QUANTUM DOT SPINS AND MICROCAVITIES
FOR QUANTUM INFORMATION PROCESSING

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Abstract

Semiconductor quantum dots are attractive building blocks for scalable quantum information processing systems. The spin-up and spin-down states of a single electron, trapped inside a quantum dot, form a single quantum bit (qubit) with a long decoherence time. The electron spin qubit can be coupled to excited states using an optical field, which provides an opportunity to quickly manipulate the spin with optical pulses, and to interface between a stationary matter spin qubit and a ‘flying’ photonic qubit for quantum communication. The quantum dot’s interaction with light may be enhanced by placing the quantum dot inside of an optical microcavity. Finally, the entire system is monolithically integrated, and modern semiconductor fabrication technology offers a path towards scalability.

This work presents experimental developments towards the utilization of single quantum dot electron spins in quantum information processing. We demonstrate a complete set of all-optical single-qubit operations on a single quantum dot spin: initialization, an arbitrary gate, and measurement. First, initialization into a pure spin state (logical 0) is accomplished on a nanosecond timescale by optical pumping. Next an ultrafast single-qubit gate, consisting of a series of broadband laser pulses, rotates the spin to any arbitrary position on the Bloch sphere within 40 picoseconds. Finally, the spin state is measured by photoluminescence detection.

We then combine these operations to perform a ‘spin echo’ sequence on the quantum dot. The spin echo extends the qubit’s coherence (memory) time from a few nanoseconds to a few microseconds, more than $10^5$ times longer than the single-qubit gate time. We next investigate a feedback mechanism between the electron spin and nuclear spins within the quantum dot, which leads to dynamical pumping of the
quantum dot’s nuclear magnetization.

Finally, we take the first step towards interfacing a stationary matter quantum bit with a flying photonic qubit by strongly-coupling a quantum dot exciton to a pillar microcavity. An anti-crossing of the cavity and excitonic modes indicates that the exciton and cavity modes are coupled more strongly to each other than to the rest of their environment, while photon statistics prove that we have successfully isolated and coupled a single quantum dot exciton.
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Twenty years from now you will be more disappointed by the things that you didn’t do than by the ones you did do. – Mark Twain

and

If you don’t do it this year, you will be one year older when you do. – Warren Miller
# Contents

**Abstract**

1 **Introduction**

1.1 Quantum Computing Ingredients ............................................ 3

1.1.1 Single-qubit gates .................................................. 3

1.1.2 Two-qubit gates ....................................................... 5

1.2 Quantum Computing Architecture ........................................... 5

1.2.1 Surface Code Requirements .......................................... 6

1.2.2 Surface Code Resources Estimate ..................................... 8

1.3 Physical Qubit Candidates .................................................. 8

1.3.1 Trapped Ion Qubits ................................................... 9

1.3.2 Trapped Neutral Atom Qubits ....................................... 9

1.3.3 Superconducting Qubits ............................................. 10

1.3.4 Diamond Nitrogen Vacancy Center Qubits .......................... 11

1.3.5 Electron Spin in Quantum Dot Qubits .............................. 11

1.3.6 Qubit Comparison .................................................... 12

1.4 Quantum Computing Criteria .............................................. 12

1.5 Thesis Outline ............................................................ 13

2 **Quantum Dots**

2.1 Quantum Dot States in Magnetic Fields ................................. 18

2.1.1 Neutral Quantum Dots .............................................. 19

2.1.2 Charged Quantum Dots ............................................. 21
2.2 Charged QD Samples .............................................. 25
2.3 Magneto-Photoluminescence Experimental Setup .................. 26
2.4 Magneto-Photoluminescence Experiments ......................... 30

3 Coherent Single Qubit Control ........................................ 34
  3.1 Ultrafast Rotation Theory - Raman Transition Picture ............. 35
    3.1.1 3-level System ............................................. 35
    3.1.2 4-level System ............................................. 38
  3.2 Ultrafast Rotation Theory - Stark-shift Picture ................... 41
  3.3 Initialization and Measurement Theory ................................ 43
  3.4 Experimental Setup ............................................. 44
  3.5 Initialization Experiment ....................................... 47
  3.6 Rabi Oscillations ............................................... 49
  3.7 Ramsey Interference ............................................. 50
  3.8 Arbitrary Single-Qubit Gate ..................................... 54

4 Spin Echo ............................................................ 56
  4.1 Nuclear Hyperfine Interaction ...................................... 57
  4.2 Racetrack Analogy for $T^*_2$, $T_2$, and Spin Echo ............... 59
  4.3 Experimental Setup ............................................... 59
  4.4 Sample Structure ............................................... 62
  4.5 Rabi Oscillations and Ramsey Fringes ................................ 63
  4.6 Spin Echo and $T^*_2$ ............................................. 64
  4.7 Spin Echo and $T_2$ ............................................... 66

5 Nuclear Spin Pumping .................................................. 71
  5.1 Experimental Setup ............................................... 72
  5.2 Experimental Results ............................................. 74
  5.3 Modeling .......................................................... 74
    5.3.1 Trion-driven Nuclear Spin Flips ................................ 76
    5.3.2 Nuclear Spin Relaxation ...................................... 77
    5.3.3 Mathematical Model ........................................... 78
<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.3.4 Model Results and Comparison to Data</td>
<td>79</td>
</tr>
<tr>
<td>5.4 Discussion</td>
<td>81</td>
</tr>
<tr>
<td>6 Strong Coupling in a Pillar Microcavity</td>
<td>82</td>
</tr>
<tr>
<td>6.1 Theory of Quantum Dot-Microcavity Coupling</td>
<td>83</td>
</tr>
<tr>
<td>6.1.1 Energy Levels and Decay Rates</td>
<td>85</td>
</tr>
<tr>
<td>6.1.2 Weak Coupling Regime</td>
<td>87</td>
</tr>
<tr>
<td>6.1.3 Strong Coupling Regime</td>
<td>88</td>
</tr>
<tr>
<td>6.2 Semiconductor Microcavities</td>
<td>89</td>
</tr>
<tr>
<td>6.2.1 Photonic Crystal Cavities</td>
<td>89</td>
</tr>
<tr>
<td>6.2.2 Microdisk Cavities</td>
<td>89</td>
</tr>
<tr>
<td>6.2.3 Pillar Microcavities</td>
<td>90</td>
</tr>
<tr>
<td>6.3 History of Strong Coupling</td>
<td>90</td>
</tr>
<tr>
<td>6.4 Pillar Microcavity Sample Design</td>
<td>91</td>
</tr>
<tr>
<td>6.5 Micropillar PL: above-band versus resonant excitation</td>
<td>92</td>
</tr>
<tr>
<td>6.6 Strong coupling PL</td>
<td>95</td>
</tr>
<tr>
<td>6.7 Photon Statistics</td>
<td>95</td>
</tr>
<tr>
<td>6.7.1 Photon Correlation Setup</td>
<td>97</td>
</tr>
<tr>
<td>6.7.2 Autocorrelation Results</td>
<td>98</td>
</tr>
<tr>
<td>6.7.3 Cross-Correlation Results</td>
<td>99</td>
</tr>
<tr>
<td>6.8 Single Photon Source</td>
<td>102</td>
</tr>
<tr>
<td>7 Conclusions and Future Directions</td>
<td>104</td>
</tr>
<tr>
<td>7.1 Current Status</td>
<td>104</td>
</tr>
<tr>
<td>7.2 Future Work</td>
<td>106</td>
</tr>
<tr>
<td>7.2.1 Site-controlled Quantum Dots</td>
<td>106</td>
</tr>
<tr>
<td>7.2.2 Single-shot Qubit Measurement</td>
<td>107</td>
</tr>
<tr>
<td>7.2.3 Further Extension of Decoherence Time by Dynamical Decoupling</td>
<td>108</td>
</tr>
<tr>
<td>7.2.4 Two-qubit Gate</td>
<td>109</td>
</tr>
<tr>
<td>A Qubit Rotation Details</td>
<td>110</td>
</tr>
<tr>
<td>A.1 Bloch Vector Trajectory Reconstruction</td>
<td>110</td>
</tr>
</tbody>
</table>
## List of Figures

1.1 (a) A simple 2-level qubit, with the ground state $|0\rangle$ and excited state $|1\rangle$ connected by an optical transition. (b) A three-level Λ-system. Two long-lived states, $|0\rangle$ and $|1\rangle$, are connected to an excited state by optical transitions. ................................. 3

1.2 Bloch sphere representation of a single qubit state $|\Psi\rangle$. .............. 4

1.3 Notation for a single-qubit gate, or rotation R. It rotates the input qubit $x$ into the output qubit $x'$. .................................................. 5

1.4 (a) Notation for a CNOT gate, with input qubits $x$ and $y$ and output qubits $x'$ and $y'$. (b) Truth table for the CNOT gate. ............... 6

1.5 A small section of a huge two-dimensional array of qubits necessary for a topological surface code quantum computer. Half of the physical qubits store data, while the other half are measured to detect errors called the syndrome. ................................. 7

1.6 A table summarizing the current state-of-the-art of the five qubit candidates discussed. .......................................................... 13

2.1 (a) A schematic of QD energy levels. (b) Different pumping schemes to generate a QD exciton, and relaxation channels for electrons and holes. .................................................. 17
2.2 (a) Atomic force micrograph of self-assembled InGaAs QDs before a GaAs capping layer is grown. Image by courtesy of Bingyang Zhang. (b) PL spectrum of an ensemble of InGaAs QDs. i - GaAs free and impurity-bound exciton emission, ii - QD emission. (c) PL spectrum of a 600 nm mesa showing individual QD emission lines.

2.3 Neutral and charged QDs in a Faraday magnetic field. (a) Eigenstates of a neutral QD. The eigenstates at zero field are listed in the left column, eigenstates at high magnetic field are listed to the right of the diagram. Optically active transitions are shown in red arrows, with their polarization labeled. The crystal ground state (empty QD) is denoted $|0\rangle$. (b) Eigenstates of a charged QD. Normalization factors have been dropped for compactness.

2.4 Neutral and charged QDs in a Voigt magnetic field. (a) Eigenstates of a neutral QD. (b) Eigenstates of a charged QD. Normalization factors have been dropped for compactness.

2.5 (a) The planar structure of charged QD sample M471 as-grown. (b) The planar sample is etched into mesas containing 10-100 QDs each.

2.6 (a) The planar structure of charged QD sample M471 as-grown. (b) The planar sample is etched into mesas containing 10-100 QDs each.

2.7 Experimental setup for magneto-PL.

2.8 Solid immersion lenses, showing how the collected solid angle is increased. (a) No SIL, (b) hemispherical SIL, (c) hyper-hemispherical SIL.

2.9 PL spectra of a mesa at various magnetic fields, showing a charged QD and a neutral QD. Horizontal and vertical polarized spectra have been summed to make the figure.
2.10 Line splittings as a function of magnetic field for (a) neutral and (b) charged QD from Figure 2.9. The square data points indicate PL emission of one polarization, the circular points indicate the orthogonal polarization. The two polarizations have arbitrarily been named H and V. The solid lines in (a) are the eigenvalues of equation 2.3, using $g_{e,z} = 0.3$ and $g_{h,z} = 0.37$ (or vice-versa). The solid lines in (b) are the eigenvalues of (equation 2.6) $\pm$ (equation 2.7), using $g_{e,z} = 0.35$ and $g_{h,z} = 0.25$ (or vice-versa).

2.11 (a) PL intensity (encoded in color map) as a function of wavelength for various polarization angles, showing one charged QD and one neutral QD. (b) Higher-resolution PL intensity versus wavelength and polarization for another charged QD, showing four well-resolved peaks. Note that the outer peaks are polarized orthogonally to the inner peaks, as predicted in Figure 2.4(b).

3.1 A general 3-level Λ-system, with ground-state splitting $\delta_e$. An optical field, detuned by $\Delta$ from the excited state, couples the two transitions with Rabi frequencies $\Omega_{0,1}$.  

3.2 The 4-level system of a negatively-charged QD in a Voigt magnetic field, with electron splitting $\delta_e$ and trion splitting $\Delta$. The four transitions are coupled by Rabi frequencies $\Omega_{1,2,3,4}$.  

3.3 (a) The polarization of the two optically active transitions of a charged QD in no magnetic field. (b) The states rewritten in the eigenstates of a Voigt magnetic field, showing the relative matrix elements of the optical transitions between the four states and corresponding polarizations.  

3.4 (a) The polarization of the two optically active transitions of a charged QD in no magnetic field. (b) The states rewritten in the eigenstates of a Voigt magnetic field, showing the relative matrix elements of the optical transitions between the four states and corresponding polarizations.  

3.5 The spin initialization and measurement scheme performed by optical pumping.
3.6 Experimental setup. One or two rotation pulses may be sent to the sample during each experimental cycle, to observe Rabi oscillations or Ramsey interference, respectively. The time delay $\tau$ between pairs of pulses is controlled by a retroreflector mounted on a computer-controlled translation stage.

3.7 Details of the double-monochromator. The first stage consists of the pinhole, G1, and S1. The second stage consists of S1, G2, and S2. G1: grating 1, holographic 2000 lines/mm; G2: grating 2, ruled 1716 lines/mm. S1 and S2: slit 1 and slit 2; SPCM: single-photon counting module.

3.8 Measured photoluminescence spectrum of the charged QD excited by an above-bandgap 785 nm laser. The rotation pulse is detuned by $\Delta/2\pi = 290$ GHz below the lowest transition.

3.9 (a) Saturation of the spin initialization process via optical pumping, showing single photon signal versus optical pump power for a fixed rotation angle $\Theta = \pi$. The operating power $P_{\text{op}}$ for the optical pump in all subsequent experiments is indicated. (b) Time-resolved measurement of optical pumping following a fixed rotation by angle $\Theta = \pi$. The count rate is fit by an exponential decay with a 3.4 ns time constant.

3.10 (a) Rabi oscillations between the spin states are evident in the oscillating photon signal as rotation pulse power $P_{\text{rf}}$ is increased. (b) The rotation angle as a function of rotation pulse power, showing an empirical fit to a power-law dependence. (c) Amplitude of measured Rabi oscillations as a function of rotation angle, with an empirical exponential fit.
3.11 Experimental trajectory of the Bloch vector. The curves trace out the tip of the Bloch vector in the one-pulse (Rabi oscillation) experiment over the range of rotation angles Θ from 0 to 3π. The color scale indicates the length of the Bloch vector, which shrinks exponentially with Θ. Views are from the perspective of: (a): the x-axis, and (b): the −y-axis of the Bloch sphere. The length of the Bloch vector and rotation angle is extracted from the extrema of the Rabi oscillation data shown in Figure 4.6, while the azimuthal position of the Bloch vector is revealed by the phase of the Ramsey fringes shown in Figure 3.13.

3.12 (a) Ramsey interference fringes for a pair of π/2 pulses, showing photon count rate versus time delay between pulses. (b) Destructive Ramsey interference for a pair of π pulses. The data in (a) and (b) are fit to an exponentially-decaying sinusoid with a linear offset (see Supplementary Information for details).

3.13 (a) Photon count rate is color-mapped as a function of rotation angle Θ and delay time between pulses τ. (b) The amplitude of Ramsey fringes for various rotation angles. Fringe amplitudes are determined by fitting the data shown in (b) with decaying sinusoids.

4.1 Runners around a circular racetrack. (a) Start of the race, (b) runners switch direction after a time T, (c) the runners all cross the starting line in phase after a total time of 2T.

4.2 Experimental setup. One arm of the rotation laser’s path generates π/2 pulses, the other arm generates π pulses. QWP: quarterwave plate, PBS: polarizing beamsplitter, EOM: electro-optic modulator.

4.3 EOM set up in double-pass configuration. PBS: polarizing beamsplitter, NPBS: non-polarizing beamsplitter.

4.4 Second generation spin-echo sample. The silicon δ-doping layer was 10 nm below the QD layer.

4.5 Third generation spin-echo sample, used for the experiments presented in this chapter.
4.6 Rabi oscillations in the photon count rate as the power of a single rotation pulse is varied. ........................................ 65
4.7 Ramsey fringes as the time offset between a pair of \(\pi/2\) pulses is varied. 65
4.8 Experimental demonstration of spin echo and single spin dephasing. 
   a, Spin-echo signal as the time offset \(2\tau\) is varied, for a time delay of \(2T = 264\) ns and magnetic field \(B_{\text{ext}} = 4\) T. Single-spin dephasing is evident at large time offset. 
   b, Decaying Fourier component of fringes. Red line is exponential fit, green line is Gaussian fit. One standard deviation confidence interval described in the text is determined by bootstrapping. ........................................ 67
4.9 Measurement of \(T_2\) using spin echo. 
   a, Spin-echo signal as the time offset \(2\tau\) is varied, for a time delay of \(2T = 132\) ns. Magnetic field \(B_{\text{ext}} = 4\) T. 
   b, Spin-echo signal for a time delay of \(2T = 3.1\) \(\mu\)s. 
   c, Spin-echo fringe amplitude on a semilog plot versus time delay \(2T\), showing a fit to an exponential decay. Error bars represent one standard deviation confidence intervals estimated by taking multiple measurements of the same delay curve. ........................................ 68
4.10 Magnetic field dependence of \(T_2\). Decoherence time \(T_2\) at various magnetic fields \(B_{\text{ext}}\). Error bars represent 1 standard deviation confidence intervals estimated from 3 independent measurements of the \(B_{\text{ext}} = 4\) T experiment, combined with bootstrapped uncertainties from each coherence decay curve. Black dashed lines are guides to the eye which indicate an initial rising slope, and then saturation for high magnetic field. The inset shows the linear dependence of the Larmor precession frequency \(\delta_e\) on the magnetic field. The slope uncertainty is determined by bootstrapping. ........................................ 69

5.1 (a) Hahn spin-echo sequence used to measure \(T_2^*\) (b) Ramsey pulse sequence (or FID sequence) ................................. 73
5.2 Sequence of initialization/measurement pulses and rotation pulses used for the FID experiment. ................................. 73
5.3 (a) Experimental Ramsey fringe count-rate as a function of two-pulse time delay $\tau$. (b) Average electron polarization as a result of the periodic pulse sequence used to generate this data. Optical pumping increases the polarization for a duration $T = 26$ ns. The saturation polarization, is $S_{z}^{p}$; in time $T$ only $S_{z}^{f}$ is reached. After pumping and a short delay, a picosecond pulse indicated by a green arrow nearly instantaneously rotates the electron spin to the equator of the Bloch sphere ($\langle S^z \rangle = 0$); a time $\tau$ later a second pulse rotates the spin to achieve electron polarization $S_{z}^{i}$, depending on the amount of Larmor precession between the pulses. The theoretical count-rate $C(\omega, \tau)$ of Eq. (5.1) is found as $S_{z}^{f} - S_{z}^{i}$ in steady-state conditions. (c) Experimental Ramsey fringe count-rate as $\tau$ is continuously scanned longer and then shorter, showing clear hysteresis.

5.4 Count-rate $C(\omega, \tau)$ as a function of Overhauser shift $\omega$ and two-pulse delay $\tau$. The green areas indicate where a higher count-rate is expected. Oscillations in the horizontal directions at frequency $\omega_{0} + \omega$ are due to Ramsey interference; the Gaussian envelope in the vertical direction is due to the reduction of optical pumping with detuning. The superimposed black line indicates stable points where $\partial \omega / \partial t = 0$ according to Eq. (5.4). Superimposed on this line are the solutions to this equation which result as $\tau$ is scanned longer (yellow) and shorter (white).

5.5 (Color online) The modeled (a) countrate or Ramsey amplitude $C(\omega_{f}, \tau)$, (b) Overhauser shift $\omega_{f}$, and (c) Optical pumping rate $\beta(\omega_{f})$. The dotted line in (a) is the expected Ramsey fringe in the absence of nuclear effects. The traces in (b) are the same as those in Fig. 5.4. The blue (red) line corresponds to scanning $\tau$ longer (shorter).

6.1 Schematic of a two-level emitter coupled to a microcavity.
6.2 Energies and linewidths of the two coupled modes of a QD-microcavity system. QD: QD exciton mode, cav: cavity mode, LP: lower polariton, UP: upper polariton. .......................................................... 86

6.3 (a) Scanning electron micrograph of a sample of uncapped InGaAs quantum dots. (b) Scanning electron micrograph of a 1.8 µm diameter pillar microcavity. .......................................................... 92

6.4 Experimental setup for micro-photoluminescence and photon correlation measurements. NPBS: non-polarizing beamsplitter, BS: beamsplitter, LPF: long-pass filter, ML: modelocked, SPCM: single-photon counting module. .......................................................... 93

6.5 Above-band pumping compared to resonant pumping of a chosen QD in Pillar 1. With above-band pump (725 nm, 0.4 µW), the chosen QD exciton (X) emits, but so do the cavity (C) and many other QDs. With 937.1 nm (3 µW) pump, the chosen QD is selectively excited and its PL dominates an otherwise nearly flat spectrum. ......................... 94

6.6 Temperature dependent PL from Pillar 2 with (a) above-band CW pump (725 nm), and (b) resonant CW pump (936.25 – 936.45 nm). Each spectrum is rescaled to a constant maximum since tuning the QD changes excitation efficiency. Resonance occurred at lower temperature for resonant pump case (10.5 K vs. 12 K) due to local heating. ........ 96

6.7 Emission wavelength and FWHM of upper (circles) and lower (squares) lines as a function of temperature, based on double-Lorentzian fits to resonantly-excited spectra of Pillar 2 (Fig. 6.6b). ......................... 96

6.8 Intensity autocorrelation function of the resonantly-coupled QD-cavity system Pillar 2, $g_{r,r}^2(\tau)$, pumped with a pulsed resonant laser. ........ 98

6.9 Intensity autocorrelation function of the resonantly-coupled QD-cavity system Pillar 3, pumped with a pulsed above-bandgap laser. ........ 99

6.10 PL spectrum of Pillar 2 with the QD detuned from the cavity. Shaded regions indicate the pass-bands for the spectral filters used for subsequent photon correlation measurements. ......................... 100
6.11 Correlation functions of the detuned QD-cavity system. (a) Autocorrelation function of QD emission only, $g^{(2)}_{xx}(0) = 0.19$. (b) Autocorrelation function of cavity emission only, $g^{(2)}_{cc}(0) = 0.39$. (c) Cross-correlation function of QD and cavity, $g^{(2)}_{xc}(0) = 0.22$.

6.12 Lifetime measurement of QD only, detuned 0.7 nm from cavity.

7.1 One possible design for an optically-active gate-defined QD. A top metal gate electrode is suspended away from the QWs by SiO$_2$, except above the QD region. The electric field between the top gate and bottom n-doped mirror is stronger than the surrounding region, which leads to an increased quantum-confined Stark effect that traps electrons and trions.

7.2 The CPMG DD rotation sequence.

A.1 The angle conventions used in this work. Rotations through angle $\Theta$ are left-handed about an axis tipped from the north pole by $\theta$. 
Chapter 1

Introduction

Quantum computers promise to solve certain problems which are too complex for classical computers. Originally proposed as a means to simulate other quantum systems [1], quantum computers have also been shown theoretically superior to classical computers at factoring large numbers [2] and searching databases [3]. Although quantum computers have so far only solved trivial problems, more basic quantum information systems have already been demonstrated which provide unconditionally secure communication [4, 5] over distances up to several hundred kilometers [6]. Quantum communication over longer distances will require quantum repeaters [7], which are essentially simple quantum computers.

The basic unit of information in a quantum information system is a quantum bit or qubit, which can be written as a state vector

\[ |\Psi\rangle = a |0\rangle + b |1\rangle \]  \hspace{1cm} (1.1)

where \( |0\rangle \) and \( |1\rangle \) are the two basis states that form the qubit, and take the place of logical 0 and 1 of a classical bit. Unlike classical computing, in which a bit must be either in state 0 or 1, the qubit may be in a coherent superposition of states \( |0\rangle \) and \( |1\rangle \), as \( a \) and \( b \) may be any complex numbers satisfying \( |a|^2 + |b|^2 = 1 \), where \( |a|^2 \) and \( |b|^2 \) respectively give the probabilities of measuring the qubit in states \( |0\rangle \) and \( |1\rangle \). Similarly, a quantum register composed of several qubits can be placed in a
superposition of all possible register states. These superposition states are the key to quantum computing’s power over classical computing.

Unfortunately, no single implementation of a qubit has proven ideal for all applications. Matter-based qubits are ideal for storing and processing quantum information, because long memory times and strong interactions between qubits are possible. Unfortunately, matter qubits are difficult to transport making them impractical for quantum communication. Photonic qubits are better suited to communication since they travel great distances while barely interacting with their environment. Long-distance quantum communication protocols [7], and some proposed quantum computing architectures [8], require both stationary matter qubits and ‘flying’ photonic qubits. It is therefore desirable to interface between matter and photonic qubits.

The most basic level structure for a qubit is a simple 2-level system, such as that shown in figure 1.1(a). Unfortunately, if the qubit interacts strongly with light (as is desirable for interfacing with photonic qubits) then the qubit’s lifetime is limited by spontaneous emission. A 3-level Λ-system, shown in figure 1.1(b), is a powerful level structure for quantum computing. The qubit consists of the two metastable ground states $|0\rangle$ and $|1\rangle$, which both couple to a common excited state $|e\rangle$ via optical transitions. Because no optical transition is possible directly between $|0\rangle$ and $|1\rangle$, the qubit can be long-lived. However, $|0\rangle$ and $|1\rangle$ may still be coupled and manipulated by a Raman transition.

A single electron spin confined in a semiconductor quantum dot is a promising matter qubit candidate that can implement a Λ-system. It offers memory storage times of at least several microseconds [9, 10], fast single-qubit operation times of several tens of picoseconds [11], and strong interaction with light. Furthermore, it may be possible to leverage existing semiconductor manufacturing techniques to aid in large-scale integration of devices.
1.1. QUANTUM COMPUTING INGREDIENTS

It has been shown that a complete set of single-qubit gates, plus a two-qubit gate called the Controlled NOT gate (CNOT) form a universal set for quantum computation [12]. We will now briefly describe these gates.

1.1.1 Single-qubit gates

As introduced previously, a single qubit state may be represented by the state vector $|\Psi\rangle = a|0\rangle + b|1\rangle$. However, because it is inconvenient to write down the complex coefficients $a$ and $b$, we may instead rewrite the state vector in terms of angles $\theta$ and $\phi$

$$|\Psi\rangle = \sin(\theta/2)|0\rangle + \cos(\theta/2)e^{i\phi}|1\rangle$$

where we have dropped the overall phase factor $\angle a$. We can then associate the qubit state with a vector of unit length on the surface of the Bloch sphere, as shown in

Figure 1.1: (a) A simple 2-level qubit, with the ground state $|0\rangle$ and excited state $|1\rangle$ connected by an optical transition. (b) A three-level $\Lambda$-system. Two long-lived states, $|0\rangle$ and $|1\rangle$, are connected to an excited state by optical transitions.
Figure 1.2. The north pole of the Bloch sphere is associated with the state $|1\rangle$, and the south pole is associated with state $|0\rangle$. The polar angle $\theta$ gives us information about the relative probability amplitudes of measuring the qubit in state $|0\rangle$ versus $|1\rangle$, and the azimuthal angle $\phi$ gives the phase difference between the $|0\rangle$ and $|1\rangle$.

There are an infinite set of single-qubit gates possible on the state vector $|\Psi\rangle$, given by the complete set of SU(2) rotations. We can then visualize a single qubit gate as rotating the Bloch vector from one position on the Bloch sphere to another arbitrary point. The symbol for a single-qubit gate, also called a rotation gate, is shown in Figure 1.3.
1.2. QUANTUM COMPUTING ARCHITECTURE

Figure 1.3: Notation for a single-qubit gate, or rotation R. It rotates the input qubit $x$ into the output qubit $x'$.

1.1.2 Two-qubit gates

Although many different two-qubit gates exist, quantum computing theorists often work in terms of the CNOT gate. This gate has two inputs and two outputs as shown in Figure 1.4(a). The CNOT gate is mathematically convenient because two back-to-back CNOT gates form the identity matrix. The top qubit of the CNOT gate is called the control qubit, and it is unaffected by the gate. The bottom qubit is called the target qubit, and it is XOR'd with the control qubit (i.e. it is flipped if the control qubit is 1, unaffected if the control qubit is 0). The CNOT gate’s truth table is shown in Figure 1.4(b).

1.2 Quantum Computing Architecture

We will now briefly look at one particularly promising quantum computing architecture proposal: the two-dimensional topological surface code. This architecture is one of many proposed for quantum computation, and an in-depth explanation of this complicated architecture is beyond the scope of this thesis. The architecture was proposed in 2007 by Raussendorf and Harrington [13], and was then reviewed and explained in more depth by Fowler et al. [14]. The surface code architecture offers many attractive features, including a relatively high error tolerance threshold of 0.75%, and the requirement of CNOT gates only between nearest-neighbor physical qubits.

The surface code quantum computer contains a two-dimensional array of qubits,
as shown schematically in Figure 1.5. A single logical qubit is topologically encoded amongst a large number (1000 for example) of physical qubits in the array. Half of the physical qubits within a logical qubit are used to encode data, while the other half are called syndrome qubits, and are repeatedly measured in order to detect when and where physical errors occur. As long as a limited number of physical errors occur within a logical qubit, the logical qubit will be topologically protected from a logical error occurring.

### 1.2.1 Surface Code Requirements

The surface code requires the following operations on the physical qubits:

- initialization of individual qubits into either $|0\rangle$ or $|1\rangle$
- arbitrary single-qubit gates on individual qubits
- CNOT gates between nearest neighbor qubits
- measurement of individual qubits
1.2. QUANTUM COMPUTING ARCHITECTURE

Figure 1.5: A small section of a huge two-dimensional array of qubits necessary for a topological surface code quantum computer. Half of the physical qubits store data, while the other half are measured to detect errors called the syndrome.
CHAPTER 1. INTRODUCTION

Note that initialization can often be accomplished with a measurement followed by a single qubit gate.

One other requirement of any quantum computer is that the decoherence time of the individual qubits must be long in comparison to the operations. The decoherence time, often called $T_2$, is the amount of time that the phase $\phi$ between $|0\rangle$ and $|1\rangle$ is maintained. This time is also interpreted as the memory time of the qubit.

1.2.2 Surface Code Resources Estimate

If these requirements are met, then we can make a few more assumptions to get an idea of how our surface code quantum computer will look. Let us assume we want to use 1000 physical qubits to encode 1 logical qubit. We will then require an error rate for initialization, 1- and 2-qubit gates, and measurement of $\sim 0.1\%$, and a ratio of the decoherence time to operation time on the order of $T_2/T_{op} \sim 10^4$.

If we want to use our quantum computer to factor a 1024-bit number using Shor’s algorithm, we will need on the order of $10^8$ physical qubits. Let us now proceed with these targets in mind:

- $10^8$ physical qubits
- error rate per operation 0.1%
- $T_2/T_{op} \sim 10^4$

1.3 Physical Qubit Candidates

We will now briefly describe several candidates for physical qubits. We will restrict ourselves to discussing only matter qubits, because these are most promising for the surface code architecture discussed above. However, for any matter qubit system, it is extremely desirable if the qubit can interface with a ‘flying’ photonic qubit which could transmit quantum information between different quantum processors for computation, or across the continent for quantum communication.
We will only discuss a small subset of the many matter-qubit candidates that are under investigation, choosing only candidates which have been shown viable as single qubits (rather than ensembles), and which show some prospects for scalability. Ladd et al. have written an excellent review article which covers these qubits and more in depth [15].

1.3.1 Trapped Ion Qubits

Single atomic ions can be suspended in a vacuum with nanometer precision by the electric fields from nearby electrodes [16, 17]. Single-qubit gates can be accomplished with 99.5% fidelity within about 3 $\mu$s by directly driving atomic transitions with microwave fields [18]. Two-qubit gates can be achieved with 99.3% fidelity on a 50 $\mu$s timescale by driving common vibrational sidebands of two ions within a trap [17]. Ions can exhibit extremely long coherence times of 15 seconds [19], and can be interfaced with optical-frequency photons for quantum communication [20].

Based on the excellent gate fidelities and long decoherence times listed above, it appears that trapped ions are currently leading the pack of potential qubit candidates, and up to eight ions have been entangled together in a trap [17]. However, it will be extremely difficult for the ion trap system to scale orders of magnitude larger, because the densely packed motional spectrum of ions in a trap leads to crosstalk and nonlinearities that degrade gate fidelities [16].

1.3.2 Trapped Neutral Atom Qubits

Neutral atoms offer many of the same advantages as trapped ions. An atom can be held in place by an optical trap, where a far off-resonance laser is used to Stark shift the atomic levels creating an attractive potential. An array of atoms can be suspended by an optical lattice consisting of crossed laser beams [21]. Perhaps the most exciting aspect of neutral atoms is the prospect of loading hundreds of millions of individual atoms into such an optical lattice. Nearest-neighbor interactions have been achieved by adjusting the optical lattice lasers to move pairs of atoms closer to each other to induce exchange interactions, allowing entanglement between neighboring atom
pairs [22]. Single-qubit gates have been demonstrated with 95% fidelity by driving atomic transitions of a single atom with microwaves in a single-atom trap [23].

1.3.3 Superconducting Qubits

Superconducting qubits are generally made from a superconducting LC circuit which acts as a harmonic oscillator, with a Josephson junction introduced to cause anharmonicity of the oscillator levels. This allows two levels to be chosen as the qubit states. An important characteristic of the qubit is the ratio of the Josephson energy $E_J$ to the capacitor charging energy $E_C = e^2/2C$. Superconducting qubits are often classified as one of three types of superconducting qubits: charge, flux, and phase qubits.

Charge qubits omit the inductor, and are sometimes called Cooper pair boxes. They were originally developed in the regime of $E_J/E_C \ll 1$ [24], and were later extended to $E_J/E_C \gg 1$ and given new names of quasitronium [25] and transmon [26]. In both flux and phase qubits, $E_J/E_C \gg 1$, but the fluxes through their inductors are biased differently creating different potential profiles. Flux qubits [27] make qubit states out of the lowest two energy states in a symmetric double-well potential, while phase qubits [28] have highly asymmetric potentials and use two states within one well as their qubit states.

In a current state-of-the-art device, single-qubit operations can be accomplished in 4 ns with 99.3% fidelity by driving the qubit transition with microwaves, and 2-qubit gates can be performed in 30 ns with 90% fidelity by tuning a microwave cavity into resonance with both of the qubits [29]. Decoherence times as long as 4 $\mu$s have been demonstrated [26]. These achievements make superconducting qubits extremely attractive candidates for a scalable solid-state quantum computer. One drawback is that the footprint of each qubit is on the order of 10 $\mu$m square, so $10^8$ such qubits would fill a 10 cm x 10 cm chip before any of the necessary control electronics and cavities are introduced.
1.3.4 Diamond Nitrogen Vacancy Center Qubits

A nitrogen-vacancy (NV) center occurs in diamond when a substitutional nitrogen atom sits next to a missing carbon atom. The negatively charged state of this optically-active impurity forms a spin triplet which can implement a qubit. Single-qubit gates can be achieved by coherently manipulating the qubit states with microwave fields [30] on a microsecond timescale. Decoherence times up to 3 ms have been demonstrated in isotopically-purified diamond [31], an extremely long time for a solid-state qubit. Although the NV center couples to visible photons, it is extremely difficult to fabricate optical cavities in diamond, which may pose a challenge to creating quantum photonic links between NV centers.

1.3.5 Electron Spin in Quantum Dot Qubits

Quantum dots (QDs), sometimes called ‘artificial atoms’, occur when a semiconductor nanostructure confines an electron or hole into a localized potential trap with discretized energy levels. QDs can be either electrostatically defined by metallic gates around a two-dimensional electron gas, which can trap only electrons [32, 33], or they can be self-assembled, where a random growth process involving two different semiconductor materials creates a potential that can trap both electrons and holes [34]. In each case, a qubit can be encoded in the two spin states of an electron (or hole) confined in the QD.

Electrostatically-defined QD spins were first coherently controlled about five years ago. Single qubit gates can be achieved either by microwave drive [33], or more rapidly (350 ps) by shifting the electrostatic trap for the case of two electrons in a double QD [32]. The decoherence time of the electron spin is limited to $\sim 1 \mu s$ [32] by fluctuating nuclear spins in the QD which couple to the electron spin through the hyperfine interaction. Because holes are not simultaneously confined with electrons in the electrostatically-defined QD, it may be challenging to interface the these matter-spin qubits with photonic qubits.

Self-assembled QDs can trap an electron whose spin makes a good qubit, and also confine an additional electron-hole pair to create a composite particle called a
charged exciton or trion. This trion state allows the electron spin to be optically manipulated, and can ultimately lead to an energy level structure similar to that shown in Figure 1.1(b).

The main result of this thesis work is to bring optically-controlled QD spin qubits ‘up to speed’ with their other competitors. In the chapter on Coherent Spin Control, we will demonstrate a complete set of single-qubit operations, including single-qubit gates with up to 98% fidelity in under 40 picoseconds [11]. Later, in the Spin Echo chapter, we use a spin-echo technique to extend the spin’s decoherence time to nearly 3 μs [10].

1.3.6 Qubit Comparison

In Figure 1.6 we summarize the state-of-the-art for the various qubit candidates discussed above. The error rates (1 minus the fidelity) of demonstrated single-qubit and two-qubit gates, as well as approximate gate times normalized by the coherence time are given. Normal typeface values are experimental demonstrations from other works, as listed previously. **Bold** values are experimentally demonstrated in this thesis work. *Italicized* values are theoretical predictions from Reference [35]. Note that none of the qubit systems have achieved the necessary error rates of 0.1%, but several of the systems have reached the necessary ratio of operation time to decoherence time of $10^4$ for the surface code architecture.

1.4 Quantum Computing Criteria

Below is a modified version of the original DiVincenzo criteria [36], which summarizes much of the previous discussion by listing seven criteria that a qubit candidate system must possess. Although meeting all of these criteria does not guarantee that a quantum computer can be built from a given physical qubit system, the criteria can serve as a checklist for evaluating and pursuing any given system.

1. Qubits must be implemented in a scalable physical system

2. Individual qubits must be initialized into a pure state
Figure 1.6: A table summarizing the current state-of-the-art of the five qubit candidates discussed.

3. Individual qubits must be measured
4. Single-qubit (rotation) gates must be demonstrated
5. Two-qubit (CNOT) gates must be demonstrated
6. The qubit must have a long decoherence time
7. The qubit should interface with a ‘flying’ photonic qubit

### 1.5 Thesis Outline

In Chapter 2, we will address criterion #1 by describing our physical qubit system: a single electron spin confined in a QD. We will discuss the basics of semiconductor quantum dots, and show how an electron-charged quantum dot can implement a 3-level Λ-system.

Chapter 3 will address criteria #2-4. We use a technique called optical pumping to initialize an electron spin in the QD into a pure spin state with ~92% fidelity. We measure the spin state of the electron after the optical pumping step by counting the
photons emitted from the QD. By applying a sequence of detuned ultrafast optical pulses to the QD, we can implement a single-qubit gate with up to 98% fidelity within 40 ps.

Relevant publication:


In Chapter 4, we deal with criterion #6 by implementing an all-optical version of a ‘spin echo’ technique to extend the decoherence time of the QD electron spin from about 2 ns to 3 µs. The spin-echo technique was first developed 60 years ago [37] and is still finding new applications today. The decoherence time is limited by the electron spin’s hyperfine interaction with background nuclear spins in the QD.

Relevant publication:


In Chapter 5 we investigate the electron spin’s interaction with the nuclear spin background in more depth, finding a surprising new hysteresis effect. This effect results from a feedback between the electron and nuclear spins, and leads to a controllable polarizing of the nuclei and dynamic tuning of the electron Larmor frequency.

Relevant publication:


Chapter 6 introduces optical micropillar cavities, which are promising devices to increase the interaction of the QD with light as needed to implement criterion #7, the spin/photon interface [7, 8, 38]. We then experimentally probe a single QD which is strongly coupled to a pillar microcavity, meaning that the optical mode of the cavity
and exciton mode of the QD interact more strongly with each other than with the environment.

Relevant publication:

Chapter 2

Quantum Dots

A great number of advances in experimental quantum information and quantum opt- 
cics have been achieved using neutral or charged atomic systems [39, 40, 41, 42]. 
However, solid-state systems are desirable for quantum computers because of their 
stability and scalability. One natural solid-state candidate is the quantum dot, which 
is often referred to as an artificial atom. A quantum dot is a nanometer-scale blob 
of narrow-bandgap semiconductor (such as indium arsenide, InAs), often surrounded 
by a wider-bandgap semiconductor (such as gallium arsenide, GaAs). The narrow-
bandgap material creates a potential well in both the conduction and valence bands, 
which can trap electrons and holes at discrete energy levels. In general, several bound 
states may be created for both electrons and holes. The lowest energy electron/hole 
states have s-like envelope-wavefunction symmetry, and are referred to as the s-shell 
electrons and holes (labeled $e_s$ and $h_s$, respectively). The next higher energy states 
have p-like symmetry, and so on. Figure 2.1(a) shows an energy level diagram of a 
QD.

The simplest way to excite a QD is above-bandgap pumping, using a laser with 
higher energy than the bandgap of the surrounding semiconductor matrix (see Fig-
ure 2.1(b)(i)). The above-band laser creates free electron-hole pairs, which may be 
captured by all the QDs on the sample. The electrons and holes relax into the QD en-
ergy levels by giving up energy to acoustic and optical phonons. Once the exciton has 
relaxed to the s-shell in the QD, it recombines radiatively in roughly a 1 ns timescale,
emitting a single photon. Alternatively, a QD may be excited by resonantly pumping the p-shell exciton, called quasi-resonant excitation (Figure 2.1(b)(ii)). The p-shell exciton relaxes to the s-shell exciton by interacting with optical phonons in roughly a 10 ps timescale. Quasi-resonant excitation has numerous advantages in certain applications. For example, in QD ensembles where many QDs have different sizes, it may be possible to selectively excite only one QD with a unique p-shell resonance energy. The fast relaxation to the s-shell exciton may also be advantageous for lifetime measurements. Finally, an s-shell QD exciton may also be created resonantly as shown in Figure 2.1(b)(iii).

The QDs used in this work were formed of indium gallium arsenide on a gallium arsenide substrate, grown by molecular beam epitaxy (MBE) using the self-assembled Stransky-Krastanow growth method [43]. Depending on growth conditions, the QDs are typically 20-50 nm in diameter and 2-4 nm in height, with densities on the order of $10^{10}$ cm$^{-1}$. An atomic force micrograph of some typical InGaAs QDs is shown in Figure 2.2(a). Figure 2.2(b) shows a typical photoluminescence (PL) spectrum of an ensemble of InGaAs QDs when excited by an above-bandgap laser. The peak around 820 - 830 nm (i) is from bulk GaAs free and impurity-bound exciton emission, while
CHAPTER 2. QUANTUM DOTS

Figure 2.2: (a) Atomic force micrograph of self-assembled InGaAs QDs before a GaAs capping layer is grown. Image by courtesy of Bingyang Zhang. (b) PL spectrum of an ensemble of InGaAs QDs. i - GaAs free and impurity-bound exciton emission, ii - QD emission. (c) PL spectrum of a 600 nm mesa showing individual QD emission lines.

The broad peak centered around 950 nm (ii) is from QDs. The ∼50 nm linewidth of the QDs is due to size and shape inhomogeneities amongst the different QDs resulting in different emission energies. In order to investigate single QDs, the bulk planar sample may be etched into mesa structures containing just a few QDs. The spectrum of a typical 600 nm diameter mesa is shown in Figure 2.2(c). The individual emission lines correspond to discrete energy levels of different QDs.

The p-shell hole states are at least several meV higher in energy than the s-shell hole state, and the electron p-shell states are split even further. Thus one may eliminate thermal excitations of p-shell states by cooling the sample cryogenically. Typically, narrow emission peaks may be observed in self-assembled InGaAs QDs for temperatures less than about 50 K.

2.1 Quantum Dot States in Magnetic Fields

A quantum dot may trap both electrons and holes. We will label the particles according to their angular momentum projections along the growth direction, which we will label the x direction. (Note that this definition is different from most other QD
works, but will be consistent with notation when a magnetic field is applied perpendicular to the growth direction.) The electrons, which occupy conduction band states with s-like atomic orbitals, are labeled $|S, S_x\rangle = |1/2, \pm 1/2\rangle$ where $S$ and $S_x$ are the total and $x$-projection of the angular momentum (growth-direction projections). The holes occupy valence band states with p-like atomic orbitals, and thus have both spin and orbital degrees of freedom. This leads to the existence of heavy holes with $|J, J_x\rangle = |3/2, \pm 3/2\rangle$, light holes $|3/2, \pm 1/2\rangle$, and a split-off hole band $|1/2, \pm 1/2\rangle$, where $J$ is the hole’s angular momentum. The split-off holes are separated from the light and heavy holes by several hundred meV even in bulk semiconductors, and thus don’t play a role in our experiments. The light holes are split by several tens of meV from the heavy holes by confinement in self-assembled QDs [44]. We will thus only consider electrons and heavy holes in this work. This discussion will follow the excellent review found in reference [44].

2.1.1 Neutral Quantum Dots

In a neutral QD, the ground state is an ‘empty’ QD (full valence band, empty conduction band). If we inject an electron-hole pair, four exciton states may be formed. These excitons may be characterized by their total angular momentum $M = S_x + J_x$. Because a single photon can only carry one quantum of angular momentum, states with $|M| = 2$ are unable to couple to the optical field and are called dark excitons. States with $|M| = 1$ are optically active and called bright excitons. In the absence of an applied magnetic field, the energies of the excitons may be found using the matrix representation of the exchange Hamiltonian, using the basis of exciton states ($|+1\rangle, |-1\rangle, |+2\rangle, |-2\rangle$):

$$H_{exchange} = \frac{1}{2} \begin{pmatrix} +\delta_0 & +\delta_1 & 0 & 0 \\ +\delta_1 & +\delta_0 & 0 & 0 \\ 0 & 0 & -\delta_0 & +\delta_2 \\ 0 & 0 & +\delta_2 & -\delta_0 \end{pmatrix}$$

(2.1)

The electron-hole exchange energy $\delta_0$ separates the bright and dark exciton states.
The dark excitons are always hybridized into states $\frac{1}{\sqrt{2}}(|+2\rangle \pm |-2\rangle)$, which are split by an amount $\delta_2$ [44]. In QDs that lack circular symmetry (as is the case for most self-assembled QDs) the bright excitons are also hybridized into states $\frac{1}{\sqrt{2}}(|+1\rangle \pm |-1\rangle)$, which are split by an amount $\delta_1$. These bright excitons emit linearly polarized light.

**Faraday Magnetic Field**

We now analyze the effect of a magnetic field applied in the x-direction (growth direction, called Faraday geometry). We will ultimately show that Faraday geometry is not appropriate for our experiments because we will be unable to differentiate between charged and neutral QDs, and we cannot create a fully-connected Lambda system.

The Hamiltonian describing the Zeeman splitting induced in a neutral QD by a Faraday magnetic field is given by [44]

$$H_{\text{zeeman}}^F = \frac{\mu_B B_x}{2} \begin{pmatrix}
+ (g_{e,x} + g_{h,x}) & 0 & 0 & 0 \\
0 & -(g_{e,x} + g_{h,x}) & 0 & 0 \\
0 & 0 & -(g_{e,x} - g_{h,x}) & 0 \\
0 & 0 & 0 & + (g_{e,x} - g_{h,x})
\end{pmatrix}$$ (2.2)

Where $g_{e,x}$ and $g_{h,x}$ are the electron and hole $g$-factors in the x-direction, $\mu_B$ is the Bohr magneton, and $B_x$ is the applied Faraday magnetic field. The neutral QD eigenstates of the combined Hamiltonian $H_{\text{total}}^F = H_{\text{exchange}} + H_{\text{zeeman}}^F$ are shown schematically in Figure 2.3(a). It is clear from this Hamiltonian that the Faraday field does not mix bright and dark excitons, and there are only two optically-active transitions in Figure 2.3(a). For zero magnetic field, the two bright excitons are mixed and split by an amount $\delta_1$, and the optically active transitions are linearly polarized. However, $\delta_1$ is often on the order of $\sim 10 \mu\text{eV}$ or less for a typical self-assembled QD, which is very difficult to resolve using a spectrometer. As the magnetic field is increased, the bright and dark excitons are ‘purified’ by the Faraday field into their original eigenstates, and the optical transitions are circularly polarized.
Voigt Magnetic Field

Next we analyze the effect of a magnetic field applied in the $z$-direction (perpendicular to the growth direction, called Voigt geometry). This magnetic field orientation will allow us to differentiate between neutral and charged QDs, and allows us to create a fully-connected $\Lambda$-system with a charged QD.

The Hamiltonian describing the Zeeman splitting induced by a Voigt magnetic field is given by

$$H_{\text{zeeman}} = \frac{\mu_B B_z}{2} \begin{pmatrix} 0 & 0 & g_{e,z} & g_{h,z} \\ 0 & 0 & g_{h,z} & g_{e,z} \\ g_{e,z} & g_{h,z} & 0 & 0 \\ g_{h,z} & g_{e,z} & 0 & 0 \end{pmatrix}$$ (2.3)

Where $g_{e,z}$ and $g_{h,z}$ are the electron and hole $g$-factors in the $x$-direction, $\mu_B$ is the Bohr magneton, and $B_z$ is the applied magnetic field. The Voigt magnetic field mixes the bright and dark excitons, allowing the dark excitons to be measured spectroscopically. The neutral QD eigenstates of the combined Hamiltonian $H_{\text{total}} = H_{\text{exchange}} + H_{\text{zeeman}}$ are shown schematically in Figure 2.4(a). Figure 2.4(a) also shows the optically active transitions, which are labeled by $\pi_H$ and $\pi_V$ to indicate that the transitions are linearly polarized and orthogonally polarized in our simple model which neglects light holes. In reality, the transitions may be somewhat elliptically polarized and non-orthogonal, because strain and shape effects can mix light holes with the heavy and add higher order terms to $H_{\text{zeeman}} [45, 46]$. In the actual lab experiments, ‘horizontal’ and ‘vertical’ linear polarization axes are set by the QD’s strain axis.

2.1.2 Charged Quantum Dots

We will focus our attention on negatively charged QDs. However, we may understand positively charged QDs by simply swapping the electrons and holes. In a negatively charged QD, the ground state is given by a single electron, whose spin may be up or down: $|1/2, \pm 1/2\rangle$. The excited states consist of the initial electron plus an additional electron-hole pair. This three-particle system is called a trion or negatively-charged
CHAPTER 2. QUANTUM DOTS

Figure 2.3: Neutral and charged QDs in a Faraday magnetic field. (a) Eigenstates of a neutral QD. The eigenstates at zero field are listed in the left column, eigenstates at high magnetic field are listed to the right of the diagram. Optically active transitions are shown in red arrows, with their polarization labeled. The crystal ground state (empty QD) is denoted $|0\rangle$. (b) Eigenstates of a charged QD. Normalization factors have been dropped for compactness.

Figure 2.4: Neutral and charged QDs in a Voigt magnetic field. (a) Eigenstates of a neutral QD. (b) Eigenstates of a charged QD. Normalization factors have been dropped for compactness.
exciton. The lowest energy trion has both of the electrons in the $e_s$ state. Because the electrons occupy the same state their spins must form a spin-less singlet configuration: $\frac{1}{\sqrt{2}}(|+1/2⟩−1/2⟩−|−1/2⟩+1/2⟩)$. The x-projection of the angular momentum of the lowest-energy trions is thus given by the angular momentum of the unpaired hole, $|±3/2⟩$. Higher energy trion states also exist with at least one of the electrons in a higher energy QD state such as $e_p$ (see Figure 2.1), in which the electrons may form a triplet. However, because these states are several tens of meV higher in energy than the ground state trion [47], we will neglect them.

**Faraday Magnetic Field**

There is no exchange splitting in any particle with an odd number of fermions [44], such as the trion. Thus we are only concerned with the Zeeman Hamiltonian, which may be written separately for the electron ground states in the $([-1/2⟩, |+1/2⟩)$ basis and for trions in the $([-3/2⟩, |+3/2⟩)$ basis:

$$H_{e,F}^{\text{zeeman}} = \frac{\mu_B B_z}{2} \begin{pmatrix} -g_{e,z} & 0 \\ 0 & g_{e,z} \end{pmatrix}$$

(2.4)

$$H_{h,F}^{\text{zeeman}} = \frac{\mu_B B_z}{2} \begin{pmatrix} g_{h,z} & 0 \\ 0 & -g_{h,z} \end{pmatrix}$$

(2.5)

Thus the trion and electron states are simply split without being mixed. There will be two optically active transitions, for the two cases where the change in angular momentum is one: $|\Delta M| = 1$.

Figure 2.3(b) shows the eigenstates of the Zeeman Hamiltonians for the electrons and trions, and the two optically active transitions. It is extremely difficult to differentiate between neutral and charged QDs in Faraday geometry: because $\delta_1$ is very small ($< 10 \mu eV$), for any appreciable magnetic field the neutral QD exhibits the same optical transitions and splittings as the charged QD in Figure 2.3. It is also clear from the figure that no $\Lambda$-system is possible for a changed QD in Faraday geometry - there is no excited state that is optically connected to both ground states. We are thus unable to coherently optically control the electron spin in Faraday geometry.
Voigt Magnetic Field

The Zeeman Hamiltonians for electrons and trions in Voigt geometry, again in the basis of \((|+1/2\rangle, |-1/2\rangle)\) for electrons and \((|+3/2\rangle, |-3/2\rangle)\) for trions, are given by:

\[
H_{e,V}^{\text{zeeman}} = \frac{\mu_B B_z}{2} \begin{pmatrix} 0 & g_{e,z} \\ g_{e,z} & 0 \end{pmatrix}
\]

\[
H_{h,V}^{\text{zeeman}} = \frac{\mu_B B_z}{2} \begin{pmatrix} 0 & g_{h,z} \\ g_{h,z} & 0 \end{pmatrix}
\]

Thus, for any applied magnetic field, the electron states will be diagonalized into

\[
|\uparrow\rangle \equiv \frac{1}{\sqrt{2}} (|+1/2\rangle + |-1/2\rangle)
\]

\[
|\downarrow\rangle \equiv \frac{1}{\sqrt{2}} (|+1/2\rangle - |-1/2\rangle)
\]

The trion states meanwhile will be diagonalized into

\[
|\uparrow\rangle \equiv \frac{1}{\sqrt{2}} (|+3/2\rangle + |-3/2\rangle)
\]

\[
|\downarrow\rangle \equiv \frac{1}{\sqrt{2}} (|+3/2\rangle - |-3/2\rangle)
\]

All four transitions are optically active, and in our simple model have equal oscillator strengths and orthogonal linear polarizations, as shown in Figure 2.4(b). Chapter 3 gives a detailed calculation of the matrix elements and polarization selection rules of the four active transitions. In a real sample however, strain and shape may again cause non-orthogonal transitions with non-equal oscillator strengths [45, 46].

Figure 2.4(b) shows how we will implement a Λ-system in using a charged QD. The two electron-spin ground states, split by the Zeeman effect, are connected to a common excited state by optical transitions. In fact, a charged QD contains two Λ-systems because there are two excited states. We will therefore perform our optical experiments on charged QDs in Voigt geometry.
2.2 Charged QD Samples

Even in a nominally undoped sample, both charged and uncharged QD PL may be observed. When the sample is excited by an above-bandgap pump laser, unpaired electrons and holes are present throughout the sample substrate. A QD may thus capture an unpaired electron (hole), becoming negatively (positively) charged. If it next captures an unpaired hole (electron), then the exciton will recombine giving emission as a neutral QD. However, if it next captures an exciton, then a trion will be present in the QD and a charged emission spectrum will be produced. Studies have shown that both unpaired-carrier capture, and electron-hole pair capture, are relevant processes in undoped QDs using above-bandgap pumping [48].

However, above-bandgap injection of unpaired electrons is unreliable for the purposes of quantum information, since the electron qubit will only be present in the QD until it recombines with an unpaired hole. It is preferable to have an electron permanently present in the QD. The simplest method is to dope the host material surrounding the QD. A thin layer of dopants, called modulation doping or $\delta$-doping, is grown a short distance from the QD layer. If one of these dopant impurities is close enough to a QD, its electron (or hole) may become trapped in the potential well of the QD at low temperature, creating a permanently charged QD. The $\delta$-doping layer is typically separated from the QD layer by 2 - 20 nm. We used $\delta$-doped QD samples for much of the work throughout this thesis.

An even more deterministic way to create a permanently charged QD is to embed the QD in a Schottky-diode structure [49]. The QD is separated by a tunnel barrier from a doped layer which sets the Fermi level. By adjusting the bias on the Schottky-diode, the s-shell electron state may be brought just below the Fermi level and an electron will tunnel from the doped layer into the QD. We have begun to work with Schottky-diode samples, but have not yet succeeded in loading a single electron into a QD using this technique. Our efforts will continue in this direction in the future.

Our first sample containing charged QDs, M471, was a simple $\delta$-doped structure grown by Bingyang Zhang at Stanford. As shown in Figure 2.5, it contained a 300 nm buffer layer of GaAs is grown on top of a GaAs substrate. An n-type $\delta$-doping
layer of silicon was grown next, followed by a 20 nm barrier of GaAs and then a layer of nominally InAs QDs, and finally a 90 nm capping layer of GaAs to protect the QDs from surface charges. Atomic force microscopy on similar samples indicated an areal QD density of approximately $10^{10}$ cm$^{-2}$. The areal density of the silicon dopants was approximately the same.

The QD density in M471 was too high to allow single QD studies as-grown: even a small laser excitation spot of around 1 $\mu$m would excite about 100 QDs. One method to isolate just a few QDs is to deposit a metal mask over the planar sample, then etch small holes in the mask through which PL may be collected. Instead, we chose to etch the planar sample into mesas containing a few QDs each. Our sample was etched into mesas varying from 100 nm to 1 $\mu$m, and most experiments in this work were performed on a 600 nm mesa which contained a few tens of QDs.

### 2.3 Magneto-Photoluminescence Experimental Setup

In order to Zeeman split the QD emission lines enough to resolve them using our PL detection setup, we required a magnetic field of at least several Tesla. We used an Oxford Spectromag magnetic cryostat as pictured in Figure 2.6(a), which can achieve
fields as high as 10 T and sample temperatures down to 1.6 K. The Spectromag has four windows for optical access: two are parallel to the magnetic field axis (Faraday geometry) and two are perpendicular to the field axis (Voigt geometry). Our experiments were performed in one of the Voigt windows.

One disadvantage of the Spectromag is the low numerical aperture (NA) of its optical windows. The Voigt windows have $\text{NA} \sim 0.1$. This low NA may be enough for some basic QD PL studies, but for experiments where signal-to-noise ratio is critical, the collection efficiency of such an objective lens may be too low. It is possible to embed the QDs in microcavities so that their emission is directional and may be collected efficiently by a low-NA objective. A more general solution to the problem, however, is to place a high-NA objective lens inside the cryostat. The Spectromag has an internal sample-space bore of 25-30 mm (depending on whether the system includes a radiation shield or not), which allows a small lens to be placed inside the sample space. We used an aspheric singlet lens, with a focal length of 3.1 mm and NA of 0.68 (Thorlabs part #352330-B) as our objective inside the Spectromag. It principle it collects roughly 45 times more uniformly-emitted photons than an objective with $\text{NA} = 0.1$.

Because our samples contain many mesa structures that we wish to image, we must be able to position the sample relative to the objective lens inside the Spectromag. We used a stack of Attocube positioners, which are unique in that they offer long travel ($\sim 2.5$ mm), high resolution ($\ll 1$ $\mu$m), a small footprint (15 mm), and are designed to operate at low temperatures and high magnetic fields. Each positioner allows linear motion in one axis. We used two ANP-x51 horizontal positioners and one ANP-z51 vertical positioner to achieve full three-dimensional control over the sample position.

The sample holder we constructed is shown in Figure 2.6(b). It holds the sample on a finger connected to the Attocube positioner stack, which sits behind the objective lens. The sample holder attaches to a long sample holder rod which is inserted into the top of the Spectromag. The sample holder was originally machined out of titanium in order to minimize thermal expansion mismatch with the titanium Attocube positioners. However, any high-purity non-magnetic metal could be used instead of titanium. Tellurium copper is another good choice; aluminum and brass
may be poor choices because of the typically high amounts of magnetic impurities in commercially available alloys.

The experimental setup for basic magneto-PL is shown in Figure 2.8. The QDs are excited using a fiber-coupled 785 nm above-bandgap laser diode. The pump light is polarized by a polarizing beam splitter (PBS) before passing through a half-wave plate (HWP) and being focussed onto the sample by the objective lens. The PL is collected through the same objective and passes through the HWP and another PBS which is cross-polarized to the first. This second PBS blocks much of the reflected pump light, and allows the polarization of the detected PL to be defined by the HWP. The PL then passes through a non-magnifying telescope and a pinhole, which acts as a spatial filter to further remove scattered laser light. Finally, the PL is dispersed by a grating spectrometer and detected by a liquid-nitrogen cooled charge-coupled device (CCD). In order to locate a particular position on the sample, a wide-field white-light imaging mode may be used by flipping out the pinhole and sending the image to a second uncooled CCD.
2.3. MAGNETO-PHOTOLUMINESCENCE EXPERIMENTAL SETUP

Figure 2.7: Experimental setup for magneto-PL.
Figure 2.8: Solid immersion lenses, showing how the collected solid angle is increased. (a) No SIL, (b) hemispherical SIL, (c) hyper-hemispherical SIL.

The collection efficiency was further enhanced for sample M471 by placing a hemispherical solid immersion lens (SIL) on the sample surface. In principle, a hemispherical SIL boosts the NA of the collection optics by a factor of $n$, where $n$ is the SIL’s refractive index. (A hyper-hemispherical SIL can boost the NA by up to $n^2$, but only up to a limit of about $NA = 1$, and has more limited field-of-view). The cubic zirconia ($\text{ZrO}_2$) SIL was 2 mm in diameter with a refractive index of $n = 2.2$, and had an aberration-free region in the center of the SIL of about 100 $\mu$m. The SIL must be mounted flush to the sample surface in order to improve collection efficiency, and can be held onto the sample surface by ‘soldering’ it in place with mounting wax. We observed a 2-4 time improvement in collection efficiency of sample M471 using the SIL.

### 2.4 Magneto-Photoluminescence Experiments

A set of PL spectra of a 600 nm mesa on sample M471, taken at magnetic fields ranging from 0 to 7 T, are shown in Figure 2.9. Two QDs can be seen in the figure. The line at shorter wavelength splits into a symmetric quadruplet at high field, which is indicative of a charged QD. The line at longer wavelength splits into an asymmetric doublet,
Figure 2.9: PL spectra of a mesa at various magnetic fields, showing a charged QD and a neutral QD. Horizontal and vertical polarized spectra have been summed to make the figure.

which is characteristic of the bright exciton in a neutral QD. At high magnetic fields, the neutral QD’s dark exciton becomes visible (roughly 0.2 nm red of the neutral bright exciton).

We plot the line centers of the neutral and charged QDs from Figure 2.9 in Figure 2.10, after subtracting off the quadratic diamagnetic shift common to all the lines of each QD. The neutral QD’s lines split as predicted by Figure 2.4(a), and the charged QD’s line splits as predicted by Figure 2.4(b). Note that from this data we may determine the magnitude of the electron and hole $g$-factors $g_{e,z}$ and $g_{h,z}$, but we are unable to determine the sign of the $g$-factors or which $g$-factors belongs to the electron versus hole.

Figures 2.9 and 2.10 illustrate that by ramping the magnetic field, neutral and charged QDs may be identified by looking for symmetry in their split lines. However, ramping the magnetic field consumes both time and liquid helium. An alternative method to identify charged QDs is to fix the field and vary the polarization of the collected PL. The charged QD can be identified by its symmetric splitting as the
Figure 2.10: Line splittings as a function of magnetic field for (a) neutral and (b) charged QD from Figure 2.9. The square data points indicate PL emission of one polarization, the circular points indicate the orthogonal polarization. The two polarizations have arbitrarily been named H and V. The solid lines in (a) are the eigenvalues of equation 2.3, using $g_{e,z} = 0.3$ and $g_{h,z} = 0.37$ (or vice-versa). The solid lines in (b) are the eigenvalues of (equation 2.6) $\pm$ (equation 2.7), using $g_{e,z} = 0.35$ and $g_{h,z} = 0.25$ (or vice-versa).
2.4. MAGNETO-PHOTOLUMINESCENCE EXPERIMENTS

Figure 2.11: (a) PL intensity (encoded in color map) as a function of wavelength for various polarization angles, showing one charged QD and one neutral QD. (b) Higher-resolution PL intensity versus wavelength and polarization for another charged QD, showing four well-resolved peaks. Note that the outer peaks are polarized orthogonally to the inner peaks, as predicted in Figure 2.4(b).

The polarization angle of collected PL is varied, while the neutral QD is asymmetric.

Although charged and neutral QDs can be identified by varying the magnetic field as in Figure 2.9, this experiment is time consuming and burns a lot of liquid helium. A faster and less costly way to identify charged and neutral QDs is to take multiple PL spectra as the HWP angle is rotated, making a polarization-resolved PL map as shown in Figure 2.11. A charged QD will appear as a symmetric triplet (Figure 2.11(a)) or quadruplet (Figure 2.11(b)) pattern, while neutral QDs appear as a wandering line (Figure 2.11(a)).

In all, roughly half of the QD lines on sample M471 were found to be charged. Unfortunately there is no method to definitely identify a charged QD line as positively or negatively charged. Throughout this work, we assume that the charged QDs are negatively charged because of our n-type \( \delta \)-doping.
Chapter 3

Coherent Single Qubit Control

A basic building block for quantum information processing systems is the ability to completely control the state of a single qubit\cite{50, 51, 52, 53, 54, 55}. For spin-based qubits such as our single electron spin in a QD, a universal single-qubit gate is realized by a rotation of the spin by any angle about an arbitrary axis. Driven coherent Rabi oscillations between two spin states can be used to demonstrate control of the rotation angle. Ramsey interference, produced by two coherent spin rotations separated by a variable time delay, demonstrates control over the axis of rotation. Full quantum control of an electron spin in a QD has previously been demonstrated using resonant radio-frequency pulses that require many spin precession periods\cite{33, 56, 57, 58}. However, because solid-state systems typically suffer from fast $T_2$ decoherence times, it is desirable to perform single-qubit operations as quickly as possible.

Optical manipulation of the spin allows quantum control on a picosecond or femtosecond timescale\cite{59, 60, 9, 61, 62, 63, 64, 65}, permitting an arbitrary rotation to be completed within one spin precession period\cite{55}. Other recent works in optical single-spin control have demonstrated the initialization of a spin state in a QD,\cite{66, 67, 68} as well as the ultrafast manipulation of coherence in a largely unpolarized single-spin state\cite{64}.

In this chapter we demonstrate complete coherent control over an initialized electron spin qubit using picosecond optical pulses. First we describe theoretically how a single broadband optical pulse can rotate the spin of a single electron in a QD. We
then experimentally demonstrate optical pumping as a means to initialize the qubit with high fidelity. Next we vary the intensity of a single optical pulse to observe over six Rabi oscillations between the two spin-states. We then apply two sequential pulses to observe high-contrast Ramsey interference. Such a two-pulse sequence realizes an arbitrary single-qubit gate completed in a picosecond timescale. These results demonstrate a complete set of all-optical single-qubit operations.

3.1 Ultrafast Rotation Theory - Raman Transition Picture

3.1.1 3-level System

We begin by analyzing the 3-level Λ-system shown in Figure 3.1. In atomic physics it is common to control the state of such a system by applying a pair of lasers, separated in frequency by $\delta_e$, on or off resonance with the excited state to induce a Raman transition between $|1\rangle$ and $|0\rangle$. We will show that a single laser can be used instead to induce Raman transitions between $|1\rangle$ and $|0\rangle$, provided that the bandwidth of the laser pulse is broader than the splitting $\delta_e$. This may be intuitively understood by imagining that the broadband pulse contains many pairs of Raman sidebands with a well-defined phase relationship between them. Alternatively we may interpret the system in time-domain: if all the timescales (including the laser pulse duration) are fast compared to $1/\delta_e$, then the qubit rotation will be complete before any phase can accumulate between the two ground states, and we may therefore neglect $\delta_e$ entirely.

We now mathematically analyze the spin rotation illustrated in Figure 3.1, following [69]. The laser pulse is red-detuned from the excited state by $\Delta$, and the ground states $|0\rangle$ and $|1\rangle$ are split by $\delta_e$. The purpose of the detuning $\Delta$ is to prevent real excitation of the radiatively lossy excited state. The laser field couples states $|0\rangle - |2\rangle$ with Rabi frequency $\Omega_0(t) = \mu_{02}E_0(t)/\hbar$, and $|1\rangle - |2\rangle$ with Rabi frequency $\Omega_1(t) = \mu_{12}E_1(t)/\hbar$. Here $\mu_{ab}$ is the dipole matrix element of the $|a\rangle - |b\rangle$ transition, and $E_a$ is the complex field amplitude of the laser’s electric field.
The Hamiltonian for the Λ-system in the rotating wave approximation and interaction picture, written in the basis of \( \{ |0\rangle, |1\rangle, |2\rangle \) is given by

\[
H_{\text{int}} = \begin{pmatrix}
-\delta_e & 0 & -\Omega_0(t)/2 \\
0 & 0 & -\Omega_1(t)/2 \\
-\Omega_0^*(t)/2 & -\Omega_1^*(t)/2 & \Delta
\end{pmatrix}
\]  

(3.1)

where we have set \( \hbar = 1 \). The equation of motion for the three-level system is

\[
\frac{d |\Psi\rangle}{dt} = -iH_{\text{int}} |\Psi\rangle
\]  

(3.2)

where \( |\Psi(t)\rangle = a_0(t) |0\rangle + a_1(t) |1\rangle + a_2(t) |2\rangle \). The coefficients \( a_0, a_1, \) and \( a_2 \) evolve...
3.1. ULTRAFAST ROTATION THEORY - RAMAN TRANSITION PICTURE

According to

\[ \dot{a}_0(t) = \frac{i}{2} \Omega_0(t) a_2(t) + i \delta_e a_0(t) \] (3.3)

\[ \dot{a}_1(t) = \frac{i}{2} \Omega_1(t) a_2(t) \] (3.4)

\[ \dot{a}_2(t) = \frac{i}{2} \Omega_0^*(t) a_0(t) + \frac{i}{2} \Omega_1^*(t) a_1(t) - i \Delta a_2(t) \] (3.5)

If we are far red-detuned, such that \( \Delta \gg \Omega_{0,1} \), then we may adiabatically eliminate \( |2\rangle \) from the equations by neglecting the population in \( |2\rangle \). This allows the system to be effectively reduced to a 2-level system (following [70]) governed by the effective Hamiltonian

\[ H_{\text{eff}} = \begin{pmatrix} \delta_e & \Omega_{\text{eff}}(t) / 2 \\ \Omega_{\text{eff}}^*(t) / 2 & 0 \end{pmatrix} \] (3.6)

In the special case where \( |\Omega_0(t)| = |\Omega_1(t)| \), we may subtract off the common diagonal terms which simply AC Stark shift both states by the same amount, arriving at the simplified Hamiltonian

\[ H_{\text{eff}} = \begin{pmatrix} \delta_e - \frac{|\Omega_0(t)|^2}{4\Delta} & \Omega_{\text{eff}}(t) / 2 \\ -\Omega_{\text{eff}}^*(t) / 2 & \delta_e - \frac{|\Omega_1(t)|^2}{4\Delta} \end{pmatrix} \] (3.7)

where we have defined

\[ \Omega_{\text{eff}} = \frac{\Omega_0(t) \Omega_1^*(t)}{2\Delta} \] (3.8)

as the effective Rabi frequency of the Raman transition.

If the Rabi frequencies are unequal \( |\Omega_0| \neq |\Omega_1| \), then the rotation will have a component parallel to the magnetic field. We may rewrite equation 3.7 in the frame precessing with the electron spin at frequency \( \delta_e \) in order to understand how it can lead to spin rotations:

\[ H_{\text{eff}} = \begin{pmatrix} 0 & -\Omega_{\text{eff}}(t) e^{-i\delta_e t} / 2 \\ -\Omega_{\text{eff}}^*(t) e^{i\delta_e t} / 2 & 0 \end{pmatrix} \] (3.9)
We may use equation 3.9 to derive the equations of motion for the Pauli spin operator vector \( \vec{S}(t) = (\langle \hat{S}_x(t) \rangle, \langle \hat{S}_y(t) \rangle, \langle \hat{S}_z(t) \rangle) \) following [71]. With the magnetic field parallel to the z-axis, the spin-vector \( \vec{S}(t) \) is rotated according to

\[
\dot{\vec{S}} = \vec{R} \times \vec{S}
\]  

(3.10)

where the axis about which the spin rotates is given by

\[
\vec{R} = \begin{pmatrix}
|\Omega_{\text{eff}}| \cos(\theta - \delta_e t) \\
|\Omega_{\text{eff}}| \sin(\theta - \delta_e t) \\
0
\end{pmatrix}
\]  

(3.11)

where \( \theta = \angle \Omega_{\text{eff}} \). The instantaneous axis of rotation in the X-Y plane is given by \((\theta - \delta_e t)\). If the laser pulse duration is short compared to \(2\pi/\delta_e\), then the rotation will be about a single axis in the X-Y plane. We have thus shown that a single pulse may rotate the electron spin, provided the pulse bandwidth is broader than the splitting \(\delta_e\). Further, we may control the axis of rotation (in the frame precessing with the spin) by controlling the arrival time of the pulse. If we move back to the stationary lab frame, then the spin rotation will always be about the same axis, but the spin precesses at the Larmor frequency \(\delta_e\).

### 3.1.2 4-level System

As described in the previous chapter, our system in fact contains not 3 levels, but 4, as shown in Figure 3.2. We can see that now the electron spin states \(|\uparrow\rangle\) and \(|\downarrow\rangle\) are now connected by two \(\Lambda\)-systems. In order to ensure that the probability amplitude from the two \(\Lambda\)-systems interfere constructively rather than destructively, we will analyze the dipole matrix elements of the four transitions.

First, we consider the optical transitions in a charged QD with no applied magnetic field, as shown in Figure 3.3(a). The \(|+3/2\rangle_x\) trion couples to the \(|+1/2\rangle_x\) electron by emitting a \(\sigma_+ = \frac{1}{\sqrt{2}}(\vec{X} + i\vec{Y})\) circularly polarized photon which carries angular momentum +1, while the \(|-3/2\rangle_x\) trion couples to the \(|-1/2\rangle_x\) electron by emitting a \(\sigma_- = \frac{1}{\sqrt{2}}(\vec{X} - i\vec{Y})\) circularly polarized photon with angular momentum -1.
3.1. ULTRAFAST ROTATION THEORY - RAMAN TRANSITION PICTURE

Figure 3.2: The 4-level system of a negatively-charged QD in a Voigt magnetic field, with electron splitting $\delta_e$ and trion splitting $\Delta$. The four transitions are coupled by Rabi frequencies $\Omega_{1,2,3,4}$.

Figure 3.3: (a) The polarization of the two optically active transitions of a charged QD in no magnetic field. (b) The states rewritten in the eigenstates of a Voigt magnetic field, showing the relative matrix elements of the optical transitions between the four states and corresponding polarizations.
We may re-diagonalize the states into the eigenstates of the Voigt magnetic field, as shown in Figure 3.3(b). In this basis we have four optically-active transitions. We may calculate the polarization and phase of each transition:

\[ |⇑⟩ \rightarrow |↑⟩ \]  \hspace{1cm} (3.12)

\[ \frac{1}{\sqrt{2}}(|+3/2⟩_x + |-3/2⟩_x) \rightarrow \frac{1}{\sqrt{2}}(|+1/2⟩_x + |-1/2⟩_x) \]  \hspace{1cm} (3.13)

\[ \Rightarrow \frac{1}{2} \sigma_+ + \frac{1}{2} \sigma_- = \frac{1}{2\sqrt{2}}(\vec{X} + i\vec{Y}) + \frac{1}{2\sqrt{2}}(\vec{X} - i\vec{Y}) \]  \hspace{1cm} (3.14)

\[ = \frac{1}{\sqrt{2}} \vec{X} \]  \hspace{1cm} (3.15)

Thus the \( |⇑⟩ \rightarrow |↑⟩ \) transition couples to \( \vec{X} \) polarized light with a relative (normalized) matrix element strength of \( \mu = \frac{1}{\sqrt{2}} \). Similarly, the \( |⇓⟩ \rightarrow |↓⟩ \) transition also couples to \( \vec{X} \) with matrix element \( \mu = \frac{1}{\sqrt{2}} \), while \( |⇓⟩ \rightarrow |↑⟩ \) and \( |⇑⟩ \rightarrow |↓⟩ \) couple to \( \vec{Y} \) light with matrix element \( \mu = \frac{i}{\sqrt{2}} \).

We denote the effective Rabi frequency of the \( \Lambda \)-system involving the \( |⇑⟩ \) and \( |⇓⟩ \) trion states respectively as \( \Omega_{⇑} \) and \( \Omega_{⇓} \), and the rotation pulse’s electric field as \( E_x \vec{X} + E_y \vec{Y} \). Using \( \Omega = \mu E \) with \( |\mu| = \frac{1}{\sqrt{2}} \) from before (and setting \( h = 1 \)), we find that

\[ \Omega_{⇑} = \frac{\Omega_2 \Omega_1^*}{2(\Delta + \delta_h)} = \frac{E_y E_x^*}{4(\Delta + \delta_h)} \]  \hspace{1cm} (3.17)

\[ \Omega_{⇓} = \frac{\Omega_4 \Omega_3^*}{2\Delta} = \frac{-iE_x E_y^*}{4\Delta} \]  \hspace{1cm} (3.18)

Under the approximation that \( \delta_h \ll \Delta \), we find the total effective Rabi frequency to be

\[ \Omega_{\text{eff}} = \Omega_{⇑} + \Omega_{⇓} = \frac{E_y E_x^* - iE_x E_y^*}{4\Delta} \]  \hspace{1cm} (3.19)

The magnitude of the effective Rabi frequency \( |\Omega_{\text{eff}}| \) may thus be maximized by
setting $E_y = \pm i E_x \equiv \frac{|\vec{E}|}{\sqrt{2}}$, namely using a circularly-polarized rotation laser pulse with total electric field strength $|\vec{E}|$. In this case

$$\Omega_{\text{eff}} = \frac{\Omega^2}{\Delta} = \frac{|\vec{E}|^2}{4\Delta}$$

(3.20)

where $\Omega$ is the Rabi frequency of each of the four individual transitions. The total angle through which the electron spin is rotated is given by

$$\Theta = \int_{\text{pulse}} \Omega_{\text{eff}}(t) dt \propto \frac{\varepsilon}{\Delta}$$

(3.21)

where $\varepsilon$ is the total energy in the laser pulse. Thus we expect the spin to Rabi oscillate between $|\uparrow\rangle$ and $|\downarrow\rangle$ periodically as rotation pulse energy is varied.

### 3.2 Ultrafast Rotation Theory - Stark-shift Picture

Another more intuitive way to understand why a circularly-polarized rotation pulse is ideal is to view the pulse’s effect as an AC Stark shift, as shown in Figure 3.4. From the ultrafast pulse’s point of view, the splittings $\delta_e$ and $\delta_h$ are extremely small and may be ignored, so we are free to re-diagonalize the electron and trion states into the x-basis (growth direction, Faraday geometry, optical axis direction). The circularly-polarized pulse couples only one transition, and AC-Stark shifts the electron state $|\pm 1/2\rangle$ relative to $|\mp 1/2\rangle$.

We can calculate the amplitude of the AC Stark shift $\delta_S$ as follows. In the previous section we defined $\Omega$ to be the Rabi frequency of each of the four transitions in Figure 3.2 in the presence of circularly polarized coupling light. Each transition had relative dipole strengths of $|\mu| = \frac{1}{\sqrt{2}}$, and was coupled by an electric field $E_{X,Y} = \frac{|\vec{E}|}{\sqrt{2}}$. Because in Figure 3.4 we have re-diagonalized the states back into the x-basis, the relative dipole strength of the two optically active transitions is $|\mu| = 1$, and is coupled by the full electric field strength $|\vec{E}|$ so we have $\Omega_{\pm} = 2\Omega$. Only two of the states in Figure 3.4 are coupled, and their Hamiltonian in the rotating frame and interaction
Figure 3.4: (a) The polarization of the two optically active transitions of a charged QD in no magnetic field. (b) The states rewritten in the eigenstates of a Voigt magnetic field, showing the relative matrix elements of the optical transitions between the four states and corresponding polarizations.

The polarization is given by

\[ H_{\text{Stark}} = \begin{pmatrix} 0 & \Omega_+/2 \\ \Omega_+/2 & 0 \end{pmatrix} = \begin{pmatrix} 0 & \Omega \\ \Omega^* & 0 \end{pmatrix} \] (3.22)

We may diagonalize this Hamiltonian to find the new Stark-shifted energies of the system in the presence of the rotation laser:

\[ H_{\text{Stark}} = \begin{pmatrix} \frac{\Delta}{2} - \frac{1}{2} \sqrt{\Delta^2 + 4|\Omega|^2} & 0 \\ 0 & \frac{\Delta}{2} + \frac{1}{2} \sqrt{\Delta^2 + 4|\Omega|^2} \end{pmatrix} \] (3.23)

The Stark shift is therefore given by

\[ \delta_S = \frac{\Delta}{2} - \frac{1}{2} \sqrt{\Delta^2 + 4|\Omega|^2} \] (3.24)

\[ = \frac{|\Omega|^2}{\Delta} - \frac{|\Omega|^4}{\Delta^3} + \ldots \] (3.25)
Now consider what happens to a superposition spin state when the Stark shift occurs. If the spin began in state

\[ |\Psi\rangle = a |\uparrow\rangle + b |\downarrow\rangle = \frac{1}{\sqrt{2}} (a + b) |+1/2\rangle_x + \frac{1}{\sqrt{2}} (a - b) |-1/2\rangle_x \] (3.26)

and the pulse Stark shifts $|+1/2\rangle$ by an amount $\delta_s$ for a pulse duration time $T_P$, then after the pulse the state will be

\[ |\Psi'\rangle = \frac{1}{\sqrt{2}} (a + b)e^{-i\delta_s T_P} |+1/2\rangle_x + \frac{1}{\sqrt{2}} (a - b) |-1/2\rangle_x \] (3.27)

\[ = \left( a \cos \frac{\delta_s T_P}{2} - b \sin \frac{\delta_s T_P}{2} \right) |\uparrow\rangle \] (3.28)

\[ + \left( b \cos \frac{\delta_s T_P}{2} - a \sin \frac{\delta_s T_P}{2} \right) |\downarrow\rangle \]

where we have dropped the overall phase factor. Thus we see that the spin has been rotated by an angle $\Theta = \delta_s T_P$. We can therefore identify that the Stark shift gives the effective rotation frequency, or $\delta_s = \Omega_{\text{eff}} = |\Omega|^2 / \Delta$, in agreement with equation 3.20. Thus the Raman transition picture and AC Stark shift picture are equivalent.

### 3.3 Initialization and Measurement Theory

In addition to rotations, a complete set of single qubit operations also requires initialization and measurement. We perform both of these tasks by optical pumping (see Fig. 1b). A narrow-band continuous-wave laser optically drives the $|\downarrow\rangle \leftrightarrow |\uparrow\downarrow, \downarrow\rangle$ transition with rate $\Omega_p$. The optical pumping laser has negligible effect on the spin rotation because $\Omega_p \ll \Omega_{\text{eff}}$. Spontaneous decay at rate $\Gamma/2$ into the two spin states quickly initializes the electron into the $|\uparrow\rangle$ state, where $\Gamma$ is the trion’s total spontaneous emission rate. After spin rotation, the population in the $|\downarrow\rangle$ state is measured by the same optical pumping process. If the spin was rotated to $|\downarrow\rangle$, the QD will emit a single photon from the $|\uparrow\downarrow, \downarrow\rangle \rightarrow |\uparrow\rangle$ transition, which can be detected using a single-photon counter.

Our single-spin measurement technique has been proposed for use in quantum
CHAPTER 3. COHERENT SINGLE QUBIT CONTROL

Figure 3.5: The spin initialization and measurement scheme performed by optical pumping.

computation[50], and offers the experimental convenience of including measurement and initialization in the same step. However, the fidelity of a single-shot readout is limited by the photon collection efficiency. An optical microcavity would boost the measurement scheme’s efficiency, and could also enable coherent conversion of spin-qubits into photon-qubits for quantum networking[72]. Resonant absorption measurements[66, 67, 68] offer similar advantages, but also require a microcavity-enhanced absorption cross-section to enable single-shot readout. Quantum non-demolition measurements based on dispersive Kerr rotation[73], Faraday rotation[74], or a recycling transition[75] use many photons to measure the spin and are therefore more robust to photon loss, but require a separate initialization step.

3.4 Experimental Setup

A simplified diagram of the experimental setup is shown in Figure 3.6. The rotation pulses are generated by a Spectra-Physics Tsunami mode-locked laser. The laser emits picosecond-duration pulses with a 13.2 ns repetition period, and is tunable in emission wavelength from 700 - 1000 nm. The optical pump was performed by
3.4. EXPERIMENTAL SETUP

Figure 3.6: Experimental setup. One or two rotation pulses may be sent to the sample during each experimental cycle, to observe Rabi oscillations or Ramsey interference, respectively. The time delay $\tau$ between pairs of pulses is controlled by a retroreflector mounted on a computer-controlled translation stage.

The QD emission was spectrally dispersed and filtered using a double-monochromator with 0.02 nm resolution. Scattered laser light was further rejected by double-passing through a quarter-wave plate (QWP) and polarizing beam splitters (PBS). The first stage of the double-monochromator utilized the pinhole spatial-filter as its "input slit", a 2000 groove-per-mm holographic grating as its dispersive element, and the spectrometer’s input slit as its "output slit". The second stage of the double-monochromator was the spectrometer itself, using the side-exit as its "output slit". The double-monochromator had roughly double the spectral resolution of just the

continuous-wave (CW) a Spectra-Physics Matisse ring laser. The Matisse is tunable with one optics set from about 850 - 950 nm. The charged QD sample M471 was held in the Spectromag magneto-optical cryostat at 1.6 K temperature and 7 T magnetic field.

The QD emission was spectrally dispersed and filtered using a double-monochromator with 0.02 nm resolution. Scattered laser light was further rejected by double-passing through a quarter-wave plate (QWP) and polarizing beam splitters (PBS). The first stage of the double-monochromator utilized the pinhole spatial-filter as its "input slit", a 2000 groove-per-mm holographic grating as its dispersive element, and the spectrometer’s input slit as its "output slit". The second stage of the double-monochromator was the spectrometer itself, using the side-exit as its "output slit". The double-monochromator had roughly double the spectral resolution of just the
CHAPTER 3. COHERENT SINGLE QUBIT CONTROL

Figure 3.7: Details of the double-monochromator. The first stage consists of the pinhole, G1, and S1. The second stage consists of S1, G2, and S2. G1: grating 1, holographic 2000 lines/mm; G2: grating 2, ruled 1716 lines/mm. S1 and S2: slit 1 and slit 2; SPCM: single-photon counting module.

The single photons were detected after the double-monochromator by a single-photon counting module (SPCM). The count rate from the signal photons was roughly equal to the count rate from ring-laser leakage photons, on the order of 1000 counts per second each. The SPCM output a single 5 V transistor-transistor logic (TTL) pulse, roughly 30 ns in duration, each time it detected a photon. Each TTL pulse triggered a 1 V, 1 µs-duration electrical pulse from a pulse generator in order to ‘clean up’ the relatively noisy TTL signal. The electrical pulses were then filtered by a low-pass RC circuit with a 50 µs time constant before being detected by a Stanford Research Systems SR830 lock-in amplifier. The mode-locked rotation pulse laser was chopped using an optical chopper at 500 Hz, and the lock-in amplifier was synchronized to this.
3.5 Initialization Experiment

The optical initialization is calibrated by measuring the single photon count rate as a function of optical pumping power $P_{op}$ following a fixed rotation through angle
Θ = π in Figure 3.9(a). The signal saturates around $P_{\text{op}} \sim 15 \mu W$ as the population in the $|\downarrow\rangle$ state is nearly completely initialized to $|\uparrow\rangle$. For all remaining experiments, $P_{\text{op}}$ was fixed just above the saturation of the optical pumping curve. To quantify the initialization fidelity, a time-resolved measurement of photon count rate following a rotation of Θ = π is shown in Figure 3.9(b). The count rate is proportional to the instantaneous population in $|\downarrow\rangle$. Immediately following the rotation pulse, the population in $|\downarrow\rangle$ is near unity and the signal is maximized. The signal drops as the spin is pumped back to $|\uparrow\rangle$ in a characteristic time of 3.4 ns, orders of magnitude faster[68] than optical pumping schemes involving a dipole-forbidden transition[66]. The minimum count rate, occurring just prior to the next rotation pulse, corresponds to the remnant population in $|\downarrow\rangle$ due to imperfect initialization.

The time-resolved photon count rate data (Figure 3.9(b)) has had a background subtracted to remove photons scattered directly from the optical pump laser. To estimate this background, four measurements of the scattered laser count rate were performed, at positions on the sample approximately 1 µm above, below, left, and right of the QD. Spherical aberrations in the solid immersion lens lead to varying background count rates at the four positions, and estimating the background is the largest source of uncertainty in the fidelity calculation. The mean of these backgrounds was subtracted from the measured data, and the standard deviation of the backgrounds was used to calculate the uncertainty in the initialization fidelity. By comparing the count rates immediately before and after the rotation pulse, we estimate (see below) the spin initialization fidelity to be $F_0 = 92 \pm 7\%$.

To estimate the initialization fidelity, we assume that the Bloch vector is optically pumped to an initial length $L_0$. We take the minimum count rate of Fig. 2b to correspond to $(1 - L_0)/2$. Immediately after a rotation by angle Θ = π, the Bloch vector has length $L_0D_\pi$ and is tilted by $2\theta = 0.34$ radians away from the Bloch sphere’s north pole (this angle $\theta$ is calculated in Appendix A). The maximum signal is thus $[1 + L_0D_\pi \cos(2\theta)]/2$. By equating the calculated and measured ratios of the maximum to minimum signal we determine $L_0 = 0.83 \pm 0.14$, where the uncertainty is dominated by the background subtraction. The initialization fidelity is given by $F_0 = (1 + L_0)/2 = 0.92 \pm 0.07$. 
3.6. RABI OSCILLATIONS

Rabi oscillations between the two spin states are evident in the photon count rate as the rotation pulse power $P_{rp}$ is varied in Figure 4.6(a). In contrast to the adiabatic elimination model discussed earlier which predicts $\Theta \propto P_{rp}$, we empirically determine that $\Theta \propto P_{rp}^{0.68}$ over the range of $\pi \leq \Theta \leq 13\pi$ (Figure 4.6(b)). This sub-linear dependency is a consequence of the breakdown of the adiabatic approximation $\Omega_{v,h} \ll \Delta$, as non-negligible virtual population is present in the excited states during the rotation pulse.

The amplitude of the Rabi oscillations shrinks due to incoherent processes such as trion dephasing. This may be understood as a decrease of the length of the Bloch vector of the two-state system as $\Theta$ increases. This decreasing length is well fit (excluding the first data point) by an empirical exponential decay proportional to $\exp(-\Theta/8.6\pi)$ as shown in Figure 4.6(c). These incoherent processes transform the virtual population in the excited states during the rotation pulse into real population,
which contributes to the photon count rate as background noise. The increasing background is responsible for the overall upwards slope of the data in Figure 4.6(a).

In order to find the rotation angle $\Theta$ and Rabi oscillation amplitude shown respectively in Figures 4.6(b) and (c), the raw data of Figure 4.6(a) was smoothed with a five-point moving-average filter. The locations of the maxima (minima) of the smoothed data were taken to be where the rotation angle equaled an odd (even) multiple of $\pi$. The first oscillation amplitude was excluded from the exponential fit (Figure 4.6(c)) because the first Rabi oscillation peak is reduced by the tilted rotation axis for small angles $\Theta$.

The experimentally determined trajectory of the Bloch vector as it undergoes Rabi oscillations is parametrically plotted in Figure 3.11 as a function of rotation pulse power. The methods to generate this trajectory are described in Appendix A. For small rotation angles $\Theta \lesssim \pi$ the vector rotates about a tilted axis because the Larmor precession frequency $\delta_e$ is non-negligible compared to the effective Rabi frequency $\Omega_{\text{eff}}$. This tilted axis of rotation causes the reduced height of the first peak in Figure 4.6(a) and the lowered first oscillation amplitude in Figure 4.6(c). For larger rotation angles, $\Omega_{\text{eff}} \gg \delta_e$, and the rotation is very nearly about the $x$-axis.

### 3.7 Ramsey Interference

Rabi oscillations demonstrate the rotation of a qubit by an arbitrary angle about a single axis, i.e. $U(1)$ control. Full control over the Bloch sphere, i.e. $SU(2)$ control, requires rotation about a second axis. The natural Larmor precession of the spin about the $z$-axis accomplishes this rotation, and can be investigated by Ramsey interferometry.

In a Ramsey interferometer, the spin population is measured following a pair of $\pi/2$ rotations about the $x$-axis separated by a variable free precession time $\tau$ about the $z$-axis. Ramsey fringes are shown in Figure 3.12.
Figure 3.10: (a) Rabi oscillations between the spin states are evident in the oscillating photon signal as rotation pulse power $P_{RP}$ is increased. (b) The rotation angle as a function of rotation pulse power, showing an empirical fit to a power-law dependence. (c) Amplitude of measured Rabi oscillations as a function of rotation angle, with an empirical exponential fit.
Figure 3.11: Experimental trajectory of the Bloch vector. The curves trace out the tip of the Bloch vector in the one-pulse (Rabi oscillation) experiment over the range of rotation angles $\Theta$ from 0 to $3\pi$. The color scale indicates the length of the Bloch vector, which shrinks exponentially with $\Theta$. Views are from the perspective of: (a): the $x$-axis, and (b): the $-y$-axis of the Bloch sphere. The length of the Bloch vector and rotation angle is extracted from the extrema of the Rabi oscillation data shown in Figure 4.6, while the azimuthal position of the Bloch vector is revealed by the phase of the Ramsey fringes shown in Figure 3.13.

The Ramsey interference data are fit at each rotation angle $\Theta$ by:

$$y(\tau, \Theta) = B(\Theta) \left[ 1 - \exp \left( -\frac{\tau}{b(\Theta)} \right) \right] + A(\Theta) \exp \left( -\frac{\tau}{T_2^*(\Theta)} \right) \cos[\delta_e \tau + \phi(\Theta)],$$

where the first term accounts for optical pumping from $|\downarrow\rangle$ to $|\uparrow\downarrow, \downarrow\rangle$, $A$ is the initial amplitude of the Ramsey fringe, $T_2^*$ is the fringe decay time, $\delta_e$ is the non-adjustable Larmor frequency, and $\phi$ is the phase of the fringe. Figure 5d shows the peak-to-peak fringe height $2A(\Theta) \exp[-\tau_0/T_2^*(\Theta)]$ versus $\Theta$, evaluated at the the minimum measured time delay $\tau_0 = 16$ ps.

The fringe amplitude decays with a time constant $T_2^* = 185$ ps. This short $T_2^*$ is a consequence of the optical pumping laser remaining on between the two rotation pulses, and could be extended by switching the optical pump off between pulses using a fast electro-optic modulator. We determine the electron $g$-factor magnitude to be $|g_e| = 0.267$ from the Larmor frequency $\delta_e/2\pi = 26.3$ GHz.

In order to calculate the fidelity of a $\pi/2$ pulse, we assume that the Bloch vector
begins with length \( L_0 \) and shrinks by a factor of \( D_{\pi/2} \) with each \( \pi/2 \) rotation, so the vector has length \( L_0 D_{\pi/2}^2 \) after two \( \pi/2 \) rotations. The population in state \( |\downarrow\rangle \) oscillates between \( (1 + L_0 D_{\pi/2}^2)/2 \) and \( (1 - L_0 D_{\pi/2}^2)/2 \), and our lock-in detection technique automatically subtracts a background corresponding to \( (1 - L_0)/2 \). The measured signal therefore oscillates between \( C(L_0 - L_0 D_{\pi/2}^2)/2 \) and \( C(L_0 + L_0 D_{\pi/2}^2)/2 \), where \( C \) is a scale factor to convert from population to the measured arbitrary units. The initialized length \( L_0 \) and scale factor \( C \) thus factor out of the expressions; they simply set the overall scale of the signal. We equate these two expressions to the maximum and minimum of the fit to Eq. (1), evaluated at the minimum measured delay \( \tau = 16 \) ps, to determine \( CL_0 = 3.1 \) and \( D_{\pi/2} = 0.87 \). The \( \pi/2 \)-pulse fidelity is estimated to be \( F_{\pi/2} = (1 + D_{\pi/2})/2 = 94\% \).

To investigate the quality of our \( \pi \)-pulses, we perform a similar experiment with two \( \pi \)-pulses separated by a variable time delay, as shown in Fig 5b. Ideally, the signal would remain constant at \( L_0(1 - D_{\pi}^2)/2 \) with no oscillations. The signal shows an overall upwards slope, again due to the optical pump remaining on between the two \( \pi \)-pulses and pumping population from the \( |\downarrow\rangle \) state into \( |\uparrow\downarrow, \downarrow\rangle \) where it is later detected. Small oscillations remain in the signal due to the fact that our \( \pi \)-pulse is not exactly around the \( x \)-axis as discussed earlier.

We assume that the Bloch vector rotates about an axis with polar angle \( \theta \) below the Bloch sphere’s north pole. The measured signal should oscillate between \( CL_0(1 - D_{\pi}^2)/2 \) and \( CL_0[1 - D_{\pi}^2 \cos(4\theta)]/2 \). However, experimental noise and errors in the relative intensity of the two pulses lead to an unreliable estimate of the rotation axis \( \theta \) using the \( \pi \)-pulse fringe amplitude alone. Instead, we take \( \theta = 1.4 \) radians from the phase of the Ramsey fringe (discussed in the Appendix), i.e. the rotation axis is tilted 0.17 radians from the Bloch sphere’s equator. If we simply model the rotation pulse as a rectangular pulse with constant \( \Omega_{\text{eff}} \) applied over 4 ps, we would expect to rotate around an axis tilted by roughly \( \delta_e/\Omega_{\text{eff}} = 0.21 \) radians, in reasonable agreement with experiment.

We use the minimum of the fit to the Ramsey fringe data evaluated at \( \tau = 16 \) ps to estimate \( CL_0(1 - D_{\pi}^2)/2 \), and assume that the scale factor \( CL_0 \) is unchanged from the \( \pi/2 \)-pulse case. This yields \( D_{\pi} = 0.88 \). The \( \pi \)-pulse fidelity is then estimated by
CHAPTER 3. COHERENT SINGLE QUBIT CONTROL

Figure 3.12: (a) Ramsey interference fringes for a pair of $\pi/2$ pulses, showing photon count rate versus time delay between pulses. (b) Destructive Ramsey interference for a pair of $\pi$ pulses. The data in (a) and (b) are fit to an exponentially-decaying sinusoid with a linear offset (see Supplementary Information for details).

$$F_\pi = \frac{1 + \cos(2\theta)D_\pi}{2} = 91\%.$$  

3.8 Arbitrary Single-Qubit Gate

In order to construct a general SU(2) single-qubit gate, we may adjust the intensities of the first and second rotation pulses and the precession duration $\tau$, thus applying rotations through three Euler angles about the $x$, $z$, and $x$ axes. In Figure 3.13(a) we explore the entire surface of the Bloch sphere by varying the rotation angle of both rotation pulses as well as the delay time $\tau$. The fringe amplitude is shown versus rotation angle in Figure 3.13(b). High contrast Ramsey fringes are visible when each rotation angle is a half-integer multiple of $\pi$, while the fringes vanish when each rotation angle is a full-integer multiple of $\pi$.

Our single-qubit gate, consisting of three independent rotations about different axes, is accomplished in less than one Larmor period of 38 ps. $T_2$ coherence times of 3.0$\mu$s have been reported for QD electron spins$^9$, so nearly $10^5$ gate operations may be possible within the qubit’s coherence time. The rotation pulses are of sufficient fidelity to be applied to a simple spin-based quantum information processing system.
Figure 3.13: (a) Photon count rate is color-mapped as a function of rotation angle \( \Theta \) and delay time between pulses \( \tau \). (b) The amplitude of Ramsey fringes for various rotation angles. Fringe amplitudes are determined by fitting the data shown in (b) with decaying sinusoids.
Chapter 4

Spin Echo

The preservation of phase coherence of a physical qubit is essential for quantum information processing because it sets the memory time of the qubit. The decoherence time is not a fundamental property of the qubit, but rather it depends on how the qubit states are manipulated and measured. Although the intrinsic decoherence of an individual spin can be quite slow, electron spin coherence in an ensemble of QDs is typically lost in a nanosecond timescale due to inhomogeneous broadening[76, 77]. In a single, isolated QD, there is no inhomogeneous broadening due to ensemble averaging. Instead, a single spin must be measured repeatedly in order to characterize its dynamics, which can lead to a temporally-averaged fast dephasing if the spin evolves differently from one measurement to the next. In an InGaAs QD, the electron’s hyperfine interaction with nuclear spins causes a slowly-fluctuating background magnetic field, which leads to a short dephasing time $T_{2}^*$ on the order of 1-10 ns[32, 78, 79] after temporal averaging. This dephasing can be largely reversed using a spin echo[37, 32, 62, 78, 80], which rejects low-frequency nuclear-field noise by refocussing the spin’s phase, allowing a qubit of information to be stored throughout the spin’s decoherence time $T_2$. This $T_2$ decoherence time is limited by dynamical processes, such as nuclear spin diffusion and electron-nuclear spin feedback[81, 82]. It is possible to even further extend the decoherence time, or enhance coherence during a desired time interval, by applying a ”dynamical decoupling” sequence of rapid spin rotations[83, 84, 85, 86] to filter out higher-frequency nuclear noise. Dynamical
decoupling and spin refocussing techniques benefit greatly from having the rotation operations performed as fast as possible[85].

A spin-echo pulse sequence consists of a first $\pi/2$ rotation to generate a coherence between $|\uparrow\rangle$ and $|\downarrow\rangle$ states, followed by a time delay $T$ during which the spin dephases freely. Next, a $\pi$ rotation is applied which effectively reverses the direction of the spin’s dephasing. The spin rephases during another time delay $T$, at which point another $\pi/2$ rotation is applied to read out the spin’s coherence.

In this chapter we extend the ultrafast spin-rotation techniques developed in Chapter 3 to implement a spin-echo pulse sequence. We observe $T_2$ decoherence times up to $\sim 3 \, \mu$s, which is more than 1000 times longer than the spin’s $T_2^*$ dephasing time of $\sim 2$ ns. We find that $T_2$ improves with increasing applied magnetic field for low fields, then saturates for high fields.

4.1 Nuclear Hyperfine Interaction

We start by briefly analyzing the interaction between a single electron spin and the nuclear spins in a QD. The nuclear hyperfine interaction between the single electron with spin $m_e$ and a single nucleus with spin $m_n$ is given by [69]

$$E_{HF} = m_em_n \frac{2\mu_0}{3} g_0 \mu_B \gamma_n \hbar |\psi_e(x_n)|^2 \quad (4.1)$$

where $\mu_0$ is the permeability of free space, $g_0$ is the electron g-factor, $\mu_B$ is the Bohr magneton, $\gamma_n$ is the nuclear gyromagnetic ratio, and $\psi_e(x_n)$ is the electron’s wavefunction evaluated at the nuclear site. Because the electron in the semiconductor’s conduction band has an s-like Bloch wavefunction, $|\psi_e(x_n)|$ is sizeable and there is a ‘contact’ hyperfine interaction between the electron and nuclear spins. Gallium and arsenic nuclei each have spin 3/2, while indium nuclei have spin 9/2.

The Zeeman splitting of a nuclear spin at the highest fields used in our experiments (10 T) is on the order of $\hbar \times 10$ MHz or a few 10s of neV. Meanwhile, the thermal energy at 1.5 K is $\hbar \times 30$ GHz or 100 µeV, more than three orders of magnitude higher than the nuclear Zeeman splitting. The nuclear spins may therefore be thermally excited.
to any spin state, and the nuclei are expected to be completely unpolarized with a net magnetization of zero. However, for a collection of $N$ nuclei, one expects fluctuation of the total nuclear polarization on the order of $\sqrt{N}$.

Typical dimensions for a self-assembled InGaAs QD are 30 nm diameter and 2.5 nm in height, and we may assume that the electron’s wavefunction penetrates slightly into the surrounding GaAs barrier to say 40 nm diameter and 3 nm height. We can then estimate the number of nuclear spins $N$ interacting with the electron by approximating the electron using a hard-edged pancake-shaped wavefunction and using a nuclear density of $4.5 \times 10^{22}$ cm$^{-3}$, leading to $N \sim 10^5$.

The contact hyperfine interaction will increase or decrease the electron spin’s Larmor frequency $\delta_e$ depending on the nuclear spin configuration during a given experiment. For this reason the hyperfine interaction is often said to give rise to an ‘effective magnetic field’ $\Delta B_{HF}$. The magnitude of the hyperfine field can be estimated by [87]

$$\Delta B_{HF} = b_0 \sqrt{I_0(I_0+1)/N}$$

where $b_0 = 3.5$ T and $I_0 = 3/2$ for pure GaAs. Although our QD contains some indium nuclei as well, the pure GaAs case will get us an order-of-magnitude estimate. Using $N \sim 10^5$, we find $\Delta B_{HF} = 20$ mT. If the effective magnetic field varies by 20 mT then the electron Larmor frequency will vary by $\Delta \delta_e = g_e\mu_B \Delta B_{HF} \sim 100$ MHz.

The nuclear spins flip relatively slowly compared to the other timescales in our experiments, so we can consider the nuclear magnetization to be nearly constant within one experimental trial. However, because we repeat each experiment hundreds of millions of times to collect photon statistics, the nuclear magnetization may change from one experimental trial to the next, leading to the time-averaged dephasing $T_2^*$ time discussed earlier. If the nuclear magnetization has a Gaussian distribution as we are assuming here, then the Larmor frequency will also be Gaussian distributed with a width on the order of 100 MHz, and we would expect the electron spin to dephase with a Gaussian profile on a timescale of a few nanoseconds.
4.2 Racetrack Analogy for $T_2^*$, $T_2$, and Spin Echo

We can use an analogy of runners on a track to illustrate the ideas of $T_2^*$, $T_2$, and Spin Echo. Let’s imagine the electron spin precessing around the Bloch sphere in each different experimental trial as a different runner racing on a circular track. The runners each run at a different constant speed (due to different nuclear magnetization values in the QD). The runners are all in phase when the starting gun goes off (initial $\pi/2$ pulse that tips the Bloch vector onto the equator) in Figure 4.1(a). After a time $T_2^*$, the runners are all spread uniformly around the track and no longer have any particular phase relationship. However, at some even later time $T$, another gun goes off which tells all the runners to switch direction ($\pi$ refocussing pulse) as shown in Figure 4.1(b). After another time $T$, the runners will all cross the starting/finish line at the exact same time - they will naturally re-phase after a total time of $2T$ - as shown in Figure 4.1(c). (We can then measure the position of the ‘runners’ by applying the final $\pi/2$-pulse of the spin-echo sequence.)

We can always re-phase our runners, no matter how different their running speeds are, as long as they are each able to maintain a constant running speed. However, if the runners cannot maintain a constant speed over the entire duration of the race $2T$ and some speed up while others slow down, then they will not cross the starting line in phase as shown in Figure 4.1(c). The amount of time over which the runners can maintain a constant speed is called $T_2$, and is generally longer than the dephasing time $T_2^*$.

In the QD, the electron spin won’t maintain a constant Larmor precession frequency $\delta_e$ forever because of dynamical changes in the nuclear magnetization during a single experimental trial, leading to the microsecond-timescale $T_2$ decoherence.

4.3 Experimental Setup

In order to measure decoherence times up to a microsecond timescale, it was necessary to block the optical pump laser and rotation pulse laser while the electron spin undergoes free precession. The experimental setup used to demonstrate arbitrary
single-qubit gates (Figure 3.6) was modified as shown in Figure 4.2 in order to accomplish the spin-echo pulse sequence. One arm of the rotation laser's path is used to generate $\pi/2$ pulses, while the other arm generates $\pi$ pulses. Each arm is gated by an electro-optic modulator (EOM) which acts as pulse-picker to apply rotation pulses with a coarse spacing of an integer multiple of modelocked-laser repetition periods $T = nT_r$. The two EOMs used for pulse-picking were free space Conoptics modulators (model 100 and model 25-D). Each EOM had an extinction ratio of approximately 100 when used in single-pass configuration. In order to increase the extinction ratio of the EOMs to allow the setup to measure decoherence times as long as possible, the EOMs were each double-passed as shown in Figure 4.3. By adding an external mirror and polarizing beamsplitter, the extinction ratio of each EOM could be nearly squared to roughly $10^4$.

The coarse spacing between subsequent rotation pulses must be an integer multiple of modelocked-laser repetition periods $T = nT_r$. A computer-controlled stage allows a fine time offset $\tau$ between the train of $\pi/2$ and $\pi$-pulse rotations.

A fiber-based EOM (EOSpace AZ-6K5-10-PFU-PFUP-900-R4-S) with roughly $10^4$ extinction ratio was used to gate the optical pump laser into 26 ns duration pulses. The fiber-EOM's voltage bias was controlled by a modulator-bias controller (YY-Labs Mini-MBC-1) in order to maintain maximum extinction ratio. A typical
4.3. EXPERIMENTAL SETUP

Figure 4.2: Experimental setup. One arm of the rotation laser’s path generates $\pi/2$ pulses, the other arm generates $\pi$ pulses. QWP: quarterwave plate, PBS: polarizing beamsplitter, EOM: electro-optic modulator

Figure 4.3: EOM set up in double-pass configuration. PBS: polarizing beamsplitter, NPBS: non-polarizing beamsplitter
CHAPTER 4. SPIN ECHO

The rotation laser was chopped at 1 kHz by electronically gating the EOMs, and the single-photon counts were detected by a digital lock-in counter synchronized to this frequency. In order to exclude SPCM dark counts during the spin’s precession from the photon counting, the SPCM’s output was electrically gated. The 5 V positive-logic pulses from the SPCM were converted to fast (< 5 ns) -1 V negative logic pulses, then sent to a fast AND-gate. The other AND-gate input, as well as all three EOMs, were controlled by an electrical pulse-pattern generator that was clocked to the modelocked laser repetition frequency.

4.4 Sample Structure

The first generation of sample used for spin-echo experiments was the same 600 nm diameter mesa structure as used in the previous chapter on Coherent Single Qubit Control, with the SIL in place. The $T_2$ decoherence time was measured to be only $65 \pm 10$ ns for this particular QD. This short decoherence time might be attributed to charge- and spin-fluctuations of paramagnetic surface states on the etched mesa sidewalls.

Subsequent generations of sample structures attempted to increase the decoherence time by either passivating surface states or moving surfaces far from the QD layer. The second generation of spin-echo samples is shown in Figure 4.5. The sample contained about $2 \times 10^9$ cm$^{-2}$ self-assembled InAs QDs at the center of a GaAs pillar microcavity. A $\delta$-doping layer of roughly $4 \times 10^9$ cm$^{-2}$ Si donors was grown 10 nm below the QD layer to probabilistically dope the QDs. Approximately one-third of the QDs were charged, and these could be identified by their splitting into a symmetrical quadruplet at high magnetic field[44]. The lower and upper cavity mirrors contained 24 and 5 pairs of AlAs/GaAs $\lambda/4$ layers, respectively, giving the cavity a quality factor of roughly 200. The cavity increased the signal-to-noise ratio of the measurement in two ways: first, it increased collection efficiency by directing most of the QD emission towards the objective lens, and second, it reduced the laser power required to achieve optical pumping, thereby reducing reflected pump-laser
4.5. **RABI OSCILLATIONS AND RAMSEY FRINGES**

Noise. The SIL was not used with the microcavity samples because it would interfere with the resonance condition. The planar cavity was etched into pillars of 1-4 $\mu$m diameter. Roughly 100 nm of silicon nitride was deposited on the sample surface in an attempt to passivate surface states. Spin-echo measurements determined the $T_2$ decoherence time of this sample to be $660 \pm 70$ ns.

The third generation sample was used for the experiments presented in this chapter. The same planar structure as the second generation above, but was left as a planar microcavity in order to ensure no etched surfaces were near the QDs. A 100 nm thick aluminum shadow mask was deposited on top, and large 10 $\mu$m diameter windows were etched in the mask, to help navigation and position marking on the sample. Again, no SIL was used on the planar samples. QDs on this sample showed spin-echo $T_2$ times up to roughly 3 $\mu$s.

### 4.5 Rabi Oscillations and Ramsey Fringes

The improved experimental setup described in the previous section allows the optical pumping laser to be gated off when the rotation pulses are applied, which reduces
optical-pump induced decoherence during spin manipulation. The decoherence is even further improved by reducing the detuning $\Delta$ of the rotation laser below the excitonic transitions, from 270 GHz in the previous chapter to 150 GHz in this chapter.

In order to observe Rabi oscillations, a single rotation pulse is applied between the 26 ns optical pumping pulses, as demonstrated in Figure 4.6. The oscillations return much closer to 0 signal than those shown in the previous chapter.

The reduced rotation-induced decoherence and lack of optical pumping between rotations also greatly improved the visibility of Ramsey fringes as the time delay between a pair of $\pi/2$ pulses is varied, as shown in Figure 4.7. From these fringes we may estimate the fidelity of each $\pi/2$ rotation following the same method outlined in the previous chapter to be $F_{\pi/2} = 98\%$.

4.6 Spin Echo and $T_2^*$

The photon count rate following a spin-echo pulse sequence as the time offset $\tau$ is varied is shown in Figure 4.8a, for a total delay time of $2T = 264$ ns at magnetic field of $B_{\text{ext}} = 4$ T. For short time offsets, coherent sinusoidal fringes are observed.
Figure 4.6: Rabi oscillations in the photon count rate as the power of a single rotation pulse is varied.

Figure 4.7: Ramsey fringes as the time offset between a pair of $\pi/2$ pulses is varied.
Because our experimental apparatus lengthens the first time delay by $\tau$ while simultaneously shortening the second delay by $\tau$, we observe the spin-echo signal oscillating at twice the Larmor frequency with respect to $\tau$: $\cos[2\pi\delta_e(2\tau)]$. For longer time offsets however, $T_2^*$ dephasing becomes apparent as the phase of the fringes becomes incoherent and random due to the background nuclear field slowly fluctuating between one measurement point and the next. We are able to directly observe this phase randomization because our measurement time per data point ($\sim 2$ s) is shorter than the nuclear field’s fluctuation timescale. Note that this behavior is different from that observed by averaging over a spatial ensemble of spins or using slower time-averaging: in these cases, the data would resemble a damped sinusoid with decaying amplitude but well-defined phase. In order to quantify the dephasing time $T_2^*$, we perform a running Fourier transform on blocks of the data four Larmor-periods in length, and plot the amplitude of the appropriate Fourier component in Figure 4.8b. The data are slightly better fit by a Gaussian decay (green line) than a single exponential (red line). The timescale for the Gaussian dephasing $\propto \exp(-t^2/T_2^{*2})$ of $T_2^* = 1.71 \pm 0.08$ ns.

4.7 Spin Echo and $T_2$

In order to investigate $T_2$ decoherence, we vary the time delay $T$ of the spin-echo sequence and observe the coherent fringes for small time offset $\tau \ll T_2^*$. The count rate as a function of time offset $2\tau$, at a magnetic field of $B_{\text{ext}} = 4$ T and echo time delay $2T = 132$ ns, is shown in Figure 4.9a. The data for these short time offsets $\tau$ are well-fit by a sinusoid. Figure 4.9b shows that the amplitude of the spin-echo fringes for a delay time of $2T = 3.2$ $\mu$s are much smaller compared to the shorter delay. The spin-echo fringe amplitude versus total time $2T$ is shown in Figure 4.9c, and is well fit by a single exponential decay with decoherence time $T_2 = 2.6 \pm 0.3$ $\mu$s. This decoherence time is in close agreement with that measured for an ensemble of QD electron spins by optical spin-locking at high magnetic field[9].

The decoherence time $T_2$ is plotted as a function of magnetic field $B_{\text{ext}}$ in Figure 4.10. The coherence decay curves over a wide range of magnetic fields were well
4.7. SPIN ECHO AND \( T_2 \)

Figure 4.8: Experimental demonstration of spin echo and single spin dephasing. 

a, Spin-echo signal as the time offset \( 2\tau \) is varied, for a time delay of \( 2T = 264 \) ns and magnetic field \( B_{\text{ext}} = 4 \) T. Single-spin dephasing is evident at large time offset.

b, Decaying Fourier component of fringes. Red line is exponential fit, green line is Gaussian fit. One standard deviation confidence interval described in the text is determined by bootstrapping.
Figure 4.9: Measurement of $T_2$ using spin echo. **a**, Spin-echo signal as the time offset $2\tau$ is varied, for a time delay of $2T = 132$ ns. Magnetic field $B_{\text{ext}} = 4$ T. **b**, Spin-echo signal for a time delay of $2T = 3.1$ $\mu$s. **c**, Spin-echo fringe amplitude on a semilog plot versus time delay $2T$, showing a fit to an exponential decay. Error bars represent one standard deviation confidence intervals estimated by taking multiple measurements of the same delay curve.
Figure 4.10: Magnetic field dependence of $T_2$. Decoherence time $T_2$ at various magnetic fields $B_{\text{ext}}$. Error bars represent 1 standard deviation confidence intervals estimated from 3 independent measurements of the $B_{\text{ext}} = 4$ T experiment, combined with bootstrapped uncertainties from each coherence decay curve. Black dashed lines are guides to the eye which indicate an initial rising slope, and then saturation for high magnetic field. The inset shows the linear dependence of the Larmor precession frequency $\delta_e$ on the magnetic field. The slope uncertainty is determined by bootstrapping.
fit by single exponentials, but sizable error bars prevent us from conclusively excluding other decay profiles. The data show that $T_2$ increases with magnetic field at low fields $B_{\text{ext}} < 4$ T. $T_2$ appears to saturate at high magnetic field. Theory predicts that decoherence should be dominated by magnetic field fluctuations caused by random inhomogeneities of the nuclear magnetization inside the QD diffusing via the field-independent nuclear dipole-dipole interaction. Higher-order processes such as hyperfine-mediated nuclear spin interactions are not expected for spin-echo at these magnetic fields [38]. Consequently, the spin-echo decoherence time is predicted to be invariant with magnetic field, and to be in the range of $1 - 6 \, \mu$s for an InAs QD of our dimensions [38, 82, 88]. Our high-field results of $T_2 \sim 3 \, \mu$s for $B_{\text{ext}} \gtrsim 4$ T are consistent with these predictions, and also consistent with experimental results measured by spin-locking [9].

For low fields, $T_2$ may be limited by spin fluctuations in paramagnetic impurity states close to the QD, whose spin states become frozen out at high field. Our observation of shorter $T_2$ times for QDs close to etched surface interfaces is also consistent with this picture. The figure inset shows that the Larmor precession frequency $\delta_e$ increases linearly with magnetic field $B_{\text{ext}}$ as expected, with a slope corresponding to an electron g-factor $|g_e| = 0.442 \pm 0.002$. Using our optical manipulation techniques, an arbitrary single-qubit gate operation may be completed within one Larmor precession period: $T_{\text{gate}} = 1/\delta_e$ [11]. At the highest magnetic field of 10 T, the ratio of decoherence time to gate time is $T_2/T_{\text{gate}} = 150,000$, which is an order of magnitude longer than necessary for the topological surface code architecture.
Chapter 5

Nuclear Spin Pumping

Optically controlled quantum dots (QDs) are in many ways similar to atomic systems, and are therefore often regarded as strong candidates for solid-state quantum information processing. However, one key feature distinguishing QDs in group III-V semiconductors from atomic systems is the presence of a large nuclear-spin ensemble [89].

Nuclear spins cause adverse effects such as inhomogeneous broadening and non-Markovian decoherence processes. However, nuclear spins may play useful roles as well. Although methods to use QD nuclear spins directly as a quantum memory remain challenging due to the difficulty of achieving sufficiently high levels of nuclear polarization, nuclear spins may provide novel methods for the dynamic tuning and locking of electron spin resonances for electrons trapped in QDs.

Several examples of manipulating nuclei to improve electron spin coherence have recently been observed. In electrically-controlled double QDs, transition processes between electron singlet and triplet states allow the manipulation of interdot nuclear spin polarizations, improving coherent control [90, 91, 92]. In single QDs under microwave control, nuclear effects dynamically tune the electron spin resonance to the applied microwave frequency [93]. Tuning effects are also observed in two-color continuous-wave (CW) laser experiments, in which the appearance of coherent electronic effects such as population trapping are modified by nonlinear feedback processes with nuclear spins [94, 95]. Finally, nuclear spins have been shown to dynamically
CHAPTER 5. NUCLEAR SPIN PUMPING

bring ensembles of inhomogeneous QDs into spin-resonance with a train of ultrafast pulses [96, 97].

We now describe a related but different manifestation of the non-Markovian dynamics occurring between a single electron in a QD and the nuclear bath with which it interacts, with new possibilities for use in controlling nuclear effects. The effect occurs when measuring the familiar “free-induction decay” (FID) of a single spin in a single QD under pulsed control. The Larmor frequency of the electron spin is dynamically altered by the hyperfine interaction with QD nuclei; the nuclear polarization is in turn altered by the measurement results of the FID experiment. The result is a feedback loop in which the nuclear hyperfine field stabilizes to a value determined by the timing of the pulse sequence. In what follows, we show the experimental manifestation of this feedback loop and present a numerical model for the effect.

5.1 Experimental Setup

Figure 5.1(a) shows the Hahn spin-echo pulse sequence that was used in the previous Spin Echo chapter to measure the $T_2^*$ dephasing time of the single electron spin. By increasing the time offset $\tau$ and making the spin-echo sequence increasingly asymmetric, we may directly observe the dephasing effects caused by various nuclear spin configurations. However, one would also expect it should be possible to measure the $T_2^*$ dephasing time more directly by simply using the Ramsey pulse sequence (or FID) shown in Figure 5.1(b). In this chapter we will investigate the surprising behavior observed when the time delay $\tau$ between the pair of $\pi/2$ pulses shown in Figure 5.1(b) is varied to larger values than probed in the previous two chapters.

The sample and experimental setup are identical to those of the previous chapter on spin echo.

We perform an FID (Ramsey interferometer) experiment by applying the pulse sequence shown in Figure 5.2. After initialization into $|\uparrow\rangle$ by an optical pumping step, the spin is manipulated by a pair of $\pi/2$ rotations separated by a variable time delay $\tau$ before being measured by another optical pumping pulse. In between the two rotations, the spin freely precesses around the equator of the Bloch sphere at
its Larmor precession frequency $\delta_e$. In the absence of any electron-nuclear spin feedback mechanisms, the nuclear spins would be expected to fluctuate randomly on a timescale slow compared to the Larmor precession, leading to random Overhauser shifts of the electron’s Larmor frequency due to contact-hyperfine interaction. Repeated measurements of the same spin with different Larmor frequencies would lead to expected sinusoidal fringes with frequency $\delta_e$, with a Gaussian decay on a timescale $T_2^*$ of a few nanoseconds due to this nuclear-induced dephasing.
5.2 Experimental Results

However, such a Gaussian decay was not observed. Figure 5.3 shows the result of the FID experiment. The top three traces show the fringes seen as the delay $\tau$ is increased, and the bottom three correspond to decreasing $\tau$. The oscillatory fringes, rather than decaying, evolve into a sawtooth pattern at high values of $\tau$, and show hysteresis depending on the direction in which $\tau$ is scanned. Figure 5.3(c) illustrates the result of switching the scan direction. The signal initially traces back its same path in the reverse direction, then overshoots to the maximum signal level before continuing its sawtooth pattern in the opposite direction.

5.3 Modeling

Thaddeus Ladd, a former post-doctoral scholar in the Yamamoto group, developed a model to explain these results in collaboration with this thesis’ author. We now present the results of Thaddeus’ model.

These data result from two competing processes: changes in the average nuclear hyperfine (Overhauser) shift $\omega$ due to trion emission, in conjunction with the motion of that magnetization due to spin diffusion. In what follows, we first qualitatively describe these physical processes and explain how they lead to our data, and then we present equations to formally model the dynamics quantitatively.

One important assumption is a separation of dynamics into three very distinct timescales. The fastest timescale is the pulse sequence and resulting electron-spin dynamics, repeated continuously with a repetition period of 143 ns, shown in Figure 5.3(b). This is much faster than the nuclear dynamics we consider, which are presumed to occur on millisecond timescales. Finally, the averaging timescale of the measurement is much longer still, on the order of several seconds, allowing the nuclei substantial time to reach quasi-equilibrium.

Processes that change the total nuclear magnetization at the high magnetic fields used here (4 T) are unlikely to be due to the flip-flop terms of the contact hyperfine interaction of the ground-state electron in the QD, as its energy levels are known
Figure 5.3: (a) Experimental Ramsey fringe count-rate as a function of two-pulse time delay $\tau$. (b) Average electron polarization as a result of the periodic pulse sequence used to generate this data. Optical pumping increases the polarization for a duration $T = 26$ ns. The saturation polarization, is $S_{zf}^p$; in time $T$ only $S_{zf}^p$ is reached. After pumping and a short delay, a picosecond pulse indicated by a green arrow nearly instantaneously rotates the electron spin to the equator of the Bloch sphere ($\langle S_z \rangle = 0$); a time $\tau$ later a second pulse rotates the spin to achieve electron polarization $S_{zi}$, depending on the amount of Larmor precession between the pulses. The theoretical count-rate $C(\omega, \tau)$ of Eq. (5.1) is found as $S_{zi} - S_{zf}^p$ in steady-state conditions. (c) Experimental Ramsey fringe count-rate as $\tau$ is continuously scanned longer and then shorter, showing clear hysteresis.
to be narrow (on the order of $\hbar/T_2$, with $T_2 \sim 3 \mu s$) leaving few viable pathways for energy-conserving nuclear-spin flips. In contrast, the dipolar interaction between a trion’s unpaired hole and a nuclear spin may induce a spin-flip with the nuclear Zeeman energy compensated by the broad width of the emitted photon ($\gamma/2\pi \sim 0.1 \text{ GHz}$). Fermi’s golden rule allows an estimate of the rate at which a trion hole (at position $\vec{r}_h$, with gyromagnetic ratio $g_h$) polarized along the sample growth axis (orthogonal to the magnetic field) randomly flips a nuclear spin at position $\vec{r}$ in a spatially flat QD during spontaneous emission, with the photon energy density of states negligibly changed by the Zeeman energy of the nucleus. The result is

$$\Gamma(\vec{r}) \approx \left(9\mu_0^2/128\pi\right)\left(\mu_B g_h / B_0\right)^2 \gamma \langle |F - \vec{r}_h|^{-3}\rangle^2 \sim 1/(20 \text{ ms}),$$

where the brackets refer to an average over the hole wavefunction. Nuclear polarization due to this process has been considered before in the modeling of similar effects [97, 94].

### 5.3.1 Trion-driven Nuclear Spin Flips

A trion may cause nuclear spin flips to occur in either direction, leading to a random walk in magnetization. However, the rate of spin flips is proportional to the probability that a trion is created by the FID pulse sequence. The trion creation probability is plotted in the green and blue contours of Figure 5.4 as a function of pulse delay $\tau$ and Overhauser shift $\omega$, and is described by the equation

$$C(\omega, \tau) = S_p^2 \left[ 1 - \exp(-\beta(\omega) T) \right] \left[ 1 - \cos[(\omega_0 + \omega) \tau] \exp(-\beta(\omega) T) \right].$$

(5.1)

Here, $\omega_0$ is the electron Larmor frequency in the absence of nuclear shifts, $\omega$ is the Overhauser shift, $\beta(\omega)$ is the rate of optical pumping, $T$ is the pumping time, and $S_p$ is the saturation value of the polarized spin, equal to 1/2 for perfect pumping.

The trion creation probability $C(\omega, \tau)$ oscillates sinusoidally with increasing $\tau$ due to the spin’s Larmor precession and would lead to Ramsey fringes for fixed $\omega$: the Overhauser shift $\omega$ affects the frequency of Larmor precession. The trion creation probability drops to 0 for large values of $|\omega|$ because the trion transition shifts away from resonance with the optical pumping laser, leading to reduced pumping efficiency ($\beta(\omega) \to 0$) and trion creation. The optical pumping efficiency $\beta(\omega)$ is taken to be a
Figure 5.4: Count-rate $C(\omega, \tau)$ as a function of Overhauser shift $\omega$ and two-pulse delay $\tau$. The green areas indicate where a higher count-rate is expected. Oscillations in the horizontal directions at frequency $\omega_0 + \omega$ are due to Ramsey interference; the Gaussian envelope in the vertical direction is due to the reduction of optical pumping with detuning. The superimposed black line indicates stable points where $\partial \omega / \partial t = 0$ according to Eq. (5.4). Superimposed on this line are the solutions to this equation which result as $\tau$ is scanned longer (yellow) and shorter (white).

Gaussian corresponding to the optical absorption lineshape.

### 5.3.2 Nuclear Spin Relaxation

The second process which counters this drift is the presence of nuclear spin diffusion. When trion emission pushes $\omega$ to too large a value, nuclear dipolar interactions “flatten” the nuclear magnetization. As a result, the shift $\omega$ is “pulled” back to a low value, countering the tendency of trion emission to push $\omega$ away from zero. The stable quasi-equilibrium value of $\omega$ resulting from the balance of these processes lives
on the edge of the fringes shown in Figure 5.4; the nuclear polarization “surfs” along the edge of this function as $\tau$ is changed. As $\tau$ is increased, $|\omega|$ increases causing the observable photon count to decrease due to the reduced degree of optical pumping. When $|\omega|$ is so high that pumping is ineffective ($\beta(\omega) \to 0$) and the trion-induced walk stops, spin-diffusion causes the system to drift back to a new stable magnetization at a lower value of $|\omega|$, and the process continues.

5.3.3 Mathematical Model

These processes may be formally modeled by a diffusion equation for the nuclear distribution. In the model, the nuclear magnetization at each nuclear site $j$ is a random variable, $M_j$. A probability distribution function (pdf) $f(m_1, m_2, \ldots; t) = f(\vec{m}; t)$ gives the joint probability that the nuclear magnetization at each position is $M_j = m_j$ at time $t$. The Overhauser shift is then also a random variable $\Omega$, defined by $\Omega = \sum_j A(\vec{r}_j)M_j$, where $A(\vec{r}_j)$ is the electron hyperfine field at the position $\vec{r}_j$ of nucleus $j$. The average value of $\Omega$ at time $t$ is written $\langle \Omega \rangle = \omega(t)$ and is found by averaging over all possible values of each $M_j$, weighted by the joint pdf $f(\vec{m}; t)$. This joint pdf obeys the equation

$$\frac{\partial f}{\partial t} = \sum_j \left\{ -\sum_k D_{jk} \left( \frac{\partial f}{\partial m_k} - \frac{\partial f}{\partial m_j} \right) + \left[ F_j + \Gamma(\vec{r}_j)C(\Omega, t) \right] \frac{\partial^2 f}{\partial m_j^2} \right\}.$$  \hspace{1cm} (5.2)

The first term, in which the sum over $k$ is the sum over neighbors of $j$, describes the dissipative component of nuclear spin diffusion with diffusion rates $D_{jk}$. The second term describes the random walk of the magnetization at each location $\vec{r}$ due to stochastic nuclear spin-flips from the trion hole-spin; the constant $F_j$ models the fluctuating component of nuclear spin diffusion, a term needed to understand single QD $T_2^*$ effects in the absence of the nonlinearities we consider here.

Remarkably, the data of Figure 5.3 can be understood by examining just the average shift $\omega(t)$. This results in the equation

$$\frac{\partial \omega}{\partial t} = -\kappa \omega + \alpha \left\langle \frac{\partial^2}{\partial \Omega^2} [\Omega C(\Omega, t)] \right\rangle.$$  \hspace{1cm} (5.3)
5.3. MODELING

The constant $\kappa$ depends on the electronic wavefunction and the rate of nuclear diffusion, but we treat this parameter as adjustable rather than attempting a microscopic description. The constant $\alpha$ is formally given by $\sum_j \Gamma(\vec{r}_j) A^2(\vec{r}_j)$.

Unfortunately, Eq. (5.3) is not a closed system of equations, because it still requires full knowledge of $f(\vec{m}; t)$ to solve. However, if we assume $C(\Omega, \tau)$ to be a sufficiently flat function of $\Omega$ in comparison to the width of $f(\vec{m}; t)$, then we may treat $C(\Omega, \tau)$ as roughly constant at $C(\omega(t), \tau)$ over the small width of $f(\vec{m}; t)$.

$$\frac{\partial \omega}{\partial t} = -\kappa \omega + \alpha \frac{\partial^2}{\partial \omega^2}[\omega C(\omega, t)].$$  

(5.4)

Invoking our separation of timescales, we presume $\omega(t)$ evolves from its initial value (set by the last chosen value of $\tau$) to a quasi-equilibrium final value $\omega_f$. This final value determines the expected count rate $C(\omega_f, \tau)$ at this value of $\tau$. We solve by assuming $\omega(0) = 0$ at the first attempted value of $\tau$, and then we scan $\tau$ up and then down as in the experiment, finding the steady-state solution of Eq. (5.4) at each value.

5.3.4 Model Results and Comparison to Data

Figure 5.5 shows the modeled $C(\omega_f, \tau)$, $\omega_f$, and $\beta(\omega_f)$ as a function of $\tau$. This particular model used $\kappa/\alpha = 10^4$, which reproduces the qualitative shape of the data quite well, and quantitatively reproduces the location where sinusoidal fringes evolve into sawtooth-like fringes.

Qualitative differences are dominated by the random conditions that develop when the stage is moved on its rail, forming the breaks between data sets in Figure 5.3(a). Details of the shape of the waveform are related to the assumed form of the optical absorption. For simplicity, we have used $\beta(\omega) = \beta_0 \exp(-\omega^2/2\sigma^2)$, with $\sigma/2\pi = 1.6$ GHz and $\beta_0 = 3/T$ for known pumping time $T = 26$ ns, which roughly matches the experimentally observed count-rate when scanning the pump laser across the resonance. The real absorption shape is difficult to observe directly since hysteretic nuclear pumping effects also appear in absorption experiments with scanning CW lasers, as reported elsewhere [94, 95].
Figure 5.5: (Color online) The modeled (a) countrate or Ramsey amplitude $C(\omega_f, \tau)$, (b) Overhauser shift $\omega_f$, and (c) Optical pumping rate $\beta(\omega_f)$. The dotted line in (a) is the expected Ramsey fringe in the absence of nuclear effects. The traces in (b) are the same as those in Fig. 5.4. The blue (red) line corresponds to scanning $\tau$ longer (shorter).
5.4 Discussion

This effect may be useful for future coherent technologies employing QDs. This pulse sequence may serve as a “preparation step” for a qubit to be used in a quantum information processor, as it tunes the qubit to a master oscillator [55] and narrows the random nuclear distribution, assisting more complex coherent control [90, 91, 93, 92, 96, 97]. In particular, the ability to control a single electron with effectively δ-function-like rotation pulses introduces strong potential for dynamical decoupling [98, 85], but many schemes, especially those that compensate for pulse errors such as the Carr-Purcell-Meiboom-Gill (CPMG) sequence, require some method to tune the QD’s Larmor period to an appropriate division of the pulse-separation time.
Chapter 6

Strong Coupling in a Pillar Microcavity

Optical emitters coupled to cavities form the basic component of many quantum networking and communication proposals [7, 99, 100]. These schemes generally rely on generating entanglement between distant qubit nodes by interfering photons on beamsplitters that were emitted into well-defined cavity modes by identical but separate matter-spin qubits. Another scheme is proposed to directly interconvert between a stationary matter-spin qubit and a flying photonic qubit [38, 101]. By applying a carefully-designed optical pulse to a spin qubit in an optical cavity, a phase-coherent photon can be emitted or trapped, making a direct mapping between the spin-qubit and photon-qubit states. One common feature of nearly all quantum networking schemes is that the emitter must be well-coupled to an optical microcavity.

An optical emitter such as a QD may be placed inside of an optical microcavity to strengthen the emitter’s interaction with light. This may be understood qualitatively by considering that light may recirculate within a cavity and interact with the QD several times before leaking through the cavity mirrors. For a fixed cavity size, increasing the quality factor $Q$ of the cavity will increase the number of optical roundtrips in the cavity, thereby strengthening the QD-light interaction. Complementarily, for a fixed cavity $Q$, reducing the cavity size will result in higher optical fields for given optical energy, giving another means to strengthen QD-light interaction. In
order to maximize a QD’s interaction with light, it is therefore desirable to embed
the QD inside of a high-$Q$, low volume cavity.

In this chapter, we first introduce the theory of coupling between a QD exciton and
a microcavity. We will then experimentally probe such a system in the strong-coupling
regime, meaning that the QD exciton and cavity modes couple more strongly to each
other than to their environment. We analyze the photon statistics emitted from the
coupled QD-cavity system, and find a high degree of antibunching, which proves
that only a single QD is coupled to the cavity. Finally, we may also interpret our
experimental results as the first demonstration of a solid-state single-photon source
operating in the strong-coupling regime.

Throughout this chapter we will deal with neutral QDs, which in these circum-
stances can be viewed as simple two-level systems, rather than charged QDs. How-
ever, the results will naturally generalize and be applicable to electron-spin qubits in
charged QDs.

6.1 Theory of Quantum Dot-Microcavity Coupling

A schematic of a generic two-level quantum emitter inside of an optical microcavity
is shown in Figure 6.1. The two-level emitter is representative of a single atom, ion,
molecule, or in our case, a QD. The energy in the optical cavity mode leaks from the
cavity at a rate $\kappa$, while the QD exciton and the cavity mode exchange energy at a
rate $g$. The QD exciton may also radiatively decay into non-cavity modes (ie leaky,
free-space modes) at a rate $\gamma$.

The cavity decay rate $\kappa$ is related to the cavity’s quality factor $Q$ through $Q = \omega_c/\kappa$, where $\omega_c$ is the cavity’s resonance frequency. The cavity’s full-width at half-
max (FWHM) linewidth is also given by $\kappa$. The QD exciton’s decay rate into leaky
modes $\gamma$ may be modified by the presence of the cavity compared to a QD in bulk
semiconductor.

The coupling strength $g$ is given by [102]:
Figure 6.1: Schematic of a two-level emitter coupled to a microcavity.

\[ g = \sqrt{\frac{e^2}{4\epsilon_0\epsilon_m}} \sqrt{\frac{f}{V_m}} \]  

(6.1)

where \( e \) is the charge of an electron, \( \epsilon_0 \) is the permittivity of free space, \( \epsilon \) is the relative dielectric constant of the material, \( m_0 \) is the free-space mass of an electron, \( f \) is the QD exciton’s oscillator strength, and \( V_m \) is the mode volume of the cavity. \( V_m \) is calculated as [103]:

\[ V_m = \frac{\int_V \epsilon(\vec{r})|\vec{E}(\vec{r})|^2d^3\vec{r}}{\max[\epsilon(\vec{r})|\vec{E}(\vec{r})|^2]} \]  

(6.2)

where \( \vec{E}(\vec{r}) \) is the electric field distribution of the microcavity, which may be calculated by finite-difference time-domain (FDTD) simulation. If the dipole of the QD exciton is not spatially aligned with the maximum of the cavity mode’s electric field, then the coupling constant \( g \) will be reduced from the expression given in equation 6.1 by a factor of

\[ g = g_{\text{max}} \frac{\vec{E}(\vec{r}) \cdot \vec{\mu}}{|\vec{E}_{\text{max}}||\vec{\mu}|} \]  

(6.3)

where \( \vec{\mu} \) is the QD exciton’s dipole vector.
6.1.1 Energy Levels and Decay Rates

Next we calculate the energy levels and decay rates of the coupled QD-microcavity modes. The interaction of a two-level emitter and a harmonic oscillator such as a microcavity is governed by the Jaynes-Cummings Hamiltonian:

\[ H = \hbar \left[ \omega_x \sigma_z^2 + \omega_c a\dagger a + g(a\dagger \sigma_- + a\sigma_+) \right] \]  \hspace{1cm} (6.4)

where \( \omega_x \) and \( \omega_c \) are the resonant frequencies of the QD exciton and cavity, respectively, \( \sigma_z = |e\rangle \langle e| - |g\rangle \langle g| \) is the exciton population operator (\(|g\rangle \) and \(|e\rangle \) are the excitonic ground and excited states), \( a\dagger \) and \( a \) are the photon creation and annihilation operators, \( \sigma_- = |g\rangle \langle e| \) is the exciton lowering operator and \( \sigma_+ = |e\rangle \langle g| \) is the exciton raising operator.

The eigenstates of this Hamiltonian are an anharmonic ladder of states known as the Jaynes-Cummings ladder, which has been directly investigated using Rydberg atoms in a microwave cavity [104]. If the system is excited only weakly, then it will stay within the lowest manifold of states. Without coupling, the lowest two states may be written as \((|1\rangle_c |g\rangle , |0\rangle_c |e\rangle)\) where the first ket represents the number of photons in the cavity and the second ket gives the QD state. When we turn on the QD-cavity coupling, the lowest eigenstates may be solved using the aforementioned basis simply by diagonalizing the following Hamiltonian:

\[ H = \hbar \begin{pmatrix} \omega_c - i\frac{\kappa}{2} & g \\ g & \omega_x + i\frac{\gamma}{2} \end{pmatrix} \]  \hspace{1cm} (6.5)

where we have accounted for the lifetime of the QD exciton and cavity photon states by including their decay rates as an imaginary part of their resonance frequencies. The eigenvalues of this Hamiltonian are:

\[ \omega_{1,2} = \frac{\omega_c + \omega_x}{2} - i\frac{\kappa + \gamma}{4} \pm \sqrt{g^2 - \left(\frac{\kappa - \gamma - 2i\Delta}{4}\right)^2} \]  \hspace{1cm} (6.6)

where \( \Delta = \omega_x - \omega_c \) is the detuning between the exciton and cavity modes.

The real and imaginary parts of the eigenvalues from equation 6.6, corresponding
Figure 6.2: Energies and linewidths of the two coupled modes of a QD-microcavity system. QD: QD exciton mode, cav: cavity mode, LP: lower polariton, UP: upper polariton.

to the real energies and linewidths of the coupled modes, are plotted as a function of cavity quality factor $Q$ in Figure 6.2 for the case of zero detuning: $\Delta = 0$. The figure is plotted in term of energy $E = \hbar \omega$ in $\mu$eV. For QDs in semiconductor microcavities, the cavity decay rate is typically very fast compared to the QD exciton decay rate: $\kappa \gg \gamma$. The parameters chosen for the explanatory plot are: $\hbar g = 35 \ \mu$eV, $\hbar \gamma = 35 \ \mu$eV. Two distinct regions of the figure are observed: the regime to the left of the dashed line is known as the weak coupling regime, while the regime to the right is the strong coupling regime.
6.1. THEORY OF QUANTUM DOT-MICROCAVITY COUPLING

6.1.2 Weak Coupling Regime

In the weak coupling regime, the QD exciton mode (labeled QD) and cavity exciton mode (labeled cav) maintain their own natural resonance energies, but their linewidths or decay rates are modified by the coupling rate $g$. Of particular interest is the linear increase in the QD exciton’s decay rate, which can be observed in the lower left corner of Figure 6.2.

We may understand this increase in the QD exciton’s decay rate by expanding the square-root term in equation 6.6 to first order assuming $\kappa \gg \gamma, g$ and neglecting terms of order $\gamma^2$. The coupled decay rate of the QD exciton mode then becomes

$$
\gamma' = \gamma + \frac{4g^2}{\kappa}
= \gamma (1 + F_P)
$$

(6.7)

(6.8)

where we have defined the Purcell factor as

$$
F_P = \frac{4g^2}{\kappa\gamma}
$$

(6.9)

The regime in which $g^2 \gg \kappa\gamma$ is often referred to as the Purcell regime, where the QD exciton’s decay rate is greatly enhanced by its coupling to the cavity mode. We may also rewrite the Purcell factor in terms of only parameters of the cavity [105]:

$$
F_P = \frac{3}{4\pi^2} \left( \frac{\lambda_c}{n} \right)^3 \frac{Q}{V_m}
$$

(6.10)

where $\lambda_c$ is the cavity’s resonance wavelength and $n$ is the cavity’s index of refraction at the location of the QD. In order to enter the Purcell regime of weak coupling, it is therefore necessary to maximize the cavity’s ratio of $Q/V_m$: we want a small cavity with a large quality factor.
6.1.3 Strong Coupling Regime

As the cavity quality factor $Q$ is increased past a certain threshold, a distinctive change in the eigenstates is observed in Figure 6.2. This threshold occurs when

$$g > \frac{\kappa - \gamma}{4}$$  \hspace{1cm} (6.11)

or, taking the approximation that $\kappa \gg \gamma$,

$$g > \frac{\kappa}{4}$$  \hspace{1cm} (6.12)

This is the condition for the onset of the strong coupling regime. In the strong coupling regime, the cavity and QD exciton coherently exchange energy back and forth at a rate called the vacuum Rabi frequency $\Omega_R$:

$$\Omega_R = 2\sqrt{g^2 - \frac{(\kappa - \gamma)^2}{16}}$$  \hspace{1cm} (6.13)

Thus the system is better described by a pair of new eigenstates, each of which is half-cavity-photon and half-QD-exciton at resonance, called the upper polariton (UP) and lower polariton (LP). These two new eigenstates are split in frequency by $\Omega_R$, as shown in Figure 6.2. If the detuning $\Delta$ between the QD exciton and cavity photon is varied, the two modes will anti-cross with each other, with a minimum separation given by $\Omega_R$.

In order to reach the strong coupling regime, it is necessary to maximize the ratio of $g/\kappa$. By combining the definition of $Q = \omega_c/\kappa$ with equation 6.1, we find that strong coupling depends on maximizing the ratio of $\sqrt{f\frac{Q}{\sqrt{V_m}}}$: we again want cavities with small mode volumes and high quality factors, and also QDs with large oscillator strengths.
6.2 Semiconductor Microcavities

At the time of writing, three main types of semiconductor microcavities are most commonly used to confine light to small volumes: the planar photonic crystal cavity, the microdisk cavity, and the pillar microcavity. QDs may be monolithically integrated inside of all three of these cavity designs, allowing strong coupling between the confined light and QD exciton.

6.2.1 Photonic Crystal Cavities

A photonic crystal consists of a periodic arrangement of optical materials with different refractive indexes [106]. Coherent interference on reflection from the various interfaces creates an optical band-gap, whereby certain frequencies of light are unable to propagate in certain directions within the crystal. A popular geometry is to etch a two-dimensional triangular array of holes into a thin slab of semiconductor. By leaving out some of the holes (commonly 3 holes in a line), a defect cavity may be created. Light is trapped in the cavity in the plane of the slab by reflection from the band-gap structure, and is confined in the direction normal to the slab by total internal reflection.

The mode volume of such a so-called ‘L3’ photonic crystal cavity is often as small as $\sim \frac{1}{2} \left( \frac{\lambda}{n} \right)^3$ [103, 107]. The light emission pattern can be designed through careful engineering to be normal to the slab [103, 108], increasing optical collection efficiency into a microscope objective. Alternatively, the cavity may be coupled to a photonic crystal waveguide fabricated on the same sample, allowing for integrated devices.

6.2.2 Microdisk Cavities

A microdisk cavity also consists of a thin slab of semiconductor material, but it is etched into a circle [109]. Light traveling around the edge of the disk experiences repeated total internal reflections from the smooth outer edge of the disk, leading to the formation of so-called whispering gallery modes which concentrate light near the disk’s perimeter. Mode volumes on the order of $\sim 5 \left( \frac{\lambda}{n} \right)^3$ are commonly achieved.
The cavity emission of a free-standing microdisk is uniformly distributed around the
perimeter of the disk, and mostly in the plane of the slab, making collection using a
objective lens difficult. However, the disk may be coupled to a tapered fiber [110] or
integrated waveguide [111] to more efficiently extract light.

6.2.3 Pillar Microcavities

Pillar microcavities confine light in the vertical direction by reflection between a
pair of distributed Bragg reflector (DBR) mirrors, essentially creating a Fabry-Perot
cavity. These are stacks of alternating layers of two different semiconductors with
contrast indexes of refraction, with each layer generally being one-quarter wave-
length of light in that material. Such a stack leads to coherent reflections from the
layers, and is sometimes called a one-dimensional photonic crystal. The DBR mirror
reflectivity is limited by the number of mirror pairs, the index contrast between the
layers, and the quality of growth.

The planar cavity is etched into pillars, which confine light in the horizontal di-
rection by total internal reflection. The mode volume of a pillar cavity may be on the
order of $\sim 10 \left( \frac{\lambda}{n} \right)^3$ [112]. The pillar cavity may be designed to have efficient emission
in the vertical direction with a clean gaussian mode shape. Their use in quantum
information processing in the long-term may be limited by the difficulty of coupling
them to an integrated waveguide structure. However, demonstrated collection effi-
ciencies into objective lenses in excess of 22% have been demonstrated[113], making
pillar cavities ideal for performing free-space quantum information experiments in the
short-term.

6.3 History of Strong Coupling

Strong coupling between a single atom and a cavity was first achieved more than
a decade ago [114]. More recently, several groups have achieved strong coupling
between a single (In,Ga)As QD and either micropillar [112], photonic crystal [107],
or microdisk [109] resonators. Strong coupling can also occur between a single cavity
mode and a collection of degenerate emitters, such as an ensemble of atoms or a quantum well [115]. However, in the latter case the behavior is classical: adding or removing one emitter or one photon from the system has little effect.

In the initial studies of QD-cavity strong coupling [112, 107, 109] it was argued that the spectral density of QDs was sufficiently low that it is unlikely that several degenerate emitters contributed to the anticrossing. However, it was not verified that the system had one and only one emitter. There was a surprisingly large amount of emission from the cavity mode when the QD was far detuned. It was unclear whether this emission originated from the particular single QD or from many background emitters. It was therefore important to verify that the double-peaked spectrum originates from a single quantum emitter, not a collection of emitters, interacting with the cavity mode.

In the remainder of this chapter we show that the emission from a strongly-coupled QD-microcavity system is dominated by a single quantum emitter. Photons emitted from the coupled QD-microcavity system at resonance showed a high degree of antibunching. Away from resonance, emission from the QD and cavity modes was anticorrelated, and the individual emission lines were antibunched. The key to these observations was to resonantly pump the selected QD via an excited QD state to prevent background emitters from being excited. These background emitters, which are usually excited by an above-band pump, prevent the observation of antibunching by emitting photons directly into the cavity mode and by repeatedly exciting the QD after a single laser pulse. With pulsed resonant excitation, the device demonstrates the first solid-state single photon source operating in the strong coupling regime. The Purcell factor exceeds 60 and implies very high quantum efficiency, making such a device interesting for quantum information applications.

6.4 Pillar Microcavity Sample Design

Planar cavities were grown with DBR mirrors consisting of 26 and 30 pairs of AlAs/GaAs layers above and below a one-wavelength-thick GaAs cavity. A layer of InGaAs QDs, with an indium content of about 40% and a density of $10^{10} \text{ cm}^{-2}$, was grown in the
Figure 6.3: (a) Scanning electron micrograph of a sample of uncapped InGaAs quantum dots. (b) Scanning electron micrograph of a 1.8 µm diameter pillar microcavity.

central antinode of the cavity. A scanning electron micrograph of typical uncapped InGaAs QDs is shown in Fig. 6.3a. The QDs in this sample typically showed splittings between the s-shell and p-shell transition energies of $25 - 30$ meV, suggesting lateral QD dimensions of $20 - 30$ nm [116]. The cavities were etched into circular micropillars with diameters varying from 1 to 4 µm. An electron microscope image of a 1.8 µm diameter micropillar is shown in Fig. 6.3b. Further details on fabrication can be found in Ref. [117].

6.5 Micropillar PL: above-band versus resonant excitation

Photoluminescence measurements were performed while the sample was cooled to cryogenic temperatures. Increasing the sample temperature caused the QD excitons to red-shift faster than the cavity mode, allowing the QDs to be tuned by nearly 1.5 nm relative to the cavity between 6 K and 40 K. The sample was optically pumped by a tunable continuous wave (CW) or mode-locked pulse Ti:sapphire laser, focused
6.5. **MICROPILLAR PL: ABOVE-BAND VERSUS RESONANT EXCITATION**

Figure 6.4: Experimental setup for micro-photoluminescence and photon correlation measurements. NPBS: non-polarizing beamsplitter, BS: beamsplitter, LPF: long-pass filter, ML: modelocked, SPCM: single-photon counting module.

to a 2 μm spot through a 0.75 NA objective. PL was detected by a 750mm grating spectrometer with N₂-cooled CCD (spectral resolution 0.03 nm). The experimental setup is shown in Fig. 6.4. The PL may be also be directed towards an intensity correlation setup (described later) by moving a flip mirror.

In the simplest picture, above-band pumping creates electron-hole pairs that can radiatively recombine to emit photons at the QDs’ quantized energy levels. The cavity should be nearly dark if no QD level is resonant with it. However, in previous studies of QD SC the cavity emission was much brighter than the QD emission even when no QD was resonant with the cavity [112, 107, 109]. It was unclear whether the cavity emission resulted from coupling to the specific QD involved in SC, or to a broad background of emitters such as spectrally far-detuned QDs and wetting layer states. These background emitters might contribute to the cavity emission by simultaneously...
Figure 6.5: Above-band pumping compared to resonant pumping of a chosen QD in Pillar 1. With above-band pump (725 nm, 0.4 µW), the chosen QD exciton (X) emits, but so do the cavity (C) and many other QDs. With 937.1 nm (3 µW) pump, the chosen QD is selectively excited and its PL dominates an otherwise nearly flat spectrum.

emitting a cavity photon and one or more phonons.

In order to eliminate any background emitters, the laser can be tuned to resonantly pump the excited state (p-shell) exciton in a selected QD [118]. The exciton quickly thermalizes to the QD ground state (s-shell) where it can interact with the cavity. Ideally, resonant pumping creates excitons only in the selected QD, eliminating all extraneous emitters coupled to the cavity.

The PL spectrum of a typical weak-coupling device called Pillar 1, excited by CW above-band pumping, is shown in the lowest trace in Fig. 6.5. The cavity mode ($Q = 17300$) could be identified amongst the various QD lines by its broader linewidth, slower tuning with respect to temperature, and lack of saturation at high pump powers. The cavity emits strongly even though there is no QD resonant. The higher traces in Fig. 1(b) show how tuning the pump laser towards an excited state in a chosen QD (937.1 nm in this case) can selectively excite the QD with greatly reduced background cavity emission. Resonant pumping suppresses the cavity emission relative to the QD emission by roughly a factor of ten in this particular pillar. The resonant pump was
nearly ten times as intense as the above-band pump to achieve the same PL intensity, which caused local heating and lead to a slight red shift (0.01 – 0.03 nm) of the QD line.

6.6 Strong coupling PL

The temperature dependent PL for a device exhibiting strong coupling called Pillar 2 is presented in Fig. 6.6. A clear anticrossing of the QD line and the cavity mode at resonance is evident. When the device was pumped above-band (725 nm), the cavity was significantly brighter than the QD and many QDs lines were visible. Resonant pumping of the particular QD involved in SC eliminated the other QD lines and reduced the cavity background emission. The vacuum Rabi splitting at resonance is more pronounced with resonant pumping, possibly because the above-band pump creates background excitons and trapped charges that interact with the QD exciton to broaden its emission.

The line centers and linewidths of the resonantly-pumped QD-cavity system (Fig. 6.6b) are shown in Fig. 3. For the lowest temperatures the lower line is narrower and exciton-like, and the upper line is broader and cavity-like. Increasing the temperature causes the lines to switch character as they anticross. From Fig. 3 we determine the cavity linewidth of Pillar 2 is $\kappa = 85\ \mu eV$ ($Q = 15200$) and the vacuum Rabi splitting is $56\ \mu eV$. Using formula (1) we calculate $g = 35\ \mu eV$. This gives a ratio of $g/\kappa = 0.41 > \frac{1}{4}$ as required to satisfy the strong coupling condition. Fits to the above-band pumped spectra yield a similar value for $\kappa$, and slightly smaller values for the vacuum Rabi splitting (50 $\mu eV$) and $g$ (33 $\mu eV$).

6.7 Photon Statistics

To verify the quantum nature of the system and determine whether a single emitter is responsible for the photon emission, we measured the photon intensity autocorrelation function:
Figure 6.6: Temperature dependent PL from Pillar 2 with (a) above-band CW pump (725 nm), and (b) resonant CW pump (936.25 – 936.45 nm). Each spectrum is rescaled to a constant maximum since tuning the QD changes excitation efficiency. Resonance occurred at lower temperature for resonant pump case (10.5 K vs. 12 K) due to local heating.

Figure 6.7: Emission wavelength and FWHM of upper (circles) and lower (squares) lines as a function of temperature, based on double-Lorentzian fits to resonantly-excited spectra of Pillar 2 (Fig. 6.6b).
\[ g^{(2)}(\tau) = \frac{\langle : I(t)I(t + \tau) : \rangle}{\langle I(t) \rangle^2} = \frac{\langle a^\dagger(t)a^\dagger(t + \tau)a(t + \tau)a(t) \rangle}{\langle a^\dagger a \rangle^2} \]  

(6.14)

(6.15)

where \( a^\dagger \) and \( a \) are the photon creation and annihilation operators, \( I = a^\dagger a \) is the intensity operator, and : denote normal ordering dots. For single photons, the intensity autocorrelation function evaluated at zero time delay should be zero: \( g^{2}(0) = 0 \) indicating there is no probability of detecting two photons simultaneously.

A single quantum emitter, such as an atom, molecule, or quantum dot, can only emit one photon at a time and should in principle exhibit \( g^{2}(0) = 0 \). In practice however, a finite probability for emitting two photons in close succession always exists, and a threshold is chosen to be \( g^{2}(0) = 0.5 \) as the definition of a system that is dominated by a single quantum emitter.

### 6.7.1 Photon Correlation Setup

The intensity correlation function can be measured by a Hanbury-Brown and Twiss (HBT) setup containing a beamsplitter and two detectors. The setup is shown above in Fig. 6.4. A grating disperses the PL according to its frequency. A lens focusses the PL onto two multi-mode fibers in two fiber couplers via a non-polarizing beamsplitter. The 50\( \mu \)m core of each fiber acts as an input slit of a monochromator, allowing out setup to act as two independent monochromators each with \( \sim 0.2 \) nm resolution. Each fiber coupler may be moved independently, allowing intensity autocorrelation measurements if both fibers are spatially aligned to the same frequency of PL or cross-correlation measurements if the fibers are aligned to different frequencies. Each fiber is connected to a single-photon counting module (SPCM) with \( \sim 1 \) ns timing resolution. The correlation function \( g^{2}(\tau) \) is formed by creating a histogram of the time delays between subsequent detection events on the two SPCMs.

With weak excitation, the width of the dip in \( g^{(2)}(\tau) \) near \( \tau = 0 \) is given by the lifetime of the emitter, which is roughly 15 ps (i.e. twice the cavity lifetime) for
the resonantly-coupled QD-cavity system. The emitter’s extremely fast decay rate necessitates a pulsed excitation scheme using a modelocked laser since the SPCMs cannot resolve such a short time scale.

### 6.7.2 Autocorrelation Results

The autocorrelation function of photons collected from the coupled QD-cavity system at resonance when excited by a pulsed resonant laser is shown in Fig. 6.8. The periodic peaks reflect the 13 ns periodicity of the pulsed laser, while antibunching in the photon statistics is evidenced by the suppressed area of the peak at zero time delay. The observed value of $g^{(2)}_{\text{rr}}(0) = 0.19 < \frac{1}{2}$ proves that the emission from the coupled QD-cavity is dominated by the single QD emitter. Increasing pump power yielded higher values for $g^{(2)}(0)$ as the QD saturated but the cavity emission continued to rise.

When another strongly-coupled micropillar (Pillar 3) was pumped with above-band pulses, $g^{(2)}_{\text{rr}}(0)$ of the resonantly-coupled QD-cavity system remained between 0.85 and 1 even for the lowest pump powers as shown in Fig. 6.9. Antibunching could not be observed with an above-band pump for two reasons. First, the above-band pump creates many background emitters that couple to the cavity mode. Second,
6.7. PHOTON STATISTICS

the free excitons created by the pump have lifetimes much longer than the coupled QD-cavity lifetime, allowing multiple capture and emission processes after a single laser pulse. Resonant pumping solves both of these problems.

6.7.3 Cross-Correlation Results

Next the QD in Pillar 2 was red detuned by 0.4 nm from the cavity mode so that photon statistics could be collected from the cavity and QD emission lines separately. Surprisingly, even with the resonant pump tuned to selectively excite the chosen QD, the cavity emission was \( \sim 3.5 \) times brighter than the QD (see Fig. 6.10). (Note that with above-band pumping, background emitters were excited and the cavity emission grew another five times brighter relative to the QD). The QD emission was antibunched as expected with \( g^{(2)}_{xx}(0) = 0.19 \) (Fig. 6.11(a)). Interestingly, the cavity emission was also antibunched with \( g^{(2)}_{cc}(0) = 0.39 < \frac{1}{2} \) (Fig. 6.11(b)), showing that the cavity emission is dominated by a single quantum emitter. This slightly higher value of \( g^{(2)}(0) \) suggests that some background emitters were still weakly excited and contribute to the cavity emission. Finally, the cross-correlation function between the QD exciton and cavity emission \( g^{(2)}_{xc}(\tau) \) was measured (Fig. 6.11(c)). Strong
antibunching was observed with $g_{x,c}^{(2)}(0) = 0.22$, conclusively proving that the single QD emitter is responsible for both peaks in the PL spectrum.

The bright cavity emission cannot be explained by radiative coupling to the QD due to their large detuning. This suggests that another, unidentified mechanism couples QD excitations into the cavity mode when off-resonance. At the relatively small detunings investigated in this work, it is plausible that this coupling could be mediated by the absorption or emission of thermally-populated acoustic phonons [119, 120].

Similar off-resonant cavity-QD coupling has been reported by the Imamoglu group as well, but at larger detunings of up to 18 nm [121]. At such large detunings, phonons are thermally unpopulated and cannot be responsible for the coupling. These same authors later claimed that the coupling was due to transitions between a quasi-continuum of excited multi-excitonic states in the QD [122]. Other researchers, however, claim that considering pure-dephasing of the QD alone is enough to explain the off-resonant coupling [123].
Figure 6.11: Correlation functions of the detuned QD-cavity system. (a) Autocorrelation function of QD emission only, $g_{x,x}^{(2)}(0) = 0.19$. (b) Autocorrelation function of cavity emission only, $g_{c,c}^{(2)}(0) = 0.39$. (c) Cross-correlation function of QD and cavity, $g_{x,c}^{(2)}(0) = 0.22$. 
6.8 Single Photon Source

Under pulsed resonant excitation at the resonance temperature, Pillar 2 emits a pulse train of photons, demonstrating the first solid-state single-photon source (SPS) operating in the strong coupling regime. A useful figure of merit for a SPS is the Purcell factor $F_P$. As previously explained, in the weak coupling limit, $F_P$ gives the enhancement of the QD’s emission rate $\gamma$ due to the cavity: $\gamma' = (1 + F_P)\gamma$. This relation no longer holds in the strong coupling regime, where the decay rates of the coupled QD-cavity states are fixed at $(\gamma_c + \gamma_x)/2$. We define the Purcell factor more generally as $F_P = \frac{4g^2}{\kappa\gamma}$ (also called the cooperativity parameter in atomic physics), where $\gamma$ is the QD’s emission rate in the limit of large detuning from the cavity. This Purcell factor is often used to quantify the performance of CQED-based quantum information processing schemes [124, 101], and is related to the quantum efficiency of the resonantly-coupled SPS [125]:

$$\eta = \frac{F_P}{1 + F_P} \frac{\kappa}{\kappa + \gamma}$$  \hspace{1cm} (6.16)

The efficiency $\eta$ gives the probability that a photon will be emitted into the cavity mode given that the QD is initially excited. We measured the QD lifetime to be $620 \pm 70$ ps when the QD was detuned by 0.7 nm from the cavity mode, as shown in Fig. 6.12. At this moderate detuning, the QD’s emission rate was slightly enhanced from $\gamma$ by coupling to the cavity. We may calculate the decay rate $\gamma = 1/\tau_x$ from formula the eigen-energy formula of the previous chapter using $\gamma_{1,2}(\Delta) = 2\text{Im}\{E_{1,2}\}$. From this expression and the measured lifetime, we determine the QD’s lifetime in the large detuning limit to be $\tau_x = 700 \pm 80$ ps. This lifetime agrees with measurements of bulk QDs showing an ensemble lifetime of 600 ps when we consider that a pillar microcavity may quench the emission rate of a far-detuned QD by roughly 10% [126]. Using $\tau_x$ we determine a Purcell factor of $61 \pm 7$ and quantum efficiency of $97.3 \pm 0.4\%$.

The high quantum efficiency and short single-photon pulse duration make this device directly applicable to high speed quantum cryptography. However, the incoherent nature of the resonant pump likely results in moderate photon indistinguishability of
Figure 6.12: Lifetime measurement of QD only, detuned 0.7 nm from cavity.

around 50%. Indistinguishability could be improved using a coherent pump scheme, such as one involving a cavity-assisted spin flip Raman transition [124, 127, 101], to make the device ideal for quantum information processing with single photons.
Chapter 7

Conclusions and Future Directions

7.1 Current Status

We summarize the work presented in this thesis by revisiting our checklist that must be demonstrated in a qubit system. We remind the reader that our goal is to achieve \( \sim 99.9\% \) fidelity operations and construct \( \sim 10^8 \) physical qubits. Note that although these particular targets are based on one quantum computing architecture, the 2-D topological surface code quantum computer, the experimental work presented in this thesis generally architecture-agnostic, and would work equally well with another computer architecture.

Qubits must be implemented in a scalable physical system

We have implemented our qubit using the two spin-states of a single electron trapped in a semiconductor QD, as described in Chapters 2-3. The size of the QD is a few 10s of nanometers in diameter, and a few nanometers in height. In principle, one can imagine using semiconductor microfabrication techniques to scale the single QD system into an integrated quantum computer containing millions of qubits. However, it will be necessary to replace the current self-assembled QDs with a different technique to better control their position and emission energy, as will be discussed in the future work section.
7.1. CURRENT STATUS

Individual qubits must be initialized into a pure state

In Chapter 3, we demonstrated the use of optical pumping to initialize our electron-spin qubit into a pure spin state with 92% fidelity within 13 ns.

Individual qubits must be measured

We measure the spin state by detecting the single photon that is emitted with the QD is optically pumped. A click on our single-photon counter tells us that the spin was flipped since the previous initialization. Because of the limited collection efficiency, we must repeat the experiment $\sim 10^4$ times to determine the state of the spin.

Single-qubit gates must be demonstrated

We demonstrated an arbitrary single-qubit gate in Chapter 3 using a pair of rotation pulses separated by a variable time delay. Each pulse rotates the spin around the Bloch sphere’s x-axis, while the time delay provides a z-axis rotation. By combining these three rotations, an arbitrary SU(2) rotation can be accomplished. The total time to accomplish an arbitrary gate is at most 40 ps, which is one Larmor period of the electron spin. The gate fidelity is as high as 98% (see Chapter 4), and can potentially be increased by further reducing the detuning of the rotation pulse from the excitonic transitions.

Two-qubit gates must be demonstrated

We have not yet discussed the implementation of a two-qubit gate. This remains the single largest open challenge facing our system.

The qubit must have a long decoherence time

In Chapter 4 we used an optical spin-echo sequence to extend the qubit’s decoherence time from $T_2^* \sim 2$ ns to $T_2^* \sim 3$ $\mu$s. The decoherence time is roughly constant for applied magnetic fields greater than about 4 T, but decreases at low magnetic field. At high field it is likely that nuclear spin diffusion limits the decoherence time, while
at low fields it is possible that spin fluctuations in paramagnetic impurity states near the QD become ‘unfrozen’ and lead to faster decoherence. We also showed in Chapter 5 how attempting to measure $T_2^*$ with a usual Ramsey interferometer led to the discovery of an electron-nuclear spin feedback mechanism, and presented a numerical model for this feedback.

**The qubit should interface with a ‘flying’ photonic qubit**

Coupling a single quantum emitter to a cavity is the first required step for many quantum communication and entanglement schemes [7, 99, 38]. We discussed strong coupling between a single QD exciton and a pillar microcavity in Chapter 6. An anti-crossing between the exciton and cavity modes demonstrated that our device was in the strong coupling regime. We showed that the background emission from the cavity mode can be suppressed by exciting the QD with a resonant rather than above-bandgap pump laser. The remaining cavity emission was proven to originate from the single QD by observing antibunched photon statistics. These results may also be interpreted as the first demonstration of a solid-state single-photon source operating in the strong coupling regime.

### 7.2 Future Work

#### 7.2.1 Site-controlled Quantum Dots

Self-assembled QDs have provided a superb test-bed for our optically-controlled single-qubit experiments. However, self-assembled QDs’ characteristics of random location and emission energy will make it impossible to scale to devices containing $10^8$ QD qubits. Even for work involving two or more qubits, the ability to precisely control the position and emission wavelength of the QDs becomes essential.

One promising route towards controlling the position and emission energy of QDs is using optically-active gate defined QDs. Confinement for a single electron spin, and an additional electron-hole pair, could be provided in the semiconductor growth direction by a double quantum well (QW), while in-plane confinement could be provided
7.2. FUTURE WORK

Figure 7.1: One possible design for an optically-active gate-defined QD. A top metal gate electrode is suspended away from the QWs by SiO$_2$, except above the QD region. The electric field between the top gate and bottom n-doped mirror is stronger than the surrounding region, which leads to an increased quantum-confined Stark effect that traps electrons and trions.

by the quantum-confined Stark effect induced by gate electrodes (see Figure 7.1). This design could allow for highly reproducible qubits with precisely positioned QDs, deterministic loading of an electron spin from a nearby Fermi sea, and wavelength tuneability by the DC electric field. The QWs could be integrated into a planar microcavity consisting of a pair of distributed Bragg reflector mirrors. The presence of the QW outside of the QD regions could also prove beneficial by allowing the virtual excitation of exciton-polaritons for two-qubit gate operations, as will be discussed later.

7.2.2 Single-shot Qubit Measurement

Our current qubit measurement scheme, based on optical pumping and single-photon counting, requires many experimental cycles to complete. However, most quantum computing architectures, including the topological surface code, require a single-shot measurement scheme with high fidelity. We hope to implement a fast single-shot quantum non-demolition (QND) measurement scheme based on a dispersive phase shift measurement of the QD electron spin state. A $\sim$ 1 ns optical probe pulse,
detuned below an excitonic transition, would receive a spin-dependent phase shift on reflection from a high-Q ($Q \sim 10000$) pillar microcavity containing the QD. The probe pulse’s phase shift is then measured by a polarization interferometer [49].

### 7.2.3 Further Extension of Decoherence Time by Dynamical Decoupling

As mentioned briefly in Chapter 4, the $T_2$ decoherence time of any qubit may in principle be extended by a magnetic resonance technique called dynamical decoupling (DD). The first DD scheme, CPMG, was demonstrated in the 1950s. After an initial $\pi/2$ pulse about the x-axis, a period sequence of $\pi$ pulses are applied to flip the spin about the y-axis [128, 129], as shown in Figure 7.2. The coherence is refocussed after each $\pi$ pulse, and if the refocussing pulses are applied faster than the decoherence, then $T_2$ is extended.

The sequence of refocussing $\pi$ pulses may be viewed as a function that filters the decoherence noise. The spacing between refocussing pulses may be modified to tailor the filter function to the particular noise spectrum experienced by the qubit. One example is the UDD refocussing sequence [84], which is analytically derived to suppress errors that occur on short timescales. These DD sequences, as well as empirically-optimized refocusing sequences, have recently been implemented in trapped ion qubits [86] and should be achievable in our QD spin system.
7.2. FUTURE WORK

7.2.4 Two-qubit Gate

As mentioned, the two-qubit gate is a necessary and challenging next step in pursuing a QD-spin based quantum computer. We have recently proposed an optical two-qubit gate for spins in QDs embedded in planar microcavity [130]. Two neighboring QDs (separated by \( \sim 1 \mu m \)) are coupled by an optical pulse driving a common planar microcavity mode, resulting in a spin-state dependent geometric phase rotation. However, the fidelity of this gate may be limited far below the required 99.9%.

Another scheme worth investigating is based on the virtual excitation of exciton-polaritons, which is analogous to the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. A planar microcavity encloses not only the gate-defined QDs but also two unperturbed QWs beside the QDs (see Figure 7.1). When a delocalized polariton mode is optically excited in the QW-microcavity system, the two QD spin qubits interact through an indirect exchange coupling mediated by the electron-spin component of the polariton, potentially allowing a higher fidelity two-qubit gate to be accomplished.

In conclusion, we note that numerous daunting yet fascinating challenges remain along the roadmap to constructing a semiconductor-spin based quantum computer. We hope that the work presented here demonstrates only a small part of what will be achieved in the future with optically-controlled quantum dot spins.
Appendix A

Qubit Rotation Details

A.1 Bloch Vector Trajectory Reconstruction

In order to reconstruct the trajectory of the Bloch vector as a parametric function of rotation pulse power \( p \), we assume that each pulse-induced rotation may be modeled as two manipulations on the Bloch vector of sub-unity length \( L_0 \): first a power-dependent, uniform shrinkage of the Bloch vector \( D(p) \), and second a rotation matrix \( R(\Theta, \theta) \). The rotation is defined to extend through a power-dependent angle \( \Theta(p) \) around an axis with power-dependent polar angle \( \theta(p) \), as shown in Figure A.1. The azimuthal angle of the rotation axis is tantamount to an overall phase or choice of reference frame, and so we set this angle to zero by convention. The rotation \( R(\Theta, \theta) \) is caused by both the rotation pulse and Larmor precession during the pulse’s finite duration. The combination of these contributions to the rotation give the expressions

\[
\Theta^2(p) = [\Omega^2(p) + 2\Omega(p)\delta_e \cos \xi(p) + \delta_e^2] \tau_{\text{eff}}^2 \\
\Theta^2(p) \sin^2 \theta(p) = \Omega^2(p)\tau_{\text{eff}}^2 \sin^2 \xi(p),
\]

where \( \Omega(p) \) is the magnitude of the effective rotation vector caused by the pulse, \( \xi(p) \) is the effective rotation vector’s polar angle excluding Larmor precession (in contrast to \( \theta \), which is the polar angle of the rotation axis including Larmor precession), and \( \tau_{\text{eff}} \) is a power-independent effective pulse length. The existence of
power-variation of $\xi(p)$ is evident in approximate descriptions of the dynamics via adiabatic elimination as well as in more quantitative simulations. Note especially that at $p = 0$, $\Theta(p) = \delta\tau_{\text{eff}}$, so even at zero-power there is a non-zero rotation angle due to Larmor precession during the finite pulse duration.

The amplitude of the Rabi oscillations and Ramsey fringes is determined by the vector shrinkage $D(p)$ as well as the tilt in the rotation axis $\theta(p)$. In principle, knowing $\Theta(p)$ and $D(p)$ based on the extrema of the Rabi oscillation could allow the deduction of $\theta(p)$. However, noise and experimental imperfections make such an extraction unreliable. It is more reliable to examine the phase of the Ramsey fringes as a function of power.

The Ramsey fringe data as a function of power $p$ and interpulse delay $\tau$ is modeled
as \( [1 - M_{zz}(p, \tau)] / 2 \), where \( M(p, \tau) \) is the total matrix

\[
M(p, \tau) = D^2(p) R[\Theta(p), \theta(p)] R[\delta_e \tau, 0] R[\Theta(p), \theta(p)]
\]

\( \equiv B(p) + A(p) \cos[\delta_e \tau + \phi(p)]. \) (A.3)

The offset \( B(p) \) is not used. The extrema of the experimentally-determined amplitude \( A(p) \) is used to estimate the locations where \( \Theta = n\pi / 2 \), an approximation valid when \( \theta(p) \) is close to \( \pi / 2 \). The phase \( \phi \) may be calculated from the model to be

\[
\cos \phi(p) = \frac{\cos \Theta(p) + \sin^2 \theta(p) \sin^2[\Theta(p)/2]}{1 - \sin^2 \theta(p) \sin^2[\Theta(p)/2]}.
\] (A.4)

When \( \Theta(p) = 2\pi \) (as found from the second minimum in the Ramsey fringe amplitude), the phase \( \phi \) becomes zero. This allows us to set a phase reference for the observed phase of the Ramsey data. At \( p = 0 \), \( \theta(0) = 0 \) so the phase becomes \( \phi(0) = \delta_e \tau_{\text{eff}} \) due to Larmor precession during the pulse. This phase offset is also observed in the Ramsey data.

With such a phase reference defined, a smoothly interpolated function for \( \Theta(p) \) is determined from the extrema of \( A(p) \) in addition to the condition \( \Theta(0) = \phi(0) \). The experimentally observed Ramsey phase \( \phi(p) \) is then used in conjunction with this smoothed \( \Theta(p) \) in Eq. (A.4) to find \( \theta(p) \) at each power \( p \).

However, this result is very noisy near the minima of the Ramsey Fringe amplitude (i.e. at \( \Theta = n\pi \)), since near these points the Ramsey signal vanishes and phase determination is less reliable. In order to find a smooth trajectory, we return to our assumption that \( \xi(p) \) and \( \Omega(p) \) vary slowly and smoothly with \( p \). Smooth functions for these functions are therefore found which are consistent with the data; these in turn yield a smooth function for \( \theta(p) \). The combination of \( \theta(p) \) and \( \Theta(p) \) are used to generate smooth Ramsey curves which are checked to be consistent with the data within its noise. These functions are then used to find \( \vec{v}(p) = L_0 D(p) R[\Theta(p), \theta(p)] \cdot (-\hat{z}) \), a smoothly interpreted vector for the range of power used in the Ramsey experiment.
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