuclear spins are interacting with the electron spin with different magnitudes.

One of the methods for readout of the single electron spin qubit is spin-to-charge conversion \cite{68}. Similarly to when the electron spin is initialized a chemical potential of a nearby lead is tuned so that only one of the spin eigenstates can tunnel out of the quantum dot. If a tunneling event occurs the current of a nearby quantum point contact is changed accordingly. An alternative method for spin readout is having an adjacent quantum dot near to the spin qubit, not occupied with an electron \cite{77, 78}. The adjacent quantum dot is required to have a different Zeeman splitting compared to the spin qubit quantum dot. This can be achieved by $g$-factor engineering \cite{79} or by embedding a micromagnet on top of one of the quantum dots. The energy levels of one of the qubit states (for instance the spin down state) are aligned in the spin qubit quantum dot and the adjacent quantum dot. If the spin qubit was in the spin down state a tunneling event occurs and a nearby quantum point contact modifies the current. On the other hand, the spin up state is forbidden to tunnel to the adjacent quantum dot due to the large energy mismatch between the spin up states in the spin qubit quantum dot and the adjacent dot.

The exchange interaction produces a convenient platform for implementing two qubit gates between two single electron spin qubits \cite{18}. However, the exchange interaction is highly susceptible to electric noise. The exchange interaction can mediate $\sim 10$ coherent exchange oscillations before the quantum mechanical nature of the two qubit state is irreversibly lost \cite{58}. A “sweet spot” is such a value of the detuning between the two quantum dots, in which the exchange interactions would be first order insensitive to the detuning noise. A double quantum dot loaded with two electrons only has a trivial “sweet spot”, one in which the exchange interaction is close to zero. Possible ways of reducing the sensitivity of exchange interaction to electric noise is to control the exchange by controlling the tunneling between the dots instead of the detuning \cite{80, 81}. As shown in Chapter 6, another way of reducing the sensitivity of the exchange interaction to electric noise is to perform two qubit operations with an adjacent quantum dot between the spin qubit quantum dots. A system like this has four “sweet spots”, giving rise to a possibility of charge-noise-insensitive exchange interaction.
2.6. Two electron spin qubits in double quantum dots

An alternative to the single electron spin qubit is the two electron spin qubit [58, 59, 82–87]. The logical qubit states $|0\rangle$ and $|1\rangle$ are the singlet $S(1,1) = (|\uparrow\rangle_L |\downarrow\rangle_R - |\downarrow\rangle_L |\uparrow\rangle_R)/\sqrt{2}$ and the $m_s = 0$ triplet $T_0 = (|\uparrow\rangle_L |\downarrow\rangle_R + |\downarrow\rangle_L |\uparrow\rangle_R)/\sqrt{2}$, where the indices $L$ and $R$ stand for the left and the right quantum dot respectively. The $\uparrow$ and $\downarrow$ stand for the values of the spin projection $m_s = 1/2$, $m_s = -1/2$ respectively. Alternatively, one can use the $S = (|\uparrow\rangle_L |\downarrow\rangle_R - |\downarrow\rangle_L |\uparrow\rangle_R)/\sqrt{2}$ and one of the triplets $m_s = 1$, $T_+ = |\uparrow\rangle_L |\uparrow\rangle_R$ or $m_s = -1$, $T_- = |\downarrow\rangle_L |\downarrow\rangle_R$ as qubit states.

Similarly to the case of the single electron spin qubit, the initialization of qubit states is done via lead-to-dot tunneling [59]. One of the quantum dots (L) in the double quantum dot system is biased so that the singly occupied singlet becomes energetically favorable. When lead-to-dot relaxation occurs the only state that tunnels into the quantum dot is the doubly occupied singlet state $S(2,0) = (|\uparrow\rangle_L |\downarrow\rangle_L - |\downarrow\rangle_L |\uparrow\rangle_L)/\sqrt{2}$.

Two axes of control are given by the difference in Zeeman energies of the double quantum dot and the exchange interaction Fig. 2.3. Spin readout is achieved by detuning one of the quantum dots (for instance L) so that the doubly occupied

![Diagram](image-url)\)

Figure 2.3. Quantum control of $S - T_0$ qubit states on the Bloch sphere.
2. Spin qubits in semiconductor quantum dots

singlet $S(2, 0)$ is energetically favorable. If the system was in the singly occupied singlet state after the manipulation $S(1, 1)$, the singly occupied singlet tunnels to a doubly occupied singled thus modifying the current of a nearby QPC. If the system was in the triplet state $T_0$ a tunneling event is forbidden by the fact that the singly occupied triplet $T_0(2, 0)$ is much higher in energy, thus the current of a nearby QPC is unchanged.

Similarly to the case of the single electron spin qubit, the coherence of the $S - T_0$ qubit is limited by the uncertainties of the nuclear spin magnetic field and charge noise [85, 88]. The difference of the Zeeman fields of the double quantum dot provide one axis of rotation of the qubit, and the qubit is therefore sensitive to an uncertainty of the nuclear difference field. Feedback loops [89] and CPMG sequences [90] can extend the coherence time of the $S - T_0$ qubit up to $T_2 = 200 \mu$s. Another alternative is dynamical nuclear polarization (DNP), a transfer of polarization from the electron spin to the nuclear spin system. The efficiency of DNP can be limited by spin-orbit interaction, as shown in Chapter 3.

Two qubit operations between $S - T_0$ qubits [91, 92] can be performed by capacitively coupling the qubits. Charge noise is a limiting factor to two qubit coupling. The influence of charge noise to the efficiency of two qubit operation can be diminished by engineering strong Zeeman field gradients between the double quantum dots embedding the qubits [92].

2.7. Three electron spin qubits

The exchange interaction Hamiltonian commutes with both the square of the spin operator $S^2$ and the $z$-component of the spin operator $S_z$. Therefore, the logical states of a qubit controlled with only exchange interaction would have to have the same spin $s$ and spin projection $m_s$ quantum numbers. The most simple qubit of such sort is the so-called exchange only qubit comprising of three electrons in a triple quantum dot [93–95]. The logical qubit states comprise of the $|0\rangle = |S\rangle|\uparrow\rangle$ and $|1\rangle = (2/3)^{1/2}|T_+\rangle|\downarrow\rangle - (1/3)^{1/2}|T_0\rangle|\uparrow\rangle$ states, all having $s = 1/2$ and $m_s = 1/2$.

Initialization is achieved by pulsing one of the outer quantum dots to the $(2, 0, 1)$ or $(1, 0, 2)$ charge configurations. Only the $|S\rangle|\uparrow\rangle$ can tunnel to the $(2, 0, 1)$ charge configuration because the doubly occupied triplet state is much higher in energy. Readout is achieved by pulsing again to the $(2, 0, 1)$ charge configurations, with only the $|S\rangle|\uparrow\rangle$ state being energetically allowed to tunnel to the $(2, 0, 1)$ charge configuration.

In the triangular arrangement of the triple quantum dot three exchange operations suffice for single qubit control. In the linear arrangement minimally
2. Spin qubits in semiconductor quantum dots

\[ |0\rangle = |S\rangle|\uparrow\rangle \]

\[ J_{23} \]

\[ |0\rangle - |1\rangle / \sqrt{2} \]

\[ |0\rangle + |1\rangle / \sqrt{2} \]

\[ |1\rangle = (2/3)^{1/2} |T_+\rangle|\downarrow\rangle - (1/3)^{1/2} |T_0\rangle|\uparrow\rangle \]

Figure 2.4. Universal quantum control of three electron spin qubit states on the Bloch sphere.

four exchange operations are needed. CNOT gate is achieved by 19 exchange interaction between the two exchange only qubits or by having cavity mediated dipole-dipole coupling. The resonant exchange (RX) qubit is an alternative realization of the exchange only qubit, which can be operated close to a sweet spot with first order insensitivity to charge noise. One of the exchange couplings is operated in the oscillatory manner with the frequency of oscillation of one the exchange interactions matching the frequency of the qubit splitting. Single qubit operation of the exchange only qubit happens on \( \sim 2.5 \) ns and with a \( T_2 \sim 19 \) \( \mu \)s.

1. The hybrid qubit

An alternative to the exchange only qubit is the so-called hybrid qubit, achieved by filling the double quantum dot with three electrons \([96-99]\). The qubit states are given by \( |0\rangle = |S\rangle_L|\downarrow\rangle \), \( |1\rangle = \sqrt{1/3} |T_0\rangle|\downarrow\rangle - \sqrt{2/3} |T_-\rangle|\uparrow\rangle \) both having \( s = 1/2 \) and \( m_s = -1/2 \). The singlet state is antisymmetric with respect to electron exchange, requiring the orbital part to have a symmetric wavefunction in order to preserve the antisymmetry of the total wavefunction. In contrast to that, the triplet state is symmetric, requiring the orbital part of the wavefunction to be antisymmetric in order to preserve the antisymmetry of the total wavefunction.
2. Spin qubits in semiconductor quantum dots

\[ |E\rangle = |\downarrow \rangle |S\rangle \]

\[ |0\rangle = |S\rangle |\downarrow \rangle \]

\[ |1\rangle = \sqrt{\frac{1}{3}} |T_0\rangle |\downarrow \rangle - \sqrt{\frac{1}{3}} |T_-\rangle |\uparrow \rangle \]

Figure 2.5. The control of the hybrid qubit. The \( |0\rangle = |S\rangle_L |\downarrow \rangle \) tunnels virtually to the intermediate state \( |E\rangle = |\downarrow \rangle_L |S\rangle_R \), followed by a tunneling event to the \( |1\rangle \) state.

The confinement in electrostatically defined quantum dots is often modeled as harmonic, having a symmetric ground state and an antisymmetric first excited state.

The initialization and readout of the qubit rely on the fact that the triplet state has a much shorter lead-to-dot and relaxation time compared to the singlet state [96, 97]. This happens because the antisymmetric spatial wavefunction has a maximum of the spatial distribution close to the edge of quantum dot near the lead. In contrast to that, the spatial part of the singlet wavefunction has a maximum of the spatial distribution in the center of the quantum dot, therefore having a smaller overlap and a longer relaxation time to the lead.

The energy difference between the qubit states provides with one axis or rotation, the \( z \) axis. In the case of the hybrid qubit the leading contribution to the energy difference of the qubit states is given by the orbital splitting which is on the order of 0.5 \( \sim \) 5 meV in Si and InGaAs quantum dots. The orbital splitting can be tuned by varying the confinement potential, yielding an axis of rotation with a highly tunable rotation frequency.

The second axis of rotation is given by a virtual tunneling process to an intermediate state \( |E\rangle = |\downarrow \rangle_L |S\rangle_R \) Fig. 2.5, the \( x \) axis. Both the \( x \) and the \( z \) control happens on \( \sim 100 \) ps timescales, allowing for \( \sim 100 \) qubit rotations during the coherence time of the qubit with the maximally achieved 93\% and 95\% fidelity of the \( x \) and the \( z \) gate respectively [96, 98]. Similarly to the exchange only qubit the two qubit operations are mediated by exchange interaction or by capacitive coupling.
3. Interplay of spin-orbit and hyperfine interactions in dynamical nuclear polarization in semiconductor quantum dots

3.1. Summary

In this chapter the interplay of spin-orbit and hyperfine interactions is theoretically studied during dynamical nuclear polarization (DNP). The DNP is studied for the case of two-electron semiconductor double quantum dots near the singlet ($S$) - triplet ($T_+$) anticrossing. The goal of the scheme under study is to extend the singlet ($S$) - triplet ($T_0$) qubit decoherence time $T^*_2$ by dynamically transferring the polarization from the electron spins to the nuclear spins. This polarization transfer is achieved by cycling the electron spins over the $S - T_0$ anticrossing. Here, the influence of Rashba and Dresselhaus spin orbit-interaction to hyperfine mediated dynamical polarization is investigated. In addition to $T^*_2$, the singlet return probability $P_s$ is determined, a quantity that can be measured in experiments. The results suggest that the spin-orbit interaction establishes a mechanism that can polarize the nuclear spins in the opposite direction compared to hyperfine mediated nuclear spin polarization. In materials with relatively strong spin-orbit coupling, this interplay of spin-orbit and hyperfine mediated nuclear spin polarizations prevents any notable increase of the $S - T_0$ qubit decoherence time $T^*_2$.

This chapter is adapted from Marko J. Rančić and Guido Burkard, Phys. Rev. B 90, 245305 (2014)
3. Interplay of spin-orbit and hyperfine interactions in DNP in semiconductor QDs

3.2. Introduction

Electron spins in semiconductor quantum dots are considered to be excellent candidates for qubits [18]. In order for a full scale quantum computer to be produced, a successful fulfillment of the DiVincenzo criteria [57] is necessary. Accurate qubit manipulation [100, 66] and reliable state preparation [59] are some of the requirements that have been satisfied in the past years. Techniques for qubit identification and fast readout are also known, e.g., the spin readout for a two-electron double quantum dot is most commonly done in the regime of Pauli spin blockade [101] using spin to charge conversion measurements [83]. Still, one challenge remains - sufficiently isolating the qubit from the corruptive effects of its surroundings.

Due to the influence of its surroundings, a qubit will irreversibly lose information. Different types of information losses happen on different time scales. The time in which a qubit relaxes to a state of thermal equilibrium is the relaxation time $T_1$, whereas the time in which a qubit loses coherence due to the collective effects of its surroundings is the decoherence time $T^*_2$. Although experimental and theoretical solutions for overcoming these information losses have been steadily developed for years [85,102], overcoming qubit decoherence caused by a fluctuating nuclear spin bath is still an ongoing task.

Silicon [103] and graphene [104] have stable isotopes with a zero nuclear spin. Therefore, they can be isotopically purified leaving only spin zero nuclei which do not contribute to the electron spin qubit decoherence. On the other hand, III-IV semiconductors, and particularly In$_x$Ga$_{1-x}$As structures, only have stable isotopes with a non-zero nuclear spin. An electron confined in a typical In$_x$Ga$_{1-x}$As quantum dot interacts with $10^3 - 10^6$ nuclear spins, which contribute strongly to electron spin qubit decoherence. Optically [105-106] or electrically polarizing the nuclear spins can prolong the coherence times of electron spins. Such a polarization of nuclear spins is achieved by transferring angular momentum from the electron spins to the nuclear spins in a procedure called dynamical nuclear polarization (DNP) [107].

A suitable system for conducting DNP is a gate defined double quantum dot loaded with two electrons. There has been a variety of proposals [82, 100] to use DQDs as qubits, e.g., by focusing on the singlet $|S\rangle = 1/\sqrt{2}(|\uparrow\rangle|\downarrow\rangle - |\downarrow\rangle|\uparrow\rangle)$ and triplet $|T_0\rangle = 1/\sqrt{2}(|\uparrow\rangle|\downarrow\rangle + |\downarrow\rangle|\uparrow\rangle)$ logical subspace [90], where the generated nuclear difference field and the exchange interaction are used to perform universal control of the qubit on the Bloch sphere. Other than the already mentioned DNP, the effects of dephasing caused by a nuclear spin bath, can be canceled by applying a Hahn echo sequence [60], or the more elaborate CPMG sequences [90].

The generation of a nuclear gradient field, required to control the $S - T_0$ qubit
3. Interplay of spin-orbit and hyperfine interactions in DNP in semiconductor QDs

[90], can be achieved by cycling the electron spins over the anticrossing between the singlet $|S\rangle = 1/\sqrt{2}(|\uparrow\rangle|\downarrow\rangle - |\downarrow\rangle|\uparrow\rangle)$ and triplet $|T_+\rangle = |\uparrow\rangle|\uparrow\rangle$ states. During such a $S - T_+$ cycle, the electron spins transfer polarization to the nuclear spins [108], and a nuclear difference field is generated. Furthermore, a higher degree of nuclear spin polarization causes a longer spin coherence time of the $S - T_0$ qubit. In materials with sizable spin-orbit interaction, the spin-orbit interaction induces electron spin flips, and this mechanism competes with the hyperfine mediated electron spin flips required for DNP. In such materials, the interplay of spin-orbit and hyperfine effects on nuclear spin preparation schemes is theoretically explored in the vicinity of the $S - T_+$ anticrossing.

It is assumed that the dots are embedded in the semiconductor material In$_x$Ga$_{1-x}$As with $0 \leq x \leq 1$. 150 nuclear spins per dot are modeled fully quantum mechanically, keeping track of how the probabilities and coherences of all nuclear states change in time. Compared to the model under study, recent models treating more [108] or fewer [109] nuclear spins fully quantum mechanically do not take into account the spin-orbit interaction. Although there has been some work on the interplay of spin-orbit and nuclear effects in GaAs double quantum dots [110,84], none of these theoretical frameworks treat the nuclear spin dynamics fully quantum mechanically, nor investigate the nuclear spin dynamics when subjected to a large number ($\approx 300$) of DNP cycles. On the other hand, there has been no theoretical work to describe the $S - T_+$ DNP in materials having strong spin-orbit interaction, e.g., InAs. Experiments in InAs have been carried out with a single electron spin in a single quantum dot [111], or in a double quantum dot, by using a different, more elaborate pulsing sequence [112]. As a consequence of the fully quantum treatment a precise estimations of $T^*_2$ will be given, a comparison to known experiments in GaAs [113] will be made, and a value for $T^*_2$ in In$_x$Ga$_{1-x}$As will be predicted. The results presented here can also be extrapolated to materials with even stronger spin-orbit coupling as compared to InAs such as, e.g., InSb.

This chapter is organized as follows. In Section 3.3 the model is described, in Section 3.4 the total nuclear spin angular momentum basis is discussed. The total angular momentum basis allows for a significant reduction of the dimension of the Hilbert space. In Section 3.5 the time evolution during the DNP cycle is studied, in Section 3.6 results on In$_{0.2}$Ga$_{0.8}$As are presented, a material with an intermediate strength of spin-orbit interaction, and in Section 3.7 results for different abundances of indium in In$_x$Ga$_{1-x}$As are presented before concluding.
3. Interplay of spin-orbit and hyperfine interactions in DNP in semiconductor QDs

3.3. Model

The confinement in a quantum dot is modeled with a quadratic potential and the electronic wave functions are calculated according to the Hund-Mulliken theory [114]. The approach used here is a good approximation in the regime where half of the interdot separation $a$ is larger than the effective Bohr radius, $a \gtrsim a_B = \sqrt{\hbar/m^*\omega_0}$. Here, $\omega_0$ is the circular frequency of the confining potential, which is later assumed to be $\hbar\omega_0 = 3.0$ meV, and $m^*$ is the effective electron mass ($m^* = 0.067 m_0$ for GaAs and $m^* = 0.023 m_0$ for InAs). The interdot separation $2a$ needs to be chosen sufficiently large, due to the fact that the Hund-Mulliken theory is valid in the regime of weakly interacting quantum dots. On the other hand, the extended tunneling matrix element $t_H$ needs to be non-vanishing, so that the DNP sequence is still possible. Therefore, for In$_{0.2}$Ga$_{0.8}$As, which is the material under study in Section 3.6, $t_H \approx 0.01 U$, where $U$ is the Coulomb energy of the electrons. This is why $a = 46.3$ nm is set. A magnetic field of $B = 110$ mT is applied perpendicular to the plane spanned by the [110] and [110] crystallographic axes, see Fig. 3.1. The specific value of the magnetic field is chosen so that the $S-\bar{T}_+$ anticrossing is located at $\varepsilon \approx 3U/2$, where $\varepsilon$ is the energy difference between the quantum dots, Fig. 3.2.

All stable isotopes of gallium and arsenic have a nuclear spin $j = 3/2$, while stable isotopes of indium have a nuclear spin $j = 9/2$. Here, a simplified model is discussed, in which all of the nuclear spins are assumed to be $j = 1/2$ [115]. To compensate the approximation of using $j = 1/2$, the hyperfine constant that is used through this chapter represents an average over all present nuclear spin isotopes and all possible values of the quantum number $j$. Also, spin-orbit effects depend strongly on the homogeneity of the distribution of In and Ga atoms in In$_x$Ga$_{1-x}$As. Here, a completely homogeneous distribution of In and Ga is assumed. For numerical convenience a geometry in which the [110], [110] crystallographic axes and the interdot connection axis $p_x$ lie in plane is modeled (Fig. 3.1). A numerical method is developed for modeling up to $N = 150$ nuclear spins per dot, a constraint imposed by current computational resources.

The total Hamiltonian describing the electronic and nuclear degrees of freedom is

$$H = H_0(\varepsilon) + H_{HF} + H_{so}. \quad (3.1)$$

Here $H_0(\varepsilon)$ is the non-relativistic Hamiltonian of two electrons in a QD [114],
3. Interplay of spin-orbit and hyperfine interactions in DNP in semiconductor QDs

\[
H_{0}(\varepsilon) = \begin{pmatrix}
U - \varepsilon & X & -\sqrt{2} t_H & 0 & 0 & 0 \\
X & U + \varepsilon & -\sqrt{2} t_H & 0 & 0 & 0 \\
-\sqrt{2} t_H & -\sqrt{2} t_H & V_{+} & 0 & 0 & 0 \\
0 & 0 & 0 & V_{+} + g\mu_{B}B_{z} & 0 & 0 \\
0 & 0 & 0 & 0 & V_{-} & 0 \\
0 & 0 & 0 & 0 & 0 & V_{-} - g\mu_{B}B_{z}
\end{pmatrix}, \quad (3.2)
\]

in the basis of \{S(2,0), S(0,2), S(1,1), T_{+}(1,1), T_{0}(1,1), T_{-}(1,1)\}. The letter \(S\) denotes the singlet state, and \(T_{+}, T_{-}, T_{0}\) are triplet states with the total spin projections \(m_{s} = +1, m_{s} = -1, m_{s} = 0\). The numbers in the parentheses indicate the charge state. More specifically, \((2,0)\) denotes a state where the left dot is occupied with two electrons and the right dot is empty, \((0,2)\) denotes a state where the right dot is being occupied with two electrons and the left dot is empty, and \((1,1)\) stands for each dot being occupied with one electron. The Hamiltonian [Eq. (3.2)] acquires time dependence through the bias energy \(\varepsilon\). To describe the DNP process, the bias energy \(\varepsilon\) will be assumed to be a linear function of time \(\varepsilon = rt\), where \(r = 2U/\tau\) is set, and where \(\tau = 50 \text{ ns}\) is the duration of the bias.

Figure 3.1. Geometry of the problem. The strength of spin-orbit interaction is tuned by varying the angle \(\theta\) between the [110] crystallographic axis and the interdot connection axis \(p_{\xi}\). Spin-orbit interaction generates an effective magnetic field \(\Omega\) along the \(y\) axis. The external magnetic field is perpendicular to the [110] - \(p_{\xi}\) plane.
3. Interplay of spin-orbit and hyperfine interactions in DNP in semiconductor QDs

![Two-electron spectrum of a DQD in InAs as a function of the interdot bias $\varepsilon$, obtained by diagonalizing the Hamiltonian $H_0$ [Eq. (3.2)].](image)

The energy $E$ and the detuning $\varepsilon$ are expressed in units of the Coulomb energy $U$. The parameters of the plot are the magnetic field $B = 1$ T, the Coulomb energy $U = 4.86$ meV, the extended tunneling hopping $t_H = 0.11$ meV, the triplet matrix element $V_+ = 2.16 \mu$eV, the doubly occupied singlet matrix element $V_- = 0.42 \mu$eV, half of the interdot separation $a = 73.6$ nm. Including hyperfine interaction and/or spin-orbit interaction opens up an avoided crossing $\Delta$ [86] (inset). The magnetic field is chosen large, as compared to the value in the remainder of the chapter, for visualization purposes. The $S(2, 0)$ and $S(0, 2)$ are singly occupied singlets, $S(1, 1)$ is the doubly occupied singlet. $T_+, T_0$ and $T_-$ are triplet states corresponding to $m_s = 1$, $m_s = 0$ and $m_s = -1$. The $S_-$ and $S_+$ are the lower and the upper hybridized singlet [see Eq. (3.4) and Eq. (3.5)].

Figure 3.2. Two-electron spectrum of a DQD in InAs as a function of the interdot bias $\varepsilon$, obtained by diagonalizing the Hamiltonian $H_0$ [Eq. (3.2)]. The energy $E$ and the detuning $\varepsilon$ are expressed in units of the Coulomb energy $U$. The parameters of the plot are the magnetic field $B = 1$ T, the Coulomb energy $U = 4.86$ meV, the extended tunneling hopping $t_H = 0.11$ meV, the triplet matrix element $V_+ = 2.16 \mu$eV, the doubly occupied singlet matrix element $V_- = 0.42 \mu$eV, half of the interdot separation $a = 73.6$ nm. Including hyperfine interaction and/or spin-orbit interaction opens up an avoided crossing $\Delta$ [86] (inset). The magnetic field is chosen large, as compared to the value in the remainder of the chapter, for visualization purposes. The $S(2, 0)$ and $S(0, 2)$ are singly occupied singlets, $S(1, 1)$ is the doubly occupied singlet. $T_+, T_0$ and $T_-$ are triplet states corresponding to $m_s = 1$, $m_s = 0$ and $m_s = -1$. The $S_-$ and $S_+$ are the lower and the upper hybridized singlet [see Eq. (3.4) and Eq. (3.5)].

The value of $r$ is chosen so that $\varepsilon = 2U$ at the beginning of the sweep ($t = 0$), $\varepsilon = 0$ at the end of the sweep ($t = \tau$), as in the experiment by Petta et al. [59].

The quantities in $H_0$ are the on-site Coulomb energy $U \sim 1$ meV, the coordinated hopping from one dot to the other $X \sim 0.1$ $\mu$eV, the doubly occupied singlet and triplet matrix elements, $V_+, V_- \sim 10$ $\mu$eV, and the extended hopping parameter, $t_H \sim 0.01U$ [114]. The Zeeman energy is given as $g\mu_BB_z$, where $g$ is the electron $g$ factor ($g = -0.44$ for GaAs, $g = -14.7$ for InAs), the Bohr magneton is $\mu_B = 5.79 \times 10^{-5}$ eV/T and $B_z = 110$ mT is the magnetic field. For an electron confined in an GaAs QD the Zeeman energy at this field is $E_z = 2.8 \times 10^{-6}$ eV.

Due to the fact that the $S-T_+$ transition is of interest, the attention is focused on the energy subspace spanned by the states $\{S(2, 0), S(1, 1), T_+(1, 1)\}$. The singlet $S(0, 2)$ is high in energy with respect to the other two singlets [cf. Fig. 3.2]
(for positive values of the detuning $\varepsilon$) whereas the remaining two singlets $S(2,0)$ and $S(1,1)$ are close in energy. The triplet states $T_0(1,1)$, and $T_-(1,1)$ are split off from the $T_+(1,1)$ by the Zeeman energy. It should be mentioned that the Hamiltonian [Eq. (3.2)] is treated within the adiabatic approximation, meaning that the system will remain in its instantaneous eigenstates. This allows one to obtain the eigenenergies by diagonalizing the Hamiltonian $H_0$ in the subspace of $\{S(2,0), S(1,1)\}$. As a result of the diagonalization the two hybridized singlets $|S_+\rangle, |S_-\rangle$ [114, 86] are obtained, with energies

$$E_{S_{\pm}} = \frac{U - \varepsilon + V_+}{2} \pm \sqrt{\frac{(U - \varepsilon + V_+)^2}{4} + 2t_H^2},$$

(3.3)

and eigenvectors

$$|S_-\rangle = c(\varepsilon)|S(1,1)\rangle + \sqrt{1 - c(\varepsilon)^2}|S(2,0)\rangle,$$

(3.4)

$$|S_+\rangle = \sqrt{1 - c(\varepsilon)^2}|S(1,1)\rangle - c(\varepsilon)|S(2,0)\rangle.$$ 

(3.5)

c(\varepsilon) = \cos \psi$ denotes the charge admixture coefficient which can be expressed with the charge admixture angle $\psi$, where

$$\cos 2\psi = \frac{U - V_+ - \varepsilon}{\sqrt{(U - V_+ - \varepsilon)^2 + 8t_H^2}}.$$ 

(3.6)

Only the transitions between the lower hybridized singlet $|S_-\rangle$ and triplet $|T_+\rangle$ are taken into account, because the upper hybridized singlet $|S_+\rangle$ is higher in energy, and therefore can be neglected, as shown in Fig. 3.2.

The spin-orbit term $H_{so}$ in the Hamiltonian is a function of the angle $\theta$ [cf. Fig 3.1] between the [110] crystallographic axis and the interdot connection axis $p_\xi$ [86],

$$H_{so} = \frac{\hat{\mathbf{\Omega}}(\theta)}{2} \cdot \sum_{s,t=\uparrow,\downarrow} (\hat{c}_{Ls}^\dagger \sigma^{st} c_{Rt} - h.c.),$$ 

(3.7)

where $\mathbf{\Omega}(\theta)$ is the spin-orbit effective magnetic field defined by

$$i\mathbf{\Omega}(\theta) = \langle \Phi_L | \hat{p}_\xi | \Phi_R \rangle ((\beta - \alpha) \cos \theta e_{[110]} + (\beta + \alpha) \sin \theta e_{[110]}).$$ 

(3.8)

Here, $\alpha$ and $\beta$ are the Rashba [8] and Dresselhaus [7] coefficients, the $c_{r,s}^\dagger$ operator creates an electron with spin $s=\uparrow, \downarrow$, in the right or left dot, $r=R,L$. Further,
\(\sigma_{s,t}\) is the vector of Pauli matrices and \(\Phi_{L,R}\) are the spatial parts of the wavefunctions corresponding to the left and the right dot respectively [86] and \(\hat{p}_\xi\) is the component of the momentum operator along the interdot connection axis.

For computational simplicity, the coordinate system is chosen such that the matrix elements of the spin-orbit part of the Hamiltonian [Eq. (3.7)] are always real. This is achieved by setting the \(e_y\) axis of the coordinate system parallel with \(\Omega\) [86], as shown in Fig. 3.1. When the spin-orbit interaction is excluded, the \(x\) and \(y\) axes are parallel to the crystallographic axes.

Finally, the hyperfine part of the Hamiltonian is given by [108]

\[
H_{HF} = S_1 \cdot h_1 + S_2 \cdot h_2 = \frac{1}{2} \sum_{i=1}^{2} (2S_i^z h_i^z + S_i^+ h_i^- + S_i^- h_i^+),
\] (3.9)

where \(S_i^{(\pm)}\) are the \(i\)th electron spin ladder operators, \(S_i^z\) and \(h_i^z\) are the \(z\) components of the \(i\)th electron spin operator and Overhauser field operator. Furthermore, \(h_i^\pm = h_i^x \pm ih_i^y\) are the ladder operators of the Overhauser field,

\[
h_i = \sum_{k=1}^{n(i)} A_{ik}^k I_i^k,
\] (3.10)

where \(I_i^k\) are the nuclear spin operators for the \(k\)th nuclear spin in contact with the \(i\)th electron spin. The strength of the hyperfine coupling between the \(i\)th electron and the \(k\)th nuclear spin is labeled \(A_{ik}^k\). In general \(A_{ik}^k\) has a different value for every nuclear spin, but this is simplified this by assuming a constant hyperfine coupling \(A_{ik}^k = A_{tot}/N\) [108, 109, 110, 116].

In general, the strength of the hyperfine interaction \(A_{ik}^k\) depends on the distance between the electron spin and the nuclear spin \(r_k\) in the following way \(A_{ik}^k = A_{tot} \exp \left[ -\frac{r_k^2}{\hbar \omega_0} \right]\) [117], where \(\hbar\) is the reduced Planck constant, \(m^*\) is the effective mass of the confined electron and \(\omega_0\) is the circular frequency of the confining potential. This means that the nuclear spins closer to the electron spin have a stronger chance of being polarized, compared to those nuclear spins that are further away from the electron spin. The electron triplet probability is given by the Landau-Zener formula \(P_{T+} = \exp \left[ -\frac{2\pi|E_{HF}|^2}{\hbar r} \right]\) [118-119]. However, the selection of \(r\) is such that \(\hbar r \gg |A_{ik}^k|^2\) for most nuclear spins, so most nuclear spin dynamics is happening near the maximum of the spatial distribution of the hyperfine constants, which validates the choice of averaging it.

Performing a diagonalization in the singlet subspace spanned by \(\{S(2,0), S(1,1)\}\), bias dependent singlet eigenfunctions are found and therefore Eq. (3.4) and Eq. (3.5) become time dependent. This implies that the coupling between the lower hybridized singlet \(|S_-\rangle\) and the \(|T_+\rangle\) triplet state is time dependent as
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compared to time independent coupling between the \( |S(1,1)\rangle \) and \( |S(2,0)\rangle \) singlets and the \( |T_+\rangle \) triplet. The time dependence of the coupling originates in the fact that the coupling depends on the charge state of the hybridized singlet [Eq. (3.4) and Eq. (3.5)]. The state \( S(2,0) \) couples to \( T_+ \) only via the spin-orbit interaction and \( S(1,1) \) couples to \( T_+ \) only by means of the hyperfine interaction. By using the wavefunctions of the lower hybridized singlet (see Eq. (3.4)) the matrix element of the Hamiltonian between the lower hybridized singlet \( |S^-\rangle \) and the triplet \( |T_+\rangle \) can be calculated

\[
\langle S^- | H | T_+ \rangle = c(\varepsilon) \langle S(1,1) | H_{\text{HF}} | T_+ \rangle + \sqrt{1 - c(\varepsilon)^2} \langle S(2,0) | H_{\text{so}} | T_+ \rangle. \tag{3.11}
\]

It should be mentioned that due to time dependent interactions, the model discussed here must go beyond the Landau-Zener model [118-119].

3.4. The basis of total angular momentum

In the model used here, all nuclear spins are treated as having spin \( j = 1/2 \). This means that the total number of nuclear spin states is \( \text{dim}(\mathcal{H}) = 2^N \), where \( N \) is the number of nuclear spins in a quantum dot. Because the total number of nuclear spin states scales exponentially with \( N \) it would be difficult to treat a large number (\( N = 150 \)) of nuclear spins with the computational power available. In order to make the problem treatable a basis change is made from the product basis \( \{\uparrow, \downarrow\} \), to the basis of total angular momentum \( \{|j,m\rangle\} \). Here, \( j \) is the total nuclear spin quantum number, \( 0 \leq j \leq N/2 \), and \( m \) is the total nuclear spin projection along the \( z \) axis, \( -j \leq m \leq j \). Now the total number of states can be written as

\[
\text{dim}(\mathcal{H}) = \sum_{j=0}^{N/2} \sum_{\text{perm}} (2j + 1) = 2^N. \tag{3.12}
\]

The inner sum runs over all permutation symmetries for a given value of \( j \). The basis of total angular momentum still scales as \( \text{dim}(\mathcal{H}) = 2^N \), but now certain states in the inner sum in Eq. (3.12) do not need to be taken into account, and states with higher \( j \) in the outer sum in Eq. (3.12) can be neglected due to the low probability of their occurrence.

Neither the hyperfine nor the spin-orbit interaction mix states with different \( j \), and thus the matrix representing the Hamiltonian is block diagonal with every block corresponding to a value of \( j = j_0, j_0 + 1, \ldots N/2 \) [109]. The value of \( j_0 \) depends on the parity of \( N \), for an even \( N \), \( j_0 = 0 \) and for an odd \( N \), \( j_0 = 1/2 \). The probability distribution of nuclear spin states, with respect to the quantum
number $j$ is a Gaussian (in the limit $N \rightarrow \infty$) with its maximum located at $\approx \sqrt{N/2}$, Fig. 3.3. From now this value of $j$ is refereed as its most likely value, $j_{ml} \approx \sqrt{N/2}$. The nuclear spin probability distribution, with respect to the number of nuclear spins per dot $N$ and quantum number $j$ is given by the following formula [120]

$$p(N, j) = \frac{(2j + 1)^2 N!}{(N/2 + j + 1)!(N/2 - j)!2^N}. \quad (3.13)$$

The $j$ and $m$ quantum numbers are generally not sufficient to describe all possible nuclear spin states. Other than $j$ and $m$, the nuclear spin states are described by their permutation symmetries. For example, for three nuclear spins defined by quantum numbers $j = 1/2$ and $m = 1/2$, there are two states $|1/2, 1/2\rangle$ and $|1/2, 1/2\rangle'$ with distinct permutation symmetries. These two states are not mixed by homogeneous hyperfine or by spin-orbit interactions. Furthermore, they remain equally probable as the matrix elements of the Hamiltonian only depend on $j$ and $m$ and not on the symmetry properties. Therefore, by evaluating the system for a certain symmetry $|1/2, 1/2\rangle$ the behavior of the state with a different permutation symmetry $|1/2, 1/2\rangle'$ is known. By generalizing this simple example

![Gaussian fit](image)

Figure 3.3. Initial nuclear spin probability distribution with respect to the quantum number $j$ for $N = 150$ nuclear spins $1/2$, where $j_{ml} = \sqrt{N/2}$ and $j_{max} = 18$. Throughout the calculations only the states $0 \leq j \leq j_{max}$ are considered (blue diamonds) and the states $j > j_{max}$ are not considered (black circles).
to $N$-spin systems the number of the states which need to be considered is significantly reduced. For every value of $j$ only one state of symmetry in Eq. (3.12) needs to be evaluated, and therefore for each value of $j$ the inner sum in Eq. (3.12) can be replaced by one representing term.

The number of states can be reduced further by choosing the maximum value of $j$, taken into consideration $j_{\text{max}}$, in a manner that $\sqrt{N}/2 \ll j_{\text{max}} \ll N/2$. The omission of all states with $j > j_{\text{max}}$ is justified because these states occur with a very low probability (see Fig. 3.3 and Eq. (3.13)). Now the total number of the states which are considered scales with $j_{\text{max}}$ as

$$\dim(H') = \sum_{j=0}^{j_{\text{max}}} (2j + 1) \approx (j_{\text{max}} + 1)^2 \ll 2^N.$$  \hspace{1cm} (3.14)

Due to the fact that the states with different $j$ do not mix by any interaction under consideration, the system can be analyzed for one value of $j$ at a time and finally averaged over all included values of $j$. By doing so, the averaging is taken over close to (but not exactly) 100% of all possible states. In the case under consideration, $N = 150$ nuclear spins per dot and $0 \leq j \leq 75$. Constraining $0 \leq j \leq j_{\text{max}} = 18$, the average is taken over 97.8% of all possible nuclear spin configurations, as shown in Fig. 3.3. The efficiency of this approach can be illustrated best if the number of states in the $\{\uparrow, \downarrow\}$ basis is calculated and compared to the number in $\{|j, m\rangle\}$ basis after only one symmetry state for every $j$ is considered and $0 \leq j \leq j_{\text{max}}$ taken into account. For $N = 150$, Eq. (3.12) yields $\dim(H) \approx 1.4 \times 10^{45}$ and for $j_{\text{max}} = 18$, Eq. (3.14) yields $\dim(H') = 361$.

### 3.5. Time evolution during DNP

A totally mixed state is assumed for the initial nuclear spin state. The electrons are initialized in a singlet state $S(2,0)$, where both electrons are occupying the same dot. Afterwards, the electronic system is driven with a finite velocity through the $S - T_\uparrow$ anticrossing (see Fig. 3.2) by varying the voltage bias $\varepsilon$. The electronic state is then measured, and finally the system is reset quickly to the initial state $S(2,0)$ [108]. Accordingly, the density matrix of the system $\rho$ is computed according to the update rule

$$\rho^{(i+1)} = M_S U \rho^{(i)} U^\dagger M_S + M_T U \rho^{(i)} U^\dagger M_T.$$  \hspace{1cm} (3.15)

Here $\rho^{(i)}$ and $\rho^{(i+1)}$ are the total density matrices before and after the $i$-th DNP step, $U$ is the unitary time evolution operator and $M_S$ and $M_T$ are the singlet and triplet projection operators [22]. They satisfy the relations $M_S + M_T = I$. 

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and $M_SM_T = 0$.

After the evolution of the system, a measurement of the electronic state takes place. This measurement procedure has two outcomes: either a singlet $S$ or a triplet $T_+$ is detected. The nuclear density matrix is updated accordingly,

$$\rho_n = P_S\rho_n^S + P_T\rho_n^T,$$

where $\rho_n$ is the nuclear density matrix and $P_S = \text{Tr}[M_SU^\dagger M_S]\rho S$ and $P_T = \text{Tr}[M_TU^\dagger M_T]\rho T$ are the singlet and the triplet outcome probabilities. The superscripts $S$ and $T$ stand for a nuclear density matrix related to the singlet and the triplet measurement outcome. For a certain value of $j$ the singlet return probability $P_S$ is calculated, alongside with the standard deviation of the nuclear difference field, $\sigma(z) = \sqrt{\langle (\delta h z)^2 \rangle - \langle \delta h z \rangle^2}$ [102]. After averaging over all included $j$, the standard deviation of the nuclear difference field is used to evaluate the $S - T_0$ spin qubit decoherence time, $T^*_2 = \hbar/\sigma(z)$ [102].

The propagator $U$ is computed by discretizing the time interval $(0, \tau)$. The model describes the passage through the anticrossing with $q = 100$ equally spaced, step-like time increments. The procedure of computing the propagator is the following: For every discrete point in time $t_i$ the Hamiltonian $H(t_i)$ is computed. The propagator for the fixed time point $t_i$ is approximated,

$$U_{t_i} = e^{-iH(t_i)\Delta t/\hbar},$$

with $\Delta t = \tau/q$. By repeating the procedure for every discrete step the total time evolution operator is obtained

$$U = U_{t_q}U_{t_{q-1}} \ldots U_{t_1}.$$
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Figure 3.4. System initialization and measurement outcomes. (a) Initially, the quantum dots have an energy bias $\varepsilon$ and the two electrons rest in a singlet $(2, 0)$ state on the left dot. (b) After slowly tuning $\varepsilon$ to zero, and measuring a singlet outcome, due to the weak measurement the spin of the nuclear bath decreases. (c) In the case of a spin triplet outcome an electron spin flips and the spin of the nuclear bath is changed accordingly. (d) The electronic spin can also be flipped due to spin-orbit, and the spin of the nuclear bath is pumped in the opposing direction (up) due to the weak measurement. $\varepsilon$ denotes the voltage bias, $\theta$ is the angle between the [110] crystallographic axis and the interdot connection axis $p_\xi$, $\Omega$ is the spin-orbit effective magnetic field.

accompanied by a flip of the nuclear spin from down to up, thus changing the
nuclear spin polarization closer to positive values. This is a process that, along with the process visualized on Fig. 3.4(d), competes with the hyperfine-mediated generation of negative polarization of the nuclear spins (down pumping). These two processes combined compensate the down pumping in systems with strong spin-orbit interaction.

To make an effective comparison between different In\(_x\)Ga\(_{1-x}\)As systems, the content of indium \(x\) is varied while \(B_z\) and \(d = a/a_B = 2.186\) are kept the same. This implies that the single particle tunneling and the overlap between the quantum dots would remain the same for every value of \(x\) (see Ref. [114]). For a comparison between different materials, the relative strength of the spin-orbit interaction can be quantified by the ratio of \(\Xi = 4a/\Lambda_{so}\), where \(\Lambda_{so}\) is the spin-orbit length defined by

\[
\frac{1}{\Lambda_{so}} = \frac{m^*}{\hbar} \sqrt{\cos^2 \theta (\alpha - \beta)^2 + \sin^2 \theta (\alpha + \beta)^2}.
\]

(3.19)

Here, \(m^*\) is the effective electron mass, \(\alpha\) and \(\beta\) are the Rashba and Dresselhaus constants and \(\theta\) is the angle between the [110] crystallographic axis and the interdot connection axis \(p_\xi\) [cf. Fig.3.4].

The spin-orbit length is the distance which an electron needs to travel in order to have its spin flipped due to spin-orbit interaction. If the electrons are initialized in a singlet state the probability for flipping the tunneling electron due to spin-orbit interaction is \(P_{\text{flip}} = 1/2\) at \(2a = \Lambda_{so}/2\). This further implies that if \(\Xi < 1\), the system is more probable to remain in a singlet state. If \(\Xi = 1\) the \(S\) and \(T_+\) outcomes due to spin-orbit coupling are equally probable and finally if \(1 < \Xi < 2\) a \(T_+\) outcome due to spin-orbit is more probable, because the probability that the tunneling electron has flipped its spin is greater than \(P_{\text{flip}} > 0.5\). In this study \(\Lambda_{so}/2 \gg 2a\) which implies \(\Xi \ll 1\), thus singlet outcomes due to spin-orbit interaction are always more probable even in pure InAs with the strongest possible value of spin-orbit \((\theta = \pi/2)\). In pure InAs, with \(\theta = \pi/2\), \(\Xi \approx 0.63\) for \(d = a/a_B = 2.186\).

3.6. Results for In\(_{0.2}\)Ga\(_{0.8}\)As

The attention is now focused on In\(_{0.2}\)Ga\(_{0.8}\)As, a material with an intermediate strength of spin-orbit coupling, as compared to the relatively weak spin-orbit coupling in GaAs and relatively strong spin-orbit coupling in InAs. The system of \(N = 150\) nuclear spins per dot is evaluated for different values of the angle \(\theta\) and with \(j_{\text{max}} = 18\). States with \(j > j_{\text{max}}\) would further lower the \(T_2^*\) and \(P_\text{s}\) and increase \(\sigma^{(2)}\). Therefore, the results presented here provide an upper bound
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Figure 3.5. (a) Probability distribution in the left quantum dot with respect to the nuclear spin projection quantum number \( m \) for \( j_L = 14 \) before and after 300 DNP cycles. (b) Probability distribution in the right quantum dot with respect to the nuclear spin projection quantum number \( m \) for \( j_R = 7 \) before and after 300 DNP cycles. The included spin-orbit interaction corresponds to the angle \( \theta = \pi/2 \). Here, \( \theta \) is the angle between the [110] crystallographic axis and the interdot connection axis \( p_\xi \). The number of nuclear spins per quantum dot is \( N = 150 \).

For \( j^L = 14 \) and \( j^R = 7 \) the pumping procedure has altered the nuclear probability distribution from a uniform distribution (with respect to the quantum number \( m \)) to a probability distribution where states with negative \( m \) are more likely. In the case without spin-orbit interaction, two processes contribute to this negative pumping of the nuclear spin [108] - the singlet detection accompanied by a weak measurement of the nuclear spin state and the \( T_+ \) detection, which flips the nuclear spin down to conserve the total spin of the system [cf. Fig. 3.4(b) and Fig. 3.4(c)]. Although including spin-orbit interaction [cf. Fig. 3.5(a), Fig. 3.5(b)], changes the final distribution of nuclear spin states only slightly, spin-orbit effects still have a notable effect on the singlet return probability \( P_S = \text{Tr}[M_S U \rho^{(i)} U^\dagger M_S] \). Fig. 3.6 plots \( P_S \) as a function of the number of cycles across the \( S - T_+ \) anticrossing for In\(_{0.2}\)Ga\(_{0.8}\)As. Here, the strength of the spin-orbit interaction is tuned by varying the angle \( \theta \) between the [110] crystallographic axis and the interdot connection axis \( p_\xi \). As shown in Fig. 3.6 (solid
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![Graph showing singlet return probability as a function of cycles](image)

Figure 3.6. The singlet return probability $P_s$ as a function of the number of cycles across the $S - T_0$ anticrossing in In$_{0.2}$Ga$_{0.8}$As. Here, $\theta$ is the angle between the [110] crystallographic axis and the interdot connection axis $p_\xi$.

red line), repeatedly cycling the system across the anticrossing point polarizes the nuclear spins, which leads to $P_s = 1$ after 300 cycles [108]. The situation changes dramatically when the spin-orbit interaction is included, which competes with the hyperfine mediated down pumping of the nuclear spins.

By theoretically varying the strength of the spin-orbit interaction the following findings were obtained. When the spin-orbit interaction has the largest possible value for $\theta = \pi/2$, it significantly affects the singlet return probability $P_s \approx 0.72$ (Fig. 3.6). Including spin-orbit interaction generates a mechanism which polarizes nuclear spins in the up direction (see Section 3.5 and Fig. 3.5). As a consequence of this behavior, the nuclear preparation mechanism is not efficient when spin-orbit effects are strong. The interplay of the hyperfine and spin-orbit interactions on nuclear state preparation can be observed better if the standard deviation of the nuclear difference field $\sigma^{(z)}$ is plotted (Fig. 3.7). The spin-orbit interaction has prevented the reduction of the standard deviation of the nuclear difference field ($0 \leq \theta \leq \pi/2$, see Fig. 3.7). Spin-orbit interactions affect the efforts to increase the spin $S - T_0$ qubit decoherence time $T^{*}_2$, see Fig. 3.8. The strongest spin-orbit coupling, corresponding to $\theta = \pi/2$, slightly lowers the resulting decoherence time from $T^{*}_2 \approx 15$ ns (red line) to $T^{*}_2 \approx 13$ ns (black dashed line with black x symbols).

Without the spin-orbit interaction the findings predict that the ratio of the final decoherence time (after the cycling is complete) $T^{*}_{2,f}$ and initial decoherence time (before the cycling starts) $T^{*}_{2,i}$ is $T^{*}_{2,f}/T^{*}_{2,i} \approx 2.28$ [cf. Fig. 3.9]. The situation
3. Interplay of spin-orbit and hyperfine interactions in DNP in semiconductor QDs

Figure 3.7. Standard deviation of the nuclear difference field $\sigma^{(z)}$ with respect to the number of DNP cycles across the $S - T_+$ anticrossing and different values of angle $\theta$ in In$_{0.2}$Ga$_{0.8}$As. Here, $\theta$ is the angle between the [110] crystallographic axis and the interdot connection axis $p_\xi$.

changes when spin-orbit interaction is included. For $\theta = 0$ a value of $T_{2f}^*/T_{2i}^* \approx 2.20$ is found, while for $\theta = \pi/2$ the ratio is $T_{2f}^*/T_{2i}^* \approx 2.04$.

After the inclusion of the spin-orbit interaction the ratio $T_{2f}^*/T_{2i}^*$ decreases with $\theta$. The results suggest that the $S - T_+$ dynamical nuclear polarization is not as effective in materials with intermediate strength of spin-orbit interaction, as compared to those without spin-orbit coupling. Nevertheless, the DNP still provides a notable enhancement of the $S - T_0$ qubit decoherence time $T_{2}^*$.

The work conducted here is within the so-called giant spin model where the behavior of $10^4 - 10^6$ nuclear spins is modeled with significantly fewer spins, $\sim 10^2 - 10^3$. In general $\sigma_i^{(z)} \propto A_i^z$, which would give rise to a much higher standard deviation of the nuclear difference field than expected. Therefore, the hyperfine constant is re-scaled, such that $\sigma_{i}^{(z)}$ has the same value for $N \approx 10^6$, and $N = 150$, $j_{ml} = \sqrt{N/2}$.

The predicted decoherence time before the start of the DNP is $T_{2}^* \approx 6.2$ ns while measurements yield $T_{2}^* \approx 10$ ns for pure GaAs [59] (where excluding spin-orbit effects is a good approximation). Averaging the hyperfine constant produces another conserved quantity $j^2$ [109], whose conservation dictates the maximum possible width of the nuclear spin distribution. Since $j_{\text{max}} \ll N/2$ the choice of $j$ is fairly independent of $N$. This means that $\sigma_{i}^{(z)} \propto \sqrt{N}$, and since $\sigma_{j}^{(z)}$ does
3. Interplay of spin-orbit and hyperfine interactions in DNP in semiconductor QDs

Figure 3.8. $S-T_0$ qubit decoherence time $T_2^*$ as a function of the number of DNP cycles across the $S-T_+$ anticrossing and strength of spin-orbit interaction in In$_{0.2}$Ga$_{0.8}$As. Here, $\theta$ is the angle between the [110] crystallographic axis and the interdot connection axis $p_\xi$.

Figure 3.9. The ratio of the final $T_{2,f}^*$ and initial $T_{2,i}^*$ decoherence times in In$_{0.2}$Ga$_{0.8}$As, for different values of the angle $\theta$ between the [110] crystallographic axis and the interdot connection axis $p_\xi$. 
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not depend on $N$ (because $\sigma_f^{(z)}$ depends on $j_{\text{max}}$ and $j_{\text{max}}$ is roughly $N$ independent), the following estimation can be made $T_{2,f}^* / T_{2,i}^* \sim \sqrt{N}$ for the case of $N = 150$ and the realistic case $N = 10^6$ (for an electrically defined quantum dot in In$_x$Ga$_{1-x}$As). Therefore, the maximum possible ratio of initial and final decoherence times can be estimated, for the realistic case of $N = 10^6$ spins and spin-orbit interaction excluded and included to be $T_{2,f}^* / T_{2,i}^* \approx 175$ without spin-orbit interaction, compared to $T_{2,f}^* / T_{2,i}^* \approx 94$ for GaAs in reference [108], $T_{2,f}^* / T_{2,i}^* \approx 174$ for $\theta = 0$, $T_{2,f}^* / T_{2,i}^* \approx 163$ for $\theta = \pi/2$. It should be noted that for $10^6$ nuclear spins, the model under consideration predicts $T_{2,f}^* \approx 2.45 \, \mu$s compared to $T_{2,f}^* \approx 1 \, \mu$s in [113]. Due to the larger ensemble of nuclear spins in reality, conducting an experiment requires a larger number of cycles to achieve the level of DNP achieved here. This number of cycles is estimated to be $10^4 - 10^5$.

3.7. Results for In$_x$Ga$_{1-x}$As

In this section the $T_2^*$ results for In$_x$Ga$_{1-x}$As are compared, with varying In content $x$. The concentration of indium $x$ in the sample is varied between 0 and 1 with a 0.2 increment. For the sake of computational efficiency, and the fact that only a mere comparison between materials with different percentages of indium will be made, the computational method is slightly simplified now. Instead of averaging over all possible states ranging from $j_{\text{min}} - j_{\text{max}}$, $j_L^R = j_{\text{m}} = \sqrt{N/2}$ is set for the left and the right quantum dot. This effectively means that a situation is assumed where an experiment is performed only once with the most likely nuclear spin configuration.

From Fig. 3.10 a conclusion can be drawn that raising the concentration of indium in In$_x$Ga$_{1-x}$As sample has a detrimental effect on the efficiency of the $S-T_+$ DNP scheme. By doping the system with indium, the spin-orbit coupling is strengthened, thus reducing the overall $\Lambda_{\text{so}}$ [Eq. (3.19)], which as a consequence has more virtual and real $T_+$ outcomes due to the spin-orbit interaction. The virtual $T_+$ will relax to $S$, quickly flipping a nuclear spin from down to up in the process. The real spin-orbit mediated $T_+$ outcomes will also pump the nuclear spin towards the positive values of the polarization (up). This process can completely vain efforts to increase $T_2^*$, even at intermediate concentrations of 40% In (Fig. 3.10). At higher indium concentrations, DNP is totally suppressed for all values of $\theta$ [cf. Fig. 3.11].
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Figure 3.10. $S - T_0$ electron spin coherence time $T_2^*$ as a function of the number of DNP cycles across the $S - T_+$ anticrossing, for different abundances of indium $x$ in In$_x$Ga$_{1-x}$As and for $\theta = \pi/2$. Here, $\theta$ is the angle between the [110] crystallographic axis and the interdot connection axis $p_\xi$.

Figure 3.11. $S - T_0$ electron spin coherence time $T_2^*$ for GaAs and InAs as a function of the number of DNP cycles across the $S - T_+$ anticrossing, for $\theta = 0$, i.e. the case where the [110] crystallographic axis and the interdot connection axis $p_\xi$ are aligned.
3.8. Conclusions and final remarks

The results presented here show that pure InAs is a not a suitable candidate for $S-T_+$ DNP, due to the fact that the enhancement of $T_2^*$ is strongly suppressed even for the smallest possible strength of the spin-orbit interaction corresponding to $\theta = 0$. Dynamical nuclear polarization in InAs could still be achieved by using single spin single quantum dot systems [111] or by using a more elaborate pulsing sequence [112]. A similar behavior could be expected in materials with even stronger spin-orbit as compared to InAs and that is, e.g., InSb.

To conclude, a nuclear polarization scheme was discussed in In$_x$Ga$_{1-x}$As double quantum dots with spin-orbit interaction. In the presence of spin-orbit interaction a suppression of the enhancement of $T_2^*$ is predicted. The conclusions drawn here are also valid for materials with fewer nuclear spins, but due to the assumed constant hyperfine constant only electrostatically defined quantum dots are treatable by the formalism presented here. The $S-T_+$ DNP sequence is highly sensitive to the strength of the spin-orbit coupling, and therefore the efficiency of the $S-T_+$ DNP sequence will depend on the angle $\theta$ and the In content $x$ in In$_x$Ga$_{1-x}$As. A stronger spin-orbit interaction will establish a process that will quickly neutralize any efforts to prolong $T_2^*$. The cases of unequally coupled and/or sized dots, and different shapes of the bias [90] are in general treatable by numerics used here and will be the subject of future studies. Charge noise [58,121] is neglected in the current model. Investigating the significance of charge coherence requires an extension of the numerical tools used [122], and is planned as a forthcoming investigation.
4. Electric dipole spin resonance in systems with a valley dependent $g$-factor

4.1. Summary

In this chapter a qualitative and quantitative investigation of the electric dipole spin resonance (EDSR) is presented. The system under study is a single Si/SiGe quantum dot in the presence of a magnetic field gradient, e.g., produced by a ferromagnet. The electron spin states are controlled by applying an oscillatory electric field, inducing real-space oscillations of the electron inside the quantum dot. One of the goals of the study is to present a microscopic theory of valley dependent $g$-factors in Si/SiGe quantum dots and investigate how valley relaxation combined with a valley dependent $g$-factor leads to a novel electron spin dephasing mechanism. Furthermore, the interplay of spin and valley relaxations in Si/SiGe quantum dots is discussed. The findings presented here suggest that the electron spin dephases due to valley relaxation, and are in agreement with recent experimental studies [Nature Nanotechnology 9, 666–670 (2014)].

This chapter is adapted from Marko J. Rančić and Guido Burkard, Phys. Rev. B 93, 205433 (2016)
4.2. Introduction

Finding efficient ways to use electron spins in quantum dots (QDs) as quantum bits (qubits) has been an active field of research in condensed matter physics for many years [18, 59, 68, 123, 14, 124]. A necessary prerequisite for building qubits are long coherence times, long enough to allow for a large number of gate operations before the quantum-mechanical nature of the qubit is irreversibly lost [57]. An electron spin confined in a semiconductor quantum dot loses its quantum phase coherence due to interactions with its noisy, solid state environment. Unavoidable interactions of the electron spin with surrounding charges and nuclear spins are common mechanisms that limit the coherence time of the electron spin $T_2^*$ to as little as nanoseconds in some structures [59, 87, 85, 117].

In natural silicon only $\approx 4.7\%$ of the atomic nuclei have a non-zero spin. Therefore, Si represents a logical candidate for the implementation of spin qubits [123, 125, 126, 11]. There are two implementation strategies for spin qubits in Si, using the nuclear spin of a phosphorus donor in Si [103] and using spin states of an electron confined inside a Si quantum dot [127, 128, 129, 13]. Bulk silicon has six minima of the conduction band, known as valleys. In a Si/SiGe quantum well, four out of six valley states are higher in energy due to strain at the Si/SiGe interface [130]. The degeneracy of the remaining two valley states can be lifted by the confining potential in the $z$-direction [131, 132].

In this chapter the focus is on a situation in which a ferromagnet is embedded on top of the quantum dot, as shown in Fig. 4.1. The in-plane component of the ferromagnet stray magnetic field, leads to the existence of a valley dependent $g$-factor, as predicted in the following theoretical study [67]. The goal of the theoretical study performed here is to establish a quantitative relationship between valley dependent $g$-factors and the tilt of the Si/SiGe interface. Consequently, the ferromagnet embedded on top of the quantum dot also leads to a valley dependent Rabi frequency [65]. The valley dependent $g$-factor causes the resonance condition to be different for the two valleys, and alongside with valley dependent Rabi frequencies, leads to errors in controlling the electron spin state in one of the valleys. Furthermore, when valley relaxation is present, a novel decoherence mechanism exists which cannot be reversed by a spin echo [17]. If the electron is driven on resonance in one of the valleys, valley relaxation abruptly changes the resonance condition causing the electron spin to decohere. Another goal of this chapter is to describe the reduction of electron spin coherence due to valley relaxation, by solving a Lindblad master equation. The presence of spin relaxation, alongside with valley relaxation, leads to a rich interplay of spin and valley relaxation, which is also described by solving a Lindblad master equation.

This chapter is organized as follows. In Section 4.3 a quantitative description
4. Electric dipole spin resonance in systems with a valley dependent $g$-factor

The control of the electron spin. A ferromagnet (FM) induces a magnetic field gradient in the $x$-direction. When microwave bursts are applied the electron experiences an effectively time dependent magnetic field in the direction of oscillation. $\omega_0$ is the Larmor frequency of the microwaves.

Figure 4.1. The control of the electron spin. A ferromagnet (FM) induces a magnetic field gradient in the $x$-direction. When microwave bursts are applied the electron experiences an effectively time dependent magnetic field in the direction of oscillation. $\omega_0$ is the Larmor frequency of the microwaves.

of the valley dependent $g$-factor is given, when the valley dependent $g$-factor is induced by an in-plane stray magnetic field. The chapter is continued by discussing the existence of a valley dependent Rabi frequency in Section 4.4. Subsequently, in Section 4.5 the Hamiltonian and the Lindblad equation for the open-system dynamics of the electron spin are presented, allowing the qualitative and quantitative description of the drop of the electron spin coherence caused by valley relaxation. In Section 4.6 the interplay of valley and spin relaxations is discussed, before concluding in Section 4.7.

4.3. Valley Dependent $g$-factor in Si/SiGe quantum dots

Bulk silicon has six effective minima of the conduction band named valleys. In a Si/SiGe quantum dot four of the valleys are lifted higher in energy by the presence of strain at the Si/SiGe interface and the two low energy valleys remain degenerate. The degeneracy of the remaining two valleys is lifted by the confining potential in the $z$-direction [131, 132].
4. Electric dipole spin resonance in systems with a valley dependent \( g \)-factor

The Hamiltonian of a single electron spin confined in a Si/SiGe quantum dot in a magnetic field in the \( z \)-direction, and a magnetic field gradient in the \( x \)-direction is given by

\[
H = H_0 + H_z + H_{FM}. \tag{4.1}
\]

Here, \( H_0 \) is the Hamiltonian of the single electron confined in a Si/SiGe quantum dot

\[
H_0 = \frac{p_z^2}{2m^*_z} + \frac{p_x^2 + p_y^2}{2m^*_t} + V(x) + V(y) + V(z). \tag{4.2}
\]

Here, \( p_i \) denotes the \( i \)-th component of the momentum operator, and \( m^*_z \) the longitudinal electron mass (in a direction perpendicular to the Si/SiGe quantum well). Furthermore, \( m^*_t \) is the transverse electron mass (in the plane of Si/SiGe quantum well) and \( V(x), V(y), V(z) \) are confining potentials in the \( x, y, z \) directions respectively. The confining potentials in the \( x \)-direction and \( y \)-direction come from the electrostatic confinement and are modeled with a harmonic oscillator potential \( V(x) = m^*_t \omega^2_0 x^2 / 2, V(y) = m^*_t \omega^2_0 y^2 / 2 \). The potential in the \( z \)-direction comes from the Si/SiGe quantum well and is modeled as a finite square well potential. \( H_z \) is the Zeeman Hamiltonian

\[
H_z = g \mu_B B_0 S_z, \tag{4.3}
\]

where \( g \) is the electron \( g \)-factor, \( \mu_B \) is the Bohr magneton, \( B_0 \) is the total magnetic field (in the \( z \)-direction) and \( S_z \) is the \( z \) component of the electron spin operator. Furthermore, \( H_{FM} \) is the Hamiltonian describing the stray field in the \( x \)-direction coming from the ferromagnet

\[
H_{FM} = g \mu_B B(x) S_x, \tag{4.4}
\]

where \( S_x \) is the \( x \) component of the electron spin operator and \( B(x) \) is the \( x \) component of the magnetic field coming from the ferromagnet \( B(x) = B^0_x x / a_B \). Here \( B^0_x \) is the strength of the slanting field, \( x \) is the position operator and \( a_B = \sqrt{\hbar / m^*_t \omega^2_0} \) is the effective Bohr radius in the \( x \)-direction of the electron spin confined in a quantum dot, where \( m^*_t \) is the transverse effective electron mass and \( \omega^2_0 \) is the confining potential in the \( x \)-direction.

An in-plane magnetic field gradient \( B(x) \) modifies the Zeeman energy [67]. In the case under consideration the in-plane magnetic field gradient is caused by the ferromagnet embedded on top of the quantum dot (Fig. 4.1). Neglecting the gradient in the \( z \)-direction is a good approximation when the total magnetic field (directed along \( z \)) is much larger than the \( z \) component of the stray field.
Proceeding similar to [67], energy levels of $H_0 + H_z$ are obtained as

$$E = E_n \pm E_z/2.$$  \hfill (4.5)

Here, $E_n$ is the confinement energy and $E_z = g \mu_B B_0$ is the electron Zeeman energy. A plus sign in Eq. (4.5) stands for a spin-up state $|\uparrow\rangle$ and a minus sign for a spin-down state $|\downarrow\rangle$.

The first order energy correction coming from $H_{FM}$ is zero because of the even parity of the ground state wavefunction of the linear harmonic oscillator (LHO) and odd parity of $H_{FM}$. The second order energy correction coming from the magnetic field gradient term $H_{FM} = g \mu_B B(x) S_x$ yields

$$E_{m_s}^{(2)} = -\frac{1}{4} \sum_{n=1}^{\infty} \frac{M_n^2}{\Delta_n - 2m_s E_z},$$  \hfill (4.6)

where $m_s = \pm 1/2$ is the spin projection quantum number. The symbol $\Delta_n$ stands for the energy difference between the orbital ground state and the $n$-th state. Furthermore, $M_n$ is the matrix element between the ground state and the $n$-th orbital state of the LHO

$$M_n = \langle \Psi_0 \uparrow | H_{FM} | \Psi_n \downarrow \rangle = \frac{g \mu_B B_0^0}{2\alpha_B} \langle \Psi_0 | x | \Psi_n \rangle,$$  \hfill (4.7)

where $\Psi_0$ is the ground state LHO wavefunction and $\Psi_n$ is the LHO wavefunction of the $n$-th excited state. Because

$$\langle \Psi_0 | x | \Psi_n \rangle = \frac{1}{\sqrt{2}} \alpha_B \delta_{n,1}$$  \hfill (4.8)

and because $S_x$ couples only states with different spin projections $m_s$, for an electron in the ground orbital state the sum in Eq. (4.6) is substituted by a single term with a matrix element

$$M_1 = \frac{g \mu_B B_0^0}{2\sqrt{2}}.$$  \hfill (4.9)

Therefore, the slanting magnetic field in the $x$-direction corrects the ground state energy of the electron

$$E_{m_s}^{(2)} = -\frac{1}{4} \frac{M_1^2}{\Delta - 2m_s E_z},$$  \hfill (4.10)
4. Electric dipole spin resonance in systems with a valley dependent $g$-factor

where $\Delta = \hbar \omega_0^2$ is the orbital splitting.

In the presence of valley-orbit mixing the orbital splitting $\Delta_{v, \bar{v}}$ is valley dependent [15, 133]. This yields a valley dependent energy correction due to the slanting magnetic field Eq. (4.10), and therefore an effective electron $g$-factor which depends on the valley eigenstate,

$$g_j = \frac{g}{E_z} \left( E_z + E_{1,j}^{(2)} - E_{1,j}^{(2)} \right) = g \left[ 1 - \frac{1}{2} \frac{M_1^2}{\Delta_j^2 - E_z^2} \right]. \quad (4.11)$$

Here, $g_j$ is the effective $g$-factor corresponding to two valley eigenstates $j = \{v, \bar{v}\}$ and $\Delta_j$ is the valley dependent orbital level spacing corresponding to $j$-th valley eigenstate. The average difference of effective $g$-factors $\Delta g/\bar{g}$ is defined as

$$\Delta g/\bar{g} = 2 \frac{g_v - g_{\bar{v}}}{g_v + g_{\bar{v}}}. \quad (4.12)$$

Inserting equation Eq. (4.11) into Eq. (4.12) yields

$$\Delta g/\bar{g} = \frac{2M_1^2(\Delta_{\bar{v}}^2 - \Delta_v^2)}{(\Delta_v^2 - E_z^2)(\Delta_{\bar{v}}^2 - E_z^2) - M_1^2(\Delta_v^2 + \Delta_{\bar{v}}^2 - 2E_z^2)/4}. \quad (4.13)$$

Furthermore, $E_z$ is the Zeeman energy, and $M_1$ is the matrix element between the orbital ground state and the first excited orbital state coming from the slanting field Eq. (4.9).

Valley-orbit mixing $\Delta_v - \Delta_{\bar{v}} \neq 0$ occurs due to miscuts of the Si/SiGe quantum well [11, 134]. The valley coupling can be described by a $\delta$ function [12, 135]

$$V_v(r) = v_v \delta(z - z_0 + \theta x). \quad (4.14)$$

Here, $z_0$ is the position of the SiGe interface, the miscut is usually between $0^\circ \leq \theta \leq 2^\circ$, so it is safe to approximate $\tan(\theta) \approx \sin(\theta) \approx \theta$. Furthermore, $v_v$ is the valley coupling strength. Assuming for simplicity that the miscut occurs in the $x$-direction, the valley coupling operator Eq. (4.14) does not depend on the $y$ component.

As the wavefunction is closest to the top interface only one delta function potential is present in the theory. Treating valley coupling as a perturbation the general formula for matrix elements of the valley coupling operator Eq. (4.14) is
4. Electric dipole spin resonance in systems with a valley dependent $g$-factor

\[
\langle n', \bar{v}|V_{v}(r)|n, v \rangle = \tilde{v}_{v} \xi^{2}(z_{0}) e^{-2ik_{0}z_{0}} \int_{-\infty}^{\infty} e^{-2ik_{0}x\theta} \Psi^\ast_{n'}(x) \Psi_{n}(x) dx. \tag{4.15}
\]

Assuming that the wavefunctions $\Psi_{n}$ are those of the LHO the diagonal elements of the valley coupling operator Eq. (4.14) have the following form

\[
\langle n, v|V_{v}(r)|n, v \rangle = v_{v} \xi^{2}(z_{0}), \tag{4.16}
\]

where $n$ is the orbital quantum number corresponding to the wavefunction in the $x$-direction, $v$ is the valley quantum number, $\xi(z_{0})$ is the ground state electron wavefunction in the $z$-direction and $z_{0}$ is the position of the Si/SiGe interface. Due to the fact that the confinement in the $z$-directions comes from a sharp Si/SiGe interface, the orbital level spacing in the $z$-direction is large, so the system is always assumed to be in the ground state in the $z$-direction. The off-diagonal matrix elements of the lowest two orbital states of the valley coupling operator Eq. (4.14) have the following form

\[
\langle 0, \bar{v}|V_{v}(r)|1, v \rangle = -i\sqrt{2} \bar{v}_{v} \xi^{2}(z_{0}) k_{0} a_{B} e^{2ik_{0}z_{0}} e^{-k_{0}^{2}\theta^{2}a_{B}^{2}},
\]

\[
\langle 0, v|V_{v}(r)|0, v \rangle = \bar{v}_{v} \xi^{2}(z_{0}) e^{2ik_{0}z_{0}} e^{-k_{0}^{2}\theta^{2}a_{B}^{2}},
\]

\[
\langle 1, v|V_{v}(r)|1, v \rangle = \bar{v}_{v} \xi^{2}(z_{0})(1 - 2k_{0}^{2}\theta^{2}a_{B}^{2}) e^{2ik_{0}z_{0}} e^{-k_{0}^{2}\theta^{2}a_{B}^{2}}. \tag{4.17}
\]

Here $k_{0}$ is the reciprocal lattice constant of Si, $z_{0}$ is the position of the Si/SiGe interface, $\theta$ is the effective tilt angle, and $a_{B}$ is the effective Bohr radius in the $x$-direction. A common way of approximating a product of Bloch wavefunctions is $\phi^{\ast}_{j} \phi_{k} \approx C_{jk} \exp(i2k_{0}r)$, where $C_{jk}$ are form factors and $\exp(i2k_{0}r)$ trivial Bloch wavefunctions. The form factors in the case of Eq. (4.16) are contained in the free parameter $v_{v}$. In the case of Eq. (4.15) and Eq. (4.17) the form factors can differ in magnitude and sign compared to Eq. (4.16), yielding a different free parameter $\bar{v}_{v}$. Both $v_{v}$ and $\bar{v}_{v}$ are unknown free parameters, which can be assumed to be of similar magnitude, and here $v_{v} \approx \bar{v}_{v}$ is chosen and with an adjusted value to obtain a valley splitting of the correct order of magnitude. It should be noted that eigenvalues of a matrix constituted from Eq. (4.16) and Eq. (4.17) do not depend on the relative sign of $\bar{v}_{v}$. A comparison between different approximations of Bloch wavefunctions in Si can be found in a recent theoretical manuscript [136].

Constraining the discussion on the lowest two orbital states, and diagonalizing the matrix constituted of elements from Eq. (4.16) and Eq. (4.17) the mixed valley-orbit eigenspectrum is obtained Fig. 4.2 (and therefore $\Delta_{v}$ and $\Delta_{\bar{v}}$).
Electric dipole spin resonance in systems with a valley dependent $g$-factor

Figure 4.2. The lowest four energy states as a function of the effective miscut angle $\theta$. $\Delta E_v$ is the ground state valley splitting, and $\Delta_v$ and $\Delta_{\bar{v}}$ are orbital splittings in the $v$ and $\bar{v}$ valleys. The parameters of the plot are $\hbar \omega_0^v = 450 \, \mu eV$, $v_0 \xi^2(z_0) = 300 \, \mu eV$, $k_0 = 2\pi \cdot 0.82/a$, where $a = 5.431 \, \text{Å}$ is the lattice constant of Si, and $m^*_i = 0.19 m_e$. It should be noted that due to valley-orbit mixing the orbital quantum numbers $n = 0, 1$ and valley quantum numbers $v = \pm 1$ are not good quantum numbers anymore.

Constraining the discussion again on the lowest two orbital states, diagonalizing the matrix constituted of elements from Eq. (4.16) and Eq. (4.17), and inserting the result of the diagonalization into Eq. (4.13) the average difference of effective electron $g$-factors is obtained as a function of the confining energy $\hbar \omega_0^v$ and the effective tilt angle $\theta$ (Fig. 4.3). Fig. 4.3 (a) shows that for $\theta \approx 0.2^\circ$ the average difference of valley dependent effective $g$-factors goes to zero due to the fact that for this particular value of the effective tilt angle $\Delta_v \approx \Delta_{\bar{v}}$. Recent experimental studies [17] yield an absolute average difference of effective $g$-factors of $|\Delta g/\bar{g}| = 1.5 \cdot 10^{-4}$ and predict an absolute average difference of effective $g$-factors of $|\Delta g/\bar{g}| = 3 \cdot 10^{-5}$, given the single orbital spacing $\hbar \omega_0^v = 450 \, \mu eV$. $|\Delta g/\bar{g}| = 3 \cdot 10^{-5}$ corresponds to the values $\theta \approx 0.15^\circ$ or $\theta \approx 0.3^\circ$ for $\hbar \omega_0^v = 450 \mu eV$. When difference of the lowest two eigenvalues is plotted Fig. 4.4, one can see that the valley splitting corresponding to $\theta \approx 0.3^\circ$ is $E_v \approx 60 \, \mu eV$, in agreement with the typical value for quantum dots $\Delta E_v \approx 0.1 \, \text{meV}$ [11]. It should be noted that a recent study shows the existence of valley dependent $g$-factors in Si/SiO$_2$ which are attributed to spin-valley mixing, taking into account the large band offset of Si/SiO$_2$ [137].
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Figure 4.3. (a) The average difference of effective $g$-factors as a function of the effective tilt angle $\theta$ and the confinement energy $\hbar \omega_0^s$. (b) The average difference of effective $g$-factors for the value of the single orbital spacing $\hbar \omega_0^s = 450$ $\mu$eV (see dashed line in (a)) [17]. The parameters of the plots are the following, $v_c \xi^2(\zeta_0) = 300$ $\mu$eV, $m_t^* = 0.19 m_e$ and $k_0 = 2\pi \cdot 0.82/a$, where $a = 5.431$ Å is the lattice constant of Si, $B_x^0 = 3.4$ mT/nm, $B_z = 0.75$ T, the $z$-component of the magnetic field of the ferromagnet $B_z^{FM} = -0.12$ T, and the height of the Si quantum well is $\zeta_0 = 12$ nm.

Figure 4.4. Ground state valley splitting $\Delta E_v$ as a function of the effective tilt angle $\theta$. The parameters of the plot are, $\hbar \omega_0^s = 450$ $\mu$eV, $v_c \xi^2(\zeta_0) = 300$ $\mu$eV, $m_t^* = 0.19 m_e$ and $k_0 = 2\pi 0.82/a$, where $a = 5.431$ Å is the lattice constant of Si, $B_x^0 = 3.5$ mT/nm, and the size of the Si quantum well is $\zeta_0 = 12$ nm.
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4.4. Valley dependent Rabi frequency

When controlling the electron spin by oscillating it inside an in-plane magnetic gradient the Rabi frequency is calculated with the following formula \[65\]

$$\Omega = \frac{g\mu_B eE_{\text{gate}}}{2\hbar} \left| \frac{\partial B(x)}{\partial x} \right| \frac{a_B^2}{\Delta}. \quad (4.18)$$

Here, $E_{\text{gate}}$ is the electric field of the gate, $B(x)$ is the in-plane magnetic field, $a_B = \sqrt{\hbar/\omega_0 m^*_e}$ is the effective Bohr radius of the electron and $\Delta$ is orbital level spacing. As seen in Section 4.3, the $g$-factors corresponding to different valleys only differ by $\approx 10^{-3}$ relative to their values, so through this section it is safe to assume that $g_v = g_\theta = g = 2$. If the valley and orbit degree of freedom mix (due to, e.g., Si/SiGe interface miscut) the orbital level spacing $\Delta_{v,\theta}$, and therefore the Rabi $\Omega_{v,\theta}$ frequency become valley dependent with an average difference of Rabi frequencies

$$\frac{\Delta \Omega}{\Omega} = \frac{\Delta_{v,\theta}}{\Delta_{v} + \Delta_{\theta}}. \quad (4.19)$$

By diagonalizing a matrix whose terms are constituted from Eq. (4.16) and Eq. (4.15) and then inserting the result into Eq. (4.19) the average difference of

![Figure 4.5. Average difference of Rabi frequencies $\Delta \Omega/\Omega$ as a function of the effective tilt angle $\theta$. The parameters of the plot are, $\hbar \omega_0^5 = 450 \mu eV$, $v_s \xi^2(z_0) = 300 \mu eV$, $m^*_e = 0.19 m_e$ and $k_0 = 2 \cdot \pi 0.82/a$, where $a = 5.431$ Å is the lattice constant of Si, $B_0^5 = 3.5$ mT/nm, $B_z = 0.75$ T, and the height of the Si quantum well is $z_0 = 12$ nm.

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Rabi frequencies as a function of the effective tilt angle $\theta$ is obtained Fig. 4.5. A 50% absolute average difference of valley Rabi frequencies is measured in a recent experimental study [17]. In the case under consideration the maximum $\Delta \Omega/\bar{\Omega} = 25\%$, which corresponds to a value of the effective tilt angle $\theta \approx 0.15^\circ$ (see Fig. 4.5). The discrepancy between the findings presented here and the experiment may be due to the fact that the product of valley coupling strength and the square of the wavefunction at the position of the Si/SiGe interface $v_v \xi^2(z_0)$ is a free parameter. $v_v$ depends on the abundance of Ge $x$ in the Si/Si$_x$Ge$_{1-x}$ quantum well and can be estimated from tight binding theories [135]. On the other hand, $\xi^2(z_0)$ depends on the thickness of the Si layer and the exact type of the confinement in the Si/SiGe quantum well.

4.5. Modeling the decoherence

A situation in which an electron spin is confined in a Si/SiGe quantum dot with a ferromagnet embedded on top of the quantum dot [67] is modeled. The ferromagnet is inducing a stray magnetic field as shown in Fig. 4.1. All-electrical two-axis control of single electron spin states is achieved by oscillating the electron in real space with microwave bursts [67, 71] (Fig. 4.1). As the electron oscillates in real space it experiences a periodic, time-dependent, magnetic field.

The free evolution of the electron spin is described by the following Hamiltonian

$$H_0 = \sum_{\sigma=\downarrow,\uparrow} \sum_{j=v,\bar{v}} E_{\sigma j} \hat{c}^\dagger_{\sigma j} \hat{c}_{\sigma j}. \quad (4.20)$$

Microwave induced oscillations of the electron in real space, combined with the stray field of the ferromagnet, alter the state of the electron spin, while leaving the valley degree of freedom unchanged

$$H'(t) = \sum_{j=v,\bar{v}} \hbar \Omega_j \cos (\omega t) (\hat{c}_{\downarrow j}^\dagger \hat{c}_{\uparrow j} + H.c.). \quad (4.21)$$

Applying the rotating wave approximation to the Hamiltonian $H_0 + H'(t)$, the time-independent Hamiltonian in the rotating frame is obtained,

$$H = \frac{1}{2} \begin{pmatrix}
E_z - \hbar \omega_0 & h \Omega_v & 0 & 0 \\
h \Omega_v & -E_z + \hbar \omega_0 & 0 & 0 \\
0 & 0 & E_z + \delta E - \hbar \omega_0 & h \Omega_\bar{v} \\
0 & 0 & h \Omega_\bar{v} & -E_z - \delta E + \hbar \omega_0
\end{pmatrix}, \quad (4.22)$$
in the \( \{ v \uparrow, v \downarrow, \bar{v} \uparrow, \bar{v} \downarrow \} \) basis, where the \( \{ v, \bar{v} \} \) represent valley eigenstates, and \( \{ \uparrow, \downarrow \} \) stand for spin states. \( E_{\sigma j} \) is the energy of the \( j \)-th valley eigenstate with spin \( \sigma \), \( c_{\sigma j} \) and \( c_{\sigma j}^\dagger \) are electron creation and annihilation operators. Furthermore, \( E_z \) is the Zeeman energy of the electron, \( \omega_0 \) is the Larmor frequency, \( \Omega_{v, \bar{v}} \) is the valley dependent Rabi frequency and \( \delta E = (g_v - g_{\bar{v}})\mu_B B_z \) is the difference of valley Zeeman energies Fig. 4.6.

One of the goals of the study conducted in this chapter is to model the influence of valley relaxation on electron spin coherence. An electron is initialized in the \( | \downarrow \rangle \) state with valley injection probabilities \( P^0_v = 0.7 \), \( P^0_{\bar{v}} = 0.3 \). A spin echo experiment is modeled. First a \( \pi/2 \) pulse is applied, followed by a free (undriven) evolution of a duration \( t/2 \). Afterwards, a \( \pi \) pulse is applied followed by another free evolution of a duration \( t/2 \) and another \( \pi/2 \) pulse.

The valley relaxation is assumed to occur only during the free evolution stage (as the duration of the free evolution stage \( t \) is much larger than the duration of \( \pi \) pulses), and is modeled with a Lindblad equation

\[
\dot{\rho} = -\frac{i}{\hbar}[H_0, \rho] + \frac{1}{2}\Gamma(2L^\dagger \rho L - LL^\dagger \rho - \rho LL^\dagger) = \mathcal{L}\rho. \tag{4.23}
\]

Here, \( \mathcal{L} \) is the \( 16 \times 16 \) Lindblad superoperator acting on the density matrix rep-

![Diagram](image.png)

Figure 4.6. Visualizing a valley dependent effective \( g \)-factor. \( \Gamma \) is the valley relaxation rate, \( \delta E = (g_v - g_{\bar{v}})\mu_B B_z \) is the difference of valley Zeeman energies, \( E_z \) is the Zeeman energy of the confined electron and \( \omega_0 \) is the Larmor frequency.
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resented in the vector form. The explicit form of $L$ is given in the Appendix 1. Furthermore, $\Gamma$ is the phenomenological valley relaxation rate, and $L^\dagger = |v\rangle \langle \bar{v}|$ and $L = |\bar{v}\rangle \langle v|$ are Lindblad inter-valley dissipation operators.

The measure of the electron spin coherence is the echo envelope function. In order to be able to subtract the echo envelope function, instead of using the Lindblad equation in the mentioned form Eq. (4.23) the Lindblad equation in superoperator form is used

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

Writing the Lindblad equation in the superoperator form allows the inclusion of a sequence of $\pi/2 - \pi - \pi/2$ pulses, around the $x$ axis, with inter-valley scattering occurring in the free evolution stage in the following way

$$\rho(t) = R_x(\pi/2)e^{Lt/2}R_x(-\pi)e^{Lt/2}R_x(\pi/2)\rho(0).$$

Here, $R_x(\beta)$ rotates the spin $\rho(t)$ about an angle $\beta$ around the $x$ axis on the Bloch sphere. The $\pi$ and $\pi/2$ pulses are achieved by applying microwave pulses with a duration $\pi/\Omega_v$ and $\pi/2\Omega_v$, described by time evolution operators $R_x(\pi) = \exp(-iH\pi/\hbar\Omega_v)$ and $R_x(\pi/2) = \exp(-iH\pi/2\hbar\Omega_v)$, with $H$ being given by Eq. (4.22).

Finally, the echo envelope function is obtained, the probability that the electron changes spin to the $|\uparrow\rangle$ state after a total time of a free evolution $t$, when being subjected to a sequence of $\pi/2 - \pi - \pi/2$ pulses

$$P_\uparrow = \sum_{j=v,\bar{v}} \text{Tr}(M_j^\uparrow \rho(t)).$$

Here the $M_j^\uparrow$ are spin-up projection operators corresponding to $j$-th valley eigenstate.

When the electron $g$-factor is valley dependent, the $\pi$ and $\pi/2$ pulses are assumed perfect (see Fig. 4.7 black line), a valley relaxation event abruptly changes the resonance condition for $\delta E$ (see Eq. (4.22)). After the initial perfect $\pi/2$ pulse, in one half of the cases of inter-valley relaxation from $|v\rangle$ to $|\bar{v}\rangle$ the electron spin is in $|\uparrow\rangle$ state. This is why the increase of the probability $P_\uparrow$, originating from valley relaxation, saturates at $P_0/2$ (see Fig. 4.7, gray dashed line).

The red triangles in Fig. 4.7 represent the result of the simulation when the effective $g$-factors are valley dependent throughout the free evolution stage and the $\pi$ and $\pi/2$ pulses are imperfect in one of the valleys due to valley dependent effective $g$-factors and Rabi frequencies. After the imperfect initial $\pi/2$ pulse, the electron spin is not perpendicular to the magnetic field yielding rotations around the quantization axis with a frequency proportional to the Zeeman energy.
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Figure 4.7. Probability that the echo sequence yields the electron $|\uparrow\rangle$ state. Red triangles and black line are a result of a simulation with injection probabilities $P^0_{v,\uparrow} = 0.7$, $P^0_{\bar{v},\downarrow} = 0.3$. Yellow disks are a result of a simulation with injection probabilities $P^0_{v,\downarrow} = 0.21$ and $P^0_{\bar{v},\uparrow} = 0.3$. The parameters of the plot are, the external magnetic field $B_z = 0.75$ T, the $z$-component of the magnetic field of the ferromagnet $B_{z\text{FM}} = -0.12$ T, valley dependent Rabi frequencies corresponding to the miscut angle $\theta \approx 0.3^\circ$, $\Omega_v = 2\pi \cdot 3.1$ MHz, $\Omega_{\bar{v}} = 2\pi \cdot 3.7$ MHz, within the values suggested in a recent experimental study [17].

$g_v \mu_B (B_z + B_{z\text{FM}})/h$, where $B_z$ is the external magnetic field and $B_{z\text{FM}}$ is the $z$-component of the magnetic field of the ferromagnet. For $B_z = 0.75$ T and $B_{z\text{FM}} = -0.12$ T this oscillations take place on a $\sim 50$ ps timescale, with the amplitude of the oscillations being given by the valley dependent Rabi frequencies $\Omega_v$ and $\Omega_{\bar{v}}$ and $g$-factors $g_v$ and $g_{\bar{v}}$. Therefore, the probability $P_{\uparrow}$ is very sensitive to the duration of the free evolution stage. Due to the fact that the results of a recent experimental study [17] represent an average over 150-1000 experimental outcomes, the results presented here (red triangles and yellow discs, Fig. 4.7) represent an average over 1000 outcomes, randomly sampled from a 5 ns interval. When the increase in probability due to valley relaxation (black line, Fig. 4.7) and the additional effect of imperfect $\pi$ and $\pi/2$ pulses is compared one observes that imperfect rotations provide an additional mechanism that further increases $P_{\uparrow}$.

A recent experimental study [17] shows a fast initial increase by 0.25 of the probability $P_{\uparrow}$. The findings presented here explain an initial increase of $P_{\uparrow}$ by a few % due to the averaging of the amplitude of 1000 randomly selected data points of the $P_{\uparrow}$ oscillations close to $t = 0$, occurring due to imperfect $\pi$ and $\pi/2$ pulses alongside with rotations around the $z$-axis in the free evolution stage.
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One possible explanation for the remaining discrepancy between the experimental findings and theory may be the initialization to the $|\downarrow\rangle$ state with a $\approx 0.79$ fidelity (yellow disks, Fig. 4.7).

4.6. Interplay between valley and spin relaxation

In Si quantum dots orbital relaxation happens on $10^{-12} - 10^{-7}$ s scale, spin relaxation $10^{-6} - 1$ s scale and valley relaxation is somewhere between the two values [138]. In order to include spin relaxation processes an additional term is added to the Lindblad equation Eq. (4.23). The Lindblad equation now has the form

$$\dot{\rho} = -\frac{i}{\hbar}[H_0, \rho] + \frac{\Gamma}{2}(2L^\dagger \rho L - LL^\dagger \rho - \rho LL^\dagger) + \frac{\gamma}{2}(2\Lambda^\dagger \rho \Lambda - \Lambda \Lambda^\dagger \rho - \rho \Lambda \Lambda^\dagger) = \mathcal{L}' \rho,$$

where $\mathcal{L}'$ is the $16 \times 16$ Lindblad superoperator acting on the density matrix represented in vector form. The explicit form of $\mathcal{L}'$ is given in the Appendix 1. Other than the terms introduced in Eq. (4.23), the newly introduced terms are the phenomenological spin relaxation rates $\gamma$ and two new Lindblad dissipation operators related to spin relaxation $\Lambda^\dagger = |\uparrow\rangle \langle \downarrow|$ and $\Lambda = |\downarrow\rangle \langle \uparrow|$.

Because the goal of the study is to obtain the echo envelope function as a measure of the coherence drop the starting point is a Lindblad equation in a superoperator form

$$\rho(t) = e^{\mathcal{L}'t} \rho(0).$$

By repeating the procedure from Section 4.5 (Eq. (4.25) and Eq. (4.26)), the echo envelope function is obtained Fig. 4.8 (probability that the electron spin is measured in the $|\uparrow\rangle$ state after a time $t$, when being subjected to a sequence of perfect $\pi/2 - \pi - \pi/2$ pulses). When the effective $g$-factor is valley dependent, the $\pi$ pulses perfect and electron spin relaxation is occurring the increase of the echo $P_\uparrow$ probability is caused by the interplay of valley and spin relaxations (Fig. 4.8, green circles and blue squares). The exponential function $f(P_\uparrow, \Gamma, \gamma, t) = 0.5(1 + P^0_\uparrow e^{-(\Gamma + \gamma/2)t} + P^0_\bar{v} e^{-\gamma t/2})$ describes the drop of coherence. In the $|\bar{v}\rangle$ state the drop of coherence is caused by both spin and valley relaxation processes, while in the $|\bar{v}\rangle$ valley the drop of coherence is caused by spin relaxation processes. By comparing the results for $T_1 = \infty$ (black dashed dotted line, Fig. 4.8) and $T_1 = 1$ ms (green dashed line, Fig. 4.8), one observes that the spin relaxation happening on $T_1 = 1$ ms timescales is increasing the $P_\uparrow$ probability by only $\sim 0.01$ on $\sim 200 \mu$s timescales.

In Fig. 4.9 imperfect $\pi$ and $\pi/2$ pulses are assumed, with the rotation oper-
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Figure 4.8. Spin-up probability after the echo sequence, when inter-valley scattering and spin relaxation are present. The parameters of the plot are the valley injection probabilities $P_0^v = 0.7$ and $P_0^\bar{v} = 0.3$, the inter-valley scattering rate $\Gamma = 25 \text{ kHz}$, the spin relaxation time $T_1 (\gamma = 1/T_1)$, the external magnetic field $B_z = 0.75 \text{ T}$ and the $z$-component of the magnetic field of the ferromagnet $B_z^{FM} = -0.12 \text{ T}$. The fitting function $f(P_0^v, \Gamma, \gamma, t) = 0.5(1 + P_0^v e^{-(\Gamma+\gamma/2)t} + P_0^\bar{v} e^{-\gamma t/2})$ was used.

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The state $R_x(\pi)$ = $\exp(-iH\pi/h\Omega_v)$ and $R_x(\pi/2)$ = $\exp(-iH\pi/h2\Omega_v)$, where the $H$ is given by Eq. (4.22), and $\Omega_v$ is the valley dependent Rabi frequency. During the free evolution stages the electron spin precesses around the external magnetic field. After the imperfect initial $\pi/2$ pulse, the electron spin is not perpendicular to the magnetic field, yielding rotations around the quantization axis with a frequency proportional to the Zeeman energy $g_v\mu_B (B_z + B_z^{FM})/h$, where $B_z$ is the external magnetic field and $B_z^{FM}$ is the $z$-component of the magnetic field of the ferromagnet. For $B_z = 0.75 \text{ T}$ and $B_z^{FM} = -0.12 \text{ T}$ this oscillations happen on $\sim 50 \text{ ps}$ timescale, with the amplitude of the oscillations being given by the valley dependent Rabi frequencies $\Omega_v$ and $\Omega_{\bar{v}}$ and Rabi dependent effective $g$-factors $g_v$ and $g_{\bar{v}}$. Therefore, the $P_\uparrow$ probability is very sensitive to the duration of the free evolution stage. The relaxation time $T_1 = 1 \text{ ms}$ is within the value suggested in a recent experimental study.

By comparing experimental data points (blue circles) with the result of the model presented here (purple diamonds) the conclusion is drawn that the saturation value of the $P_\uparrow$ probability $P_\uparrow(t \rightarrow \infty) \approx 0.39$ and the $P_\uparrow$ probability close to $t = 0$, $P_\uparrow(t = 0) \approx 0.25$ are all within the values measured in a recent experimental study [17] and that the findings presented here give the correct functional form of $P_\uparrow$ probability increase.
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Figure 4.9. Probability $P_\uparrow$ for the echo sequence yielding the electron $|\uparrow\rangle$ state. The blue circles represent experimental findings [17] and the purple diamonds are the results theoretical findings discussed in this chapter when the $\pi$ and $\pi/2$ pulses are imperfect and inter-valley and spin relaxations are present. The spin and valley injection probabilities are assumed to be $P_{0,\downarrow}^{\uparrow} = 0.49$, $P_{0,\uparrow}^{\uparrow} = 0.21$ and $P_{0,\downarrow}^{\uparrow} = 0.3$. The parameters of the plot are the external magnetic field $B_z = 0.75$ T, the $z$-component of the magnetic field of the ferromagnet $B_{FM}^z = -0.12$ T, valley relaxation rate $\Gamma = 25$ kHz, spin relaxation time $T_1 = 1$ ms, valley dependent Rabi frequencies corresponding to the miscut angle $\theta \approx 0.3^\circ$, $\Omega_v = 2\pi \cdot 3.1$ MHz and $\Omega_v = 2\pi \cdot 3.7$ MHz, all within the values suggested in a recent experimental study [17].

4.7. Conclusion

To conclude, the control of the electron spin inside a Si/SiGe quantum dot with a ferromagnet embedded on top was presented in this chapter. The stray magnetic field of the ferromagnet combined with Si/SiGe interference imperfections consequently leads to a valley dependent effective $g$-factor. When a valley dependent $g$-factor, alongside with valley relaxation is present, a novel decoherence mechanism exists, further limiting the coherence of the electron spin. Furthermore, the control of the electron spin state on the Bloch sphere is influenced by a valley dependent $g$-factor and Rabi frequency. The findings obtained in this chapter give a good qualitative and quantitative description of recent experimental studies. Further research on this topic will move towards including the drop of coherence due to the presence of nuclear spins.
5. Coherent manipulation of single electron spins with Landau-Zener sweeps

5.1. Summary

In this chapter a novel method to manipulate the state of a single electron spin in a semiconductor quantum dot (QD) is presented. The manipulation is achieved by tunnel coupling a QD, labeled $L$, and occupied with an electron to an adjacent QD, labeled $R$, which is not occupied by an electron but having an energy linearly varying in time. A parameter regime is identified in which a complete population transfer between the spin eigenstates $|L\uparrow\rangle$ and $|L\downarrow\rangle$ is achieved without occupying the adjacent QD. This method is convenient due to the fact that manipulation can be done electrically, without the precise knowledge of the spin resonance condition, and is robust against Zeeman level broadening caused by nuclear spins.

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5. Coherent manipulation of single electron spins with Landau-Zener sweeps

5.2. Introduction

Efficient initialization, manipulation and readout of single electron spins are crucial for the implementation of single electron spin qubits [18]. Spin-orbit interaction and stray magnetic fields provide a necessary toolkit to control the single electron spin [17, 65–67, 69, 139]. In Electric Dipole Spin Resonance (EDSR) microwaves drive an electron to oscillate in the spin-orbit field and/or the magnetic field gradient, producing a coherent spin rotation.

The Landau-Zener-Stückelberg-Majorana (LZSM) model [28–31] is one of the few analytically solvable time dependent problems in quantum mechanics. It has found applications modeling nano-electro-mechanical systems [140], optomechanical systems [141], Bose liquids [142], molecular magnets [143], Rydberg atoms [144], superconducting qubits [140, 145–148] and semiconductor singlet-triplet qubits [59, 108]. In the LZSM model the energy difference between two coupled states is varied linearly in time, while the coupling between the states is time independent. This results in a transition between the states with a probability determined by the coupling constant and the rate of the sweep.

Unlike the two level LZSM problem, multilevel LZSM problems are not exactly analytically solvable for a general case [34–38, 149, 150]. Chirped Raman Adiabatic Passage (CHIRAP) [151–154] and similar techniques [155–161] allow for efficient transfer of populations between two uncoupled levels. In order to utilize CHIRAP the energy of the radiatively decaying state is varied linearly in time with laser pulses having chirped frequencies.

Equivalently to CHIRAP, the goal of the scheme presented here is to transfer the population between two uncoupled levels $|L\uparrow\rangle$ and $|L\downarrow\rangle$ by coupling the levels of the $L$ quantum dot in a time-independent manner to an adjacent quantum dot $R$, with an energy linearly varying in time [162]. It should be noted that, as the probability to occupy the adjacent $R$ quantum dot remains negligible in this scheme, the states in the $R$ QD can be extremely susceptible to relaxation without influencing the efficiency of the scheme.

Two possible realizations of the scheme are discussed. In the first realization the $R$ quantum dot has significantly larger Zeeman splitting than the $L$ quantum dot. Then, the scheme operates even when the rate of spin-non-conserving tunneling events is significantly smaller than the rate of spin-conserving events. This regime is often present in GaAs double quantum dots. In the second realization the Zeeman splittings of the $L$ and $R$ quantum dots are comparable in magnitude but the rates of spin-conserving and spin-non-conserving tunneling events must be comparable. This regime can be reached in InAs double quantum dots.
5. Coherent manipulation of single electron spins with Landau-Zener sweeps

5.3. The Hamiltonian

An electron spin localized in the $L$ quantum dot coupled to a quantum dot $R$ with an energy of varied linearly in time (see Fig. 5.1) is modeled by,

$$H(t) = \sum_c \sum_\sigma E_{c,\sigma}(t)|c\sigma\rangle\langle c\sigma| + \tau \sum_{c \neq \bar{c}} \sum_\sigma |c\sigma\rangle\langle \bar{c}\sigma| + \tau_\Delta \sum_{c \neq \bar{c}} \sum_{\sigma \neq \bar{\sigma}} |c\sigma\rangle\langle \bar{c}\bar{\sigma}|.$$ (5.1)

The sum over the charge states runs over the $L$ and the $R$ quantum dots, $c = L, R$, and the sum over spin states runs over spin-up and spin-down states $\sigma = \uparrow, \downarrow$. Furthermore, $E_{c,\sigma}$ represents the energy with charge state $c$ and spin state $\sigma$. The energies of the $L$ quantum dot are time independent $E_{L\uparrow} = \Delta E_L/2$, $E_{L\downarrow} = -\Delta E_L/2$, where $\Delta E_L$ is the Zeeman splitting in the $L$ QD. The energies of the $R$ QD are time dependent with a linear time dependence, $E_{R\uparrow} = \Delta E_R + \beta t$, and $E_{R\downarrow} = \beta t$, where $\Delta E_R$ is the Zeeman splitting in the $R$ quantum dot, $t$ is time and $\beta$ the Landau-Zener velocity (see Fig. 5.1).

The off-diagonal terms in the Hamiltonian are the spin-conserving tunneling amplitude $\tau$, and the spin-non-conserving tunneling amplitude $\tau_\Delta$. The spin non-conserving tunneling can appear due to spin-orbit interaction or can be induced by the stray field of the micro-magnet, which is inhomogeneous in the tunneling direction [163, 164].

Figure 5.1. The energy diagram. The electron is initialized in the $|L\uparrow\rangle$ state, with the $R$ quantum dot being higher in energy. The energies of the states in $R$ quantum dot are ramped with a Landau-Zener velocity $\beta$. In the figure $\beta < 0$. The goal of the scheme is to find a parameter regime in which the adiabatic evolution path is followed (red dashed arrow). The Zeeman splittings of the $L$ and $R$ quantum dots are marked as $\Delta E_L$ and $\Delta E_R$ respectively.
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5.4. Different Zeeman splittings

The system is initialized in the $|L\uparrow\rangle$ state, at a negative instance of time. If the product of the Landau-Zener velocity and the total duration of the pulse $T$ is smaller than the Zeeman splitting of the $R$ quantum dot $\Delta E_R > \beta T$, and if the $R$ quantum dot is initially positively detuned with respect to the $L$ quantum dot, the system behaves like an effective three level system. Furthermore, if the evolution of the system is adiabatic ($\tau^2, \tau_\Delta^2 \gg \beta \hbar$), the system will remain in the instantaneous eigenstate of the Hamiltonian for the entire duration of the pulse $T$. Given all these assumptions, the adiabatic eigenvectors can be calculated, and therefore the time evolution of the three state probabilities

$$
\begin{align*}
P_{L\uparrow} &= \tau_\Delta^2 \frac{\left|\lambda(t) + \frac{\Delta E_L}{2}\right|^2}{N(t)^2}, \\
P_{R\downarrow} &= \frac{\left|\lambda(t)^2 - \frac{\Delta E_L^2}{4}\right|^2}{N(t)^2}, \\
P_{L\downarrow} &= \tau^2 \frac{\left|\lambda(t) - \frac{\Delta E_L}{2}\right|^2}{N(t)^2},
\end{align*}
$$

(5.2)

where $\lambda(t)$ is the appropriate adiabatic eigenvalue (see Appendix 2 for the expression for $\lambda(t)$) and $N(t)$ is the normalization of the adiabatic eigenvectors.

For simplicity it is not stated that $\lambda(t)$ is also a function of $\Delta E_L, \beta, \tau, \tau_\Delta$. Depending on the values of $\tau$ and $\tau_\Delta$, $\lambda(t) = 0$ close to $t = 0$ (for $\tau = \tau_\Delta$), $\lambda(t) = 0$ at $t > 0$ (for $\tau > \tau_\Delta$) and $\lambda(t) = 0$ at $t < 0$ (for $\tau < \tau_\Delta$). Furthermore, the adiabatic eigenvalue takes the following values $\lambda(t = \mp\infty) = \pm\Delta E_L/2$, $-\Delta E_L/2 \leq \lambda(t) \leq \Delta E_L/2$, for every $t$. Therefore, the maximal possible occupation probabilities are $P_{L\uparrow}^\text{max} \sim \tau_\Delta^2 \Delta E_L^2$, $P_{R\downarrow}^\text{max} \sim \Delta E_L^4$, $P_{L\downarrow}^\text{max} \sim \tau^2 \Delta E_L^2$. If $\tau, \tau_\Delta \gg \Delta E_L$ no significant population will occupy the $R$ quantum dot, $P_R \approx 0$ at every instance of time (see Fig. 5.2), and a complete population transfer between the spin eigenstates $|L\uparrow\rangle$ and $|L\downarrow\rangle$ occurs.

In contrast to EDSR techniques the scheme presented here does not require the precise knowledge of the spin resonance condition ($\Delta E_L$) and operates without a need of a microwave source. However, in order for the scheme to be successful a necessary requirement is that the quantum dots have significantly different Zeeman splittings $\Delta E_L \ll \Delta E_R$. For a typical double quantum dot system where the distance between the quantum dots is $\sim 200$ nm the required gradient would be $dB_z/dx \sim 10$ T/µm, which is a factor of 10 larger than the currently maximally achieved experimental value [65, 70]. A possible way to induce a large enough difference of Zeeman energies between quantum dots is to embed a micro-magnet.
5. Coherent manipulation of single electron spins with Landau-Zener sweeps

Figure 5.2. The comparison between the numerically computed probabilities (obtained from evolving the state using the Hamiltonian of Eq. (5.1)) (Num) and analytic adiabatic three level probabilities Eq. (5.2) (An). The parameters of the plot are the Landau-Zener velocity $\beta = 10^5 \text{eV/s}$, the tunnel coupling $\tau = 70 \mu \text{eV}$, the spin-non-conserving tunnel coupling $\tau_\Delta = 0.2\tau$, the external magnetic field Zeeman splitting in the L QD $\Delta E_L = 20\mu\text{eV}$ and Zeeman splitting in the R quantum dot $\Delta E_R = 250\Delta E_L$. The inset represents the magnification of the occupation probabilities of the states in the R quantum dot.

on top of the one of the QDs (for instance $R$), and further, engineer the $g$-factor of the $R$ QD [79, 165–167].

5.5. Equal Zeeman splittings

Again the system is initialized in the $|L \uparrow\rangle$ state, at a negative instance of time. Another way for the scheme to be successful is that the magnitude of spin-conserving and spin-non-conserving tunnelings are comparable $\tau \approx \tau_\Delta$. The requirement for the scheme to work $\tau/\tau_\Delta \sim 4l/3\Lambda_{\text{SO}} \approx 1$ can be fulfilled in InAs [168]. Here, $l$ is the interdot separation and $\Lambda_{\text{SO}}$ is the spin-orbit length, defined by [86, 169]

$$\frac{1}{\Lambda_{\text{SO}}} = \frac{m^*}{\hbar} \sqrt{\cos^2(\phi(\beta - \alpha))^2 + \sin^2(\phi(\beta + \alpha))^2}, \quad (5.3)$$

for a 2DEG in the [001] plane. Here, $m^*$ is the effective electron mass, $\phi$ is the angle between the [110] crystallographic axis and the interdot connection axis and
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\( \beta \) and \( \alpha \) are Dresselhaus and Rashba spin-orbit constants respectively.

Possible ways of controlling the spin-orbit interaction is the variation of angle between the external magnetic field and the spin-orbit field [170], variation of the direction in which the DQD is grown [16] (and therefore maximizing \( \cos \phi \)), isotopic control of the Indium in InGaAs, or electric field control of the Rashba constant [171, 172].

In the adiabatic limit \( (\tau^2 = \tau_\Delta^2 \gg \beta \hbar) \), the system will remain in the instantaneous eigenstate of the Hamiltonian for the entire duration of the pulse \( T \). In that limit, the adiabatic eigenvectors can be calculated, and therefore the time evolution of four states probabilities

\[
\begin{align*}
P_{L\uparrow} &= \tau^2 \frac{|\Lambda(t) + \Delta E_L/2|^2}{\tilde{N}(t)^2}, \\
P_{R\downarrow} &= P_{R\uparrow} = \frac{|\Lambda(t)^2 - \Delta E_L^2/4|^2}{2\tilde{N}(t)^2}, \\
P_{L\downarrow} &= \tau^2 \frac{|\Lambda(t) - \Delta E_L/2|^2}{\tilde{N}(t)^2},
\end{align*}
\]

where \( \Lambda(t) \) is the corresponding adiabatic eigenvalue and \( \tilde{N}(t) \) the wavefunction normalization.

The requirement that spin-conserving and spin-non-conserving tunnel couplings are equal is due to the fact that when \( \Delta E_L = \Delta E_R \) the adiabatic eigenfunctions have only a negligible contribution of the two states of the \( R \) quantum dot when \( \tau \approx \tau_\Delta \). In the case of \( \tau \gg \tau_\Delta \), the adiabatic eigenfunctions have only a small component in the \( |R\downarrow\rangle \) state when \( \Delta E_L \ll \tau, \tau_\Delta \), and the \( |R\uparrow\rangle \) state is detuned during the duration of the pulse \( T \).

Similarly to the previous implementation of the scheme, the appropriate adiabatic eigenvalue spans between \( \Lambda(t = \mp \infty) = \pm \Delta E_L/2 \), \( -\Delta E_L/2 \leq \Lambda(t) \leq \Delta E_L/2 \), for every \( t \), with \( \Lambda(t) = 0 \) for \( t \approx 0 \). The maximal possible occupations of states for the case \( \Delta E_L = \Delta E_R \) are

\[
\begin{align*}
P_{L\uparrow}^{\text{max}} &= \tau^2 \Delta E_L^2, & P_{L\downarrow}^{\text{max}} &= \tau^2 \Delta E_L^2, \\
P_{R\downarrow}^{\text{max}} &= P_{R\uparrow}^{\text{max}} \sim \Delta E^4_L/2.
\end{align*}
\]

Equivalently to CHIRAP, the probabilities to occupy the \( |R\downarrow\rangle \) and \( |R\uparrow\rangle \) states is negligible at all instances of time \( P_R \approx 0 \) in the case when \( \tau \gg \Delta E_L \) (see Fig. 5.3), and a complete population transfer between the spin eigenstates \( |L\uparrow\rangle \) and \( |L\downarrow\rangle \) occurs.
5. Coherent manipulation of single electron spins with Landau-Zener sweeps

Figure 5.3. The comparison between the numerically computed probabilities (obtained from evolving the state using the Hamiltonian of Eq. (5.1)) (Num) and analytic adiabatic four level probabilities Eq. (5.4) (An). The inset represents the magnification of the probability to occupy the $R$ quantum dot. The parameters of the plot are the Landau-Zener velocity $\beta = 4 \cdot 10^6$ eV/s, the external magnetic field $B_L = B_R = 20$ mT, the tunnel hopping $\tau = 50$ µeV, the $g$-factor in the $L$ and the $R$ quantum dot $g_L = g_R = 14.7$.

5.6. Experimental realizations

The manipulation scheme under study works optimally when the Zeeman splitting of the $L$ QD is small. Furthermore, different signs of the Landau-Zener velocity and initial detunings need to be used for different initial spin states. The problem of initializing and measuring electron spin states when the Zeeman splitting in the $L$ QD is small is addressed in the remaining part of this subsection.

If the thermal broadening of the lead is smaller than the Zeeman splitting of the electron spin states $k_B T_e \ll \Delta E_L$, the state of the spin qubit can be initialized by tuning the chemical potential of a nearby lead close to the $|\downarrow\rangle$ state of the spin qubit. When lead-to-dot relaxation occurs the only possible state to which the electron can relax from the lead is the $|\downarrow\rangle$ state. Furthermore, single shot measurement of the electron spin state can be achieved in a similar manner [68], by tuning the chemical potential of the lead in such a way so that only one of the states can tunnel out of the quantum dot to the lead.

As the scheme under study operates optimally in low magnetic fields $k_B T_e > \Delta E_L$, the initialization and readout, validating the efficiency of the scheme, must be done in an alternative way, via the $R$ QD. The chemical potential of the lead coupled to the $R$ QD can be tuned between the spin states of the $R$ QD. After
5. Coherent manipulation of single electron spins with Landau-Zener sweeps

the successful initialization the $|R \downarrow\rangle$ state, the spin is shuttled to the $|L \downarrow\rangle$ state, followed by a manipulation of the spin according to the described scheme.

After the manipulation stage the modification in the current of a quantum point contact (QPC) near to $R$ is monitored. If the current of the QPC is unchanged, this means that the manipulation stage did not produce any leakage to the $R$ quantum dot and that the spin measurement stage can follow. In the spin measurement stage states $|L \downarrow\rangle$ and $|R \downarrow\rangle$ are aligned in energy one more time. If the electron spin was in the $|L \downarrow\rangle$ state a tunneling event occurs and a nearby QPC modifies its current accordingly [77, 78]. On the other hand if the electron spin was in the $|L \uparrow\rangle$ state the current of the QPC would remain unchanged.

In the case of $\Delta E_L = \Delta E_R$ (and therefore $\tau \approx \tau_{\Delta}$) and when $\Delta E_L < k_B T_e$ the initialization could still be achieved by waiting a sufficiently long time for the electron spin to relax to the thermal equilibrium state. However, spin readout would need to be done with alternative methods, because both spin eigenstates are energetically allowed to tunnel to the $R$ QD when $|L \downarrow\rangle$ and $|R \downarrow\rangle$ are aligned in energy. This is why the case $\Delta E_L \ll \Delta E_R$ is considered to be more likely to implement in future experiments, and the influence of nuclear spin noise will only be considered for this realization.

5.7. Errors due to nuclear spins

In order to make the effects of the uncertainties in the nuclear magnetic field noticeable in the remaining part of this section the difference of Zeeman energies between quantum dots will be assumed to originate from large difference in electron $g$-factors. Therefore, the influence of nuclear spins is modeled as a distribution of the magnetic field in the $L$ and $R$ quantum dot, centered around the external magnetic field in the $L$ and the $R$ dot $\Delta E_L, \Delta E_R$, with standard deviations $\sigma = g_L \mu_B B_N$, $\chi = g_R \mu_B B_N$, where $g_L(R)$ is the electron $g$-factor in the $L (R)$ quantum dot, $\mu_B$ is the Bohr magneton and $B_N$ is the root-mean-square of the distribution of the nuclear magnetic field. The influence of nuclear spins on the manipulation scheme can be estimated by averaging the probabilities of all relevant states over a distribution of nuclear spins.
5. Coherent manipulation of single electron spins with Landau-Zener sweeps

\begin{equation}
\hat{P}_{c\sigma} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{P_{c\sigma}}{2\pi \chi \sigma} \exp \left[ -\left( \frac{\Delta E - \Delta E_L}{2\sigma} \right)^2 \right] \times \exp \left[ -\left( \frac{\beta t' - \beta t}{2\chi} \right)^2 \right] d(\Delta E) d(\beta t''), \quad (5.5)
\end{equation}

where \(c = L, R\), \(\sigma = \uparrow, \downarrow\), with the exclusion of the detuned \(|R\uparrow\rangle\) state.

Fig. 5.4 shows how the nuclear spins influence the control scheme in the case of no uncertainty of the magnetic field in the \(R\) quantum dot, \(\chi = 0\). If the random nuclear field is parallel with the external magnetic field this gives rise to more leakage into the \(|R\downarrow\rangle\) state. However, if the random nuclear field is anti-parallel with the external magnetic field this gives rise to less leakage into the \(|R\downarrow\rangle\) state, and this two effects (less and more leakage to \(|R\rangle\)) cancel first order in \(\Delta E_L\).

Fig. 5.5 presents the behavior of the control scheme under an influence of random nuclear spins in both quantum dots. Other then the already mentioned mechanism of additional leakage, the uncertainties in the nuclear field in the \(R\) quantum dot (and therefore the position of the level \(|R\downarrow\rangle\)) lead to reduced maximal probability to occupy the \(|R\downarrow\rangle\) state (Fig. 5.5, inset, gray disks versus green circles). In contrast to EDSR, a full transfer of population between the spin eigenstates is achieved, even when the uncertainty in the energy difference between spin eigenstates is large (Fig. 5.5 black empty squares and triangles).

An effective nuclear magnetic field of unknown intensity in the \(z\) direction is going to change the instance of time in which the energy of the state \(|R\downarrow\rangle\) is located between the energies of the states \(|L\uparrow\rangle\) and \(|L\downarrow\rangle\). For a nuclear magnetic field parallel with the external field the energy of the state \(|R\downarrow\rangle\) is located between the energy of the states \(|L\uparrow\rangle\) and \(|L\downarrow\rangle\) at a time \(t < 0\). In contrast to that, for a nuclear magnetic field anti-parallel with the external field the energy of the state \(|R\downarrow\rangle\) is located between the energies of the states \(|L\uparrow\rangle\) and \(|L\downarrow\rangle\) at a time \(t > 0\). A process like this is described with a Gaussian distribution \([85, 117, 173]\), centered around \(\beta t\) with a standard deviation \(\chi = g_R \mu_B B_N\) where, \(g_R\) is the \(g\)-factor in the \(R\) quantum dot, \(g_R \gg g_L\). This leads to a reduced maximal value of the occupation of the \(|R\downarrow\rangle\) state, without changing the averaged occupation of the \(|R\downarrow\rangle\) per unit time \(\bar{P}_{R\downarrow}(T) = \int_{-T/2}^{T/2} \bar{P}_{R\downarrow}(t) dt / T = \text{const.}\) for a large enough \(T\). Since the nuclear spins do not affect the final probabilities, the scheme can be operated in the presence of nuclear spin induced decoherence, as long as the total sweep time is shorter than the characteristic time of nuclear spin evolution.

It should be noted that quasi-static detuning noise yields the same effect like an uncertain nuclear spin distribution in the \(R\) quantum dot, and therefore this issue
5. Coherent manipulation of single electron spins with Landau-Zener sweeps

![Figure 5.4](image)

Figure 5.4. Spin manipulation in the presence of nuclear spins. The parameters of the plot are the Landau-Zener velocity $\beta = 10^5 \text{ eV/s}$, the tunnel coupling $\tau = 70 \mu\text{eV}$, the spin-non-conserving tunnel coupling $\tau_\Delta = 0.2\tau$, the external magnetic field $B = 0.4 \text{T}$, the standard deviation in the $R$ quantum dot $\chi = 0$, the $g$-factor in the $L$ quantum dot $g_L = 0.44$. The inset represents occupation of the states in the $R$ quantum dot.

![Figure 5.5](image)

Figure 5.5. Spin manipulation in the presence of nuclear spins. The parameters of the plot are the Landau-Zener velocity $\beta = 10^5 \text{ eV/s}$, the tunnel coupling $\tau = 70 \mu\text{eV}$, the spin-non-conserving tunnel coupling $\tau_\Delta = 0.2\tau$, the external magnetic field $B = 0.4 \text{T}$, the $g$-factor in the $L$ quantum dot $g_L = 0.44$, the $g$-factor in the $R$ quantum dot $g_R = 250g_L$. The inset represents occupation of the states in the $R$ quantum dot.

is not addressed separately in this thesis.
5.8. Conclusions and final remarks

To conclude, a novel method to manipulate a single electron spin is proposed, using Landau-Zener sweeps. The control method is robust against the uncertainties of the nuclear field and static charge noise, operates without a source of microwaves and without the precise knowledge of the spin resonance condition.
6. Charge-Noise-Insensitive Superexchange of Single Electron Spin Qubits

6.1. Summary

In this chapter superexchange is investigated, the indirect exchange interaction between two single electron spin qubits, each embedded in a single semiconductor quantum dot (QD). The exchange interaction is mediated by an intermediate, empty QD. The findings presented here suggest the existence of a double “sweet spot”, where the superexchange is first order insensitive to charge noise. Furthermore, the sign of the superexchange can be changed by varying the detunings between the QDs, and that the form of superexchange is influenced by spin-orbit interaction.

This Chapter is adapted from Marko J. Rančić and Guido Burkard, *Manuscript in preparation*
6.2. Introduction

Noise-insensitive control of qubits is an important task in quantum information science [18, 102]. The exchange interaction has been utilized to control double [59, 71, 82, 113] and triple electron spin qubits [93–95] in semiconductor quantum dots (QDs), and to perform two qubit operations on two single electron spin qubits [59]. However, overcoming the sensitivity of exchange interaction to charge noise [114, 121] and errors originating from spin-orbit interaction [174, 175] proved to be a challenging task.

Three electron spin qubits can be operated close to a “sweet spot”, where the sensitivity of exchange interaction to charge noise vanishes in first order [94, 95, 176]. On the other hand, two electron $S-T_0$ spin qubits embedded in double QDs, only have a trivial first order “sweet spot”, where the exchange interaction is close to zero. A possibility to reduce the sensitivity of the $S-T_0$ qubit to electric noise is to control the magnitude of the exchange interaction by controlling the tunnel coupling instead of controlling the detuning between the two dots (symmetric operation) [80, 81].

The spin-orbit interaction represents a powerful resource to control spin qubits [69, 73]. On the other hand, it can also reduce the coherence time of the electron spin qubit, hamper efforts to prolong the coherence time of the electron spin qubit [88, 170], and lead to errors in two qubit operations [174, 175].

Superexchange is the underlying mechanism responsible for the creation of anti-ferromagnetic order in CuO and MnO [177, 178], a possible mechanism allowing $d$-wave high $T_c$ superconductivity [179] and allows for switching between ferromagnetic and anti-ferromagnetic order in cold atomic gases [180]. Although the possibility to use mediated exchange (superexchange) was mentioned in the original Loss-DiVincenzo proposal [18], superexchange has not received significant attention from the quantum dot community [181, 182]. The reason for this lies in the fact that compared to the direct exchange superexchange requires an additional quantum dot.

In this chapter superexchange is investigated, the exchange interaction between two single electron spin qubits, each embedded in a semiconductor QD left (L) and right (R), mediated by an empty quantum dot (C). The form of superexchange is derived and a parameter regime is discovered in which the superexchange is not zero and is simultaneously insensitive in first order to fluctuations in gate detunings (a non-trivial first order double “sweet spot”). Further findings presented here suggest that the sign and the magnitude of superexchange can be controlled by varying the detunings between the QDs. Finally, the magnitude and form of superexchange is influenced by the spin-orbit interaction, giving rise to a Dzyaloshinskii-Moriya contribution to the superexchange interaction.
6.3. Superexchange in quantum dots

The superexchange is a fourth-order tunneling process, the $(1, 0, 1)$ charge state with antiparallel spin, virtually tunnels either to the $(1, 1, 0)$ or $(0, 1, 1)$ state. Furthermore, $(1, 1, 0)$ or $(0, 1, 1)$ state tunnels to the $(2, 0, 0)$, $(0, 2, 0)$ or $(0, 0, 2)$ charge state, followed by a tunneling to the $(1, 1, 0)$ or $(0, 1, 1)$ state and again to the $(1, 0, 1)$ charge state, but with the spin state of the $L$ and $R$ QD exchanged.

The system is described with a generalized Hubbard Hamiltonian $H = H_0 + H'$ for two electrons in a triple quantum dot,

$$H_0 = \sum_{i\sigma} (\varepsilon_i + E_{2\sigma}) n_{i\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \sum_{(ij)} V n_i n_j,$$

$$H' = \sum_{(ij)} \left[ \sum_{\sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} + \sum_{\sigma \neq \bar{\sigma}} t_{ij}^{\bar{\sigma}} c_{i\sigma}^{\dagger} c_{j\bar{\sigma}} \right].$$

Figure 6.1. The scheme of all possible superexchange paths in absence of spin-orbit interactions. All superexchange paths involve four tunneling events, two between the left and the center QDs ($t_{LC}$, marked with dotted blue lines) and two between the center and the right ($t_{CR}$, marked with dashed red lines) QDs. $\uparrow$ stands for a spin up state, $\downarrow$ for a spin down state and fields in the parentheses denote charge states ($L, C, R$).
Figure 6.2. Schematic representation of matrices used in the Schrieffer-Wolff transformation. The full Hamiltonian is divided into the diagonal part of the Hamiltonian $H_0$ consisting of high energy $h$ and superexchange states $s$. The interacting part $H'$ is divided into $H_1$ consisting of interactions between the high energy states $(A)$ and the part $H_2$ which describes the coupling between the low energy and high energy states. $S$ is an anti-Hermitian matrix which has the same block structure as $H_2$ and $C = C^\dagger$.

Here, $E_z$ is the Zeeman energy due to an external magnetic field, $t^{so}_{ij}$ the magnitude of spin-non-conserving tunnel hopping caused by spin-orbit interaction, $t_{ij}$ is the magnitude of spin-conserving tunnel hopping between dots $i$ and $j$. Furthermore, $\varepsilon_i$ the energy bias of the $i$-th dot, $U$ is the Coulomb penalization of the doubly occupied quantum dot and $V$ is the Coulomb energy of two neighboring dots occupied with single electrons. $n_i = n_{i\uparrow} + n_{i\downarrow} = c^\dagger_{i\uparrow} c_{i\uparrow} + c^\dagger_{i\downarrow} c_{i\downarrow}$ the number operator, with $c_{i\sigma}(c^\dagger_{i\sigma})$ being the spin creation (annihilation) operator of the $i$ charge state with spin $\sigma = \downarrow, \uparrow$. The $\langle ij \rangle$ in the index of the sum denotes that the sum goes over nearest neighbor QDs $i$ and $j$, and the index $\sigma \neq \bar{\sigma}$ denotes a double sum which runs over all possible states of different spin.

The Schrieffer-Wolff transformation divides Hamiltonians into decoupled low $s$ and high $h$ energy parts [4, 183]. In the case of interest the Schrieffer-Wolff transformation is used to derive an effective $4 \times 4$ Hamiltonian in the superexchange $s = \{ (\uparrow, 0, \downarrow), (\downarrow, 0, \uparrow), (\uparrow, 0, \uparrow), (\downarrow, 0, \downarrow), (0, \uparrow, \downarrow), (0, \downarrow, \uparrow), (0, \uparrow, \downarrow), (\uparrow, 0, \downarrow), (\uparrow, 0, \uparrow), (0, 0, \downarrow), (0, 0, \uparrow) \}$ subspace, decoupling the 11 dimensional subspace of high energy states $h = \{ (\uparrow, \downarrow, 0), (\downarrow, \uparrow, 0), (\uparrow, \uparrow, 0), (\downarrow, \downarrow, 0), (0, \uparrow, \downarrow), (0, \downarrow, \uparrow), (0, \downarrow, \downarrow), (\uparrow, 0, \downarrow), (\uparrow, 0, \uparrow), (0, 0, \downarrow), (0, 0, \uparrow) \}$ from the superexchange states $s$.

The Hamiltonian $H$ consist of a diagonal part $H_0$ Eq. (6.1), and an interaction part $H'$ Eq. (6.2). The interaction part $H'$ can further be split into a part involving interactions between different high energy states $H_1$ and a part involving interactions between the low energy $s$ states and high energy states $h \ H_2$ Fig. 6.2 (for more details see Appendix 3).

The effective $4 \times 4$ Hamiltonian is derived $\tilde{H} = e^S H e^{-S}$, where $S$ is anti-Hermitian. Because the energy separation between the low and high energy states is big compared to the spin-conserving and spin-non-conserving hopping $|E_s - E_h| \gg t_{sh}, t^{so}_{sh}$ $S$ can be expanded in a power series, $S = S_1 + S_2 + S_3...$, where each of the $S_j$ is $j$th order in perturbation, and each of the $S_j$ has the same
6. Charge-Noise-Insensitive Superexchange of Single Electron Spin Qubits

Figure 6.3. The energy diagram. $E$ is the energy and $x$ the position. $\epsilon$ is the energy difference between the outer dots ($L$ and $R$) and $\delta$ the energy between the average energy of the outer dots ($L$ and $R$) and the $C$ QD.

The Hamiltonian up to forth order in perturbation theory in the superexchange subspace $s$ (for more details see Appendix 3)

$$\hat{H} = H_0 + H_1 + \frac{1}{2!}[H_2, S_1 + S_2 + S_3] - \frac{1}{4!}[[H_2, S_1]S_1, S_1].$$  \hspace{1cm} (6.4)$$

The Coulomb repulsion of doubly occupied quantum dots is characterized by an energy of $U \sim 1 \text{ meV}$, and the Coulomb repulsion of neighboring dots being occupied $V \sim 0.1U - 0.01U$. Therefore, the Coulomb repulsion of neighboring dots being occupied can be neglected $V = 0$ for simplicity reasons.

In the remaining part of the chapter a linear triple QD arrangement is assumed, allowing one to neglect spin-conserving and spin-non-conserving tunnel hoppings between the R and the L dot, $t_{LR} = 0$, $t_{LR}^{\sigma} = 0$. Furthermore, from now on spin-conserving hoppings from left-to-center QD $t_{LC}$ and from center-to-right QD $t_{CR}$ will be assumed to have equal magnitudes and be denoted as $t$. Likewise, the spin-non-conserving hoppings from left-to-center QD $t_{LC}^{\sigma}$ and the hoppings
from center-to-right QD \( t_{so}^{CR} \) have equal magnitudes and are denoted as \( t_{so} \). For a 2DEG in the [001] plane and Rashba \( \alpha \) and Dresselhaus \( \beta \) constants of same signs, this means that the magnitude of the spin-orbit hopping \( t_{so} \) is maximal when the linear triple quantum dot is structured along the [\( \bar{1}10 \)] crystallographic axis and minimal when the triple quantum dot is structured along the [110] crystallographic axis. An assumption will be made regarding the direction in which the quantum dots and the magnetic field are oriented in the rest of the chapter. Quantum dots are assumed to be structured along the [110] crystallographic axis, giving rise to a spin-orbit field in the [\( \bar{1}10 \)] direction. Furthermore, the magnetic field (and therefore the \( z \) axis of the coordinate system) is assumed to be parallel with the [001] direction while the \( y \) axis is assumed to be parallel with [110]. The relation between the spin-conserving \( t \) and spin-non-conserving \( t_{so} \) hopping is given by

\[
t_{so} = \frac{4t}{3} \frac{l}{\Lambda_{so}},
\]

(6.5)

where, \( l \) is the interdot separation, and

\[
\Lambda_{so} = \frac{\hbar}{m^* \sqrt{\sin^2(\alpha + \beta)^2 + \cos^2(\alpha - \beta)^2}}
\]

(6.6)

is the spin-orbit length, where \( \phi \) is the angle between the [110] crystallographic axis and the interdot connection axis. Detunings in the Hamiltonian Eq. (6.1) can be expressed in terms of two detuning parameters, the detuning between the outer dots \( \epsilon \) and the detuning between the center dot and average detuning of the outer dots \( \delta \). Fig. 6.3.

### 6.4. Results

The effective Hamiltonian in the low energy subspace \( s \) is

\[
\tilde{H} = J_{SE} \mathbf{S}_1 \cdot \mathbf{S}_2 + D_{12} (\mathbf{S}_1 \times \mathbf{S}_2).
\]

(6.7)

Here, \( \mathbf{S}_1 \) and \( \mathbf{S}_2 \) are spin operators applied to the first and second electron and \( J_{SE} \) is the superexchange involving spin-conserving tunnel hoppings

\[
J_{SE} = \frac{4t^4 U (U (12\delta^2 + \epsilon^2)) - \delta (8\delta^2 + 6\epsilon^2)}{(\epsilon^2 - 4\delta^2)^2 (U - 2\delta) (U^2 - \epsilon^2)}.
\]

(6.8)
The second term in Eq. 6.7 is the antisymmetric contribution to the exchange interaction, the so-called Dzyaloshinskii-Moriya interaction [184, 185]. The quantity $D_{12}$ is the Dzyaloshinskii-Moriya vector

$$D_{12} = \frac{4tt_{so}\delta (8E_z^4 - 2E_z^2 (12\delta^2 + \epsilon^2) + (\epsilon^2 - 4\delta^2)^2)}{(\epsilon^2 - 4\delta^2) \left(4 (E_z - \delta)^2 - \epsilon^2\right) \left(4 (E_z + \delta)^2 - \epsilon^2\right)^2} \hat{y}. \quad (6.9)$$

In the process of deriving Eqs. (6.7-6.9) all terms higher than $t^4$, $t_{so}^2$ and $t_{so}t^3$ were neglected. Therefore, the results are valid when $t_{so} \ll t$, a situation often present in GaAs quantum dots.

In the remaining part of the chapter Rashba $\alpha$ and Dresselhaus constant $\beta$ are assumed to have comparable magnitudes [186]. If these constants also have the same sign in the previously assumed geometry (QDs structured along the [110] and a 2DEG in the plane perpendicular to the (001) and the external magnetic field, the contribution of spin-orbit interaction is negligible $\Lambda_{so} \to \infty$ (see Eq. 6.6) and [186]. This happens because then Rashba and Dresselhaus spin-orbit interactions cancel out. Equal $\alpha$ and $\beta$ will be assumed in the remaining part of the chapter.

A non-trivial superexchange “sweet spot” is a point in which the superexchange is first order insensitive to fluctuations of the detuning parameters $\delta$ and $\epsilon$, and furthermore the superexchange is not zero. Solving the coupled systems of equations $\partial J_{SE}/\partial \epsilon = 0$, $\partial J_{SE}/\partial \delta = 0$ and $J_{SE} \neq 0$ for $\epsilon$ and $\delta$, in the case of vanishing spin-orbit interaction four solutions for $\epsilon$ and $\delta$ are obtained in which the superexchange is first order insensitive to charge noise $\epsilon_{1,2} = 0$, $\delta_{1,2} = (5 \pm \sqrt{13})/4$, $\epsilon_{3,4} = \pm 0.745$, $\delta_{3,4} = 0.074$ in units of $U$ and “sweet spots” $J_1(\delta_1, \epsilon_1) = 0.08$, $J_2(\delta_2, \epsilon_2) = 64.65$, $J_3(\delta_3, \epsilon_3) = J_4(\delta_4, \epsilon_4) = 13.8$ in the units of $t^4/U^3$, where $t$ is the tunneling and $U$ is Coulomb repulsion Fig. 6.4.

In contrast to a double QD loaded with two electrons a linear triple QD loaded with two electrons has four points in the parameter space of $\epsilon$ and $\delta$ in which the exchange interaction is simultaneously first order insensitive in fluctuation of these two parameters. It should be noted that “sweet spots” $J_2$, $J_3$ and $J_4$ lie close to the areas in which no superexchange takes place due to leakage outside the superexchange subspace (white regions in Fig. 6.4). The width of the white areas in Fig. 6.4 is proportional to tunneling $t$, and this imposes a limit beyond which the magnitude of superexchange cannot be increased by increasing the tunnel coupling, while simultaneously performing superexchange at the double “sweet spot”.

Solving $J_{SE} = 0$ (Eq. (6.8)) $\delta_0$ is calculated for which the superexchange is zero for any value of $\epsilon$ and $\epsilon_0$ for which the superexchange is zero for any value of $\delta$ see Fig. 6.4.
\[ \delta_0 = \frac{1}{2} \left( 1 + \frac{1 - \epsilon^2}{q^{1/3}} + q^{1/3} \right) ; \quad \epsilon_0 = \pm \frac{2\sqrt{3 - 2\delta}}{\sqrt{6\delta - 1}}, \] (6.10)

where, \( q = 1 - \epsilon^2 + \sqrt{(\epsilon^2(\epsilon^2 - 1))} \) all given in the units of Coulomb repulsion \( U \). It should be noted that the result is symmetric with respect to the sign of \( \epsilon \). When \( \epsilon = -1.34U \), at large negative values of the bias \( \delta \) the main contribution of the superexchange comes from the path (6) which gives rise to negative superexchange (see Tab. I) as the bias is increased towards the positive values, the superexchange path (1) becomes more dominant yielding a positive sign of superexchange (see Fig. 6.5 (a)).

Now the dynamical evolution of spin states caused by superexchange interaction will be investigated. The starting point is the initialization of the \((\uparrow,0,\downarrow)\) state. The time evolution of the system in the superexchange subspace is mod-

![Figure 6.4. The superexchange involving spin-conserving paths \( J_{SE} \) as a function of the detuning parameters \( \epsilon \) and \( \delta \). The points represent the superexchange “sweet spots” \( J_1(\delta_1,\epsilon_1) = 0.08, J_2(\delta_2,\epsilon_2) = 64.65, J_3(\delta_3,\epsilon_3) = 13.62 \) in the units of \( t^4/U^3 \) where \( t \) is the tunneling and \( U \) is Coulomb repulsion. The black line marks \( J_{SE} = 0 \), black dashed line \( J_{SE} = 10 \) and white dashed line \( J_{SE} = -10 \). The white regions represent areas in which \( \Delta E_{ijk} \), the energy difference between the \((2,0,0)\), \((0,2,0)\), \((0,0,2)\), \((1,1,0)\), \((0,1,1)\) charge states and superexchange states becomes comparable to \( t \), and therefore no superexchange takes place. For the white regions in this plot \( t = 17.8 \mu eV \) and \( \Delta E_{ijk} > 8t \) are chosen. \]
<table>
<thead>
<tr>
<th>Nr.</th>
<th>Superexchange path</th>
<th>Superexchange expression</th>
<th>Sign of $J_{SE}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1)</td>
<td>$(\uparrow,0,\downarrow) \leftrightarrow (\uparrow,\downarrow,0) \leftrightarrow (0,\uparrow,0) \leftrightarrow (\downarrow,\uparrow,0) \leftrightarrow (\downarrow,0,\uparrow)$</td>
<td>$t^4/ [(U - 2\delta)(\epsilon/2 + \delta)^2]$</td>
<td>$J_{SE} &gt; 0$</td>
</tr>
<tr>
<td>(2)</td>
<td>$(\uparrow,0,\downarrow) \leftrightarrow (0,\uparrow,\downarrow) \leftrightarrow (0,\uparrow,\downarrow,0) \leftrightarrow (0,\downarrow,\uparrow) \leftrightarrow (\downarrow,\uparrow,0)$</td>
<td>$t^4/ [(U - 2\delta)(\epsilon/2 - \delta)^2]$</td>
<td>$J_{SE} &gt; 0$</td>
</tr>
<tr>
<td>(3)</td>
<td>$(\uparrow,0,\downarrow) \leftrightarrow (\uparrow,\downarrow,0) \leftrightarrow (0,\uparrow,\downarrow,0) \leftrightarrow (0,\uparrow,\downarrow) \leftrightarrow (\downarrow,\uparrow,0)$</td>
<td>$-t^4/ [(U - 2\delta)(\epsilon/2 - \delta)(\epsilon/2 + \delta)]$</td>
<td>$J_{SE} &lt; 0$</td>
</tr>
<tr>
<td>(4)</td>
<td>$(\uparrow,0,\downarrow) \leftrightarrow (0,\uparrow,\downarrow) \leftrightarrow (0,\uparrow,\downarrow,0) \leftrightarrow (\downarrow,\uparrow,0) \leftrightarrow (\downarrow,0,\uparrow)$</td>
<td>$-t^4/ [(U - 2\delta)(\epsilon/2 - \delta)(\epsilon/2 + \delta)]$</td>
<td>$J_{SE} &lt; 0$</td>
</tr>
<tr>
<td>(5)</td>
<td>$(\uparrow,0,\downarrow) \leftrightarrow (\uparrow,\downarrow,0) \leftrightarrow (\uparrow,0,\downarrow,0) \leftrightarrow (\uparrow,0,\downarrow) \leftrightarrow (\downarrow,0,\uparrow)$</td>
<td>$t^4/ [(U - \epsilon)(\epsilon/2 + \delta)^2]$</td>
<td>$J_{SE} &gt; 0$</td>
</tr>
<tr>
<td>(6)</td>
<td>$(\uparrow,0,\downarrow) \leftrightarrow (0,\uparrow,\downarrow) \leftrightarrow (0,0,\uparrow) \leftrightarrow (0,\uparrow,0) \leftrightarrow (\downarrow,\uparrow,0)$</td>
<td>$t^4/ [(U + \epsilon)(\epsilon/2 - \delta)^2]$</td>
<td>$J_{SE} &lt; 0$</td>
</tr>
</tbody>
</table>

Table I. Six possible superexchange paths involving spin-conserving tunneling with corresponding expressions. The parameters for which the sign of $J_{SE}$ is valid are the Coulomb repulsion $U = 1$ meV, the detuning between the outer dots $\epsilon = -1.34U$, the detuning between the middle dot and the average of the outer dots $-0.2U < \delta < 0.3U$. 

6. Charge-Noise-Insensitive Superexchange of Single Electron Spin Qubits
6. Charge-Noise-Insensitive Superexchange of Single Electron Spin Qubits

Figure 6.5. (a) Magnitude of different exchange paths in the context of Tab. I in the case of vanishing spin-orbit. The horizontal black dashed line represents the point $\delta_0$ in which the superexchange is zero. (b) The geometry of the quantum dots with respect to the crystallographic axes. (c) Coherent superexchange oscillations as a function of the detuning $\delta$ and time $T$ in the case of vanishing spin-orbit interaction. The probability to occupy the $(\downarrow, 0, \uparrow)$ state is not displayed as $P_{\uparrow0\downarrow} = 1 - P_{\downarrow0\uparrow}$. Parameters of the plots are tunneling $t = 17.8 \mu$eV, detuning $\epsilon = -1.34$ meV, the Coulomb repulsion $U = 1$ meV.

eled in a following way $\rho_{\text{SE}}(T) = U\rho_{\text{SE}}(0)U^\dagger$, where $\rho_{\text{SE}}(0)$ is the initial density matrix corresponding to the initialization of the $(\uparrow, 0, \downarrow)$ state, $\rho_{\text{SE}}(T)$ the density matrix at time $T$, and $U = \exp(-i\tilde{H}T/\hbar)$ where $\tilde{H}$ is given by Eq. (6.7). Fig. 6.5 (c) shows that superexchange oscillations are suppressed around the point $\delta = \delta_0$. Areas above and below the black line correspond to different signs of superexchange.

6.5. Conclusion

To conclude coherent superexchange was investigated and points in parameter space were found in which the superexchange is insensitive to charge noise. Furthermore, the sign of the superexchange can be changed and the form of superexchange when spin-non-conserving tunneling is present was derived. Spin-orbit
interaction induces a Dzyaloshinskii-Moriya contribution to the superexchange interaction. An experimental implementation of the findings presented here would allow for charge noise-insensitive two qubit operation of the Loss-DiVincenzo qubit and charge-noise-insensitive control of the $S - T_0$ qubit around the exchange axis.
7. Conclusion

To conclude this thesis has dealt with spin-based quantum computing in InGaAs and Si quantum dots. The nuclear spin rich environment in InGaAs limits the coherence time of the electron spin qubit and prevents the efficient manipulation of the qubit. Efforts to prolong the coherence time of the electron spin qubit are hampered by spin-orbit interaction. However, spin-orbit interaction also provides a novel method for nuclear-spin-robust preparation of the electron spin. This novel method operates without a source of microwaves, and is robust against uncertainties of the Zeeman level broadening caused by a nuclear spin environment.

Silicon represents a nuclear-spin-free alternative to InGaAs. However, the fact that Si has an additional valley degree of freedom prevents extremely long coherence times of the electron spin. This decoherence mechanism is mediated by indirect valley-spin mixing occurring due to the imperfections on the Si/SiGe interface giving rise to valley-orbit mixing and stray magnetic field of the micromagnet, giving rise to spin-orbit mixing.

Exchange interaction is a powerful resource for implementing two qubit gates, however it also couples to the electrical noise. A possible workaround is to use mediated (superexchange) instead of exchange. This would allow for the qubit to be operated at a points where the qubit does not couple to electric noise in first order.

Further studies in these topics are going to be in the direction of making realistic models incorporating charge noise into dynamical nuclear polarization, making more realistic, microscopic models of quantum wells and investigating mediated exchange schemes with more than one mediator.

To conclude the author of this thesis is going to dear to give his vision of a perfect spin qubit in a semiconductor quantum dot. It would be embedded in a single Si/SiGe quantum dot with a perfectly flat Si/SiGe interface, purified from nuclear spins. Two axis control would be achieved by an external magnetic field and a stray field of a micromagnet, while superexchange interaction would be used for two qubit operations.
A. Appendices

1. Lindblad superoperators $L'$ and $L$ in matrix representation

In this Appendix an explicit form of the Lindblad superoperator $L'$ will be given (see Eq. 4.27). The Lindblad superoperator is a $16 \times 16$ superoperator acting on the density matrix in a vector representation (a 16 dimensional column vector).

$$L' = \begin{pmatrix} A_1 & 0 \\ A_3 & A_2 \end{pmatrix}. \quad \text{(A.1)}$$

Here the $8 \times 8$ matrices $A_1$, $A_2$, $A_3$ are given by

$$A_1 = \frac{\Gamma}{2} \mathbb{1} - \begin{pmatrix} \gamma + \frac{\Gamma}{2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \gamma + \frac{\Gamma}{2} + \frac{i \delta E}{\hbar} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \gamma - \frac{i \delta E}{\hbar} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{i (\delta E + 2 E_z) \hbar + \gamma}{2} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \gamma - \frac{i \delta E}{\hbar} & 0 & 0 & 0 \\ -\gamma & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\gamma & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \frac{i \delta E / \hbar}{2} \end{pmatrix} \quad \text{(A.2)}$$

$$A_2 = -\begin{pmatrix} \gamma + \frac{\Gamma}{2} + \frac{i \delta E}{\hbar} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \gamma + \frac{\Gamma}{2} + \frac{i \delta E}{\hbar} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \gamma & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{i \delta E + E_z + \gamma}{\hbar} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{\gamma + \Gamma}{2} - \frac{i \delta E / \hbar}{2} & 0 & 0 & 0 \\ -\gamma & 0 & 0 & 0 & 0 & \frac{\Gamma - i \delta E / \hbar}{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\gamma & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -\gamma \end{pmatrix} \quad \text{(A.3)}$$

Appendix 1 is adapted from Marko J. Rančić and Guido Burkard, Phys. Rev. B 93, 205433 (2016)
Here, $E_z$ is the Zeeman energy in the $|v⟩$ valley eigenstate, $E_z + δE$ the Zeeman energy in the $|\bar{v}⟩$ valley eigenstate, $Γ$ is the phenomenological valley relaxation rate, and $γ$ the phenomenological spin relaxation rate. Note that $L$ can be obtained from $L'$ by setting $γ = 0$ (see Eq. 4.23).

2. Adiabatic eigenvectors of the three level Landau-Zener problem

Here a way to calculate adiabatic eigenvectors for a three level Landau-Zener problem will be presented. The results obtained here are used in Chapter 5. When the $|R⟩$ quantum dot has a Zeeman splitting much larger then the $|L⟩$ quantum dot (QD), is initially positively detuned with respect to $|L⟩$, and the duration of the pulse is such that one of the levels in the $|R⟩$ quantum dot does not overlap with the states in the $|L⟩$ QD, the system behaves like an effective three level system. If the evolution is adiabatic ($τ, τ_Δ ≫ βT$) the system will remain in the instantaneous eigenstate.

Non-trivial adiabatic eigenvectors $f_{ik}$ are obtained by diagonalizing the matrix of Eq. (5.1) for an unknown adiabatic eigenvalue $λ_k(t)$, equaling the equation for $f_{1k}$ and $f_{3k}$ with zero and equaling the equation for $f_{2k}$ with $\text{det} |H(t) - λ_k I|$ [38].

This procedure yields
\[
\begin{align*}
  f_{1k}(t) &= \tau \Delta \frac{\lambda_k(t) + \Delta E_L/2}{N_k(t)}, \\
  f_{2k}(t) &= \frac{\lambda_k^2(t) - \Delta E_L^2/4}{N_k(t)}, \\
  f_{3k}(t) &= \tau \frac{\lambda_k(t) - \Delta E_L/2}{N_k(t)}.
\end{align*}
\]  
(A.5)  
(A.6)  
(A.7)

where \(\tau\) is the spin conserving tunnel hopping, \(\tau_\Delta\) is the spin-non-conserving tunnel hopping, \(\Delta E_L\) is the Zeeman splitting in the left quantum dot, \(\lambda_k(t)\) is the time dependent adiabatic eigenvalue and \(N_k(t)\) are normalization factors \((k = 1, 2, 3)\).

After solving the secular equation, the adiabatic energies are obtained
\[
\begin{align*}
  \lambda_1 &= \frac{1}{3} a(t) - \frac{2}{3} s(t) \cos \frac{\theta(t)}{3}, \\
  \lambda_2 &= \frac{1}{3} a(t) + \frac{2}{3} s(t) \cos \frac{\theta(t) + \pi}{3}, \\
  \lambda_3 &= \frac{1}{3} a(t) + \frac{2}{3} s(t) \cos \frac{\theta(t) - \pi}{3},
\end{align*}
\]  
(A.8)  
(A.9)  
(A.10)

where
\[
\begin{align*}
  a(t) &= -\beta t, \\
  b(t) &= -\left(\Delta E_L^2/4 + \tau^2 + \tau_\Delta^2\right), \\
  c(t) &= \Delta E_L \left(\Delta E_L \beta t/2 + \tau_\Delta^2 - \tau^2\right)/2, \\
  s(t) &= \sqrt{a(t)^2 - 3b(t)}, \\
  \cos \theta(t) &= -\frac{2a(t)^3 - 9a(t)b(t) + 27c(t)}{2s(t)^3}.
\end{align*}
\]  
(A.11)  
(A.12)  
(A.13)  
(A.14)  
(A.15)

It should be noted that the expressions for the adiabatic eigenvalue \(\lambda_k(t)\) are written in such a way that the energy of the \(|R \downarrow\rangle\) is centered around zero at \(t = 0\). In Chapter 5 \(\lambda(t) = \lambda_2(t), \ P_{L\uparrow} = |f_{12}(t)|^2, \ P_{R\downarrow} = |f_{22}(t)|^2, \ P_{L\downarrow} = |f_{32}(t)|^2\) was used. The adiabatic eigenvalues and eigenvectors used to compute the probabilities in the case when \(\tau = \tau_\Delta\) (see Eq. (5.4)) are computed in an equivalent manner.
Appendices

3. The Schrieffer-Wolff Transformation

The full Hamiltonian describing two electrons in a triple quantum dot \( H = H_0 + H' \), comprises of the diagonal part \( H_0 \) and the off-diagonal part \( H' \)

\[
H_0 = \sum_{i\sigma} (\varepsilon_i + E_z \sigma)n_{i\sigma} + U \sum_i n_{i\uparrow}n_{i\downarrow} + \sum_{\langle ij \rangle} V n_{i\sigma}n_{j\sigma}, \quad (A.16)
\]

\[
H' = \sum_{\langle ij \rangle} \left[ \sum_{\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \sum_{\sigma \neq \bar{\sigma}} t_{ij}^{so} c_{i\sigma}^\dagger c_{j\bar{\sigma}} \right]. \quad (A.17)
\]

Here, \( E_z \) is the Zeeman energy due to an external magnetic field, \( t_{ij}^{so} \) the magnitude of spin-non-conserving tunnel hopping caused by spin-orbit interaction, \( t_{ij} \) is the magnitude of spin-conserving tunnel hopping between dots \( i \) and \( j \). Furthermore, \( \varepsilon_i \) the energy bias of the \( i \)-th dot, \( U \) is the Coulomb penalization of the doubly occupied quantum dot and \( V \) is the Coulomb energy of two neighboring dots occupied with single electrons. \( n_i = n_{i\uparrow} + n_{i\downarrow} = c_{i\sigma}^\dagger c_{i\sigma} + c_{i\sigma}^\dagger c_{i\sigma} \) the number operator, with \( c_{i\sigma}(c_{i\sigma}^\dagger) \) being the spin creation (annihilation) operator of the \( i \) charge state with spin \( \sigma = \downarrow, \uparrow \). The \( \langle ij \rangle \) in the index of the sum denotes that the sum goes over nearest neighbor QDs \( i \) and \( j \), and the index \( \sigma \neq \bar{\sigma} \) denotes a double sum which runs over all possible states of different spin. A linear arrangement is assumed, neglecting all direct couplings between the \( L \) and \( R \) QDs.

The Hamiltonian \( H \) is 15 dimensional and it comprises of the 4 dimensional superexchange subspace \( s = \{ (\uparrow, 0, \downarrow), (\downarrow, 0, \uparrow), (\uparrow, 0, \uparrow), (\downarrow, 0, \downarrow) \} \) and the 11 dimensional high energy subspace \( h = \{ (\uparrow, \downarrow, 0), (\downarrow, \uparrow, 0), (\uparrow, \uparrow, \downarrow), (\downarrow, \uparrow, \downarrow), (0, \uparrow, \downarrow), (0, \downarrow, \uparrow) \} \).

\[
\begin{array}{ccc}
H_0 & H_1 & H_2 \\
\hline
S & A & 0 \\
0 & 0 & t, t_{so}
\end{array}
\]

Figure A.1. Schematic representation of matrices used in the Schrieffer-Wolff transformation. The full Hamiltonian is divided into the diagonal part of the Hamiltonian \( H_0 \) consisting of high energy \( h \) and superexchange states \( s \). The interacting part \( H' \) is divided into \( H_1 \) consisting of interactions between the high energy states (A) and the part \( H_2 \) which describes the coupling between the low energy and high energy states. \( S \) is an anti-Hermitian matrix which has the same block structure as \( H_2 \) and \( C = C^\dagger \).

Appendices 3 and 4 are adapted from Marko J. Rančić and Guido Burkard, Manuscript in preparation.

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(0, ↑, ↑), (0, ↓, ↓), (↑, 0, 0), (0, ↑, ↓), (0, 0, ↑, ↓)\} from the superexchange states \( s \). The diagonal part of the Hamiltonian \( H_0 \) comprises of superexchange states \( s \) and high energy states \( h \) (see Fig. A.1). The interaction part of the Hamiltonian \( H' = H_1 + H_2 \) can be divided into terms containing interaction between different \( h \) states \( H_1 \) and terms containing interactions between the \( s \) and \( h \) states \( H_2 \).

The following unitary transformation is applied to the Hamiltonian \( \tilde{H} = e^{-S}He^S \), where \( S \) must be anti-Hermitian to ensure the unitarity of the transformation. Expanding \( e^{\pm S} \) in a Taylor series the Hamiltonian equals

\[
\tilde{H} = \sum_{j=0}^{\infty} \frac{1}{j!}[H, S]^{(j)},
\]

(A.18)

where \([H, S]^{(i+1)} = [[H, S]^{(i)}, S] \) and \([H, S]^{(0)} = H\). Assuming that \( S \) has the same block structure like \( H_2 \) the transformed Hamiltonian can be separated into (BD) block-diagonal part (having the block structure of \( H_0 + H_1 \)) and (OD) off-diagonal part (having the same structure like \( H_2 \))

\[
\tilde{H}_{OD} = \sum_{j=0}^{\infty} \frac{1}{(2j+1)!}[H_0 + H_1, S]^{(2j+1)} + \sum_{j=0}^{\infty} \frac{1}{(2j)!}[H_2, S]^{(2j)},
\]

\[
\tilde{H}_{BD} = \sum_{j=0}^{\infty} \frac{1}{(2j)!}[H_0 + H_1, S]^{(2j)} + \sum_{j=0}^{\infty} \frac{1}{(2j+1)!}[H_2, S]^{(2j+1)}.
\]

(A.19)

The goal of the Schrieffer-Wolff transformation is to derive the effective Hamiltonian in the (BD) form. Assuming the separation between the superexchange states \( s \) and the high energy states \( h \) is large compared to the tunnel couplings \( |E_s - E_h| \gg t, t_{so} S = S_1 + S_2 + S_3... \), where each \( S \) is \( j \)th order in perturbation. The energy separation between the \{\( (1, 1, 0) \), \( (0, 1, 1) \), \( (2, 0, 0) \), \( (0, 2, 0) \), \( (0, 0, 2) \)\} charge states and the superexchange states can be made sufficiently large by adjusting the detuning parameters.

Every order of \( S \) is determined by requiring that the \( OD \) part of the effective Hamiltonian vanishes. This gives rise to a set of coupled equations for determining orders of \( S \)

\[
[H_0, S_1] = -H_2; [H_0, S_2] = -[H_1, S_1]; [H_0, S_3] = -[H_1, S_2] - \frac{1}{3}[[H_2, S_1], S_1].
\]

(A.20)

By inserting Eq. (A.20) into Eqs. (A.19) the following expressions for the effective Hamiltonian is obtained in \( j \)th order of perturbation \( \tilde{H}^{(j)} \)
\[ \tilde{H}^{(0)} = H_0, \]
\[ \tilde{H}^{(1)} = H_1, \]
\[ \tilde{H}^{(2)} = [H_2, S_2] + \frac{1}{2}[H_0, S_1]^{(2)}, \]
\[ \tilde{H}^{(3)} = [H_2, S_2] + \frac{1}{2}[H_0, S_1]^{(2)} + \frac{1}{2}[[H_0, S_1], S_2] + \frac{1}{2}[[H_0, S_2], S_1], \]  
(A.21)

By using Eq. (A.20) this further simplifies to

\[ \tilde{H}^{(0)} = H_0, \]
\[ \tilde{H}^{(1)} = H_1, \]
\[ \tilde{H}^{(2)} = \frac{1}{2!}[H_2, S_1], \]
\[ \tilde{H}^{(3)} = \frac{1}{2!}[H_2, S_2], \]
\[ \tilde{H}^{(4)} = \frac{1}{2!}[H_2, S_3] - \frac{1}{4}[H_2, S_1]^{(3)}. \]  
(A.22)

This yields

\[ \tilde{H} = \tilde{H}^{(0)} + \tilde{H}^{(1)} + \tilde{H}^{(2)} + \tilde{H}^{(3)} + \tilde{H}^{(4)} \]
\[ = H_0 + H_1 + \frac{1}{2!}[H_2, S_1 + S_2 + S_3] - \frac{1}{4}[[H_2, S_1]S_1], S_1]. \]  
(A.23)

4. Comparison between the evolution involving the superexchange subspace and full Hilbert space

Here, a comparison is presented between the time evolution governed by an effective 4 × 4 Hamiltonian in the \( s = \{ (\uparrow, 0, \downarrow), (\downarrow, 0, \uparrow), (\uparrow, 0, \uparrow), (\downarrow, 0, \downarrow) \} \) subspace, obtained by eliminating 11 states with a Schrieffer-Wolff transformation and a time evolution governed by a 15 × 15 Hamiltonian involving all states in the \( h \) subspace Fig. A.2.
Figure A.2. Coherent superexchange oscillations of the \( (\uparrow, 0, \downarrow) \) state as a function of the detuning between the middle dot and the average of outer dots \( \delta \) and time \( T \). The dashed line represents the point \( \delta_0 \) in which the superexchange is zero. Parameters of the plot are the tunneling \( t = 17.8 \mu \text{eV} \), the Coulomb repulsion \( U = 1 \text{meV} \), detuning between the outer dots \( \epsilon = -1.34U \). (a) The probability to occupy the \( (\uparrow, 0, \downarrow) \) state when all 15 states included in the modeling of the dynamics. (b) The probability to occupy the \( (\uparrow, 0, \downarrow) \) state when only superexchange states are included in the modeling of the dynamics and other 11 states are eliminated with a Schrieffer-Wolff transformation. (c) The absolute difference of probabilities to occupy the \( (\uparrow, 0, \downarrow) \) state between the evolution when all 15 states are included and when 11 states are eliminated with a Schrieffer-Wolff transformation. The probability to occupy the \( (\downarrow, 0, \uparrow) \) state is not displayed as \( P_{\uparrow 0 \downarrow} \approx 1 - P_{\downarrow 0 \uparrow} \).

The two ways of modeling time evolution produce results which do not differ by more than 5%, and therefore the result obtained by the Schrieffer-Wolff transformation are valid in the domain of applicability of the Schrieffer-Wolff transformation.


[57] D. P. DiVincenzo, in Mesoscopic electron transport (Springer, 1997), pp. 657–677,


http://science.sciencemag.org/content/297/5585/1313.


Bibliography


Reference List


