

Quantum Sensing and Control with Color Centers in Diamond

by

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DISSERTATION ABSTRACT

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Title: Quantum Sensing and Control with Color Centers in Diamond

This thesis explores quantum sensing and control with color centers in diamond, focusing primarily on coherent-population-trapping (CPT)-based sensing using nitrogen-vacancy (NV) centers. We develop a continuous-measurement protocol in which CPT of a Λ -system maps an accumulated phase onto a time-dependent fluorescence record, and we analyze its performance using Ornstein–Uhlenbeck (OU) noise as a model. Within this framework, a Bayesian estimator is shown to approach the Cramér–Rao lower bound under realistic experimental conditions, and a neural-network-based estimator is used to benchmark its near-optimality. Building on this, we integrate CPT-based sensing with real-time feedback to both suppress environmental noise and perform spectroscopy in an OU basis, enabling operation even without detailed a priori knowledge of the noise spectrum. Experimentally, we implement a confocal microscope for addressing single color centers and demonstrate microsphere-enhanced fluorescence collection, which relaxes the trade-off between numerical aperture and working distance and improves sensitivity for practical sensing geometries. Finally, we carry out low-temperature spectroscopic studies of silicon-vacancy (SiV) centers under magnetic fields, clarifying their spin and level structure and laying the groundwork for extending CPT-based sensing to SiV centers and for future cavity QED experiments.

This dissertation includes previously published co-authored materials.

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CHAPTER I

INTRODUCTION

1.1 Background

Over the past decade, many exciting breakthroughs have been demonstrated in quantum information processing [Arute et al. \(2019\)](#); [Barredo, De Léséleuc, Lienhard, Lahaye, and Browaeys \(2016\)](#); [Bhaskar et al. \(2020\)](#); [Endres et al. \(2016\)](#); [Madsen et al. \(2022\)](#); [Yin et al. \(2017\)](#); [Zhong et al. \(2020\)](#). Since the early days when golden age physicists discovered the fundamental principles of quantum mechanics, humanity has never been closer to manipulating systems at the single-quantum level. Thanks to astonishing advances in optics and electronics, we can now use these tools to control quantum systems and explore their applications across many fields.

Quantum sensing is one such application, where quantum systems serve as sensors to probe external fields [Degen, Reinhard, and Cappellaro \(2017\)](#). Owing to their quantum nature, they can benefit from entanglement or other correlations that improve scaling beyond the standard classical limit [Pezze, Smerzi, Oberthaler, Schmied, and Treutlein \(2018\)](#), or, by virtue of their single-atom scale and well-controlled states and operations, achieve maximal sensitivity possible to the environment [Degen et al. \(2017\)](#). With these newly developed tools, we can build next-generation gyroscopes that operate for long durations with minimal recalibration [Durfee, Shaham, and Kasevich \(2006\)](#), detect tiny gravitational accelerations from ore bodies for mineral exploration [Stray et al. \(2022\)](#), characterize magnetic responses with nanoscale resolution that is useful for condensed-matter studies [Balasubramanian et al. \(2008\)](#), and detect minimal changes in path length for gravitational-wave detection [Tse et al. \(2019\)](#).

While different physical platforms offer distinct strengths, solid-state quantum systems, especially color centers in diamond, are particularly appealing for practical and nanoscale sensing, since they do not require the complex control infrastructure essential for cold atoms, SQUIDs, and related platforms. Color centers are atom-like defects trapped in a crystal lattice and thus supported and protected by the host structure. Among color centers, the nitrogen-vacancy (NV) center in diamond is one of the most studied and promising for quantum sensing [Doherty et al. \(2014\)](#); [Dolde et al. \(2011\)](#); [Kucsko et al. \(2013\)](#); [Taylor et al. \(2008\)](#). As discussed in Chapter II, diamond is transparent in the visible spectrum, and the NV center possesses a spin degree of freedom with long room-temperature coherence times thanks to its symmetry. These properties make NV-center-based sensing highly promising for its potential outside of a research lab. Its experimental setup and an approach using a microsphere to improve performance are discussed in Chapters III and IV.

At the core of many quantum-sensing applications is the readout of an accumulated phase (summarized in Chapter V). A standard approach is to use a Ramsey-type experiment, which measures the phase explicitly but in an intrinsically discrete manner, requiring high-speed repetition to track fast fluctuations. In contrast, we propose a coherent-population-trapping (CPT)–based sensing method that encodes and reads out the phase in a fundamentally different way. By coupling two lasers to a Λ -system, dark and bright states emerge, with fluorescence only from the bright state. In this configuration, CPT-based sensing effectively implements a continuous projective measurement onto the bright/dark-state manifold, so the phase is embedded in the time-dependent probability of projecting onto these states rather than in a discrete Ramsey fringe. Together with

Bayesian estimation, which extracts maximal information from this continuous measurement record, we develop a full theoretical framework in Chapter VI and demonstrate a proof-of-principle experiment in Chapter VII using Ornstein–Uhlenbeck (OU) noise as a model. A feedback experiment based on CPT sensing then shows how to extract parameters and perform spectroscopy in an OU-noise basis, enabling our CPT-based approach to operate even without prior knowledge of the underlying noise structure.

Beyond sensing, color centers are also favorable interfaces to “flying” qubits such as photons or phonons, because they are intrinsically embedded in a solid-state environment well-suited for cavity QED. Cavities confine the photon/phonon field and enhance interactions with color centers in diamond. With this approach, near-deterministic, high-fidelity swapping and entanglement between stationary and flying qubits become feasible [Evans et al. \(2018\)](#); [Nguyen et al. \(2019\)](#), which is particularly useful for building quantum networks and paves the way to scaling solid-state qubits mediated by cavities. Quantum networks are crucial for harnessing large-scale entanglement and remote connectivity, enabling applications such as large-baseline telescope [Gottesman, Jennewein, and Croke \(2012\)](#), scalable quantum computing [Monroe et al. \(2014\)](#), and quantum communications [Gisin and Thew \(2007\)](#); [Kimble \(2008\)](#).

Although NV centers have many desirable properties for quantum sensing, they also face challenges, including a low emission fraction into the zero phonon line (ZPL) and sensitivity to surface-charge fluctuations due to the lack of inversion symmetry. By contrast, silicon-vacancy (SiV^-) centers in diamond attract attention because they feature strong ZPL emission and are robust to charge noise, making them well-suited to cavity QED applications. The trade-off is that long spin

coherence of SiV^- typically requires dilution-refrigerator temperatures. Other centers are also under active study, including SnV centers [Iwasaki et al. \(2017\)](#), the neutral charge state SiV^0 (distinct from the commonly used SiV^-) [Green et al. \(2017\)](#), and certain rare-earth ions [Judd \(1962\)](#), T-center in silicon [Dhaliya, Xiong, Sipahigil, Griffin, and Hautier \(2022\)](#), each with advantages for specific applications.

To extend our CPT-based sensing to SiV centers and to explore cavity-QED studies, we must understand the spin physics of SiV centers, especially the spin-state energy structure, dynamics, and CPT behavior at millikelvin temperatures. We present these studies in Chapter IX, and preliminary results on cavity QED with SiV centers are discussed in Appendix A.

1.2 Overview of thesis

In Chapter II, we give a brief summary of the energy structures of the NV and SiV centers, and how they respond to external fields. A more detailed analysis based on symmetry and group theory is presented in Appendix A. From group representation theory, the irreducible representations of the symmetry group reflect the eigenstate structure of the Hamiltonian. This framework immediately yields the response to external fields such as strain and magnetic fields and explains why NV and SiV centers have different spin and dipole properties. Combined with optical selection rules for the relevant transitions, this gives a complete picture of the accessible quantum systems.

Regarding experimental realization, Chapter III introduces the confocal optical-microscope setup used to address and manipulate color centers with lasers. We discuss the principles and practicalities of building such an optical setup, as well as spin-manipulation experiments on NV centers. Some experimental details

and related formulas are presented in Appendix B. A key practical limitation is the trade-off between an objective’s working distance and numerical aperture (which determines fluorescence-collection efficiency). In Chapter IV, we show that this trade-off can be relaxed by placing a microsphere near the color center to collimate the emission. And the results presented in Chapter IV were published in [S.-H. Wu, Lekavicius, and Wang \(2024\)](#).

Chapter V connects quantum sensing and quantum control by recognizing their common reliance on quantum phase. The phase encodes shifts in energy levels and thus information about external fields, and it is accessible through readout. By measuring the phase faster than the fluctuation time scale, one can track time-domain behavior; by measuring averaged behavior, one can extract spectral information. In contrast, quantum control designs sequences that suppress environmental information imprinted in the phase, thereby increasing coherence time.

In Chapter VI, we develop a theoretical framework for CPT-based sensing and, in Chapter VII, we demonstrate a proof-of-principle experiment using OU noise as a model. Neural-network-based estimators are also employed to benchmark optimality. And the results presented in Chapter VI and VII were published in [Turner, Wu, Li, and Wang \(2022a\)](#); [S. H. Wu, Turner, and Wang \(2021\)](#).

In Chapter VIII, we apply CPT-based sensing to real-time feedback control, canceling noise in situ and thereby extending coherence. We further show that the feedback-enhanced coherence time can serve as a cost function to determine noise parameters. In analogy with Fourier analysis, OU noise with different correlation times forms a complete basis for modeling fluctuations; in this sense, our method performs spectral-domain quantum sensing analogous to dynamical-decoupling

spectroscopy but with an OU basis. And the results presented in Chapter VIII were published in [Turner, Wu, Li, and Wang \(2022b\)](#). Note that Chapters VI-VIII are extracted from three published papers. As such, there are redundant discussions on both theoretical and experimental details in these chapters. No attempts have been made to modify these discussions. In this way, each chapter can be read more or less independently.

Chapter IX presents fundamental studies of the SiV center’s energy structure under magnetic fields and its spin properties at millikelvin temperatures. These results allow us to validate theoretical models for SiV centers, especially dynamic-Jahn–Teller–induced orbital-Zeeman quenching and phonon-induced spin dephasing. And the results presented in Chapter IX were published in [S. Wu, Li, Gallagher, Lawrie, and Wang \(2025\)](#); [S.-H. Wu et al. \(2025\)](#).

Appendix C summarizes our initial experimental efforts toward cavity QED with SiV centers. We first show how the cooperativity C —a core figure of merit for cavity-QED systems—naturally arises from two coupled harmonic oscillators. From this, we explain why different parameter regimes favor different experimental choices. We then discuss mode characterization in both photonic and phononic platforms.

Finally, Appendix D introduces an open-loop method to extend NV coherence by continuously driving two ground states with microwaves, effectively implementing continuous dynamical decoupling. Theory and simulations show improved robustness of state transfer against energy-level fluctuations. And the results presented in Appendix D were published in [S.-H. Wu, Amezcua, and Wang \(2019\)](#).

Isaac Newton once wrote, “From the same principles, I now demonstrate the frame of the System of the World,” in *The Mathematical Principles of Natural Philosophy*. And now, from the same principles, I will demonstrate the frame of quantum sensing and control with color centers in diamond in the following chapters.

CHAPTER II

ENERGY LEVEL STRUCTURES OF NV AND SIV CENTERS

2.1 Introduction

Color centers in diamond are point defects in the diamond lattice that give rise to characteristic optical transitions in the visible and near-infrared. They were first noticed because diamonds containing such defects exhibit distinct colors, hence the name "color centers". With the development of high-purity synthetic diamond, especially chemical vapor deposition (CVD) growth, and with ion-implantation techniques, it has become possible to create specific types of color centers in a controlled manner. After implantation, an annealing step allows vacancies in the diamond lattice to migrate and combine with impurity atoms, forming a more stable defect configuration.

In principle, any point defect lowers the local symmetry and can introduce localized states in the band gap. However, in diamond most optically active centers of practical interest are not simple substitutional impurities but impurity vacancy systems (such as NV^-) or the group-IV centers, a notable exception are atom-like rare-earth dopants in diamond). The vacancy supplies unsaturated bonds that generate a localized orbital subspace inside the wide band gap, while the impurity atom sets the symmetry and the ordering of these orbitals. Consequently, the electronic wavefunctions are localized around the defect vacancy and a set of discrete energy levels emerges from the otherwise continuous bands of the host crystal.

Because diamond has a large band gap and is transparent in the visible range, diamond color centers can be optically addressed even when they are located deep in the bulk. This makes diamond color centers attractive as solid-state

quantum systems: they are embedded in and protected by a robust host material and are therefore relatively insensitive to many environmental perturbations. By contrast, systems such as trapped ions or neutral atoms require additional external trapping potentials to achieve comparable isolation and stability.

Another important advantage is that color centers are intrinsically embedded in a solid-state platform. This makes their coupling to photonic or phononic nanostructures more natural and, in many cases, more coherent, which is crucial for applications in quantum control and for interfacing stationary qubits with propagating photonic qubits. At the same time, the internal quantum degrees of freedom of the defect remain sensitive to selected external perturbations such as temperature, magnetic field, electric field, and strain. This dual nature protection by the diamond host but controllable sensitivity to specific fields underlies the now well-established use of diamond color centers as quantum sensors.

In this chapter we give a brief, experiment-oriented description of two widely used diamond color centers, the negatively charged nitrogen-vacancy center (NV^-) and the negatively charged silicon-vacancy center (SiV^-). Our main goal is to present their energy-level structures and the effective Hamiltonians that capture their response to external fields. A more detailed analysis from the group-theoretical perspective, including the derivation of the energy levels and the effective couplings to phonons and photons from representation theory, is given in the appendix.

2.2 NV symmetry and energy structure

The negatively charged nitrogen-vacancy center (NV^-) in diamond consists of a substitutional nitrogen atom adjacent to a vacancy. Together with the three nearest-neighbor carbon atoms and the next-nearest carbon along the $\langle 111 \rangle$ axis,

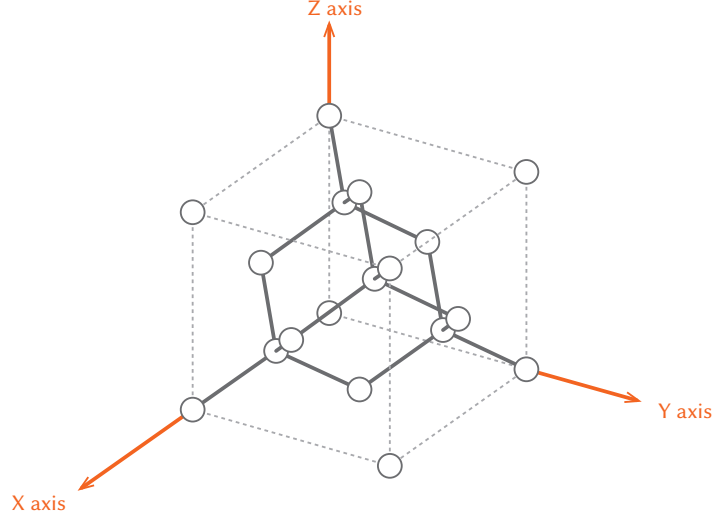


Figure 2.1. Crystal structure of diamond. Carbon atoms form a tetrahedral network of covalent bonds that can be viewed as two interpenetrating face-centered cubic sublattices, offset by $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})a$ along the $[111]$ direction.

the defect forms a quasi-molecular complex with an effective C_{3v} (often also denoted D_3) point-group symmetry (see [Figure 2.2](#)). The absence of inversion symmetry implies a permanent electric dipole moment along the NV axis, which makes the optical transition frequencies sensitive to local electric fields and charge fluctuations, especially near the surface.

In the standard molecular-orbital picture, the four dangling bonds of the three carbons and the nitrogen hybridize into one $a_1(1)$ orbital, a second $a_1(2)$ orbital, and a doubly degenerate $e = \{e_x, e_y\}$ orbital. The lowest $a_1(1)$ orbital lies deep below the valence band and is always fully occupied, while the higher $a_1(2)$ and the e doublet lie in the band gap and form the relevant defect manifold. For the NV^- charge state there are six electrons occupying these in-gap defect orbitals, which is equivalent to two holes in the manifold. Filling the levels according to Hund's rule yields a spin-triplet ground state with orbital symmetry A_2 , denoted 3A_2 . Crucially, all three spin projections $|m_s = 0, \pm 1\rangle$ share the same orbital

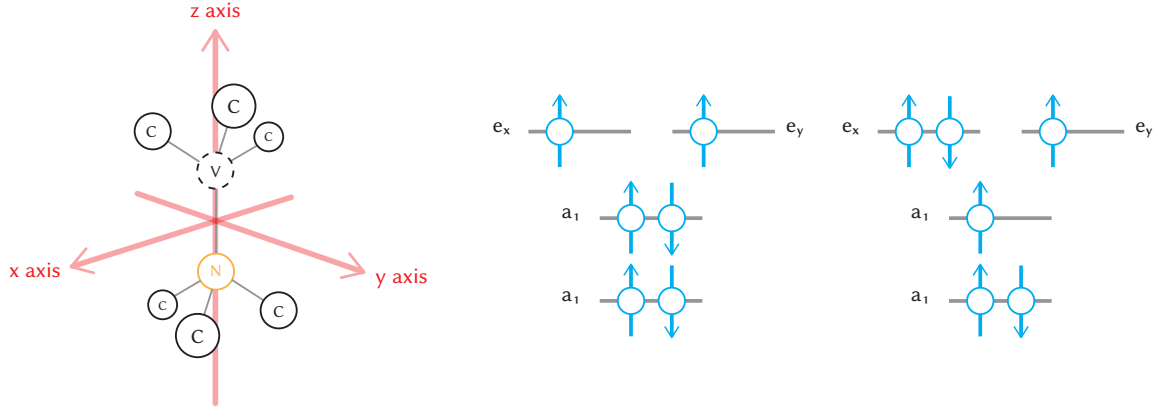


Figure 2.2. Structure of the NV center in diamond and the corresponding electronic orbital configurations of its ground and first excited states. The defect consists of a substitutional nitrogen atom adjacent to a carbon vacancy, forming a C_{3v} -symmetric center. The diagrams illustrate the occupation of the a_1 and e orbitals in the ground and excited manifolds.

wavefunction. As a result, lattice vibrations couple only weakly to the spin degree of freedom, and the NV ground state is largely protected against phonon-induced decoherence. This orbital nondegeneracy is the key reason for the long spin lifetime and coherence of NV centers, even at room temperature in high-purity samples.

Group theory alone predicts the symmetry labels and degeneracies of the low-lying states, while density-functional-theory (DFT) calculations ([Gali and Maze \(2013\)](#)) provide their absolute ordering in energy, as sketched in [Figure 2.2](#). The lowest optically bright excited state is lying about 1.95eV above the ground state and giving rise to the zero-phonon line (ZPL) at 637nm. Between the 3A_2 ground state and the excited state there are intermediate states which participate in non-radiative intersystem crossing. This intersystem crossing is responsible for optical spin polarization and spin-dependent fluorescence.

Within the 3A_2 ground-state manifold, the dominant internal interaction is the magnetic dipole-dipole (spin-spin) coupling between the electrons, which can be written as $H_{ss} = DS_z^2$ where D is the zero-field splitting parameter. This term

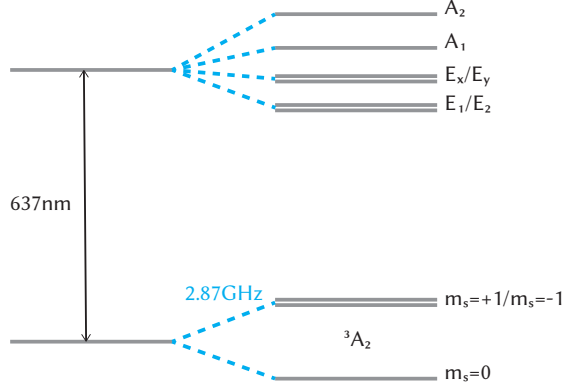


Figure 2.3. Energy-level structure of the NV center. The ground spin-triplet and excited states are shown, the intermediate metastable singlet state involved in optical spin pumping is not included in this diagram.

lifts the degeneracy between the $|m_s = 0\rangle$ and $|m_s = \pm 1\rangle$ sublevels and gives the characteristic zero-field splitting $D \approx 2.87\text{GHz}$.

2.3 SiV symmetry and energy structure

The negatively charged silicon-vacancy center (SiV^-) in diamond consists of a silicon atom situated between two adjacent vacancies along a $\langle 111 \rangle$ axis of the diamond lattice. Because the Si-C bond in diamond is longer than the C-C bond, it is energetically favorable for the Si impurity to relax to the center between two opposite vacancies. The resulting split-vacancy configuration possesses an inversion center and therefore has D_{3d} point-group symmetry (see [Figure 2.4](#)). The presence of inversion symmetry strongly suppresses the coupling of the optical transition to fluctuating electric fields, so SiV^- centers typically exhibit very narrow optical lines and reduced spectral diffusion compared to NV centers.

In contrast to the NV^- center, which effectively has two holes in its defect manifold, the SiV^- center has only one unpaired hole in its relevant orbitals. This leads to a much simpler electronic level structure, but it also means that the ground state is more susceptible to phonon-induced orbital relaxation. In

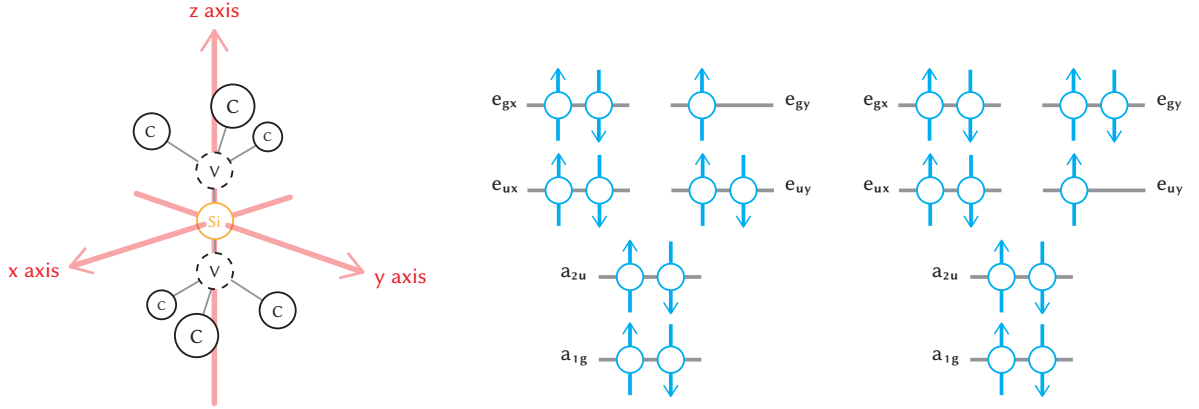


Figure 2.4. Local coordinate frame of the SiV center and the corresponding electronic orbital configurations in the ground and excited states. The diagrams indicate the occupation of the relevant orbital doublets.

the simplest picture, both the ground and excited manifolds arise from a doubly degenerate $e = \{e_x, e_y\}$ orbital occupied by a single hole. Before including spin-orbit coupling, each of these manifolds is therefore fourfold degenerate (spin and orbital). The corresponding energy-level structure is shown schematically on the right of [Figure 2.4](#).

Spin-orbit interaction mixes the spin and orbital degrees of freedom and lifts the degeneracy of both the ground and excited manifolds even in the absence of magnetic field or strain. In a local coordinate system aligned with the defect symmetry axis, the dominant contribution can be written as

$$H_{\text{SO}} = -\lambda_{\text{SO}} L_z S_z,$$

where λ_{SO} is the effective spin-orbit coupling strength, and L_z and S_z are the orbital and spin angular momentum operators projected onto the SiV axis. This interaction splits each e manifold into two degenerated doublets. Experimentally, the ground-state splitting is on the order of $\sim 50\text{GHz}$ and the excited-state splitting is on the order of $\sim 260\text{GHz}$. The optical transition between the centers of

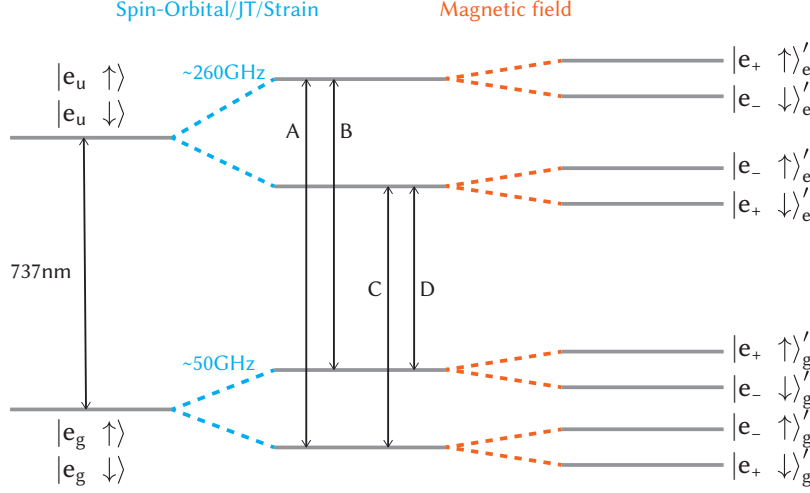


Figure 2.5. Summary of the energy-level structure of the SiV center including all relevant interactions. Spin-orbit, Jahn-Teller, and strain interactions predominantly mix and split the orbital part of the eigenstates, whereas the Zeeman interaction mainly mixes the spin degrees of freedom.

gravity of the ground and excited manifolds defines the zero-phonon line (ZPL) near 737nm, while spin-orbit coupling and strain determine the fine-structure splittings around this ZPL.

When lattice vibrations (phonons) or static strain are present, the nuclei are displaced and the electronic orbitals are distorted. From the perspective of the energy-level structure, static strain and Jahn-Teller (JT) distortions have the same symmetry and therefore cannot be distinguished at the level of a static effective Hamiltonian. It is convenient to describe these effects in terms of the strain tensor

$$\varepsilon_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right),$$

where $\mathbf{u}(\mathbf{x})$ is the displacement field. In the $\{e_x, e_y\}$ orbital basis of the SiV center, the strain interaction for a given manifold can be written as

$$H_{\text{strain}} = \begin{pmatrix} \alpha + \beta & \gamma \\ \gamma & \alpha - \beta \end{pmatrix},$$

with

$$\begin{aligned}\alpha &= t_{\perp}(\varepsilon_{xx} + \varepsilon_{yy}) + t_{\parallel}\varepsilon_{zz}, \\ \beta &= d(\varepsilon_{yy} - \varepsilon_{xx}) - f\varepsilon_{zx}, \\ \gamma &= -2d\varepsilon_{xy} + f\varepsilon_{yz}.\end{aligned}$$

Here we use the local coordinates x, y, z of the SiV center (Figure 2.4) instead of the laboratory coordinates X, Y, Z (Figure 2.1). One convenient choice of transformation from the lab frame to the local frame is $x = (-1, 1, 0), y = (-1, -1, 2), z = (1, 1, 1)$ expressed in units of the cubic axes of the diamond lattice. The parameters $t_{\perp}, t_{\parallel}, d$, and f characterize how different components of the strain tensor couple to the defect orbitals.

An external magnetic field will also mix the orbital and spin states. However, because the orbital angular momentum is partially quenched by the dynamic Jahn-Teller effect, a quenching factor q is introduced to reduce the strength of the orbital Zeeman term. The effective Zeeman Hamiltonian can be written as

$$H_{\text{Zeeman}} = q\gamma_L \mathbf{L} \cdot \mathbf{B} + \gamma_S \mathbf{S} \cdot \mathbf{B} = q\gamma_L L_z B_z + \gamma_S \mathbf{S} \cdot \mathbf{B},$$

where γ_L and γ_S are the orbital and spin gyromagnetic ratios, respectively, and $\mathbf{B}$ is the applied magnetic field. Together with the spin-orbit and strain terms, this Hamiltonian fully describes the energy structure of the SiV⁻ ground and excited states.

Finally, the coupling to light is crucial for experiments. The interaction between the defect and an optical mode is described by the electric dipole operator coupling to the electric field of the light. In the hole picture one writes

$$H_{\text{dipole}} = \mathbf{p} \cdot \mathbf{\mathcal{E}} = e(x\mathcal{E}_x + y\mathcal{E}_y + z\mathcal{E}_z) = p_0\mathcal{E}_0 + p_-\mathcal{E}_- + p_+\mathcal{E}_+,$$

where $p_0, p_{\pm}$ and $\mathcal{E}_0, \mathcal{E}_{\pm}$ denote the dipole and field components in a spherical basis.

We can predict based on group theory ([Hepp \(2014\)](#)) that the selection rules are

polarization	$ e_{g+}\rangle$	$ e_{g-}\rangle$
$\langle e_{u+} $	π	σ_-
$\langle e_{u-} $	σ_+	π

CHAPTER III

EXPERIMENTAL SETUP FOR CONFOCAL OPTICAL MICROSCOPE AND QUANTUM SPIN CONTROL

3.1 Introduction

Color centers in diamond have optical transitions in the visible/near-infrared, allowing manipulation with lasers. When the density of color centers is low, individual emitters can be optically addressed. While wide-field imaging combined with a CCD can spatially resolve emitters [Cambria, Chand, Reiter, and Kolkowitz \(2025\)](#); [Cheng et al. \(2025\)](#), it adds complexity and lacks longitudinal addressability. A confocal microscope overcomes these limitations by selectively exciting and collecting fluorescence, establishing a one-to-one mapping between a point in the sample and the detection plane. In a typical far-field confocal setup, the lateral resolution is a few hundred nanometers, and the axial resolution is worse but still on the order of $\sim \mu m$ when a single-mode fiber is used. This individual addressability enables direct interaction with multi-level quantum systems.

The basic idea is to excite the zero-phonon line (ZPL) of the transition but detect at the phonon sideband (PSB). Some experiments also use cross-polarization schemes [Müller et al. \(2014\)](#) that reject the polarization of the excitation beam but accept the emission from the color center, at the cost of some loss. As shown in [Figure 3.1](#), the color center absorbs a photon to reach the excited state and then couples to phonons and photons at the same time, eventually, after a lifetime of a few nanoseconds, it decays back to the ground state, accompanied by some phonon excitation and a photon with a longer wavelength.

By using a non-resonant excitation laser, typically 532 nm due to its common availability, we are able to locate color centers spatially, since they

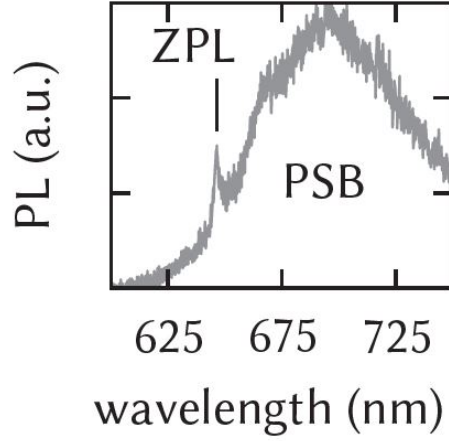


Figure 3.1. The emission spectrum of NV centers in diamond, copy from figure 3.1 of [PFAFF \(2013\)](#).

fluoresce when the laser spot overlaps with them. This can be done by scanning the galvo mirrors, which maps to scanning an area in the focal plane (i.e. the plane of the diamond sample implanted with defects). This measurement is called photoluminescence (PL). Another approach is to use a resonant laser, and by scanning the laser wavelength we can find where the laser is on resonance with the transitions of a given color center as shown in [Figure 3.2](#). This helps us determine its energy structure and also allows us to address individual transitions and individual centers by frequency. This is called photoluminescence excitation (PLE).

3.2 Excitation and collection

In [Figure 3.3](#) we show a basic optics setup for the excitation and collection part of all the experiments in this thesis. This is also a standard, so-called confocal microscope. On the excitation side, the laser is steered by a pair of galvo mirrors so that the beam enters the objective at different angles and therefore is focused to different positions on the sample. On the collection side, we use a multimode optical fiber with a small core as a pinhole, others use a physical pinhole or a

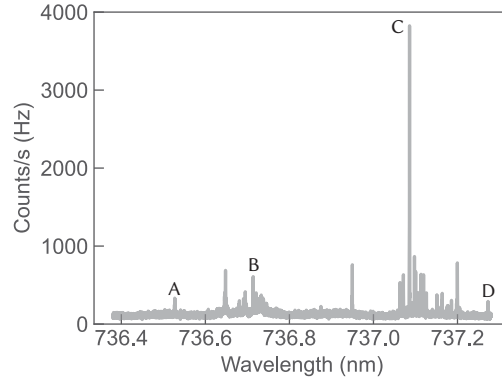


Figure 3.2. PLE scan of a single SiV center shows multiple peaks, some of which arise from other SiV centers located surrounding it. The four principal transitions of the target center, labeled A, B, C, and D, can be identified using the requirement $\omega_A - \omega_B = \omega_C - \omega_D = \lambda_{\text{so}}^g \approx 50\text{GHz}$. This equality in splittings provides a clear fingerprint that distinguishes the desired SiV from nearby centers.

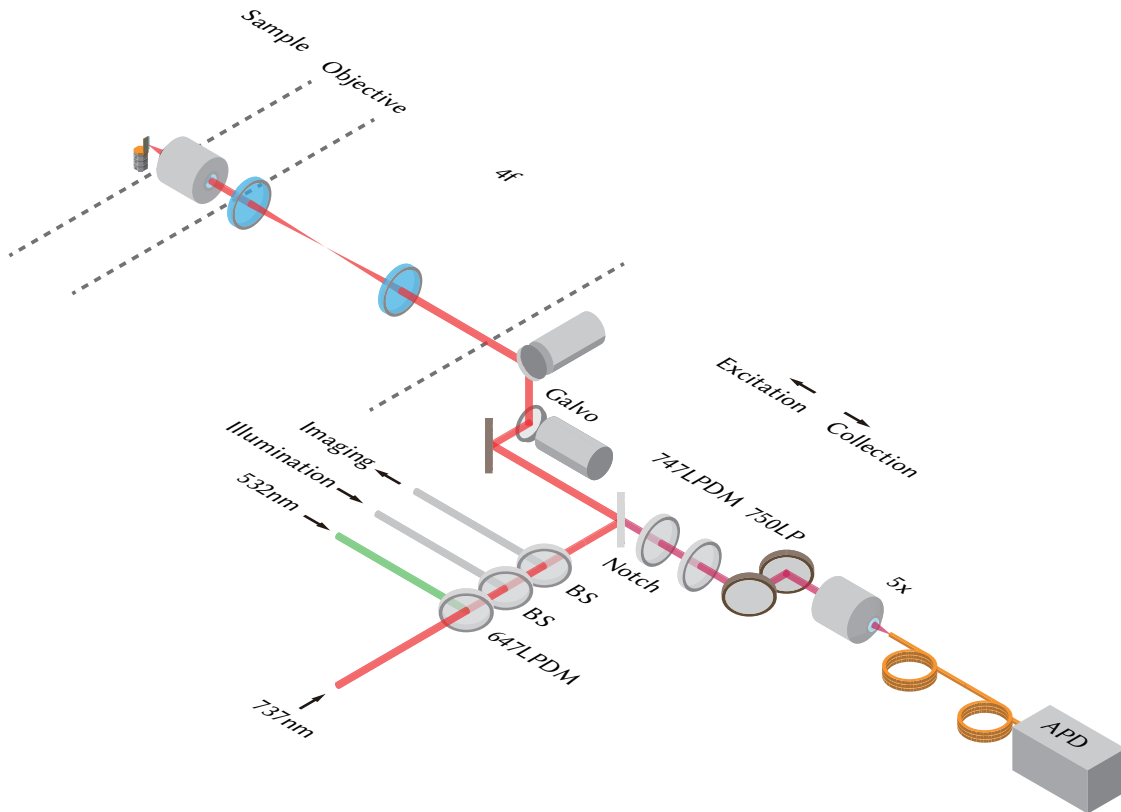


Figure 3.3. Confocal setup for a typical color-center experiment, consisting of excitation and collection paths separated by a dichroic filter. Although the specific implementations differ for room temperature, 4K, and millikelvin temperature environments, they share the same basic structure shown in the figure.

single-mode fiber as well. This confocal configuration gives the system a sharper resolution of order $\sim (2J_1(x)/x)^4$, which is the square of the Airy pattern generated by diffraction from a single circular aperture of radius r where $x = 2\pi r \sin \theta / \lambda$. It also gives an axial resolution which, in principle, allows us to scan the sample layer by layer.

In our setup, the sample is mounted on a xyz stack of nanopositioners driven by piezo actuators. This allows us to move the sample with ~ 100 nm resolution and $\sim \pm 2$ mm travel range in all directions. A copper sample mount is fixed on a copper thermal link (not shown in the figure) directly attached to the cold finger to ensure good thermal conductivity. Silver paint was used to fix the diamond sample on the mount, but it fell off after a few thermal cycles.

A 100 $\times$ objective is normally used to get the best numerical aperture. Correspondingly, the working distance is limited to ~ 0.5 mm, therefore careful alignment is needed during the experiment to move the sample to the focal plane of the objective. For dilution-fridge experiments, this objective is a special low-temperature objective with aberration correction over the required wavelength range.

A 4f system is optionally used to map the steering of the galvo mirrors to the back focal plane of the objective. In this way the incident beam always hits the center of the entrance aperture (assuming the distance between the two galvo mirrors is negligible), which results in better performance. In the dilution-fridge experiment this 4f relay is needed to delay the steering until after the long optical access inside the probe. In our typical cryogenic setup, the galvo mirrors are placed close enough to the objective that this 4f relay is not required, unless one needs to

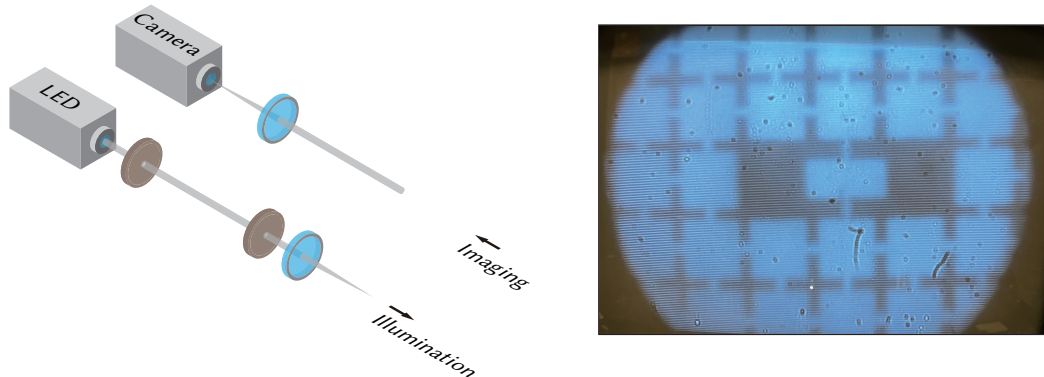


Figure 3.4. Illustration of the imaging part, a CCD image of a structured sample is shown on the right. The field of view is limited by the objective, while the image sharpness and brightness are largely determined by the illumination quality.

steer the beam by an amount smaller than the angular resolution of the galvos, in which case an unequal-magnification 4f system can be used to scale the steering.

A 745-nm long-pass dichroic mirror is used to split excitation and collection into different wavelength ranges. However, this kind of interference filter is sensitive to angle deviation, which can easily shift the edge by a few nanometers. Therefore, a 90:10 beamsplitter is also used in some experiments. It transmits 90% of the incident beam at the cost of wasting a lot of excitation power, but this is normally not a limit. The emission from the color centers then passes through a 532-nm notch filter and a 750-nm long-pass filter to further suppress unwanted wavelengths. The emission is then coupled into a multimode fiber (normally $10\mu\text{m}$) and sent to an avalanche photodiode (APD) to detect single photons. The output pulses from the APD are sent to a DAQ or a timetagger for further analysis or gating.

3.3 Imaging

For a sample with fabricated structures it is often beneficial to have a white-light image, which helps us move the sample to the correct position and quickly check whether it is on the focal plane. For the white-light part it is a wide-field

imaging configuration, so we do not use any pinhole to select the collection area. Instead, we map the focal plane onto an imaging plane (a CCD array). This can be done by placing a lens one focal length away from the CCD, since the objective is infinity-corrected and outputs a collimated beam. We did not use the tube lens that is exactly matched to the objective, because we were not aiming at optimal white-light imaging quality.

One can also add an extra iris to control the imaging area and to suppress scattered light, but since the aperture is much larger than the diffraction-limited spot size this is normally not considered confocal. For illumination, the ideal scheme is Köhler illumination, whose core idea is to generate uniform illumination at the focal plane even with a non-uniform light source. To realize it with minimal effort, we send the LED light through two irises, which greatly reduce off-axis light and its scattering, and make sure the light comes from the same area on the source, therefore sufficiently uniform. A lens after the second iris can be moved along the axis until a uniform lighting is observed on the CCD, which means the illumination is focused to the back focal plane of the objective. An example image is shown on the right of [Figure 3.4](#).

3.4 Excitation beam control

The core of the color-center experiments is to have a well-controlled laser. For an optical field, the relevant degrees of freedom are frequency, amplitude, phase, and polarization. However, most commonly available laser sources are continuous-wave and have short-term phase noise plus long-term frequency drift, both contributing to frequency noise. The short-term phase noise is typically not a problem for color-center experiments, since it is much smaller than the intrinsic linewidth of the optical transitions ($\sim 10 - 100\text{MHz}$). For some AMO experiments

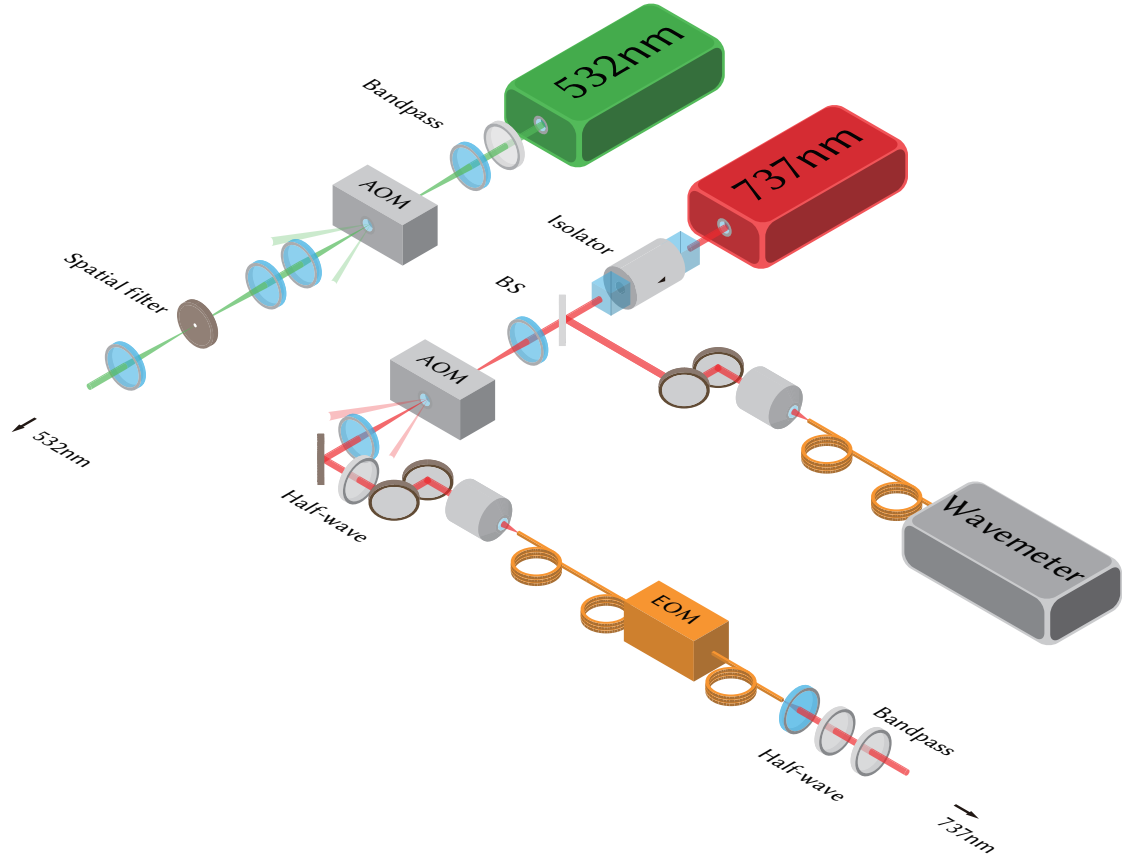


Figure 3.5. Illustration of the green and red excitation-beam control stage, where all beam-related operations such as shaping, collimation, gating, frequency stabilization, and polarization control are implemented. Coupling this module to the rest of the setup via optical fiber is recommended, as it allows flexible control over the excitation conditions while decoupling it from other components and minimizing realignment efforts.

that require sub-MHz linewidth, people lock the laser to a cavity and use PDH to reduce the linewidth down to $\sim$ Hz. But the long-term drift due to temperature or humidity fluctuations can still ruin the experiment. Additionally, for our PLE experiments it is preferred to have the ability to accurately tune the laser to the desired frequency in order to scan the wavelength range of interest.

Here we send a part of the laser to a wavemeter to monitor the wavelength in real time and send commands to control the piezo in the laser to tune the frequency in a feedback loop. This can be done at $\sim$ 10 Hz using a desktop computer and ends up with $\sim$ MHz-level fluctuations. An isolator is used to suppress reflections going back to the laser source, which can cause mode hops or destroy single-mode operation and therefore ruin the frequency feedback.

The amplitude of the laser is controlled by an AOM. The acoustic wave forms a periodic density modulation in the crystal and behaves like a diffraction grating. The laser beam after the AOM is split into different orders. By using a non-zero diffraction order and by controlling the RF drive power, we can modulate the diffraction efficiency and therefore modulate the optical amplitude; most commonly we use it to gate the laser on and off. Two lenses are used to focus the beam into the AOM to get higher diffraction efficiency and a faster rising edge.

The green laser is then sent to a spatial filter so that the beam is diffracted by a pinhole, a later iris can be placed to selectively pass only the central part, which corresponds to the diffraction of the fundamental mode (TEM_{00}), and the green beam becomes Gaussian.

The red laser is handled slightly differently. After the AOM it is coupled into a fiber and then to a fiber-pigtailed EOM, which modulates the phase of the laser and generates frequency sidebands from the carrier. Since the different

frequency components share the same optical source, their coherence is maintained. In this regard this is more ideal and easier than using two independent laser sources, and it is useful for experiments such as CPT that need two coherent excitations. On the other hand, multiple sidebands make it less ideal for tasks such as optical pumping. Because the fiber EOM uses PM fiber, we have two half-wave plates before and after the EOM to couple light into the correct polarization and also to leave room for changing polarization on demand. Eventually, both lasers are sent through a band-pass filter to remove possible extra noise outside the central wavelength. This can be crucial for single-photon level experiments, where even a tiny amount of unfiltered light at a different wavelength can easily saturate the APD.

The red power used in the experiment can be roughly estimated from the lifetime and quantum efficiency of the SiV centers. From the spontaneous decay formula

$$\Gamma_{\text{tot}} = \Gamma_{\text{rad}}/\text{QE} = \frac{\omega^3 n |\mu_{ge}|^2}{3\pi\epsilon_0 \hbar c^3 \text{QE}},$$

and the two-level steady-state excited-state population [Equation B.2](#)

$$\rho_{ee} = \frac{1}{2} \times \frac{1}{1 + \Gamma_{\text{tot}}^2/2\Omega^2},$$

with the Rabi frequency $|\Omega| = E|\mu_{ge}|/\hbar$ and the Airy-disk Gaussian approximation $\omega_0 \approx 0.42\lambda/\text{NA}$ and Gaussian-beam relations $P = \pi\omega_0^2 I_{\text{peak}}/2$, $I = n\epsilon_0 c E^2/2$. We obtain (assuming the laser spot is at the boundary so $n = 1$, while the emitter is inside diamond with $n = 2.4$)

$$P_{\text{sat}} = \frac{1.823}{(\text{NA})^2} \times \frac{n f \hbar \Gamma_{\text{tot}}}{\text{QE}},$$

that saturation power is defined as the RF power satisfied $2\Omega^2 = \Gamma_{\text{tot}}^2$. Using $f = 4 \times 10^{14} \text{Hz}$, $\Gamma_{\text{tot}} = 1/1.6 \text{GHz}$, $n = 2.4$, $\text{QE} \sim 0.1$, $\text{NA} = 0.8$, and $\hbar \approx 1 \times 10^{-34} \text{J}\cdot\text{s}$, we

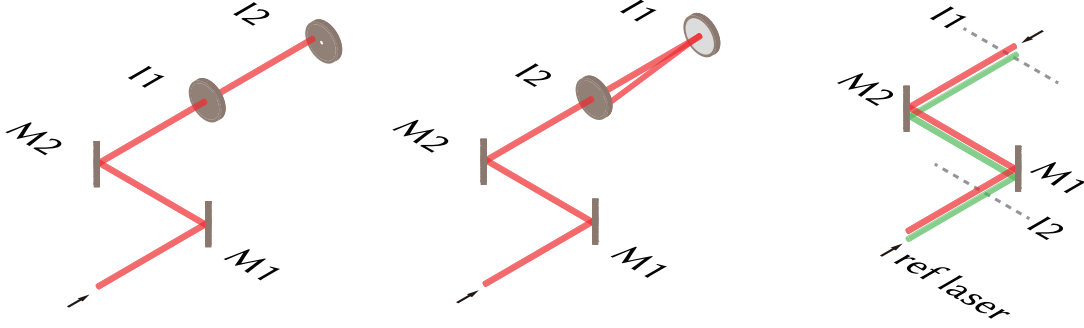


Figure 3.6. Illustration of three different alignment scenarios. The left panel shows the standard two mirror alignment. The center panel presents a variant used to achieve normal and centered incidence. The right panel illustrates a configuration often used for fiber coupling alignment, where sending the laser backwards through the fiber greatly reduces the alignment effort.

find $P_{\text{sat}} \approx 1.8\text{nW}$. Considering that we need about $\sim 100P_{\text{sat}}$ to saturate the SiV and also to compensate for alignment errors, and taking into account polarization mismatch due to the diamond cut orientation and reduced effective QE for the C transition due to decay to another transition, $\sim 1\mu\text{W}$ is the typical excitation power we should use in the experiment.

3.5 Controlling and alignment

3.5.1 Alignment approaches

Alignment is at the core of AMO experiments, and can be grouped into three different categories. The first is aligning a laser beam to overlap with a desired path, which is also the most common category. The second is aligning a beam to be normally incident on and centered with respect to a plane, e.g. a lens or objective, which greatly affects the performance of the optical component. The last is collimating a beam so that it can travel a long distance or achieve diffraction-limited performance at the objective.

To align a laser beam to overlap with a desired path, a two-mirror configuration as shown on the left of [Figure 3.6](#) is normally used, where the desired path is defined by two irises I_1 and I_2 . In this configuration we first align M_1 so that the laser beam hits the center of I_1 and then M_2 for I_2 . After a few iterations, the laser beam will overlap with the path I_1 – I_2 . We can also show that the convergence speed of this procedure depends on the ratio $d_{I_1 M_2}/d_{M_1 M_2}$ by using the ABCD-matrix formalism and solving the sequence. This can also be intuitively understood: in the limit $d_{I_1 M_2} \rightarrow 0$, the alignment on M_1/I_1 is unaffected by the M_2/I_2 operation and is therefore efficient for I_1 alignment; and vice versa, because the change on M_1 at each iteration is small, the effect on the alignment of I_2 is also small. A variant of such a configuration is shown on the right of [Figure 3.6](#), which often appears in fiber-coupling alignment, where we do not have enough space to set two irises physically and the path is defined by another laser beam coming backwards from the fiber. We could use a paper card and pick two points before M_1 as irises, but the distance $d_{I_1 I_2}$ is then not enough to distinguish a small angular error. Instead, we choose an iris I_2 near and before M_1 and an iris I_1 after and far from M_2 . In this case the convergence speed can still be guaranteed, given a small $d_{I_1 M_2}/d_{M_1 M_2}$.

Aligning the laser beam to be normally incident on and centered on a plane corresponds to the configuration illustrated in the middle of [Figure 3.6](#). We can also convert this into a variant of the two-mirror configuration in which the first iris is defined by the center on the plane, and the second iris is defined by the overlap between the incident laser beam and the reflection from the plane by using a threaded mirror mounted on that plane. The overlap between the incident and reflected beams indicates normal incidence on the plane, and, similar to the

discussion above, the second iris is not required to be placed after M_2 but can be anywhere, provided a small $d_{I_1M_2}/d_{M_1M_2}$ is satisfied. Additionally, in some cases it may be preferable to orient the plane or optical component to align to the irises when the beam path needs to be fixed.

To collimate a laser beam, we aim to transform it into a beam with a large beam waist and therefore a long Rayleigh distance, as can be seen from the Gaussian-beam formula $z_R = \pi\omega_0^2/\lambda$. For a 532~nm laser, a waist diameter $2\omega_0 = 2\text{mm}$ gives $z_R \approx 6\text{m}$, which can propagate over the optical setup without significant divergence or convergence. For a laser beam coming out of a single-mode fiber, it is easiest to use a fiber collimator that is compatible with the fiber model. When a tunable zoom lens is available, precise collimation can be obtained by tuning the convergence of the zoom lens and checking collimation with a shearing interferometry setup, in which the interference pattern generated from a tilted wedge indicates collimation. In other cases, we need to use lenses to collimate the beam. Using a single lens for collimation is relatively straightforward when the beam is significantly diverging or converging, in which case the beam waist can be identified and the desired focal length of the lens can then be estimated. However, when this is not the case, finding the correct lens can be time-consuming. A helpful preprocessing step is to use a short-focal-length lens to create a well-defined beam waist before using the collimation lens. This can be understood from the Gaussian-beam formula [Equation B.14](#), which shows that a short- f lens focuses the incident beam to approximately the focal distance regardless of the initial beam parameters.

In addition to these alignment approaches, when aligning the objective we can also use the reflection of the excitation laser from a planar surface to verify normal incidence. The reflected spot typically appears as a ring-like interference

pattern. Small tilts of the objective or the sample break the rotational symmetry of this ring, so restoring a circular and concentric pattern provides a sensitive and convenient way to detect and correct small deviations from normal incidence.

3.5.2 Optics and electronics gating

As we discussed before, in many applications we desire fast gating of the laser excitation, and this is done by sending the laser beam through an AOM. A first-order diffraction-efficiency formula for the AOM can be found in [Equation B.15](#), which gives the dependence on the driving RF power P_{RF} and the Bragg-angle error δ . The dependence on δ is

$$\eta_{0 \rightarrow 1}(\delta) \propto \text{sinc}^2 \left(\frac{\pi L \delta}{2 \lambda_a} \right),$$

where L is the thickness of the AOM crystal and λ_a is the wavelength of the acoustic wave. From this formula, we see that for a highly converging beam at the AOM the rise/fall time will be short, since $\tau_{\text{rise}} \propto \omega_0 / \lambda_a$, but the diffraction efficiency will drop due to a large average Bragg-angle error. Additionally, when an extremely short rise/fall time is required, we often use an EOM for gating instead of an AOM.

In many experiments it is also desirable to gate the RF signal, in close analogy to optical gating. As in the optical case, we care about the rise and fall time of the gate as well as the extinction ratio. A frequency mixer is often used for this purpose. It has three ports: LO (local oscillator), IF (intermediate frequency), and RF (radio frequency). The output at the RF port contains components at $f_{\text{RF}} = f_{\text{LO}} \pm f_{\text{IF}}$, corresponding to frequency mixing between the LO and IF signals. This mixing is realized by introducing diodes so that the current–voltage relation becomes nonlinear and generates products of the voltages applied on either side of the diode. A double-balanced frequency mixer, e.g. the ZAD-1-1+ from

Mini-Circuits, uses a diode ring and a balanced transformer network to provide good isolation between the ports and to suppress unwanted terms, which in turn helps to achieve a high extinction ratio when used as a gate. When the IF port is driven by a gating square pulse and the LO port is driven by a continuous-wave RF signal generator, the mixer acts approximately as a multiplier that gates the RF signal at the output port. The temporal response of such a mixer gate is limited by the bandwidth of its ports: because the device cannot faithfully transmit all high-frequency harmonics of the square-wave drive, the resulting gating waveform cannot be perfectly rectangular and has a finite rise and fall time. When available, the internal pulse-modulation function of an RF signal generator can also be used to implement RF gating, often with well-specified edge times and extinction ratios. For applications operating at higher carrier frequencies or requiring higher isolation, suitable frequency mixers may not provide sufficient isolation or may be inconvenient to use. In this case a dedicated high-speed RF switch, e.g. the ZASWA-2-50DRA+, can be used to realize nanosecond-scale edges and high isolation, at the cost of requiring external power supplies.

When we are dealing with electronic gating, impedance matching between different parts of the circuit is crucial for maintaining the rise/fall time. For the radio-frequency (RF) and microwave (MW) signals in the GHz range that we use for AMO experiments, the wavelength becomes comparable to the physical dimensions of cables and circuit components, so we can no longer use simple circuit models and must instead rely on transmission-line theory. A central concept in transmission-line theory is impedance matching between different parts of the circuit: if the characteristic impedance Z_0 of the line is not matched to the load impedance Z_L , part of the signal is reflected at the boundary and the propagating

waveform is distorted. Intuitively this is analogous to reflection at an interface between optical media with different refractive indices. In practice, RF systems are therefore designed around a well-defined characteristic impedance (typically 50Ω), and terminations and components are specified to match this value as closely as possible. One important consequence for pulsed operation is that waveforms with significant high-frequency content, such as square pulses with fast edges, are particularly sensitive to imperfect impedance matching. Because the effective impedance of cables, connectors, and components is often frequency dependent, the higher-frequency spectral components of the pulse can experience stronger reflections and dispersion. This leads to waveform distortion at the load, including overshoot, ringing, and a degradation of the effective rise/fall time, even if the nominal pulse shape at the source is ideal.

3.5.3 Spatial filter

As discussed in the appendix [Equation B.13](#), a paraxial laser beam can be expanded in the Hermite–Gaussian (HG) basis $HG_{lm}(x, y)$. In ideal single-mode operation the transverse field is well described by the fundamental Gaussian mode HG_{00} or TEM_{00} , but in practice imperfections such as imperfect beam shaping at the laser output, aberrations in the AOM, or other optical components distort the beam and populate higher-order HG components. These higher-order modes are generally not axisymmetric and exhibit poorer focusing properties and shorter effective Rayleigh ranges compared to the fundamental Gaussian mode. It is therefore desirable to suppress the higher-order content and recover a beam close to the Gaussian mode.

A standard way to achieve this is a spatial filter based on a $4f$ configuration. A collimated input field $E_{\text{in}}(x, y)$ is first focused by a lens of focal length f , and

a small pinhole is placed in the focal plane. Since higher-order modes are focused to a spot size larger than that of the Gaussian mode, the pinhole can be chosen such that it transmits most of the Gaussian beam while suppressing higher-order modes. In the scalar Fourier-optics approximation, the complex field in the focal (far-field) plane is proportional to the two-dimensional Fourier transform of the input field. As the pinhole in the focal plane can be modeled as a pupil function $P(X, Y)$, the resulting far-field distribution can be understood as the convolution between the Fourier transform of the pupil function, which gives an Airy-disk pattern, and the Fourier transform of the Gaussian mode, which is also Gaussian. The resulting intensity pattern shows a bright central lobe with surrounding rings. By using another iris to select only the central part, the transmitted beam is approximately Gaussian, since the central region is close to the product of two Gaussian distributions.

In practice, the pinhole radius is chosen to be on the order of $1-2w_0$ of the focused fundamental spot size in the Fourier plane. A too-small pinhole strongly truncates even the HG_{00} component, leading to large diffraction losses and pronounced rings, whereas a too-large pinhole transmits significant higher-order content and reduces the filtering effect. With an appropriate choice of pinhole size and careful alignment, the spatial filter provides an output beam whose central lobe is well approximated by a Gaussian TEM_{00} mode, with higher-order spatial structure suppressed to a level sufficient for most AMO applications.

3.6 Spin experiment of NV

As discussed in the theory chapter, the ground-state spin triplet ($S = 1$) of the NV center is partially split even at zero magnetic field by the spin-spin interaction, giving a zero-field splitting D between the $|m_s = 0\rangle$ and $|m_s = \pm 1\rangle$

sublevels. An applied magnetic field further lifts the degeneracy between $|m_s = +1\rangle$ and $|m_s = -1\rangle$, so that individual spin transitions can be addressed optically or with microwaves. These spin sublevels are relatively well isolated from charge and orbital degrees of freedom and therefore exhibit comparatively long coherence times, which is favorable for quantum control.

For NV centers, the most common way to manipulate the spin states is to use resonant microwave (MW) fields. A coplanar waveguide (CPW) or a metal wire is placed close to the diamond sample, and when driven at the resonance frequency it generates an oscillating magnetic field B_{driving} at the NV position. This B_{driving} field drives coherent spin rotations with a Rabi frequency $\Omega = \gamma_e B_{\text{driving}}$, where γ_e is the electron gyromagnetic ratio. Compared to optical driving, microwave control typically requires much higher input power to reach a given Ω . The main reason is that the spin transition couples via the magnetic dipole moment $\mu \sim \mu_B = e\hbar/(2m_e)$, which is several orders of magnitude smaller than typical optical electric dipole moments $d \sim ea_0$ (with a_0 the Bohr radius). In addition, microwave fields cannot be focused to a diffraction-limited spot as tightly as optical beams, the coplanar waveguide geometry does not realize a tight free-space focus, and the intrinsic diffraction-limited spot size at MW wavelengths (centimeter scale) is much larger. As a result, the achievable B_{driving} field per unit input power is relatively small, and tens of dBm of microwave power are often required to achieve fast spin rotations in NV experiments.

Spin experiments with NV ensembles are performed at room temperature, and spin initialization and readout are achieved through off-resonant 532nm excitation. When the excited state decays spontaneously, some fraction of the population is transferred to an intermediate state that does not emit fluorescence

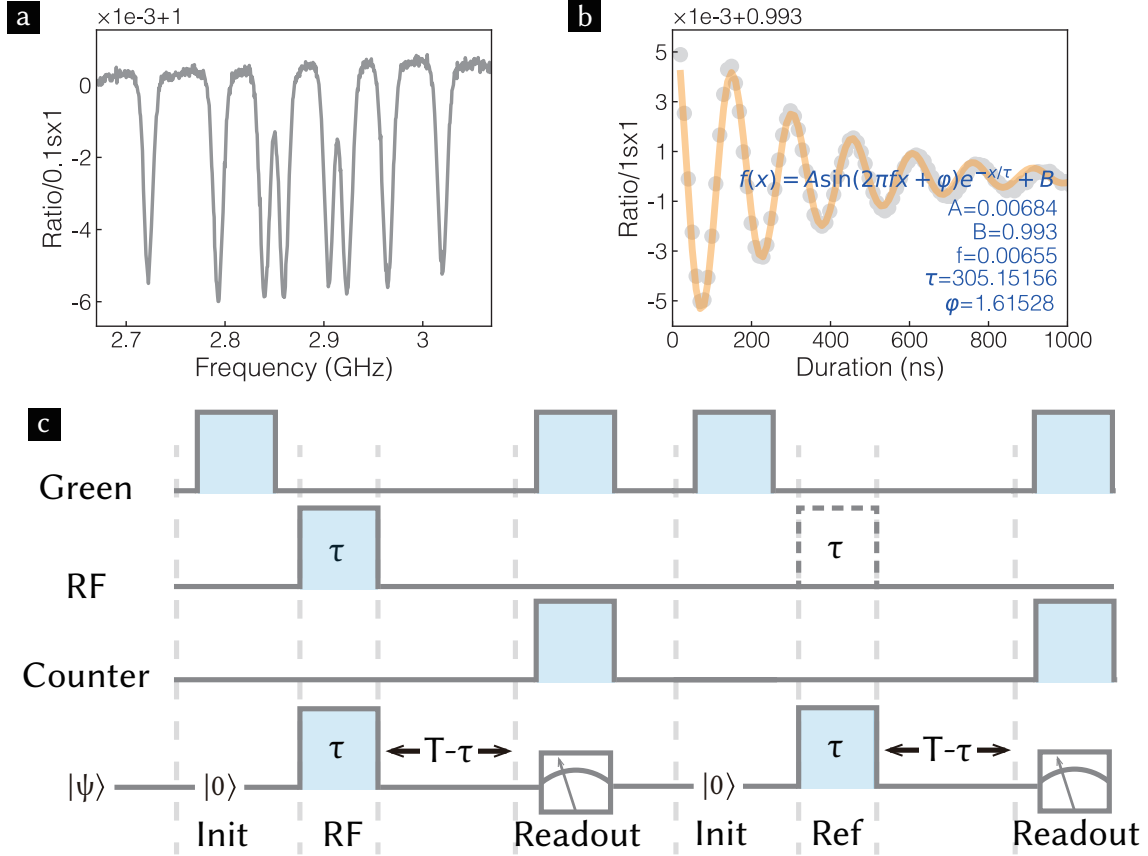


Figure 3.7. ODMR (a) and Rabi oscillations (b) of an NV ensemble at room temperature. An external magnetic field leads to the observation of eight dips in (a), corresponding to four different orientations groups of the NVs. The y -axis shows the ratio between the signal from the experimental pulse sequence and a reference pulse with the microwave turned off, allowing us to compensate slow drifts in the absolute fluorescence and to track relative changes at the 0.1% level. Subtracting the reference signal from the data is equivalent to a lock-in type technique, commonly used to enhance a small signal on top of a noisy background. The pulse sequence in (c) illustrates the experimental procedure in the time domain.

in the detection spectral range. Moreover, the process from the intermediate state to the ground state is not spin-conserving and is spin-dependent. This leads to an optical spin-pumping process that is used to pump into the $|m_s = 0\rangle$ spin state for initialization, and the unequal fluorescence intensities distinguish different initial spin states for spin readout. A MW field drives $|m_s = 0\rangle$ to another spin state, and once the frequency is on resonance with a specific spin transition, a change in the fluorescence intensity occurs, as shown in [Figure 3.7a](#). This is called optically detected magnetic resonance (ODMR). The four sets of ODMR dips correspond to four different orientations of the NV axis in a dense ensemble sample, with each group of NVs sharing the same $|m_s = 0\rangle \rightarrow |m_s = +1\rangle$ and $|m_s = 0\rangle \rightarrow |m_s = -1\rangle$ resonances. This is a consequence of the fact that, for NVs within an area of order $\sim \lambda^2$, magnetic-field gradient differences are negligible. Similarly, applying a MW drive resonant with a specific spin transition for different durations implements standard Rabi oscillations between two spin states, as shown in [Figure 3.7b](#). As the duration becomes longer, decoherence in the system smears out the contrast, and the system ends up in an equally mixed state between different spin states without interference fringes. The experimental procedure is illustrated in [Figure 3.7c](#). The first half of the sequence is used to acquire experimental data, and the second half is used to acquire reference data in the absence of the RF pulse. Taking the ratio between the signals from the experimental and reference pulse sequences allows us to compensate for slow drifts in the absolute fluorescence. We also introduce a $T - \tau$ gap after the RF pulse of duration τ so that the total sequence duration is kept fixed, which improves the stability of the experiments. The initialization and readout durations are determined separately by performing time-resolved fluorescence measurements. The initialization time is chosen to be long enough to

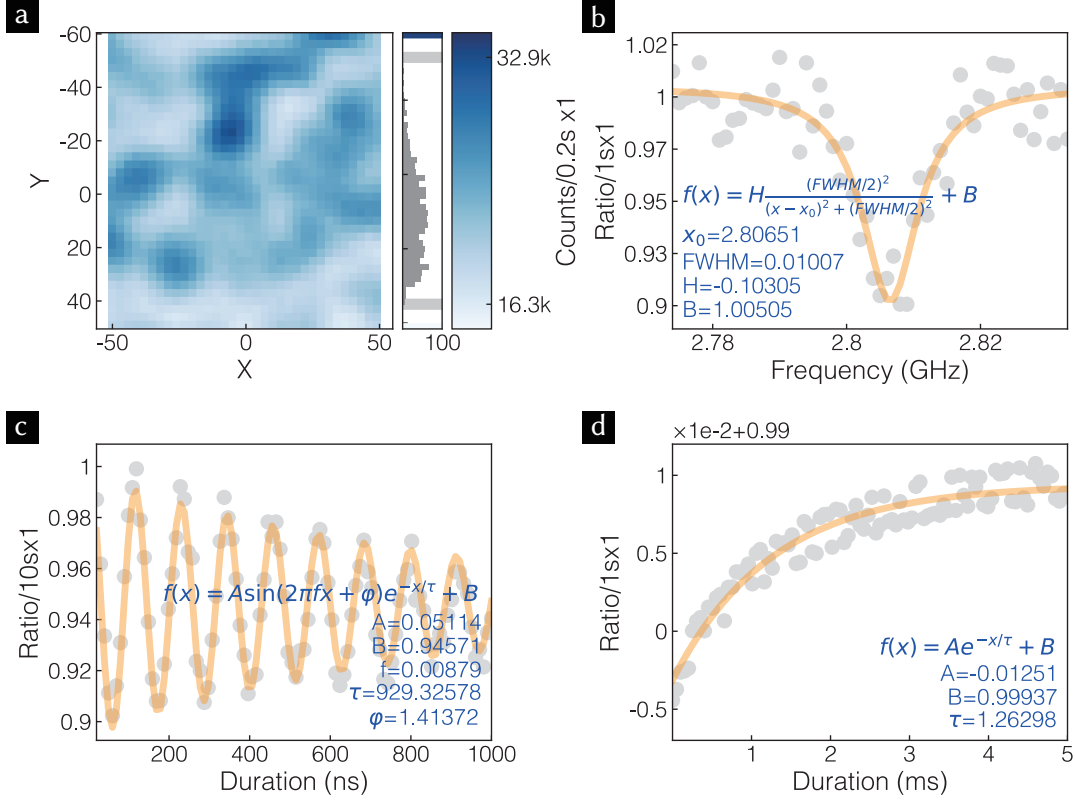


Figure 3.8. (a) Confocal PL image of NV centers in diamond. (b) ODMR spectrum of a single NV center, showing the transition $|m_s = 0\rangle \rightarrow |m_s = -1\rangle$ at 2.8065GHz. (c) Rabi oscillations of a single NV center driven on the $|m_s = 0\rangle \rightarrow |m_s = -1\rangle$ transition. (d) T_1 relaxation measurement, yielding a spin lifetime of ~ 1 ms.

ensure sufficient spin polarization without making the sequence unnecessarily long, whereas the readout window is optimized to maximize the fluorescence contrast between $|m_s = 0\rangle$ and $|m_s = \pm 1\rangle$. If the readout window is too long, continued green illumination further polarizes the spin and thereby smears out the contrast.

Although NV ensemble experiments are sufficient for many applications, they intrinsically have weaker contrast and shorter coherence times compared to single-NV experiments. Due to the four different orientations in the system and the additional broadening of the transition-frequency distribution within the same group, the spin-readout contrast is about $10\times$ weaker compared to a single

NV. The relatively high NV density in the sample also makes the environment noisier and leads to shorter spin lifetimes and coherence times. For single-NV experiments, we have optical access to an individual NV by selectively exciting one center, as shown in Figure 3.8a, and the spin transition frequencies can also be identified, as shown in Figure 3.8b. A Rabi oscillation measurement (c) and T_1 -time measurement (d) show a coherence time of $\sim 1\mu s$, compared to the $\sim 300ns$ case for the ensemble, and a T_1 time of $\sim 1ms$. The T_1 -time experiment is performed by varying the duration of the gap between initialization and readout; here we initialize to a state different from $|m_s = 0\rangle$ by applying a $\pi/2$ pulse after optical spin pumping. Strictly speaking, the coherence time measured from Rabi oscillations is not T_2 or T_2^* , which would normally require spin-echo or Ramsey-type experiments, but it still provides information on the scale of the spin coherence time.

CHAPTER IV

MICROSPHERE ENHANCED CONFOCAL EXPERIMENT

The results presented in this chapter were published in [S.-H. Wu et al. \(2024\)](#).

4.1 Introduction

Color centers in diamond, especially, nitrogen vacancy (NV) centers are one of the most widely used systems for quantum sensing [Degen et al. \(2017\)](#); [Giovannetti, Lloyd, and Maccone \(2011\)](#). NV-based quantum sensing has been employed for measurements of magnetic field, electric field, temperature, and strain with nanometer spatial resolution [Doherty et al. \(2014\)](#); [Dolde et al. \(2011\)](#); [Kucsko et al. \(2013\)](#); [Taylor et al. \(2008\)](#). The poor collection efficiency of fluorescence from these emitters, however, limits their real-world applications. This inefficiency primarily arises from the substantial mismatch in the refractive index between diamond and air such that a large fraction of the emission from color centers undergoes total internal reflection at the interface. A variety of optical techniques, such as diamond solid immersion lense (SIL) [Siyushev et al. \(2010\)](#), diamond nanopillars [Neu et al. \(2014\)](#), and metalenses [Huang et al. \(2019\)](#), have been exploited to overcome this limitation. These techniques require costly nanofabrication and/or precise positioning of the color centers. For diamond SIL, which is the standard approach for enhancing fluorescence collection, the color center needs to be at or near the center of a hemisphere with a radius of several μm , making it unsuitable for applications where the color center needs to be within tens or at least hundreds of nm from the diamond surface. Furthermore, the non-directional nature of fluorescence necessitates the use of a high numerical aperture (NA) objective, which is expensive and has a working distance (WD) of a few

mm or shorter, to achieve the necessary collection efficiency, limiting practical applications.

Dielectric microspheres have been used in super-resolution imaging, conveying high-frequency information to the far field when placed near the object [Krivitsky, Wang, Wang, and Luk'yanchuk \(2013\)](#). These studies also suggest the potential for microsphere-enhanced fluorescence collection. Enhanced collections using microspheres have been demonstrated for quantum dots [Biccari et al. \(2020\)](#); [L. Yang et al. \(2019\)](#) as well as NV centers in diamonds [Moon et al. \(2021\)](#), demonstrating significant improvements in collection efficiency as well as a reduction in saturation power for optical excitation of NV centers. Nevertheless, a few important issues remain to be addressed. There have been no investigations on the microsphere size dependence. More importantly, it is not clear whether near optimal performance of microsphere-enhanced fluorescence collection can be achieved with low-NA objectives, which are relatively inexpensive and can feature WD exceeding several centimeters. A quantitative theoretical description of the enhancement has also been lacking.

In this chapter, we present experimental studies of microsphere-enhanced fluorescence collection of NV centers using silica microspheres with diameters ranging between 15 and 50 micrometers and employing 40x and 20x objectives. The experimental results are compared with a theoretical model based on Mie scattering. Photoluminescence excitation (PLE) saturation counts as high as 95 kHz have been observed with the 20x objective, which surpasses the maximum saturation counts of 60-80 kHz typically achieved with a 0.95 NA objectives and with no enhanced collection [Irber et al. \(2021\)](#); [Lovchinsky et al. \(2016\)](#); [Siyushev et al. \(2010\)](#). We show that due to effective collimation of fluorescence by a

microsphere, relatively low NA objectives can be used without sacrificing collection enhancement. Our theoretical analysis confirms the experimental observation and indicates that for the size of the microspheres used, the enhancement is primarily due to geometric optical processes, with minor contributions from evanescent coupling between whispering gallery modes (WGMs) and the NV centers.

4.2 Experimental results

We fabricated silica microspheres by melting the tip of an optical fiber using a CO₂ laser. By controlling the laser power and the duration of melting, we can achieve a desired diameter ranging from 10 μm to 100 μm . The remaining fiber stem attached to the microsphere helps transport it to any position on the diamond sample with a vacuum pickup tweezer. A tight contact between the microsphere and the sample surface can be confirmed with the observation of a Newton ring pattern, as shown in the lower right corner of [Figure 4.1](#). Van der Waals forces secure the microsphere in place throughout the experiment. More elaborate methods involve gluing the fiber stem to a fixed position and moving the sample with nanopositioners or using an optical tweezer to position a microsphere without stems [Moon et al. \(2021\)](#), showcasing the flexibility for in-situ positioning of a microsphere. The type IIa [100] diamond sample used is from Element 6, Inc. The NV centers were created about 100nm below the surface through ion implantation, followed by stepwise high temperature thermal annealing, and wet chemical oxidation.

A standard confocal optical setup is employed for photoluminescence (PL) and photoluminescence excitation (PLE) measurements of the NV center, as illustrated in [Figure 4.1](#). The diamond sample is cooled to cryogenic temperatures (near 5 K) and excited with a non-resonant 532 nm laser for PL or a resonant 637

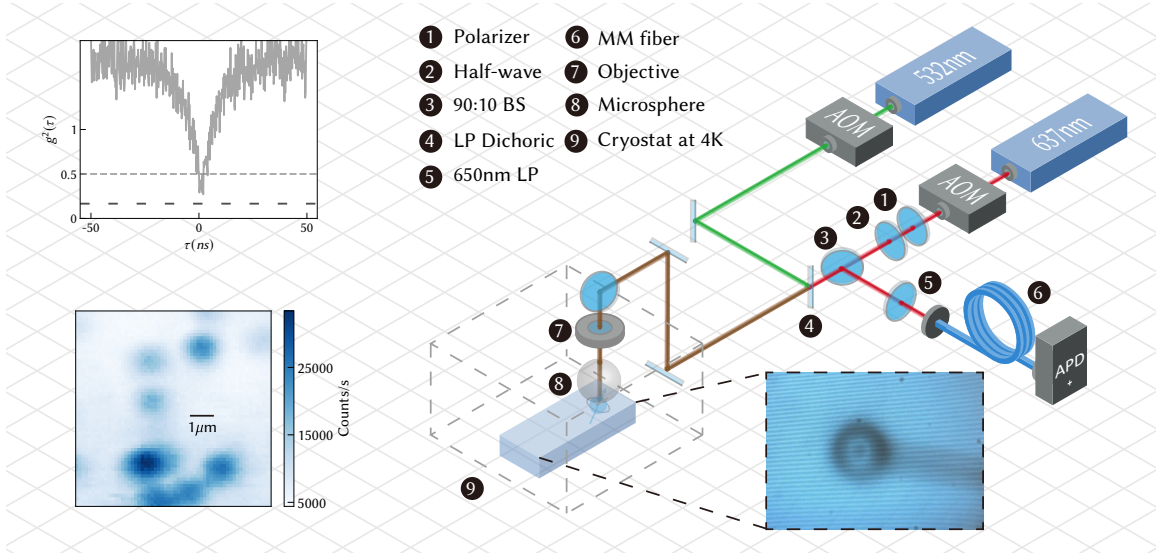


Figure 4.1. Experimental setup for microsphere-enhanced fluorescence collection. Right: A confocal optical setup. A microsphere is positioned above the diamond membrane. The observation of a Newton’s ring pattern confirms a tight contact. A 637nm laser is used for the resonant excitation of the E_x transition of the NV centers, while a 532nm laser is pulsed occasionally to initialize the NV centers and to stabilize the charge state. The fluorescence’s phonon sidebands are collected through a 650nm long-pass filter. Lower left: Confocal PL imaging of NV centers under the microsphere, obtained under non-resonant optical excitation. Upper left: Time-correlation measurement, carried out with a 50:50 fiber splitter and two APDs.

nm laser for PLE. For PLE studies, we scan the 637 nm laser across E_x transition of the NV center by tuning a voltage based on feedback from a wavemeter.

The fluorescence from the NV centers is sent through a 90:10 beamsplitter and a 650 nm long-pass filter, then coupled into a multi-mode (MM) optical fiber connected to an avalanche photodiode (APD). While dependences on microsphere sizes, objective NAs, as well as MM fiber core diameters are investigated in this work, a 50 μm MM fiber, 25 μm microsphere, and 40x objective are used in the experimental results presented here unless otherwise specified.

A confocal PL imaging of NV centers obtained with the use of a silica microsphere is shown in the lower left of [Figure 4.1](#), where individual NV centers

are identified within the contact area. We also conducted a $g^{(2)}$ time correlation experiment for one of the NV centers using a 50:50 fiber splitter and two APDs, the result of which is shown in the upper left of [Figure 4.1](#). The dashed grey line indicates the 0.5 criterion for single-photon emitters. The dashed black line denotes the estimated background correlation arising from background counts. Note that PL counts of NV centers depend on the relative position of the NV centers to the center of the contact area. For studies of microsphere size dependence, we chose two individual NV centers under each sphere, which exhibit relatively high count rates.

To quantify the enhanced collection provided by a microsphere, the most common method is to use PL saturation counts [Biccari et al. \(2020\)](#); [Moon et al. \(2021\)](#). For our studies, this approach is complicated by the background counts from nearby NV centers, especially when relatively high optical excitation powers are used. In comparison, PLE saturation counts of the NV center are unaffected by background counts, as we can always perform curve fitting in PLE spectra and extract the background-free peak PLE counts, as shown in [Figure 4.2\(a\)](#). [Figure 4.2\(b\)](#) shows the saturation of the peak PLE counts with increasing optical excitation powers. The saturation follows the equation

$$I = I_{\text{sat}} \left(\frac{P/P_{\text{sat}}}{1 + P/P_{\text{sat}}} \right), \quad (4.1)$$

where I_{sat} and P_{sat} represent the saturation counts for PLE and the saturation optical power, respectively. Theoretically, the saturation counts are determined by spontaneous decay rate of the NV center, Γ , which should be uniform among different samples, and collection efficiency, η , by $I_{\text{sat}} = \eta\Gamma/2$, thus providing a robust approach to extract and compare collection efficiency from the saturation curve. For comparison, [Figure 4.2\(b\)](#) presents the saturation curves obtained with

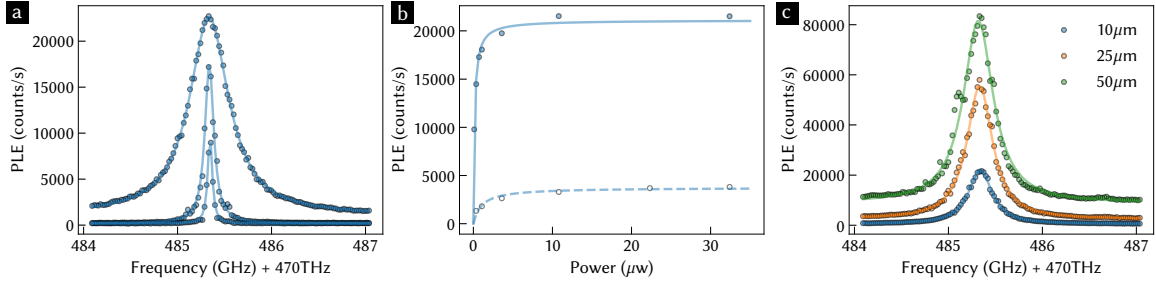


Figure 4.2. Photoluminescence excitation spectra and saturation photon counts. (a) PLE spectra with increasing optical power (0.11 μW , 1.1 μW , 32.4 μW) showing power broadening, saturation, and increasing background contributions. Solid lines are the least square fit to a Lorentzian. (b) Peak PLE counts extracted from the corresponding PLE spectra vs the optical excitation power. Solid (open) circles are data obtained with (without) a microsphere. Solid and dashed lines are numerical fits to the equation in the text. (c) PLE spectra obtained with an optical power of 10.8 μW and with a MM fiber featuring a diameter of 10 μm , 25 μm , and 50 μm , respectively, showing reduced background contribution as well as reduced collection efficiency with decreasing fiber diameters.

and without the use of a microsphere, indicating that the use of a microsphere enhances the collection efficiency by a factor of 5.7 and reduces saturation power by a factor of 7. The reduction in the saturation power, or in other words, the super-focusing or photonic nanojet effect, is related to the microsphere's ability to focus a laser beam beyond the diffraction-limited spot size. This effect, discussed in Darafsheh (2021), plays a significant role in super-resolution imaging using microspheres Krivitsky et al. (2013). Note that different NV centers were used for the two curves in Figure 4.2(b) because of the difficulties of tracking the same NV center during positioning of the microsphere. It should also be added that the background counts in PLE can be reduced with the use of MM fibers of smaller diameters, though with a corresponding decrease in the overall collection efficiency (which is in large part due to the nonoptimal optical alignment for coupling fluorescence into the MM fiber), as shown in Figure 4.2(c).

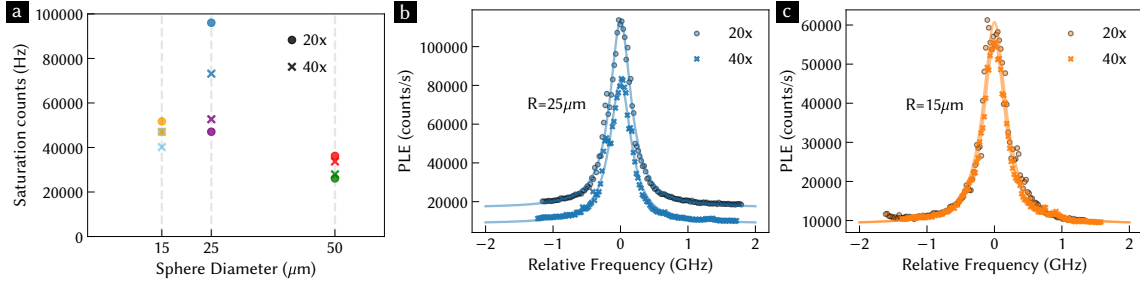


Figure 4.3. Dependence of saturation photon counts on microsphere size and objective NA. (a) Saturation PLE counts obtained with microspheres with three Different diameters and objectives with two different NAs, with each color representing a different NV center. (b) and (c) PLE spectra obtained with an optical power of 10.8 μ W and with a microsphere with 25 μ m and 15 μ m in diameter, respectively. Saturation counts extracted from the PLE spectra are also shown in (a).

Figure 4.3 shows the saturation PLE counts as well as PLE spectra obtained with microspheres featuring three different sizes and objectives featuring two different NAs. As shown in Figure 4.3(a), saturation counts as high as 95 kHz are observed with a 25 μ m sphere and a 20x objective (NA=0.42). For comparison, typical saturation PL counts are around 60 to 80 kHz with a 0.95 NA objective and without enhanced collection Irber et al. (2021); Lovchinsky et al. (2016); Siyushev et al. (2010), and the corresponding PLE saturation counts are expected to be even lower. Our maximum saturation counts represent an 11.4-fold enhancement over the saturation counts obtained without a microsphere under otherwise the same conditions, surpassing the theoretical enhancement of 6.11 provided by diamond SIL under otherwise the same conditions Siyushev et al. (2010). Because of the different lateral offsets of NVs to the center of the respective contact area as well as the variations in the optical alignment when different microspheres are used, Figure 4.3(a) does not provide conclusive results on the dependence of the enhancement in florescence collection on the microsphere diameter. Nevertheless, there does not seem to be evidence for strong size dependence.

Given that the enhancement could be affected by non-optimal alignment without a microsphere, a comparison of absolute saturation counts is beneficial. From the state-of-the-art saturated PL counts of near 1 MHz for NV centers in [111] orientation diamond and the theoretical simulation of diamond SIL [Robledo et al. \(2011\)](#), we estimate a 133 kHz PL saturation count under a 20x objective with the SIL. Considering the differences between PL and PLE, the orientation of the diamond, and potential alignment issues in our experiment, the 95 kHz PLE saturation count achieved with a silica microsphere and a 20x objective is noteworthy.

In addition, [Figure 4.3\(a\)](#) shows that despite variations due to misalignment during changes of objectives, both the 20x and 40x objectives yield similar saturation counts. For a more detailed comparison, [Figure 4.3\(b\)](#) and (c) show saturated PLE spectra obtained with an optical power of 10.8 μW and with a microsphere of 25 μm and 15 μm in diameter, respectively, which indicate that the 20x objective enables similar collection efficiency as the 40x. In addition to the much-reduced cost, the use of a 20x objective with a WD of several centimeters is especially important for applications that require a long WD. Overall, these studies suggest that after collimation by the microsphere, optical emissions from a NV center become highly directional, which we will discuss in more detail in the following theoretical analysis based on Mie scattering.

We have developed a theoretical model to describe microsphere-enhanced fluorescence collection by using Mie scattering. Earlier theoretical analysis has largely relied on finite element (FEM) numerical simulations. The Mie scattering approach provides a rigorous solution to Maxwell's equations, describing the scattering of a plane wave by a sphere. In this context, we follow the notations

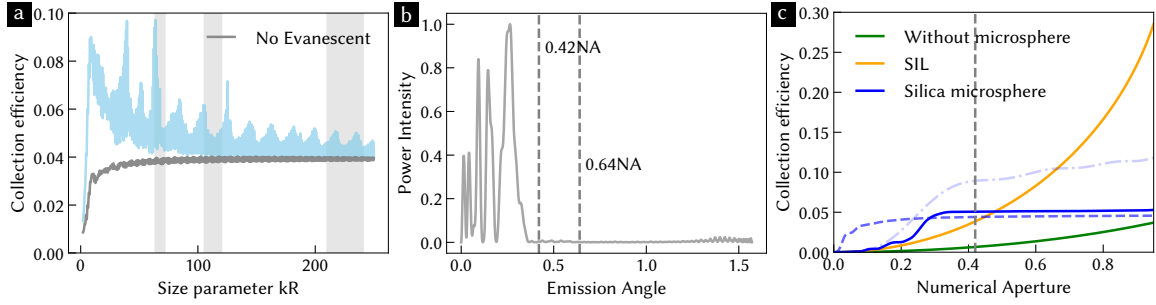


Figure 4.4. Theoretically calculated collection efficiency and angular distribution. (a) Theoretical collection efficiency (blue curve) obtained with the use of a silica microsphere and a 0.42 NA objective. The size parameter kR is the product of the wavevector and microsphere radius. The grey curve denotes the same simulation but removes contributions from evanescent waves. For a NV center with an emission spectrum from 650 to 750 nm, the collection efficiency can be obtained by averaging a given shaded area in the figure, which corresponds to microspheres with diameters of 15 μm , 25 μm , and 50 μm , respectively. (b) Simulated angular distribution of the emission power after collimation by a microsphere. Most of the emission falls within the collection range of the 0.42 NA objective, consistent with the experimental observations. (c) Comparison of theoretical collection efficiencies for various collection approaches. The solid blue, green, and orange lines denote the collection efficiencies obtained with a silica microsphere of 25 μm in diameter, without a microsphere, and with a diamond SIL, respectively. The dot-dashed line denotes the collection efficiency for a silica microsphere with a diameter of 1.8 μm . The dashed line denotes the collection efficiency for a microsphere with a diameter of 25 μm and a refractive index of 1.8.

from [Ma and Li \(2010\)](#); [Wriedt and Doicu \(1998\)](#), which include a modified Mie scattering problem with reflection from a nearby planar surface. To simplify the Mie solution without losing generality, we model the emission from the NV center as a spherical wave with radial polarization. By applying the plane wave decomposition (Weyl expansion) to the spherical wave and considering the Fresnel reflection at the interface, we derive the plane wave expression of the emission from the NV center after diamond-air surface as

$$\mathbf{E}_i(\mathbf{r}) = \int \mathbf{e}_\theta e^{i\mathbf{k}\mathbf{r}} d\theta d\varphi = \sum_{n=1}^{\infty} \frac{(-i)^{n+1}(2n+1)2\pi}{n(n+1)} a_n^i N_{0n}^1(k\mathbf{r}), \quad (4.2)$$

where

$$a_n^i = \int \sin(\theta) P_n^1(\cos \theta_t) e^{ikR \cos \theta_t} \frac{2n_d \cos \theta}{\cos \theta + n_d \cos \theta_t} d\theta, \quad (4.3)$$

$$\theta_t = \sin^{-1}(n_d \sin \theta).$$

The corresponding scattered field is,

$$\mathbf{E}_s(\mathbf{r}) = \sum_{n=1}^{\infty} \frac{(-i)^{n+1}(2n+1)2\pi}{n(n+1)} a_n^s N_{0n}^3(k\mathbf{r}), \quad (4.4)$$

where

$$a_n^s = -a_n^i \frac{n_s^2(\rho j_n(\rho))' j_n(\rho_1) - (\rho_1 j_n(\rho_1))' j_n(\rho)}{n_s^2(\rho h_n(\rho))' j_n(\rho_1) - (\rho_1 j_n(\rho_1))' h_n(\rho)}, \quad (4.5)$$

$$\rho = kR, \rho_1 = n_s kR,$$

$N_{0n}^{(i)}(k\mathbf{r})$ denotes the vector spherical harmonics with i-kind radial function, and k, n_d, n_s, R denote the wave vector, refractive index of diamond, refractive index of the sphere, and sphere radius, respectively. By estimating the total field $\mathbf{E}_i + \mathbf{E}_s$ at a far distance, we can determine the angular distribution of the optical emission power. Although the above analysis is valid for complex angles and the associated Legendre function has its complex extension, the scattering of evanescent waves resulting from total internal reflection at the surface does not converge to

a physically meaningful result. This indicates that we cannot overlook multiple reflections from the surface, especially for evanescent waves. Considering the modified Mie scattering solution mentioned above, we can incorporate the effect of the surface by evaluating it using an additional inverse matrix [Wriedt and Doicu \(1998\)](#). The scattering of evanescent waves exhibits peaks at whispering gallery mode (WGM) resonances or morphology dependent resonances (MDR) [Le Thomas, Woggon, Langbein, and Artemyev \(2006\)](#) as shown in [Figure 4.4\(a\)](#), where a considerable amount of evanescent wave energy is converted into propagating waves and brought to the far field. This, along with averaging the collection over the emission spectrum of the NV center, leads to an additional enhancement of 20%-30% over that allowed by geometrical optical processes. Our theoretical model agrees with FEM simulations and is also capable of predicting the nearly 40% enhancement observed in an earlier experiment [Moon et al. \(2021\)](#).

While the contribution from evanescent waves may be crucial for describing super-resolution imaging using a microsphere, the calculations shown in [Figure 4.4\(a\)](#) indicates that the enhancement of fluorescence collection is primarily due to optical lensing or optical collimation, especially for the microsphere sizes used in our experiments. Specifically, the grey and blue curves in [Figure 4.4\(a\)](#) denote the collection efficiency due to the geometric optical process and the overall collection efficiency (including contributions from both geometric optical and evanescent processes), respectively. The difference between these two curves thus represents the evanescent wave contribution. For the three microsphere sizes used in our experiment, contributions from the geometrical optical process still dominate, as shown in [Figure 4.4\(a\)](#). The theoretical results in [Figure 4.4\(a\)](#) are also consistent with the weak microsphere size dependence shown in [Figure 4.3\(a\)](#).

In Figure 4.4(b), we plot the angular dependence of the optical emission after its propagation through the microsphere. The numerical results reveal a highly directional emission pattern, with most of the emission falling within the 0.42 NA of the 20x objective, in good agreement with the experimental observation of Figure 4.3. This behavior can also be understood from a geometrical optics perspective, for which fluorescence from a NV center is effectively collimated by a silica microsphere, resulting in a relatively narrow cone angle. Mie scattering simulations predict that the microsphere can redirect 95% of the emission within the critical angle into a narrow cone angle compatible with the 20x objective, with an additional contribution coming from the evanescent wave conversion. Consequently, with the use of a silica microsphere, the 20x objective can effectively outperform a 0.95 NA objective by about 50%, significantly enhancing collection efficiency, as discussed earlier and confirmed by the calculation in Figure 4.4(c).

Figure 4.4(c) compares theoretically calculated collection efficiencies for various collection approaches, providing additional physical insights. For a microsphere with a diameter of 25 μm and a refractive index of 1.8 (dashed line in the figure), optical emissions can be collimated into a narrow cone such as that an objective with a NA as small as 0.1 and a WD up to 10 cm can be used without sacrificing collection efficiency. This calculation also indicates that there is no clear advantage in using high index microspheres as long as an objective with $\text{NA} > 0.3$ is used. For silica microspheres with a diameter of 1.8 μm (dot-dashed line in the figure), contributions from evanescent waves lead to an overall collection efficiency, which can be nearly twice that of microspheres with a diameter of 25 μm . Furthermore, for $\text{NA} < 0.45$, the performance of the microsphere approach can surpass that of the diamond SIL approach.

4.3 Conclusion

By carrying out a systematic experimental study and developing a theoretical model based on Mie scattering, we have gained significant insights into microsphere-enhanced fluorescence collection. Due to the effective collimation of fluorescence by a dielectric microsphere, large enhancement in fluorescence collection can be achieved with objectives that feature relatively low NA, extra-long working distance, as well as relatively small cost. With the use of a silica microsphere, a 20x objective with NA=0.42 can outperform a 0.95 NA objective (with no microsphere) by a considerable margin. Microsphere-enhanced fluorescence collection, which is not only easy to implement but also substantially lowers the total cost of the experimental setup, overcomes many of the limitations of other widely used enhancement approaches, potentially enabling wider applications of color-center based quantum sensing in real world.

CHAPTER V

PRINCIPLES OF QUANTUM SENSING AND CONTROL

5.1 Introduction

Quantum sensing exploits the sensitivity of a simple quantum system, such as solid-state spins, cold atoms, or superconducting circuits, to a given physical quantity to derive an estimate for the physical quantity. The near-term prospect for emerging quantum technologies and the potential for discoveries in wide ranging research areas have stimulated intense research efforts in this area [Degen et al. \(2017\)](#); [Giovannetti et al. \(2011\)](#). One of the most promising systems for quantum sensing is negatively charged single nitrogen vacancy (NV) centers in diamond, which can enable magnetic field, electric field, temperature, and strain sensing with nanometer spatial resolution [Doherty et al. \(2014\)](#); [Dolde et al. \(2011\)](#); [Kucsko et al. \(2013\)](#); [Taylor et al. \(2008\)](#). From the NV structure introduced in Chapter II, its electronic configuration consists of two unpaired holes that naturally form a spin triplet sharing the same orbital. This makes the NV center insensitive to phonon interactions, because selection rules require phonons to couple states with different orbital configurations. As a result, NV centers exhibit exceptional coherence times even at room temperature. This is why using NV centers in diamond for sensing is really appealing, as they do not require cryogenic conditions for long-coherence operation compared to other defect centers.

While quantum sensing with NV centers is conceptually appealing, we still need to investigate the underlying physics to give a more quantitative description. In short, how does an NV center respond to an external field, and how do we extract information from it in real time (time domain) or from its average properties (spectral domain)? Another relevant topic is how to extend its

coherence time in the presence of external noise, there are open-loop and closed-loop approaches to realize this. In this chapter we will frame these two concepts using the same principle, and we will see how they are related and how one can affect the other.

5.2 Quantum sensing

Noise in a quantum system reduces the coherence of quantum states. Among many types of noise, phase noise lies at the heart. This follows from a simple mathematical fact, if the phase $\phi \sim \mathcal{N}(\mu, \sigma^2)$, then

$$\langle e^{i\phi} \rangle = e^{i\mu} \left\langle \sum_{n=0}^{\infty} (i\phi - i\mu)^n / n! \right\rangle = e^{i\mu} e^{-\sigma^2/2}.$$

Hence a larger variance leads to smaller coherence, and in many relevant models this dephasing accumulates over time $\sigma^2(t) \propto t$. A more sophisticated description can be framed as follows. Under the Hamiltonian

$$H(t) = \hbar \frac{\omega(t)}{2} y(t) (|e\rangle \langle e| - |g\rangle \langle g|),$$

with $\omega = \omega_0 + \delta\omega$ and $\delta\omega$ Gaussian, the relative phase is

$$\phi(T) = \int_0^T \omega(t) y(t) dt,$$

and the corresponding phase factor is

$$\begin{aligned} \langle e^{-i\phi(T)} \rangle &= e^{-i\phi_0} e^{-\langle \delta\phi^2 \rangle / 2} \\ &= e^{-i\phi_0} \exp \left(-\frac{1}{2} \int_0^T \int_0^T y(t_1) y(t_2) \langle \delta\omega(t_1) \delta\omega(t_2) \rangle dt_1 dt_2 \right) \\ &= e^{-i\phi_0} \exp \left(-\frac{1}{2\pi} \int_0^\infty S(\omega) |F(\omega, T)|^2 d\omega \right), \end{aligned}$$

where $F(\omega, T) = \int_0^T y(t) e^{i\omega t} dt$ is the filter function determined by the control sequence $y(t)$, $S(\omega)$ is the power spectral density (square of fourier transformation of noise in time domain), $C(\tau) = \langle \delta\omega(t) \delta\omega(t + \tau) \rangle_t$ is the autocorrelation function,

and $\phi_0 = \langle \phi(T) \rangle$. We use the Wiener–Khinchin theorem that $C(\tau)$ and $S(\omega)$ Fourier-transform pairs.

From this expression, by designing $F(\omega, T)$ equivalently, the control sequence $y(t)$, we can either suppress or enhance the observed phase factor. A sequence that suppresses the phase factor selectively for certain spectral components and then reads out the coherence constitutes quantum sensing in spectral domain [cite], since $\mathcal{F}(\omega(t))$ encodes environmental quantities such as temperature, strain, or magnetic field. Moreover, if the sensing cycle is fast compared with the noise correlation time, we effectively access the real-time value $e^{-i\phi_0}$ rather than only spectral averages. Conversely, a fixed sequence that increases coherence (by attenuating $S(\omega)$ through $|F|^2$) can be viewed as passive control, while feedback control updates $y(t)$ or cancel out $\omega(t)$ based on real-time information from the system. In this way, we frame sensing and control as two sides of the same coin: the control shapes the information we can extract, and the acquired information, in turn, refines control.

The most widely used quantum-sensing approach has been Ramsey interferometry or Ramsey fringes, which probe the time evolution of a single or a collection of two-level systems [H. Lee, Kok, and Dowling \(2002\)](#); [Taylor et al. \(2008\)](#). The Ramsey interferometry is especially suitable for sensing of static as well as periodic signals. For a typical quantum-sensing task, a Ramsey-type protocol projects the phase factor onto population, enabling readout:

$$\frac{1}{\sqrt{2}} (|g\rangle + e^{-i\phi(T)} |e\rangle) \xrightarrow{\frac{\pi}{2}} \frac{1}{\sqrt{2}} ((e^{-i\phi(T)} - 1) |g\rangle + (e^{-i\phi(T)} + 1) |e\rangle),$$

thereby mapping phase information onto an observable. Further effort can be applied to optimize T and $y(t)$ to achieve the best sensitivity. As already noted, however, when the noise is fast this approach requires short cycles to mitigate the

factor $e^{-\langle\delta\phi^2\rangle/2}$, which adds complexity to the control electronics and experimental procedures.

Compared to the Ramsey approach, which reads out the phase factor from a projective measurement, the CPT approach reads out the phase factor in a continuous manner, continuously projecting the system into the $\{|D\rangle, |B\rangle\}$ manifold consisting of dark and bright states. Because CPT provides continuous readout at a rate $\sim \Omega^2/\Gamma$, this realizes an approximately instantaneous measurement of the frequency fluctuation $\omega(t)$.

5.3 Quantum control

We now examine how environmental noise affects basic experiments such as Rabi and Ramsey, and how quantum control can mitigate its impact. In a two-level system, energy relaxation processes define the longitudinal relaxation time T_1 , while dephasing processes, including slowly varying (non-Markovian) phase noise, limit the transverse coherence time T_2^* . Ideally, if quasi-static phase fluctuations are removed by an appropriate control sequence, the coherence approaches the intrinsic limit $T_2 \rightarrow 2T_1$, so that the decay is limited only by energy relaxation.

To connect this intuitive picture with the frequency-domain description used in the previous section, we recall that under phase noise the accumulated phase obeys

$$\langle e^{-i\phi(T)} \rangle = e^{-i\phi_0} \exp \left(-\frac{1}{2\pi} \int_0^\infty S(\omega) |F(\omega, T)|^2 d\omega \right), \quad (5.1)$$

where $F(\omega, T) = \int_0^T y(t) e^{i\omega t} dt$ is the filter function set by the control sequence $y(t)$, and $S(\omega)$ is the power spectral density (PSD). By carefully designing $y(t)$ we can minimize the effect of noise and extend coherence.

Following this logic, a convenient open-loop strategy is dynamical decoupling, for which a basic sequence can be written as

$$\left(\frac{\tau}{2} - \pi - \frac{\tau}{2}\right)^N,$$

that $N = 1$ is Hahn echo ($\tau/2$ denotes free evolution, and π is a π -pulse).

Sequences like CPMG and the XY family apply π -pulses along different axes to improve robustness against control errors. The corresponding filter function is

$$\begin{aligned} F(\omega, T) &= \int_0^T y(t) e^{i\omega t} dt = \frac{2 \sin^2(\omega\tau/4)}{i\omega \cos(\omega\tau/2)} (1 - (-1)^N e^{i\omega T}), \\ |F(\omega, T)|^2 &= \frac{16}{\omega^2} \sin^2\left(\frac{\omega T + N\pi}{2}\right) \frac{\sin^4(\omega\tau/4)}{\cos^2(\omega\tau/2)}, \end{aligned}$$

with $T = N\tau$. This filter suppresses low-frequency components with $\omega\tau \ll 1$, since

$$|F(\omega, T)|^2 \leq \frac{16 \sin^4(\omega\tau/4)}{\omega^2 \cos^2(\omega\tau/2)} \sim \omega^2 \tau^4 \ll \frac{1}{\omega^2},$$

making the integral in the exponent negligible. And an even N yields more favorable scaling. Thus, passive open-loop control such as dynamical decoupling can effectively suppress decoherence from low-frequency noise and extend coherence time.

Another way of implementing control is closed-loop control, where we drive the system while also using feedback from the system to adjust the control in real time. The most straightforward idea is to introduce a compensating term $-\tilde{\omega}(t)$ that tracks $\omega(t)$ and cancels its effect. In this picture, we effectively reduce the amplitude of the noise PSD and thereby prolong the coherence time. Practical realizations of this feedback idea will be discussed in a later chapter.

5.4 Cramér-Rao bound and Fisher information

5.4.1 General Cramér-Rao lower bound

A general sensing problem can be described as estimating a (possibly random) field $\mathbf{X} = \{x_1, \dots, x_n\}$ from the measurement outcomes $\mathbf{Y} = \{y_1, \dots, y_n\}$.

We first recall the classical Cramér-Rao lower bound (CRLB) for a single deterministic parameter and then its Bayesian extension for a random parameter vector.

Let x be a deterministic scalar parameter and $\hat{x}(\mathbf{Y})$ an unbiased estimator that $\mathbb{E}[\hat{x}(\mathbf{Y})] = x$. The likelihood is $p(\mathbf{Y}|x)$, and the Fisher information is defined as

$$F(x) = \mathbb{E} \left[\left(\frac{\partial}{\partial x} \ln p(\mathbf{Y}|x) \right)^2 \right] = \int p(\mathbf{Y}|x) \left(\frac{\partial}{\partial x} \ln p(\mathbf{Y}|x) \right)^2 d\mathbf{Y}.$$

Then the variance of any unbiased estimator satisfies the Cramér-Rao inequality

$$\text{var}(\hat{x}(\mathbf{Y})) \geq \frac{1}{F(x)}.$$

For completeness we sketch the standard proof. Unbiasedness implies

$$\int (\hat{x}(\mathbf{Y}) - x) p(\mathbf{Y}|x) d\mathbf{Y} = 0, \quad (5.2)$$

and differentiating with respect to x gives

$$0 = \frac{\partial}{\partial x} \int (\hat{x}(\mathbf{Y}) - x) p(\mathbf{Y}|x) d\mathbf{Y} = \int \left[(\hat{x}(\mathbf{Y}) - x) \frac{\partial p(\mathbf{Y}|x)}{\partial x} - p(\mathbf{Y}|x) \right] d\mathbf{Y}.$$

Rearranging,

$$1 = \int (\hat{x}(\mathbf{Y}) - x) \frac{\partial p(\mathbf{Y}|x)}{\partial x} d\mathbf{Y} = \int (\hat{x}(\mathbf{Y}) - x) \frac{\partial \ln p(\mathbf{Y}|x)}{\partial x} p(\mathbf{Y}|x) d\mathbf{Y}. \quad (5.3)$$

Define $u(\mathbf{Y}) = \hat{x}(\mathbf{Y}) - x$, $v(\mathbf{Y}) = \frac{\partial}{\partial x} \ln p(\mathbf{Y}|x) \sqrt{p(\mathbf{Y}|x)}$. The Cauchy-Schwarz inequality then gives

$$1^2 \leq \left(\int u(\mathbf{Y})^2 p(\mathbf{Y}|x) d\mathbf{Y} \right) \left(\int v(\mathbf{Y})^2 d\mathbf{Y} \right) = \text{var}(\hat{x}(\mathbf{Y})) F(x), \quad (5.4)$$

which yields the scalar CRLB $\text{var}(\hat{x}(\mathbf{Y})) \geq 1/F(x)$ (see also Chapter 2, equation 178 of [Van Trees \(2004\)](#)).

For a time-dependent or spatially varying random parameter, we consider $\hat{\mathbf{X}}(\mathbf{Y})$ be any estimator of $\mathbf{X}$. Similarly we have the bound (Chapter 2, equation

$$\text{var}(\hat{x}_i(\mathbf{Y})) \geq (\mathbf{F}^{-1})_{ii}, \quad (5.5)$$

where the Fisher information matrix is

$$\begin{aligned} \mathbf{F}_{ij} &= \mathbf{F}_{ij}^M + \mathbf{F}_{ij}^B, \\ \mathbf{F}_{ij}^M &= \mathbb{E} \left[\frac{\partial \ln p(\mathbf{Y}|\mathbf{X})}{\partial x_i} \frac{\partial \ln p(\mathbf{Y}|\mathbf{X})}{\partial x_j} \right], \\ \mathbf{F}_{ij}^B &= \mathbb{E} \left[\frac{\partial \ln p(\mathbf{X})}{\partial x_i} \frac{\partial \ln p(\mathbf{X})}{\partial x_j} \right]. \end{aligned}$$

5.4.2 Application to CPT-based sensing of Ornstein-Uhlenbeck noise

For Fisher information matrix, we have

$$\begin{aligned} \mathbf{F}_{ij}^M &= \int d\mathbf{Y} d\mathbf{X} p(\mathbf{X}, \mathbf{Y}) \frac{\partial \ln p(\mathbf{Y}|\mathbf{X})}{\partial x_i} \frac{\partial \ln p(\mathbf{Y}|\mathbf{X})}{\partial x_j}, \\ p(\mathbf{X}, \mathbf{Y}) &= p(\mathbf{X}) p(\mathbf{Y}|\mathbf{X}) = p(x_1) p(x_2|x_1) \cdots p(y_1|x_1) p(y_2|x_2) \cdots, \\ \frac{\partial \ln p(\mathbf{Y}|\mathbf{X})}{\partial x_i} &= \frac{\partial \ln p(y_1|x_1) p(y_2|x_2) \cdots}{\partial x_i} = \frac{1}{p(y_i|x_i)} \frac{\partial p(y_i|x_i)}{\partial x_i}, \end{aligned}$$

and we know $p(y|x)$ is Poisson distribution $p(y|x) = e^{-\lambda} \lambda^y / y!$, $\lambda = \langle n \rangle_{\delta t} = \kappa \rho_{ee}(x) \delta t \eta$, so

$$\frac{\partial \ln p(\mathbf{Y}|\mathbf{X})}{\partial x_i} = \frac{\partial \rho_{ee}(x_i)}{\partial x_i} \left(\frac{y_i}{\rho_{ee}(x_i)} - \kappa \eta \delta t \right).$$

For the case $i \neq j$, we have

$$\begin{aligned} \mathbf{F}_{ij}^M &= \int p(\mathbf{X}) \left\{ p(y_i|x_i) \frac{\partial \rho_{ee}(x_i)}{\partial x_i} \left(\frac{y_i}{\rho_{ee}(x_i)} - \kappa \eta \delta t \right) dy_i \right\} \\ &\quad \times \left\{ p(y_j|x_j) \frac{\partial \rho_{ee}(x_j)}{\partial x_j} \left(\frac{y_j}{\rho_{ee}(x_j)} - \kappa \eta \delta t \right) dy_j \right\} d\mathbf{X}, \\ &\int p(y_i|x_i) \frac{\partial \rho_{ee}(x_i)}{\partial x_i} \left(\frac{y_i}{\rho_{ee}(x_i)} - \kappa \eta \delta t \right) dy_i = 0. \end{aligned}$$

Then $\mathbf{F}_{ij}^M = 0$. For the case $i = j$, we have

$$\begin{aligned}\mathbf{F}_{ii}^M &= \int p(\mathbf{X}) \left\{ p(y_i|x_i) \left(\frac{\partial \rho_{ee}(x_i)}{\partial x_i} \right)^2 \left(\frac{y_i}{\rho_{ee}(x_i)} - \kappa \eta \delta t \right)^2 dy_i \right\} d\mathbf{X} \\ &= \int p(\mathbf{X}) \left(\frac{\partial \rho_{ee}(x_i)}{\partial x_i} \right)^2 \frac{\kappa \eta \delta t}{\rho_{ee}(x_i)} d\mathbf{X} \\ &= \kappa \eta \delta t \left\langle \left(\frac{\partial \rho_{ee}(x_i)}{\partial x_i} \right)^2 \frac{1}{\rho_{ee}(x_i)} \right\rangle,\end{aligned}$$

we can define a constant (depends on the parameters of OU process)

$$g(\sigma) = \left\langle \left(\frac{\partial \rho_{ee}(x_i)}{\partial x_i} \right)^2 \frac{1}{\rho_{ee}(x_i)} \right\rangle,$$

then the Fisher information matrix is

$$\mathbf{F}_{ij}^M = \kappa \eta \delta t g(\sigma) \delta_{ij}.$$

When we take the limitation $\delta t \rightarrow 0$ (the update time interval), we get the Fisher information matrix for the continuous-time case

$$F^M(t_i, t_j) = \lim_{\delta t \rightarrow 0} \frac{F_{ij}^M}{(\delta t)^2} = \kappa \eta g(\sigma) \delta(t_i - t_j).$$

And according the result in [Tsang, Wiseman, and Caves \(2011\)](#) or Chapter4, eq.231 of [Van Trees \(2004\)](#). (they use a different definition of Fourier transform) then (the prior part is a function of correlation function)

$$\mathcal{F} \{ F^B(t_i, t_j) \} = \mathcal{F} \left\{ \lim_{\delta t \rightarrow 0} \frac{F_{ij}^B}{(\delta t)^2} \right\} = \frac{1}{2\pi \mathcal{F} \{ \sigma^2 e^{-|t_i - t_j|/\mu} \}}.$$

And we know the equation

$$\sum_k F_{ik}^{-1} F_{kj} = \delta_{ij},$$

then with the definition $\lim_{\delta t \rightarrow 0} F_{ij}^{-1} = F^{-1}(t_i, t_j)$, we know

$$\lim_{\delta t \rightarrow 0} \sum_k F_{ik}^{-1} \frac{F_{kj}}{(\delta t)^2} \delta t = \int F^{-1}(t_i, t_k) F(t_k, t_j) dt_k = \lim_{\delta t \rightarrow 0} \delta_{ij} / \delta t = \delta(t_i - t_j),$$

then

$$\int F^{-1}(t - t'') F(t'' - t') dt'' = \delta(t - t').$$

Using Fourier tranform, we have

$$F^{-1}(\tau) = \frac{1}{2\pi} \mathcal{F}^{-1} \left\{ \frac{1}{\mathcal{F} \{ \kappa \eta g(\sigma) \delta(t) \} + \frac{1}{2\pi \mathcal{F} \{ \sigma^2 e^{-|t|/\mu} \}}} \right\},$$

then

$$\text{var}(\hat{x}_t(\mathbf{Y})) \geq F^{-1}(0) = \frac{\sigma^2}{\sqrt{1 + 2\kappa\eta\sigma^2\mu g(\sigma)}}.$$

5.4.3 Real-time estimation using historical information only

The derivation in the last section is under the assumption: we could take the update interval to infinitesimal then making CRLB for every step the same, but this assumption is not true for the estimation of the end in the sensing sequence. When we want to do a realtime sensing, we use only historical information to estimate next point rather than using the whole information. For this case, the estimated point is at the end of the sensing sequence, then can never become same as those points using the whole information even taking the limitation. So we have to solve the inverse Fisher information matrix before taking limits to continous. We could prove the Fisher information matrix is

$$\mathbf{F} = \frac{\sigma^2}{\sigma_0^4} \begin{pmatrix} 1 - \mu_0^2 & \mu_0^3 - \mu_0 & & & \\ \mu_0^3 - \mu_0 & 1 - \mu_0^4 & \ddots & & \\ & \ddots & \ddots & \ddots & \\ & & \ddots & 1 - \mu_0^4 & \mu_0^3 - \mu_0 \\ & & & \mu_0^3 - \mu_0 & 1 - \mu_0^2 \end{pmatrix} + \kappa \eta g(\sigma) \delta t \begin{pmatrix} 1 & & & & \\ & \ddots & & & \\ & & \ddots & & \\ & & & \ddots & \\ & & & & 1 \end{pmatrix},$$

where $\mu_0 = e^{-\delta t/\mu}$ and $\sigma_0^2 = \sigma^2(1 - \mu_0^2)$. Then we can prove

$$\frac{\mathbf{F}_{T,T}^{-1}}{\mathbf{F}_{T/2,T/2}^{-1}} = \frac{\det(\mathbf{F}_T^0)}{\det(\mathbf{F}_{T/2}^0)^2},$$

where

$$\mathbf{F}_T^0 = \begin{pmatrix} 1 - \mu_0^4 & \mu_0^3 - \mu_0 & & & \\ \mu_0^3 - \mu_0 & 1 - \mu_0^4 & \ddots & & \\ & \ddots & \ddots & \ddots & \\ & & \ddots & 1 - \mu_0^4 & \mu_0^3 - \mu_0 \\ & & & \mu_0^3 - \mu_0 & 1 - \mu_0^2 \end{pmatrix} + g_0 \begin{pmatrix} 1 & & & & \\ & \ddots & & & \\ & & \ddots & & \\ & & & \ddots & \\ & & & & 1 \end{pmatrix}$$

is a $(T/\delta t)$ -order matrix and $g_0 = \kappa\eta g(\sigma)\delta t\sigma_0^4/\sigma^2$. The ratio $\mathbf{F}_{T,T}^{-1}/\mathbf{F}_{T/2,T/2}^{-1}$ describes the difference between CRLB when using the whole information and only the historical information, to get this we only need to estimate the determinant of $\mathbf{F}_T^0$.

From the recurrence relation

$$D_n = (1 - \mu_0^4 + g_0)D_{n-1} - (\mu_0^3 - \mu_0)^2 D_{n-2},$$

$$D_1 = 1 - \mu_0^2 + g_0,$$

we get the expression of any order determinant

$$D_n = Ar_1^n + Br_2^n.$$

The roots r_1, r_2 are the solution of characteristic equation,

$$r_{1,2} = \frac{(1 - \mu_0^4 + g_0) \pm \sqrt{(1 - \mu_0^4 + g_0)^2 - 4\mu_0^2(1 - \mu_0^2)^2}}{2},$$

and A, B are determined by D_1 ,

$$A = \frac{1 + g_0 - 2\mu_0^2 + \mu_0^4 + \sqrt{(1 - \mu_0^4 + g_0)^2 - 4\mu_0^2(1 - \mu_0^2)^2}}{2\sqrt{(1 - \mu_0^4 + g_0)^2 - 4\mu_0^2(1 - \mu_0^2)^2}},$$

$$B = \frac{D_1 - Ar_1}{r_2}.$$

Then the ratio is

$$\frac{\mathbf{F}_{T,T}^{-1}}{\mathbf{F}_{T/2,T/2}^{-1}} = \frac{(Ar_1^n + Br_2^n)}{(Ar_1^{n/2} + Br_2^{n/2})^2},$$

where $n = T/\delta t$, when take the limitation $\delta t \rightarrow 0$, and take only the first and second order infinitesimal, we get

$$\frac{\mathbf{F}_{T,T}^{-1}}{\mathbf{F}_{T/2,T/2}^{-1}} = \frac{Ae^{TC_1} + Be^{TC_2}}{A^2e^{TC_1} + B^2e^{TC_2} + 2ABe^{T(C_1+C_2)/2}},$$

$$C_{1,2} = \frac{1}{4\mu}(-8 \pm \sqrt{1 + 32\kappa\eta g(\sigma)\sigma^2\mu}).$$

For our choice of parameters, we always have $\kappa\eta g(\sigma)\sigma^2\mu > 2$, so we know, no matter how many historical information we use (when $T \rightarrow \infty$), there exists a CRLB for the estimation,

$$\begin{aligned} \mathbf{F}_{T,T}^{-1} &= F^{-1}(0) \times \lim_{T \rightarrow \infty} \frac{Ae^{TC_1} + Be^{TC_2}}{A^2e^{TC_1} + B^2e^{TC_2} + 2ABe^{T(C_1+C_2)/2}} \\ &= F^{-1}(0) \times \lim_{\delta t \rightarrow 0} \frac{1}{A} = \frac{\sigma^2}{\sqrt{1 + 2\kappa\eta\sigma^2\mu g(\sigma)}} \times \frac{1}{\frac{1}{2} + 2/\sqrt{1 + 32\kappa\eta\sigma^2\mu g(\sigma)}}. \end{aligned}$$

5.4.4 Gaussian posterior

In [Van Trees \(2004\)](#), CRLB is achieveable bound when exists an efficient estimate, then requires

$$p(x|y) = e^{K(x)x^2 + C_1(y)x + C_2(y)}.$$

For a Taylor expansion of $\rho_{ee}(x)$, we have

$$\rho_{ee}(x) - \rho_{ee}(0) = Ax + Bx^2 + \dots.$$

Then the Bayes' theorem tells us,

$$p(x|y) = \frac{p(y|x)p(x)}{p(y)} = \frac{e^{-C\rho_{ee}(x)}(C\rho_{ee}(x))^y}{y!} \frac{p(x)}{p(y)}.$$

When the prior of x is Gaussian which is true, and the time slot we choose is small enough (then $y = 0$ for most cases), we have

$$p(x|y) = e^{K(x)x^2 + C_1(y)x + C_2(y)}.$$

CHAPTER VI

CONTINUOUS REAL-TIME MAGNETIC SENSING VIA COHERENT POPULATION TRAPPING: THEORY

The results presented in this chapter were published in [S. H. Wu et al. \(2021\)](#).

6.1 Introduction

As briefly discussed in Chapter V, the most widely used quantum-sensing approach has been Ramsey interferometry or Ramsey fringes, and repeated Ramsey interferometric measurements allow the tracking of time-dependent fields [Cooper, Magesan, Yum, and Cappellaro \(2014\)](#); [Santagati et al. \(2019\)](#); [Shulman et al. \(2014\)](#). Nevertheless, since each Ramsey interferometric measurement consists of three separate stages: initialization, coherent time evolution, and readout, Ramsey interferometry cannot provide continuous realtime sensing.

In this chapter we propose and analyze theoretically the use of coherent population trapping (CPT) in a NV center for continuous real-time sensing. In a CPT process, a Λ -type three-level system is trapped in a dark state, i.e., a special superposition of the two lower states, which is decoupled from the upper state due to destructive quantum interference [Scully and Zubairy \(1997\)](#). The formation of the dark state prevents optical emissions from the NV center. A fluctuating magnetic environment, however, can kick the NV center out of the dark state, leading to a sequence of single photon emissions from the NV center. These single photon emissions carry the information on the fluctuating magnetic fields and can in principle provide continuous real-time sensing of the magnetic fields, as illustrated schematically in [Figure 6.1](#). Nevertheless, the overall collection/detection efficiency for optical emissions from a NV center is only a few percent under typical

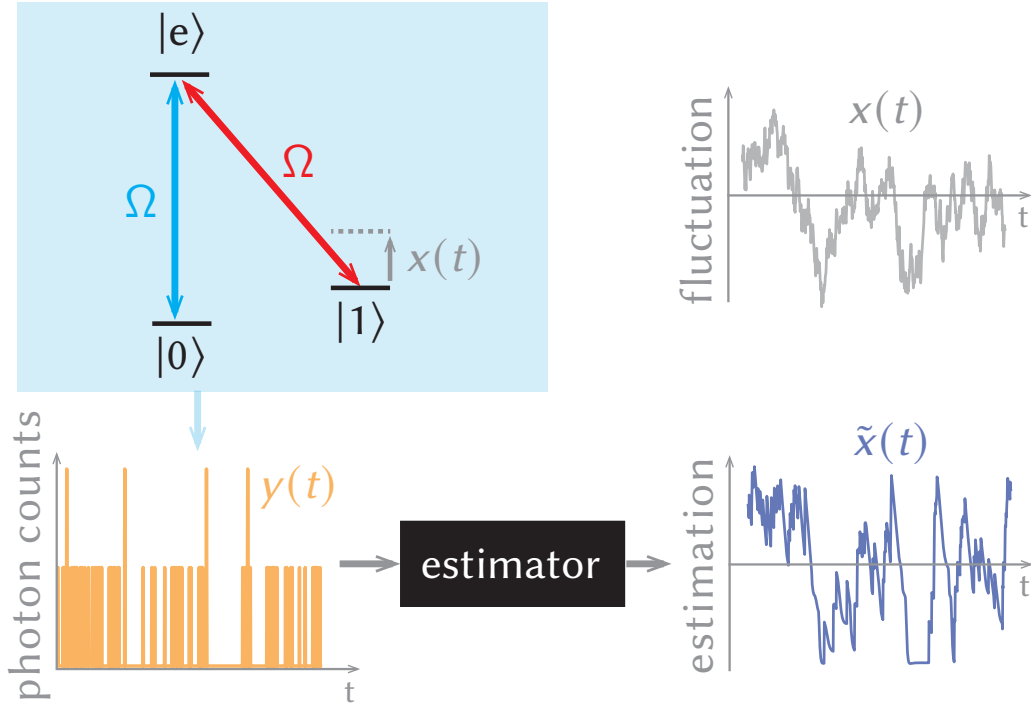


Figure 6.1. Schematic of CPT-based continuous real-time sensing of the fluctuating magnetic environment at a NV center. The fluctuating magnetic field, which induces single photon emissions from the NV center, can be estimated from the time series of the photon counts detected.

experimental conditions. For a realistic implementation, a primary challenge is to make continuous real-time estimations of the magnetic fields with the few photons detected. We show that this challenge can be overcome with the use of Bayesian inference. For the theoretical analysis, we have used the nuclear spin bath in diamond as a model fluctuating magnetic environment. We have employed the stochastic Schrödinger equation (SSE) to simulate single photon emissions from a NV center in a CPT setting [Carmichael \(1993\)](#) and have treated the nuclear spin bath as an Ornstein-Uhlenbeck (OU) process [de Lange, Wang, Ristè, Dobrovitski, and Hanson \(2010\)](#); [Dobrovitski, Feiguin, Hanson, and Awschalom \(2009\)](#); [Witzel, Carroll, Cywiński, and Das Sarma \(2012\)](#). Estimations of fluctuation magnetic fields are obtained from a time series of photon counts, for which the average

number of photons per update time interval is much smaller than 1. By taking advantage of the known statistical properties of the fluctuation environment in the implementation, we demonstrate that the Bayesian estimator can provide dynamical information on a timescale that is comparable to the inverse of the average photon counting rate. Additional theoretical analysis further indicates that the Bayesian inference-based estimations can approach the classical Cramér-Rao lower bound (CRLB).

6.2 Physical model

6.2.1 Coherent population trapping

For a NV center in diamond, several energy-level schemes have been used for the realization of CPT [Golter, Baldwin, and Wang \(2014\)](#); [Golter et al. \(2016\)](#); [Santori et al. \(2006\)](#); [Togan, Chu, Imamoglu, and Lukin \(2011\)](#); [Yale et al. \(2013\)](#). Without losing generality, here we consider two ground spin states, $m_s = 0$ and $m_s = 1$, coupling to an excited state, $|e\rangle$, through two dipole optical transitions with frequencies, ν_0 and ν_1 , respectively, as shown schematically in [Figure 6.1](#). Two external optical fields with frequencies, ω_0 and ω_1 , couple to the two respective optical transitions with equal Rabi frequency Ω . With $\omega_0 \approx \nu_0$, $\omega_1 \approx \nu_1$, and $\rho_{ee} \ll 1$, the steady-state excited-state population is given by [Golter et al. \(2014\)](#)

$$\rho_{ee} = \frac{\Omega^2}{2\Gamma\kappa} \left[1 - \frac{\Omega^2}{2\kappa} \frac{\gamma_s + \Omega^2/2\kappa}{(\delta - \omega_B)^2 + (\gamma_s + \Omega^2/2\kappa)^2} \right]. \quad (6.1)$$

where γ_s and κ are the decay rates for the spin coherence and optical dipole coherence, respectively, Γ is the spontaneous emission rate of the excited state, $\delta = \omega_0 - \omega_1$ is the laser detuning, and $\omega_B = \nu_0 - \nu_1$ is the frequency separation between the two spin states. CPT, which corresponds to the formation of a dark state for the two lower spin states, occurs near the Raman resonance with $\Delta = \delta - \omega_B = 0$. Note that the external optical fields lead to a power broadening

described by the term in [Equation 6.1](#). The CPT process can be characterized by a dimensionless cooperativity, defined as $C = \Omega^2/2\kappa\gamma_s$. CPT-based sensing will be carried out in the regime of $C \gg 1$.

6.2.2 Nuclear spin bath

We simulate the magnetic field fluctuations induced by the nuclear spin bath as an Ornstein-Uhlenbeck (OU) process characterized by a memory time τ_N , with [Gardiner \(1985\)](#)

$$dx = -\frac{1}{\tau_N}xdt + \sqrt{\frac{2\sigma^2}{\tau_N}}dW_t,$$

where $x(t) = \omega_B(t) - \langle \omega_B(t) \rangle$ represents the bath-induced fluctuation in ω_B , dW_t denotes a Wiener increment that has a Gaussian distribution with mean zero and variance dt , and the autocorrelation function for $x(t)$ is characterized by

$$R(t) = \langle x(t_0)x(t_0 + t) \rangle = \sigma^2 e^{-|t|/\tau_N}.$$

For the two bath parameters, σ can be obtained directly from the Ramsey fringes and τ_N can be obtained from additional experiments of spin echoes [de Lange et al. \(2010\)](#). For the numerical simulations in this chapter, we take $\tau_N = 1$ ms and $\sigma/2\pi = 0.13$ MHz (which corresponds to a dephasing time, $T_2^* = 1.7\mu\text{s}$) for a ^{13}C nuclear spin bath in diamond. [Figure 6.5a](#) shows an example of the simulated $x(t)$ and the autocorrelation function averaged over 1000 runs.

Additionally, according to the Bernstein–Widder theorem on completely monotone functions, if a noise has an autocorrelation function $C(t)$ that is completely monotone for $t > 0$, then there exists a nonnegative weight $\omega(\tau)$ such that

$$C(t) = \int_0^\infty \omega(\tau) e^{-|t|/\tau} d\tau, S(\omega) = \int_0^\infty \frac{2\omega(\tau)}{1 + (\omega\tau)^2} d\tau.$$

This means any process can be regarded as a mixture of OU noises with different τ . OU noise is therefore a broadly representative family, by learning how to sense it, we obtain solutions that generalize to many other noise types.

6.3 Estimators

In this section, we will discuss estimators, which we have used to output a time series of estimated frequency, $\{\tilde{\mathbf{x}}_n\} = \{\tilde{x}_1, \tilde{x}_2, \dots, \tilde{x}_n, \dots\}$, from a time series of photon counts, $\{\mathbf{y}_n\} = \{y_1, y_2, \dots, y_n, \dots\}$, where y_n is the number of photons detected during the n th time interval and the duration of the time interval τ is small compared with τ_N . We will also discuss how close these estimations can be to the actual frequency, $\{\mathbf{x}_n\} = \{x_1, x_2, \dots, x_n, \dots\}$.

6.3.1 Bayesian estimator

We have used the Bayesian inference described by the Bayes update rule,

$$p(x_n|y_n, y_{n-1}, \dots, y_1) \propto p_{\bar{y}}(y_n|x_n) \times p'(x_n|y_{n-1}, \dots, y_1).$$

to estimate $x(t)$ from the time series of photon counts, where $p'(x_n|y_{n-1}, \dots, y_1)$ is the prior probability distribution based on the previous time series of photon counts, $p(x_n|y_n, y_{n-1}, \dots, y_1)$ is the posteriori probability distribution, and $p_{\bar{y}}(y_n|x_n)$ is the likelihood of detecting y_n photons in the n th time interval given x_n . The likelihood follows a Poisson distribution,

$$p_{\bar{y}}(y_n|x_n) = \frac{\bar{y}^{y_n} e^{-\bar{y}}}{y_n!},$$

where $\bar{y} = \eta\tau\Gamma\rho_{ee}(x)$ is the average number of photons detected per time interval and the update interval τ is sufficiently short such that $\bar{y}$ is small compared with 1. The estimation of $x(t)$ as a function of time is then given by

$$\tilde{x}(t) = \int p(x, t) x dx.$$

The prior probability distribution for x_n can be improved if the statistical properties of the fluctuating fields are known. For the OU process, the probability of finding x_n at $t + \tau$ given x_{n-1} at t follows a normal distribution,

$$p_{\text{OU}}(x_n, t + \tau | x_{n-1}, t) = \mathcal{N}(x_{n-1}e^{-\tau/\tau_N}, \sigma'^2)$$

where $\sigma'^2 = \sigma^2[1 - \exp(-2\tau/\tau_N)]$. In this case, the prior probability distribution for x_n can be improved as

$$\begin{aligned} p'(x_n = x | y_{n-1}, \dots, y_1) &= \int d\omega \cdot p(x_{n-1} = x - \omega | y_{n-1}, \dots, y_1) \\ &\quad \times p_{\text{OU}}(x_n = x_{n-1} + \omega, t + \tau | x_{n-1}, t). \end{aligned}$$

We will refer this improved Bayesian estimation process as the OU Bayesian estimator.

6.3.2 Average count estimator

For comparison, we have also used the photon counts detected in a relatively long duration, τ_a , to directly estimate $x(t)$. Specifically, we carry out the estimation by using

$$y_n^{(a)} = \eta \Gamma \tau_a \rho_{ee}(x_n),$$

where $y_n^{(a)}$ is the number of photons detected between time $n\tau - \tau_a$ and $n\tau$. For achieving acceptable photon counts (of order 10), we took $\tau_a = 100$ for the results presented. This average count approach is not expected to be able to track bath fluctuations that occur in a timescale of τ .

Note that electromagnetically induced transparency (EIT) of an ensemble of NV centers, which is closely related to CPT, has been used for static sensing [Acosta, Jensen, Santori, Budker, and Beausoleil \(2013\)](#). CPT of an ensemble of NV centers, which can feature orders of magnitude more photon counts than that of a single NV, can also be exploited for time-resolved sensing. A single quantum sensor,

such as a single NV, however, is necessary for sensing of microscopic fluctuations or for sensing with nanometer resolutions.

6.3.3 Cramér-Rao lower bound

How close an estimation is to the actual variable is characterized by the average estimation variance, defined as . Theoretically, the classical Cramér-Rao lower bound (CRLB) sets a lower bound on the variance. As shown in the last chapter, the CRLB for the OU-Bayesian estimation is given by

$$\text{Var}[x_n(\mathbf{y})] \geq \frac{\sigma^2}{\sqrt{1 + 2\tau_N\eta\Gamma\sigma^2g(\sigma)}}, \quad (6.2)$$

where $g(\sigma) = \langle (\partial\rho_{ee}/\partial x)^2 / \partial\rho_{ee}(x) \rangle_x$.

For real-time sensing, only the historical data can be used. Data taken after the estimation event cannot be used for the estimation. However, the CRLB in [Equation 6.2](#) assumes that the entire data set can be used for the estimation. As shown in last section, the CRLB when only the historical data can be used is revised as

$$\text{Var}[x_n(\mathbf{y})] \geq \frac{\sigma^2}{\sqrt{1 + 2\tau_N\eta\Gamma\sigma^2g(\sigma)}} \times \frac{2}{1 + 4/\sqrt{1 + 32\tau_N\eta\Gamma\sigma^2g(\sigma)}}, \quad (6.3)$$

where we have assumed that $\tau_N\eta\Gamma\sigma^2g(\sigma) > 2$ (which is satisfied in most cases). In the limit that $\tau_N\eta\Gamma\sigma^2g(\sigma) \gg 1$, the CRLB for estimations using the historical data set is twice that using the entire data set.

Note that a necessary condition for achieving CRLB is that the posterior in the Bayesian inference is a Gaussian distribution [Van Trees \(2004\)](#). For our system, the posterior, $p(x_n = x|y_n, y_{n-1}, \dots, y_1)$, is approximately Gaussian only when ρ_{ee} depends linearly or quadratically on x .

6.4 Results and discussions

We have used the three estimators discussed for estimations of ω_B from a sequence of single photon emissions. The estimations as a function of time shown in [Figure 6.2](#) are based on the simulated fluctuations and the corresponding time series of photon counts. The update time interval used is $\tau = 10\mu s$ and the average photon count per time interval is 0.1. As shown in [Figure 6.2a](#), the estimations obtained with the average count estimator exhibit considerable deviations from the true frequency due to the relatively low photon count rate. The estimations also feature significant delays with respect to the fluctuations in the actual frequency. The delays are due to the relatively long photon counting period, τ_a , needed to avoid excessive fluctuations in the photon counts used for the estimation. In comparison, the estimations obtained with the simple Bayesian estimator provides a good estimate for the relatively longtime behavior of the frequency fluctuations. However, the photon counts are too low for the estimator to provide dynamical information on a relatively short timescale.

Much improved estimations are obtained with the OU Bayesian estimator, as shown in [Figure 6.2b](#). The statistical properties exhibited by the OU process enable the Bayesian inference to yield dynamical information on a timescale much shorter than that is achieved with the simple Bayesian estimator. The OU Bayesian estimator provides continuous real-time sensing of the fluctuating magnetic fields even though the timescale of the fluctuations is comparable to the inverse of the average photon count rate.

For a quantitative analysis of the effectiveness of the estimators, we have numerically calculated the variances of the estimations and compare these variances with those expected from the CRLB. The variances shown in [Figure 6.3](#) are the

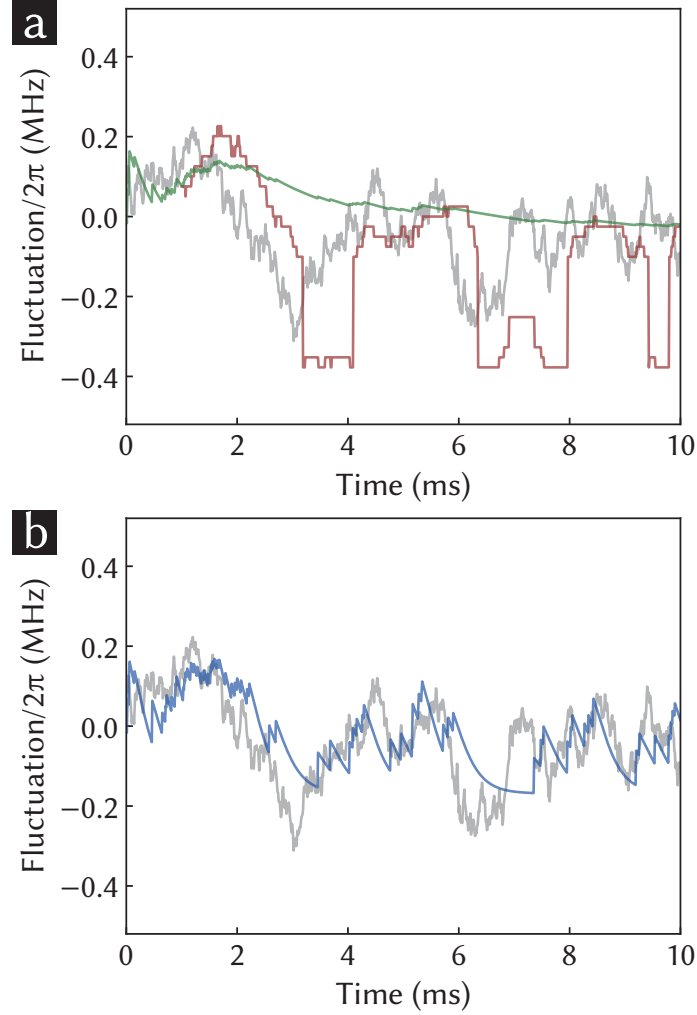


Figure 6.2. (a) Estimations obtained with the average photon account estimator (brown line) and the simple Bayesian estimator (green line). (b) Estimations obtained with the OU Bayesian estimator (blue line). For comparison, the grey line plots the actual fluctuations. The CPT parameters used are the same as in [Figure 6.5](#).

average of 100 runs. Each run covers the time duration from $t = 2$ ms to $t = 10$ ms and uses sequences of single photon emissions generated from the steady-state excited-excited population, as discussed. The parameters used for the bath and the CPT process are the same as those used for [Figure 6.2](#) unless otherwise specified.

[Figure 6.3a](#) shows the variances obtained with the three different estimators as a function of the memory time, τ_N , of the nuclear spin bath. For comparison, [Figure 6.3a](#) also plots the theoretically expected CRLB for the OU Bayesian estimator and the variance, σ^2 , of the actual fluctuations. As expected, the variances obtained with the average count estimator are far above the CRLB. These variances also exceed σ^2 . The variances obtained with the simple Bayesian estimator fall below σ^2 but are still far above the CRLB. In comparison, the variances obtained with the OU Bayesian estimator nearly approach the CRLB. Both the variances and the CRLB improve gradually with increasing τ_N . By taking advantage of the known statistical properties in the fluctuating fields, the OU Bayesian estimator extracts nearly the maximum amount of information from the detected single photons.

For the OU Bayesian estimator, the estimations are expected to depend on deviations of the statistical parameters used in the estimator from the true parameters. [Figure 6.3b](#) shows the ratio of the estimation variances over the corresponding CRLB as τ_N and σ used in the estimator move away from the true parameters. Variances close to the CRLB can still be obtained when the deviations of N and from the true parameters are smaller than 20%. The estimation variances, however, can exceed those obtained from the simple Bayesian estimator when τ_N and σ deviate from the true parameters by more than 50%. For the nuclear spin bath, τ_N and σ can be deduced from experiments such as Ramsey fringes and spin

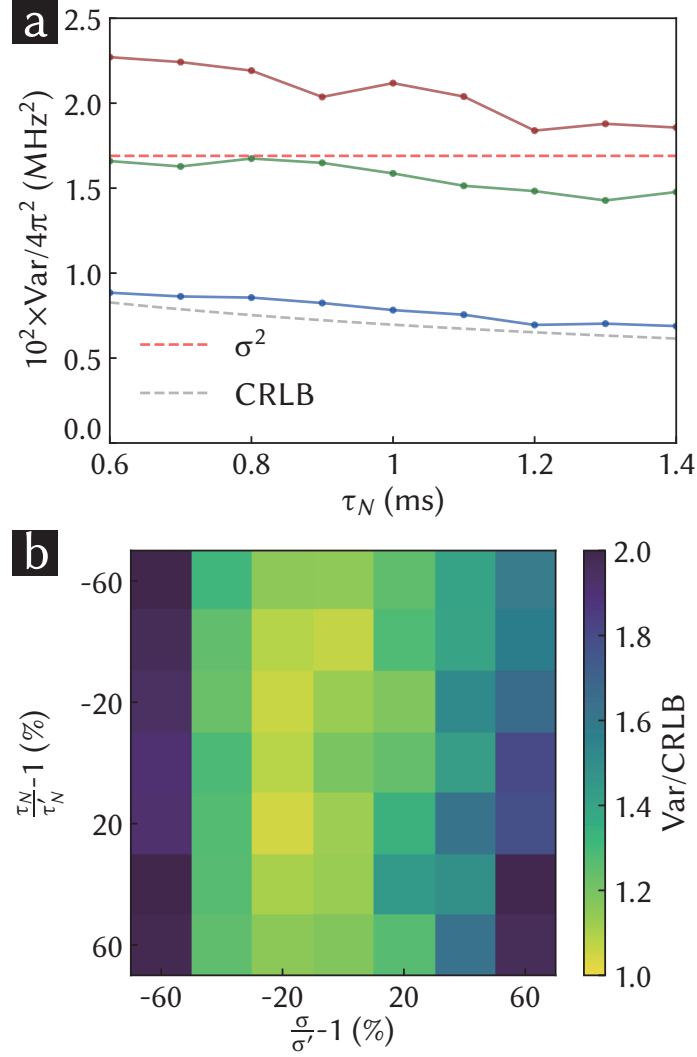


Figure 6.3. (a) The average variances for estimations obtained with the average count estimator (brown dots), simple Bayesian estimator (green dots), and OU Bayesian estimator (blue dots) as a function of the bath memory time τ_N . The estimation variances are also compared with the CRLB and the variance of the fluctuating ω_B . (b) The ratio of the estimation variance over the CRLB as τ_N and σ used for the OU-Bayesian estimation deviate from the true parameters, $\tau'_N = 1$ ms and $\sigma'/2\pi = 0.13$.

echoes. It should be noted that for the experimental implementation of the OU Bayesian estimator, feedback control, which uses the estimations to keep the NV in the dark state, can be employed for the further optimization of the statistical parameters used in the estimator. In addition, the feedback control can also serve as a verification for the estimations.

The choice of the CPT parameters, including Rabi frequency and bias $\Delta_0 = \delta - \langle \omega_B \rangle$, also strongly affects the effectiveness of the estimations. For example, little information on the magnetic field fluctuations can be obtained from a sequence of single photon emissions when Δ_0 is set to near 0. [Figure 6.4a](#) shows the variances for the OU Bayesian estimator as a function of Ω and Δ_0 , with all other parameters the same as those in [Figure 6.2](#). In this case, the optimal estimation is achieved with $\Omega/2\pi = 2.5$ MHz and $\Delta_0/2\pi = 0.2$ MHz. In comparison, the corresponding CRLBs shown in [Figure 6.4b](#) decreases with decreasing Ω and Δ_0 , which is expected from [Equation 6.3](#). As can be seen from [Equation 6.3](#), the CRLB is minimized when $g(\sigma) = \langle (\partial \rho_{ee} / \partial x)^2 / \partial \rho_{ee}(x) \rangle_x$ reaches maximum.

The optimal CPT parameters for the Bayesian estimations shown in [Figure 6.4a](#) reflect a tradeoff between a steep $\partial \rho_{ee} / \partial x$ and the need to avoid the zero bias region. This tradeoff is reached when the power-broadened CPT half linewidth, Ω^2 / κ , is comparable to σ . Note that as discussed, the CRLB is only achievable when the posterior is a Gaussian distribution [Van Trees \(2004\)](#), which occurs when ρ_{ee} depends either linearly or quadratically on x . Near zero bias, the CRLB cannot no longer been achieved with Bayesian estimations.

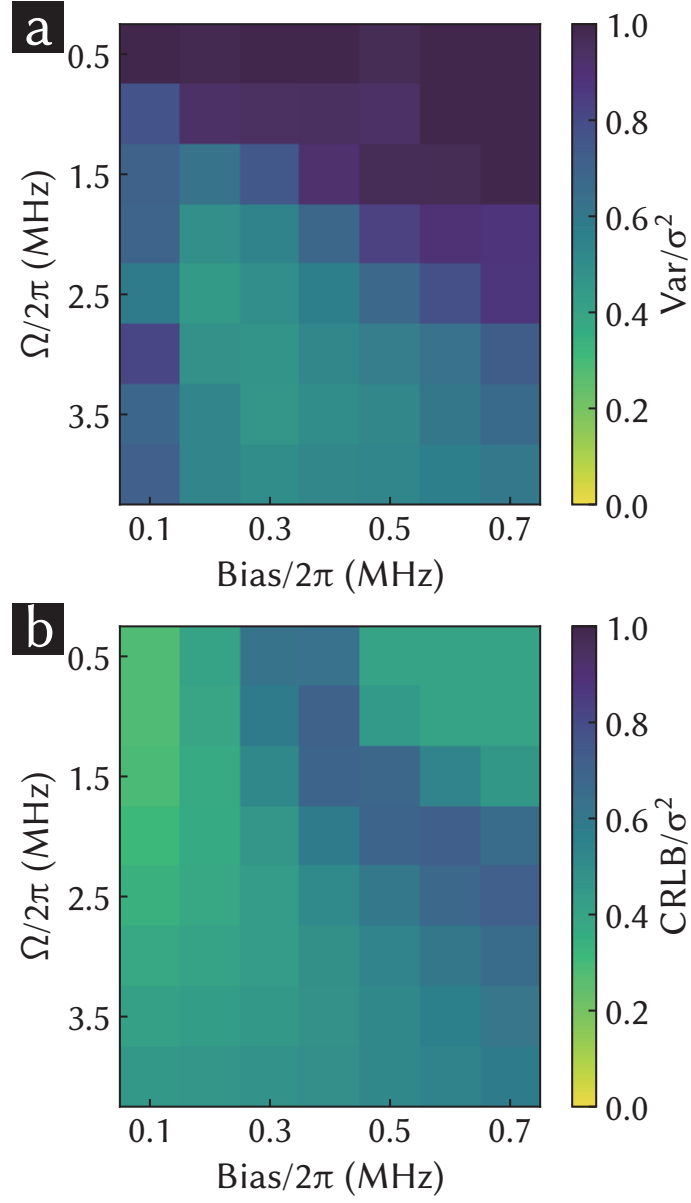


Figure 6.4. (a) Estimation variances vs and bias (i.e., Δ_0). The color bar corresponds to Var/σ^2 . (b) CRLBs for the OU Bayesian estimator vs Ω and bias. The color bar corresponds to CRLB/σ^2 .

6.5 Simulation of time series of photon counts

Magnetic field fluctuations induced by the nuclear spin bath of the diamond carbon lattice lead to corresponding fluctuations in ω_B , which can kick the NV into or out of the dark state. This in turn induces fluctuations in the excited state population and leads to a sequence of single photon emissions. It takes only a few spontaneous emission events for a NV to reach the steady state, as confirmed in an earlier experimental study [Lekavicius, Golter, Oo, and Wang \(2017\)](#). In addition, the nuclear spin bath fluctuates at a time scale many orders of magnitude slower than the NV radiative lifetime (12 ns). In this CPT setting, the single photon emissions from the excited state carry the information on the fluctuating nuclear spin bath.

For the Λ -type three-level system in [Figure 6.1](#) and with $\omega_0 = \nu_0$, the Hamiltonian in the rotating frame is given by

$$H = \frac{\hbar\Omega}{2} (|e\rangle\langle 0| + |e\rangle\langle 1| + |0\rangle\langle e| + |1\rangle\langle e|) + \hbar(\Delta_0 + x(t)) |1\rangle\langle 1|.$$

For CPT-based real-time sensing, we operate the CPT process with $C \gg 1$. In this regime, the power broadening far exceeds the intrinsic decoherence rate ($\gamma_s/2\pi \approx 0.1$ kHz). We will thus set $\gamma_s = 0$ in our simulation. The master equation is then given by

$$\dot{\rho}(t) = -\frac{i}{\hbar}[H(t), \rho(t)] + \frac{\Gamma}{2}\mathcal{D}(|0\rangle\langle e|)\rho(t) + \frac{\Gamma}{2}\mathcal{D}(|1\rangle\langle e|)\rho(t), \quad (6.4)$$

where $\mathcal{D}(O)\rho(t) = O\rho(t)O^\dagger - \{O^\dagger O, \rho(t)\}/2$ and we have also assumed $\kappa = \Gamma/2$.

We have used the SSE to simulate single photon emissions from the three-level system. The SSE, which tracks every collapse of the system, is given by

$$d|\psi\rangle = -i \left(H - \frac{i\Gamma}{2} |e\rangle\langle e| \right) |\psi\rangle dt + \frac{\Gamma}{2} \langle |e\rangle\langle e| \rangle |\psi\rangle dt \\ + \left(\frac{|0\rangle\langle e|}{\sqrt{\langle |e\rangle\langle e| \rangle}} - 1 \right) dN_0 + \left(\frac{|1\rangle\langle e|}{\sqrt{\langle |e\rangle\langle e| \rangle}} - 1 \right) dN_1,$$

where N_0 and N_1 are the accumulated photon counts from the $|e\rangle \leftrightarrow |0\rangle$ and $|e\rangle \leftrightarrow |1\rangle$ transitions, respectively. Without any optical emission, the system evolves under an effective Hamiltonian, $H - i\Gamma(|e\rangle\langle e| - \langle |e\rangle\langle e| \rangle)/2$. When a photon is emitted via the $|e\rangle \leftrightarrow |0\rangle$ (or $|e\rangle \leftrightarrow |1\rangle$) transition, the system collapse to the $|0\rangle$ (or $|1\rangle$) state. We take $\Gamma/2\pi = 13$ MHz [Tamarat et al. \(2006\)](#), a Rabi frequency of $\Omega/2\pi = 2.8$ MHz and a Raman detuning or bias of $\Delta_0 = \delta - \langle \omega_B \rangle = 2\pi \cdot 0.25$ MHz for the numerical simulation. [Figure 6.5b](#) shows the time series of the photons emitted by the three-level system, along with the corresponding excited state population. Only a small fraction of these photons is detected in a realistic sensing experiment. [Figure 6.5c](#) plots the time series of the photons detected, for which an overall collection/detection efficiency, $\eta = 1.6\%$, is used. Under these conditions, the average photon count rate is approximately 10000 per second. For comparison, the underlying fluctuations in ω_B are also plotted in [Figure 6.5c](#). Because of the small detection efficiency, it is difficult to directly discern the patterns of the fluctuations from the time series of the photon counts.

Alternatively, we can simulate the single photon emissions by using the population of the excited state and assuming that the system has a Poisson distributed probability to emit a photon with the average detection rate given by $\eta\Gamma\rho_{ee}$. For this simulation, we make the approximation that ρ_{ee} adiabatically follows the magnetic field of the bath, since the timescale of the bath fluctuation is

much longer than the NV spontaneous emission lifetime. As shown in [Figure 6.6b](#), magnetic field estimations using the single photon emissions obtained with the steady-state excited-state population exhibit the same behavior as those obtained with the SSE. For numerical calculations that require more than a few hundred runs, we have thus simulated single photon emissions using the steady-state excited-state population.

6.6 Recurrent neural network as estimator

In addition to the Bayesian based approach and the simple average approach, we also tried use the Recurrent neural network (RNN) to be the estimator of this sensing problem. RNN, especially the sequence-to-sequence structure, is widely used in the field of processing time sequence data. The input sequence will be feeded to a same RNN unit successively, then we compare the output with the ground truth then update the parameters of the NN. Finally, we will expect the RNN unit will learn the correlation inside the time sequence data. Since the sensing problem is all about give an estimation time sequence based on the information time sequence, so RNN is a natural approach for it.

For our project, the field fluctuation $\{\mathbf{X}\} = \{x_1, \dots, x_n\}$ results in the outcomes $\{\mathbf{Y}\} = \{y_1, \dots, y_n\}$, so the sensing problem is to find a RNN to realize

$$\text{RNN}(\mathbf{Y}) = \hat{\mathbf{X}} \approx \mathbf{X}. \quad (6.5)$$

But since we do the realtime sensing, we only use the historical information, a original sequence to sequence strategy is not ideal as it can not avoid the RNN use the future information. So, what we do is a modification of it. We train the RNN to meet the goal,

$$\text{RNN}(\mathbf{Y}_{t-\Delta t:t}) = \hat{x}_t, \quad (6.6)$$

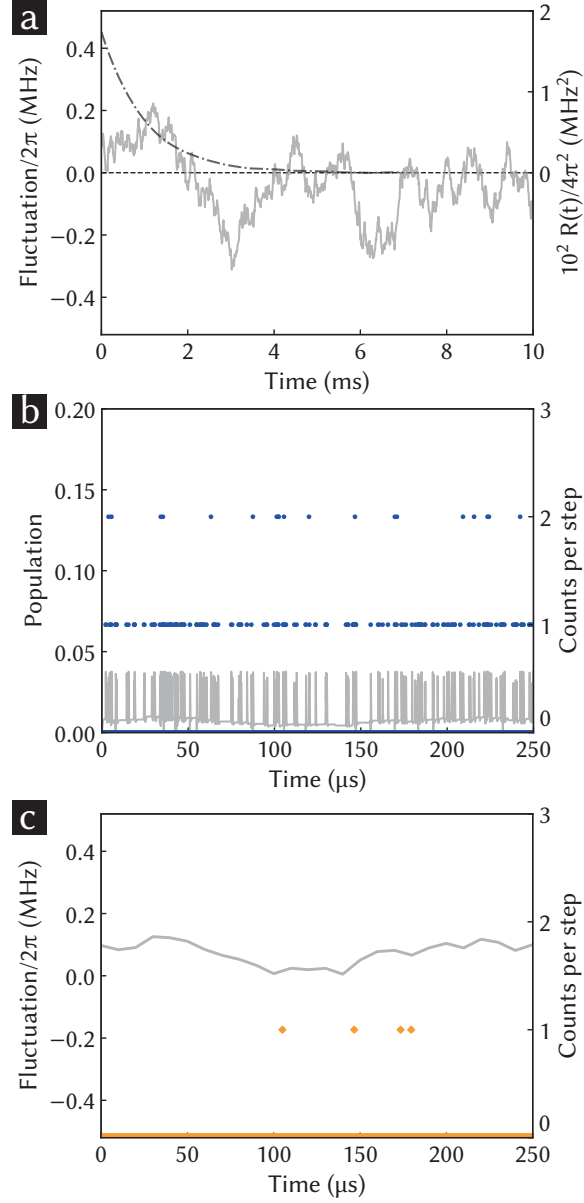


Figure 6.5. (a) Simulated fluctuations in ω_B (grey line) and the auto-correlation $R(t)$ (black line). (b) The excited-state population (grey lines) as a function of time and the corresponding time series of photon counts emitted by the NV center (blue dots). (c) The time series of the photon counts detected (orange diamond) and the underlying fluctuations in ω_B . The CPT parameters used are $(\Omega, \Gamma, \Delta_0)/2\pi = (2.8, 13, 0.25)$ MHz. The step size used for the photon counting is $0.1 \mu\text{s}$.

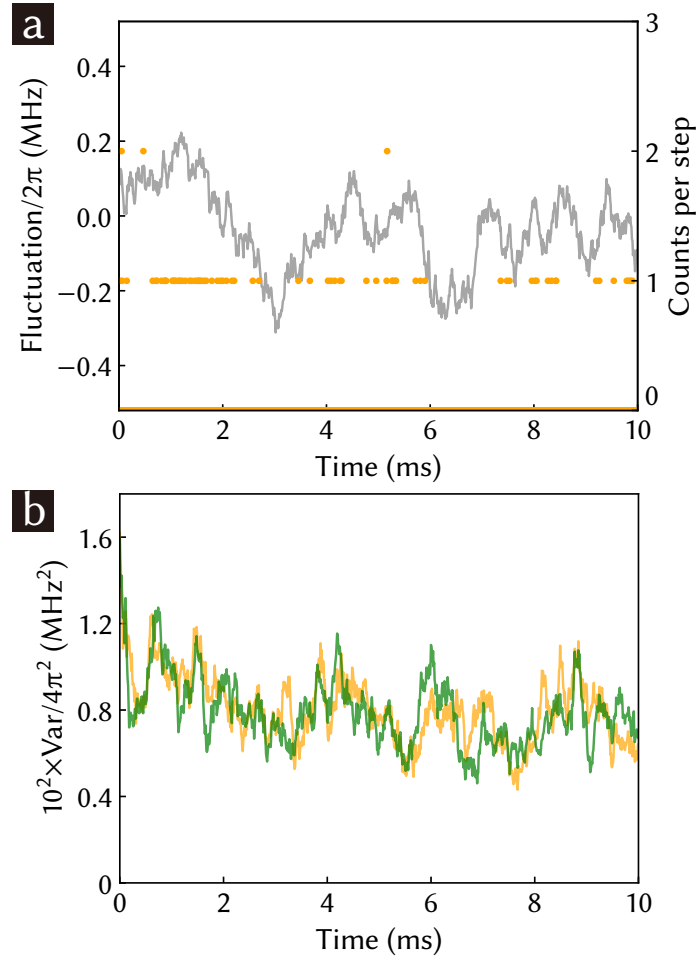


Figure 6.6. (a) Time series of photon counts generated by the SSE, along with the underlying fluctuations in ω_B . The step size for photon counting is $10\ \mu\text{s}$. (b) Estimation variances obtained from time series of photon counts generated by the SSE and those by the steady-state population of the excited state. The results shown are averages over 100 runs. The bath and CPT parameters used are the same as those for [Figure 6.5](#).

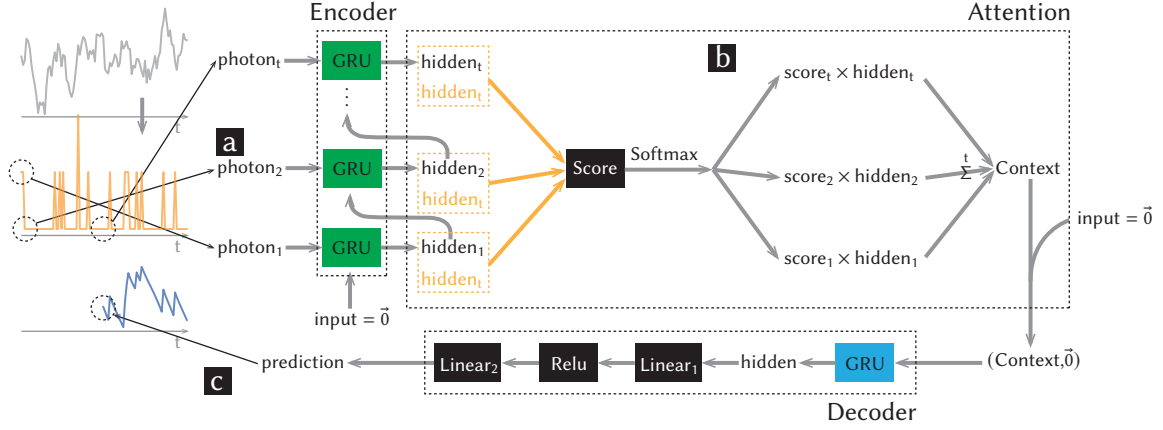


Figure 6.7. The diagram of how the RNN works.

$$\{\hat{\mathbf{X}}\} = \{\hat{x}_1, \dots, \hat{x}_n\} \approx \{\mathbf{X}\} = \{x_1, \dots, x_n\}.$$

In Figure 6.7, we show how the RNN works. In (a), we feed the photon sequence $\{y_1, \dots, y_n\}$ (here we choose $n = 50$ then $\Delta t = 1\text{ms}$) to the RNN unit GRU (Gated recurrent unit), along with the initial hidden state $\vec{0}$. The GRU unit accept two inputs, input data and input hidden state. After GRU receive y_1 and initial hidden state $\vec{0}$, it will update the hidden state according the gates and layers inside the units. Finally, GRU will output a new hidden state h_1 which is the input hidden state of next step. For the next step, the same GRU receive y_2 and hidden state h_1 then update again. This part of the estimator structure is called Encoder, it convert the input information to a vector (hidden state). After the Encoder, the Attention structure uses the whole hidden state to get the score. The score is used to do the weighted arithmetic mean of the hidden state, it evaluate which hidden or say which historical information is more important for estimation. After the evaluation, Attention outputs the context vector. This context vector is along with another initial hidden state to be feeded to a new GRU. This GRU and follow-up layers are called decoder, it decodes the context to the prediction $\hat{x}_n$. Then we

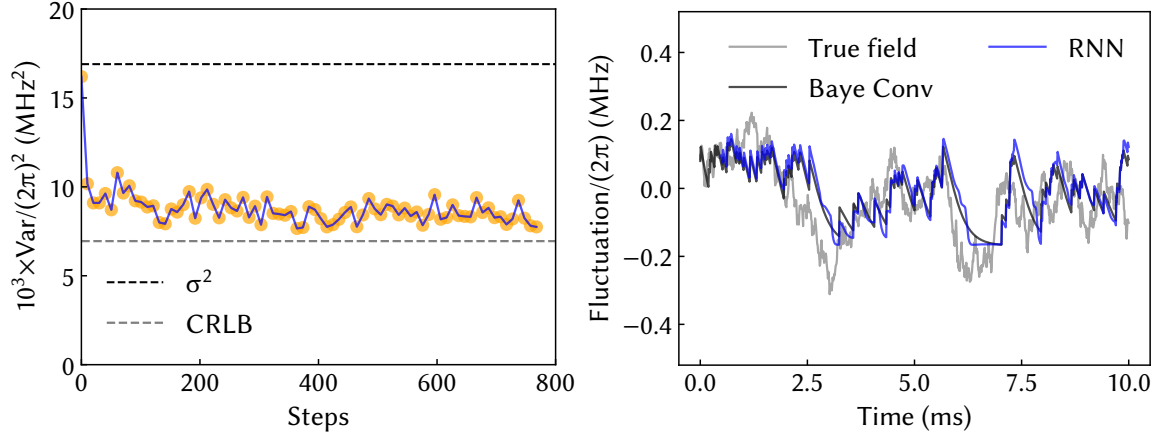


Figure 6.8. The loss against steps when training the RNN, and corresponding performance.

evaluate the loss between prediction $\hat{x}_n$ and x_n , then update the parameters of RNN accordingly.

To build the RNN, we use the Python package PyTorch, and choose a training dataset of size 8000, batch size 100 to evaluate the gradient, learning rate 0.001, hidden size 256, and an Adam Optimizer. In Figure 6.8, we show the training loss goes down after about 100 steps and become stable after hundreds of steps. When we use a trained RNN to do the estimation (1000 slots for 10ms), we feed the first 50 slots to the RNN then only call Encoder GRU one time to update the hidden and use Attention to get the new context. Finally, we feed the context to the Decoder and get the new estimation.

As shown in Figure 6.8, trained RNN basically converges to our OU Bayesian estimator, therefore verified again it's the optimal estimator we can get.

6.7 Conclusion

We have shown theoretically that CPT can enable continuous real-time sensing with a single quantum sensor such as a NV center in diamond. The use of Bayesian inference for the real-time sensing allows effective estimation of magnetic

fields from a time series of photon counts, for which the average photon count per time interval is much smaller than 1, and can lead to estimation variances that approach the CRLB. While a NV center along with the surrounding nuclear spin bath has been used as a model system for the theoretical analysis, other spin systems such as defect centers in SiC can also serve as CPT-based quantum sensors [Awschalom, Hanson, Wrachtrup, and Zhou \(2018\)](#). In addition to the nuclear spin bath, the real-time sensing can be applied to studies of time-varying magnetic field changes or fluctuations in a variety of systems, for example, two-dimensional materials on a diamond surface [Lovchinsky et al. \(2017\)](#); [Thiel et al. \(2019\)](#). Together with feedback control techniques, the real-time sensing can also be used for the protection of a spin qubit from the surrounding magnetic fluctuations [Shulman et al. \(2014\)](#). Overall, CPT-based real-time sensing can add a new and powerful tool to the emerging technology of quantum sensing.

CHAPTER VII

CONTINUOUS REAL-TIME MAGNETIC SENSING VIA COHERENT POPULATION TRAPPING: EXPERIMENT

The results presented in this chapter were published in [Turner et al. \(2022a\)](#).

7.1 Introduction

The theoretical framework of CPT-based sensing is developed in Chapter VI, where simulations show an OU-Bayesian estimator approaching the CRLB. Here we ask whether these conclusions hold in a real experiment, where noise, imperfect charge initialization, finite collection efficiency, and real-time processing latency all matter. Crucially, the achievable end-to-end latency and photon detection rate set the fastest noise dynamics we can reliably track. Quantifying these practical limits tests the robustness of the CPT-based sensing approach and indicates how to deploy it in realistic settings, including which timescales are accessible and how bias and linewidth should be chosen. These insights also guide applications of CPT-based sensing beyond the present system and suggest concrete routes for improving performance.

In this chapter we present the experimental demonstration of real-time sensing of a fluctuating magnetic field by exploiting coherent population trapping (CPT) of a NV center. For the CPT process [Golter et al. \(2014, 2016\)](#); [Santori et al. \(2006\)](#); [Togan et al. \(2011\)](#); [Yale et al. \(2013\)](#), the NV center is prepared in a special superposition of two spin states, i.e., the dark state, which prevents optical excitation and emission through destructive quantum interference [Scully and Zubairy \(1997\)](#). A time-varying magnetic field can kick the NV center out of or into the dark state, leading to a sequence of single-photon emissions from the NV center. We have used this sequence of single-photon emissions to estimate the

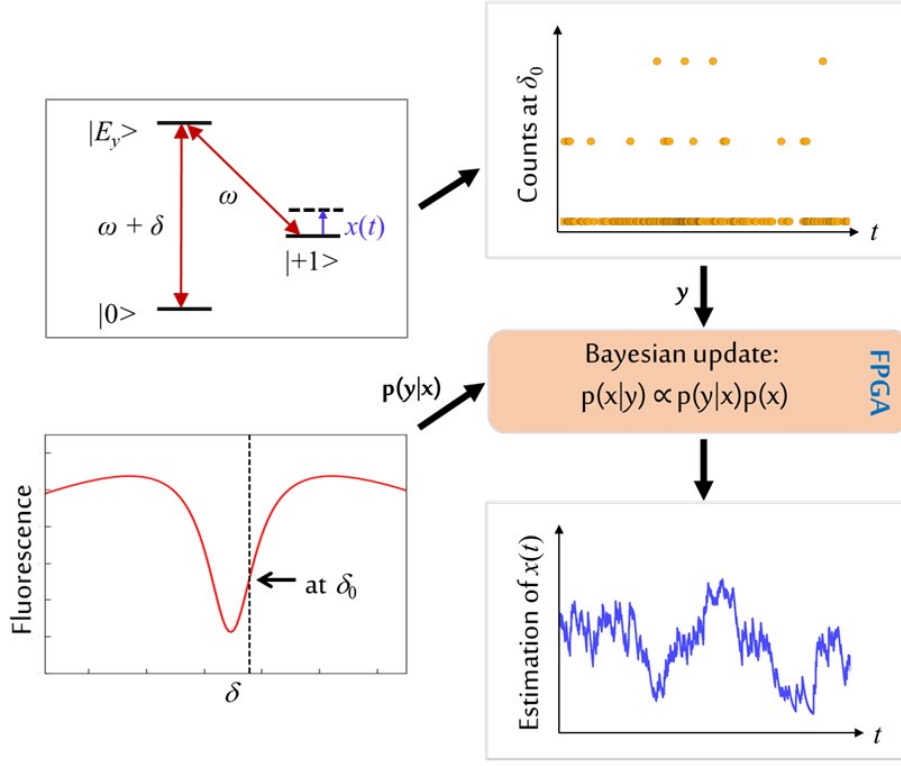


Figure 7.1. Schematic illustrating real-time sensing of magnetic fields using single-photon emissions from a NV center prepared in a CPT setting. Bayesian inference is used to estimate time-varying magnetic fields from the corresponding time series of photon counts. The upper left figure shows the Λ -type three-level system used for the CPT process. The lower left figure shows schematically the fluorescence from the excited state as a function of the detuning between the two applied optical fields. The fluorescence is quenched when the detuning equals the frequency separation between the two lower states.

time-varying magnetic field in real time, with the estimation carried out in a field programmable gate array (FPGA), as illustrated in [Figure 7.1](#).

Experimentally, the detection rate of the single-photon emissions limits the effective updating rate of the real-time estimation. Since the overall collection/detection efficiency for optical emissions from a NV center is only a few percent under typical experimental conditions, a key challenge is to obtain dynamical information with the few photons detected. We show that by taking

advantage of the statistical properties of the time-varying fields, we can use Bayesian inference to update dynamical information in real time with the detection of just a single photon, demonstrating real-time magnetometry with single detected photons. A detailed analysis further shows good agreement between the experiment and the theoretical expectation. The experimental results are also compared with the classical Cramér-Rao lower bound (CRLB) for the estimation process.

For real-time magnetic-field sensing, we couple the $m_s = 0$ and $m_s = +1$ ground spin states to the E_y excited state in a NV center through two dipole optical transitions driven respectively by two resonant optical fields (see [Figure 7.1](#)). In the limit of equal Rabi frequency, Ω , the dark state for the Λ -type three-level system can be simply written as $(|m_s = 0\rangle - |m_s = +1\rangle)/\sqrt{(2)}$. As illustrated in [Figure 7.1](#), the electron becomes trapped in the dark state and the optical emission is quenched when the Raman resonance condition $\Delta = \delta - \omega_B = 0$ is satisfied, where ω_B is the frequency separation between the two spin states and δ is the detuning between the two laser fields.

A time-varying magnetic field leads to a corresponding change in ω_B . With an average Raman detuning, $\delta - \langle\omega_B\rangle$, near the half width of the CPT spectral response as illustrated in [Figure 7.1](#), the optical excitation of the NV center and the resulting time sequence of single-photon emissions will be directly correlated with the field variation as long as the variation in ω_B does not exceed the half width. Note that it takes only a few spontaneous emission events for the CPT process to reach steady state [Lekavicius et al. \(2017\)](#). For field variations with a timescale that is long compared with the NV radiative lifetime (12 ns [Tamarat et al. \(2006\)](#)), the time series of photon counts, $\{\mathbf{y}_n\} = \{y_1, y_2, \dots, y_n, \dots\}$, where y_n is the number of photons detected during the n th time interval, each with a

duration of τ , carries the information on the corresponding change in ω_B , denoted as $\{\mathbf{x}_n\} = \{x_1, x_2, \dots, x_n, \dots\}$ [S. H. Wu et al. \(2021\)](#).

We have used three different estimators to output a time series of estimated frequency changes, $\{\tilde{\mathbf{x}}_n\} = \{\tilde{x}_1, \tilde{x}_2, \dots, \tilde{x}_n, \dots\}$, from the observed time series of photon counts, $\{\mathbf{y}_n\}$. Bayesian inference, which has been used extensively in earlier sensing studies with NV centers [Bonato et al. \(2016\)](#); [Santagati et al. \(2019\)](#), follows the Bayes update rule given by,

$$p(x_n|y_n, y_{n-1}, \dots, y_1) \propto p_{\bar{y}}(y_n|x_n) \times p'(x_n|y_{n-1}, \dots, y_1), \quad (7.1)$$

where $p'(x_n|y_{n-1}, \dots, y_1)$ is the prior probability distribution based on the previous time series of photon counts, $p(x_n|y_n, y_{n-1}, \dots, y_1)$ is the posteriori probability distribution, and $p_{\bar{y}}(y_n|x_n)$ is the likelihood of detecting photons in the n th time interval given x_n . We assume that $p_{\bar{y}}(y_n|x_n)$ follows a Poisson distribution,

$$p_{\bar{y}}(y_n|x_n) = \frac{\bar{y}^{y_n} e^{-\bar{y}}}{y_n!},$$

where $\bar{y}_n \propto \tau \rho_{ee}(x_n)$ is the expected average photon count per updating time interval, with ρ_{ee} being the excited-state population. The estimation is then given by

$$\tilde{x}(t) = \int p(x, t) x dx.$$

To achieve maximum time-resolution, we usually have $\bar{y}_n < 1$.

The prior probability distribution in [Equation 7.1](#) can be significantly improved if the statistical properties of the time-varying field is known. A variety of time-varying processes can be modeled as an Ornstein-Uhlenbeck (OU) process [de Lange et al. \(2010\)](#); [Dobrovitski et al. \(2009\)](#); [Witzel et al. \(2012\)](#); [C. Zhang and Mølmer \(2020\)](#), which is both Gaussian and Markovian and features an

autocorrelation function given by

$$R(t) = \langle x(t_0)x(t_0 + t) \rangle = \sigma^2 e^{-|t|/\tau_c}.$$

with σ^2 and τ_c being the variance and correlation time, respectively. Details of the improved prior, which takes into account the statistical properties of the OU process, are discussed as well as in an earlier theoretical study [S. H. Wu et al. \(2021\)](#).

In addition to the Bayesian inference, a more intuitive approach is to simply use photon counts detected in a relatively long duration, τ_a , to estimate $x(t)$ by using

$$y_n^{(a)} \propto \tau_a \rho_{ee}(\tilde{x}_n),$$

where $y_n^{(a)}$ is the photon count accumulated between time $n\tau - \tau_a$ and $n\tau$. To achieve an acceptable photon count (of order 10 or greater) for this average count estimator, we need $\tau_a \gg \tau$. For the experimental results presented in this chapter, we took $\tau_a = 1.4\tau_c$.

7.2 Results

Our experimental studies were carried out at 10 K with an electronic grade chemical-vapor-deposition grown diamond sample from Element Six. A solid immersion lens (SIL) fabricated on the diamond sample along with a confocal optical microscopy setup was used for the optical excitation and fluorescence collection of a single NV [Golter et al. \(2014\)](#); [Golter and Wang \(2014\)](#). A permanent magnet was used to split the $m_s = \pm 1$ states by 590 MHz. The two resonant optical fields for the CPT process, with an estimated $\Omega/2\pi$ of order 5 MHz, were derived from a 637 nm diode laser and a sideband generated by an electro-optical modulator (EOM). Under these conditions, the CPT dip obtained

from the Λ -type system depicted in [Figure 7.1](#) features a linewidth of 11.6 MHz, which includes contributions from hyperfine splitting (2.2 MHz), spin dephasing (0.6 MHz) as well as power broadening (near 5 MHz). Details of the experimental method will be presented in the next section.

For a proof-of-principle demonstration, we apply an external time-varying magnetic field to the NV center by passing an electric current through the coplanar waveguide (CPW) fabricated next to the SIL. The electrical current, which is generated by an arbitrary function generator (AFG), follows a simulated OU process with given σ and τ_c and with $\langle x(t) \rangle = 0$. The use of an external field has enabled us to investigate the dependence of CPT-based real-time sensing on key parameters such as σ and τ_c .

For the estimation experiment, the NV center is first initialized to the $m_s = 0$ ground state by a 10 μ s green laser pulse ($\lambda = 532$ nm). This is followed by the application of two resonant optical fields for CPT and the detection of fluorescence from the NV center. To avoid NV ionization due to the resonant optical excitation, we limit the CPT and the fluorescence detection to a duration of 100 μ s before reinitializing the NV with a 10 μ s green laser pulse. Numerical calculations of the real-time estimation are carried out in a FPGA in a Keysight M3302A card, which also contains a digitizer and an arbitrary waveform generator (AWG). The digitizer tallies the photon counts per update time interval and the AWG outputs the corresponding estimation. The overall process for generating a single update takes about 7 μ s. We thus set the updating time interval to be $\tau = 10\mu$ s. Details of the experimental setup are presented in the supplement.

[Figure 7.2a](#) and [b](#) show, as an example, estimations obtained with the OU-Bayesian estimator (which takes into account the statistical properties of

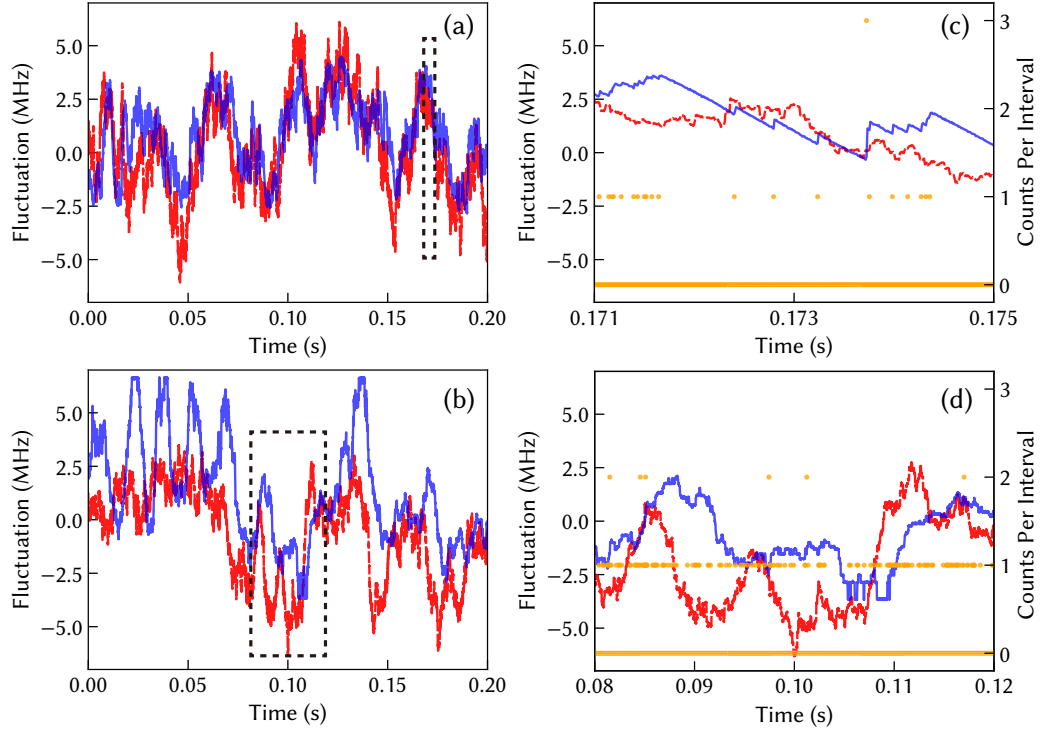


Figure 7.2. (a) and (b) Estimations (blue solid line) of the fluctuations in $\omega_B/2\pi$ obtained with the OU-Bayesian and the average count estimators, respectively, along with the actual fluctuations (red dashed line). (c) and (d) A closer look at the results in the dashed-line boxes in (a) and (b), respectively, along with the corresponding photon counts (orange dots) per updating interval.

the OU process) and with the average count estimator, respectively, as well as a direct comparison between the estimations and the actual frequency changes. For these experiments, we used $\sigma/2\pi = 2.2$ MHz, $\tau_c = 5$ ms, and an average photon count rate of 5400 per second. The Raman detuning or the bias was set to $(\delta - \langle\omega_B\rangle)/2\pi = 4$ MHz (the choice of the bias will be discussed in detail later). As can be seen from these figures, estimations obtained with the OU-Bayesian estimator closely follow the actual field dynamics, whereas estimations obtained with the average count estimator exhibit large deviations from the actual frequency changes for extended periods of time.

To further highlight the differences between these two estimators, we compare estimations obtained in relatively short durations (marked by the dashed-line boxes in [Figure 7.2a](#) and [b](#)) with the corresponding time series of photon counts. [Figure 7.2c](#) shows that the OU-Bayesian estimator can effectively update the dynamics of the frequency change in real time with the detection of just a single photon. Relatively small amount of information is acquired if no photons are detected. The saw-tooth type estimation observed in [Figure 7.2c](#), which is directly correlated with the detection of single photons, corresponds to the updating that takes place at the level of single detected photons. Note that an earlier study has used the complete CPT spectrum of a single NV for the sensing of the magnetic fluctuations induced by the nuclear spin bath [Togan et al. \(2011\)](#), for which it takes about 100 detected photons to obtain a single estimation.

In comparison with the OU-Bayesian estimator, the average count estimator is susceptible to photon number shot noise, which leads to large spikes in the estimations observed in [Figure 7.2b](#). The asymmetry in the spikes is in part due to the choice of the Raman bias, which is considerably smaller than the half width

of the CPT spectral response. As can be seen from [Figure 7.2d](#), there is also a large delay between the estimation and the actual frequency change due to the relatively long τ_a used. Note that significantly increasing τ_a and thus $y_n^{(a)}$ leads to reduced fluctuations in the estimation, but at the expense of time resolution. Average count estimators can thus work well for the sensing of static signals. For example, electromagnetically induced transparency (EIT), which is closely related to CPT, of an ensemble of NV centers has been used successfully for static sensing [Acosta et al. \(2013\)](#).

For a quantitative analysis of the estimations, we have examined the estimation variance defined as $\text{Var}[\tilde{x}_n] = \langle (\tilde{x}_n - x_n)^2 \rangle$ and in particular $\langle (\tilde{x}_n - x_n)^2 \rangle / \sigma^2$, denoted as Var/σ^2 . We have also carried out detailed comparisons between the estimation variances obtained from the experimentally observed time series of photon counts and those obtained from the theoretically simulated time series of photon counts. The theoretical model used for the simulations has been presented in our earlier study [S. H. Wu et al. \(2021\)](#) and is also discussed in detail in the supplement. For sensing of a time-varying signal with a given distribution, the estimation performance averaged over the entire distribution rather than that at a single point is important. In this regard, the sensing sensitivity as defined for static sensing is no longer applicable. Instead, we use the estimation variance as an effective measure of the sensing performance [Bonato and Berry \(2017\)](#).

[Figure 7.3a](#) plots Var/σ^2 as a function of c and compares the relative variances obtained with the OU-Bayesian estimator, the average count estimator, and the simple Bayesian estimator, which takes no account of the statistical properties of the OU process. As expected, the variances for both the average count and the OU-Bayesian estimators decrease with increasing τ_c . Nevertheless, Var/σ^2

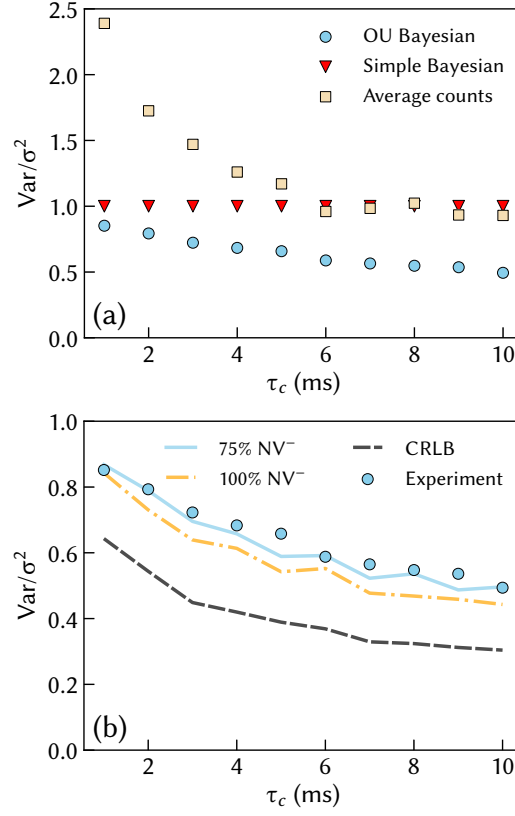


Figure 7.3. (a) Comparison of estimation variances obtained with OU-Bayesian, simple Bayesian, and average count estimators as a function of τ_c . (b) Comparison of the estimation variances obtained with the OU-Bayesian estimator with the corresponding simulated values, for which a charge initialization fidelity of 100% (dotted line) and 75% (solid line) is assumed. The dashed line shows the calculated CRLB. Experimental parameters used are the same as those for [Figure 7.2](#) unless otherwise specified.

for the average count estimator is far above 1 when τ_c is near 1 ms and only falls slightly below 1 when τ_c approaches 10 ms. In comparison, Var/σ^2 for the OU-Bayesian estimator remains significantly below 1 when τ_c approaches 1 ms.

It should be noted that with $\text{Var}/\sigma^2 \approx 1$ for the range of τ_c used in [Figure 7.3a](#), the simple Bayesian estimator essentially provides no information on the time-varying field. As shown in our earlier theoretical study [S. H. Wu et al. \(2021\)](#) and confirmed in additional experiments, estimations obtained with the simple Bayesian estimator quickly converge to the average value, with $\tilde{x}(t) \approx 0$.

[Figure 7.3b](#) compares the experimentally observed variances with the corresponding simulated variances. Both variances were obtained with the OU-Bayesian estimator. We found that the experimentally observed variances are considerably greater than the simulated variances, for which a perfect charge initialization for the negatively charged NV center is assumed. A detailed analysis shows that for our experiments, the charge initialization fidelity is about 75%. Including the non-ideal charge initialization in the model yields a good agreement between the experiment and the simulation. [Figure 7.3b](#) also shows that the experimentally observed variance is considerably above the calculated CRLB (see [S. H. Wu et al. \(2021\)](#) for a detailed discussion on the calculation of the CRLB). Theoretically, CRLB can be reached only when the relevant CPT spectral response is linear or quadratic, which is not the case for the actual experiment.

The estimation variances depend on the choice of CPT parameters, especially the bias. [Figure 7.4a](#) shows Var/σ^2 obtained with the OU-Bayesian estimator as a function of the bias, with other experimental conditions the same as those used for [Figure 7.2a](#). As expected, the estimations become ineffective when the bias approaches 0 (i.e., near the bottom of the CPT dip), in agreement with

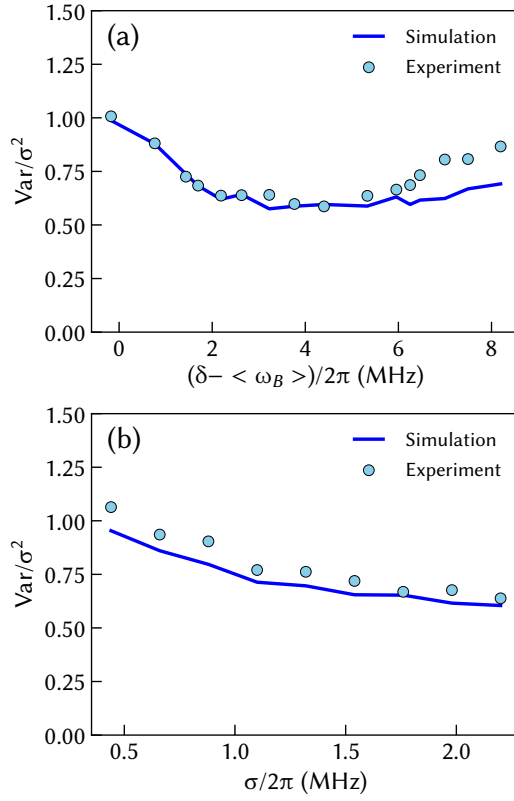


Figure 7.4. (a) Estimation variances obtained with the OU-Bayesian estimator as a function of the bias (i.e., Raman detuning). (b) Estimation variances obtained with the OU-Bayesian estimator as function of σ . The solid lines in both figures show the corresponding simulated values.

the theoretical expectation. The estimations also perform poorly when the bias significantly exceeds the half width of the CPT dip. Note that near the wings of the CPT dip, effects of hyperfine splitting of the relevant spin states, which are not included in the theoretical model, become important, leading to the observed variances that are larger than the simulated values, as shown in [Figure 7.4a](#).

The sensitivity and range of the real-time sensing process also depend on the CPT parameters. In particular, there is a trade-off between the largest and the smallest frequency changes that can be sensed via a CPT process. The smaller the CPT linewidth, the more sensitive the CPT-based sensing process becomes, whereas the CPT linewidth sets the range of the sensing process. [Figure 7.4b](#) shows Var/σ^2 obtained with the OU-Bayesian estimator as a function of σ , with other experimental conditions the same as those used for [Figure 7.2a](#). For relatively small σ , Var/σ^2 increases with decreasing σ and goes above 1 when $\sigma/2\pi$ falls below 0.5 MHz. In this case, the large CPT linewidth (11.6 MHz as mentioned earlier) used in the experiment limits the sensitivity of the real-time sensing process. The experimental results are in good agreement with the simulated values, as shown in [Figure 7.4b](#).

We can further improve the sensitivity of the real-time sensing process by reducing the CPT linewidth. For example, polarizing the ^{14}N nuclear spin with optical pumping avoids the complication of hyperfine splitting [Goldman, Patti, Levonian, Yelin, and Lukin \(2020\)](#). The use of isotopically purified diamond reduces dephasing induced by the nuclear spin bath and can lead to a CPT linewidth less than 1 kHz [Balasubramanian et al. \(2009\)](#). Additional improvements in the overall sensing performance can also be achieved through better charge

initialization, for example, through the use of real-time control techniques [Hopper, Lauigan, Huang, and Bassett \(2020\)](#).

7.3 Experimental details

7.3.1 Optical Setup

The experimental studies described in the main text were carried out with a home-built confocal microscopy setup shown in [Figure 7.5a](#). The sample used is an electronic grade chemical-vapor-deposition grown diamond (from Element Six, Inc.), which is mounted on the cold finger of an optical cryostat (from Montana Instruments, Inc.) cooled down to 10 K. The fluorescence is collected with an objective (Nikon L Plan 100x/0.85 NA) and is filtered by a 637 nm notch filter and a 150 nm bandpass filter centered at 750 nm. The collected fluorescence is launched into an optical fiber (with a core diameter of 10 μm) coupled to a Perkin Elmer SPCM-AQR-16-FC Avalanche Photodiode (APD) for single photon counting. The fluorescence collection is also enhanced by a solid immersion lens (SIL) milled on the surface of the diamond sample with the use of a focused ion beam (FIB) ([Figure 7.5b](#)) [Jamali, Gerhardt, Rezai, Wrachtrup, and Fedder \(2014\)](#).

The NV is initialized into its negative charge state, NV^- , with $m_s = 0$ via off-resonant excitation with a 532 nm (green) laser (LCS-0532-TFC-0050-05 from Laser Flow Technologies). Coherent population trapping (CPT) is driven by two optical fields near 637 nm, which are derived from a 637 nm diode laser (New Focus TLB-6700) and a sideband generated by a fiber coupled Jenoptic electro-optical modulator (EOM). The optical paths of the green and red laser beams are combined in free space with a Semrock Di02R594 dichroic mirror before reflecting off a second dichroic mirror (Semrock Di02R 635) and into a 2D-galvanometer (from Thorlabs, Inc.), which is used to steer the beam to the desired NV.

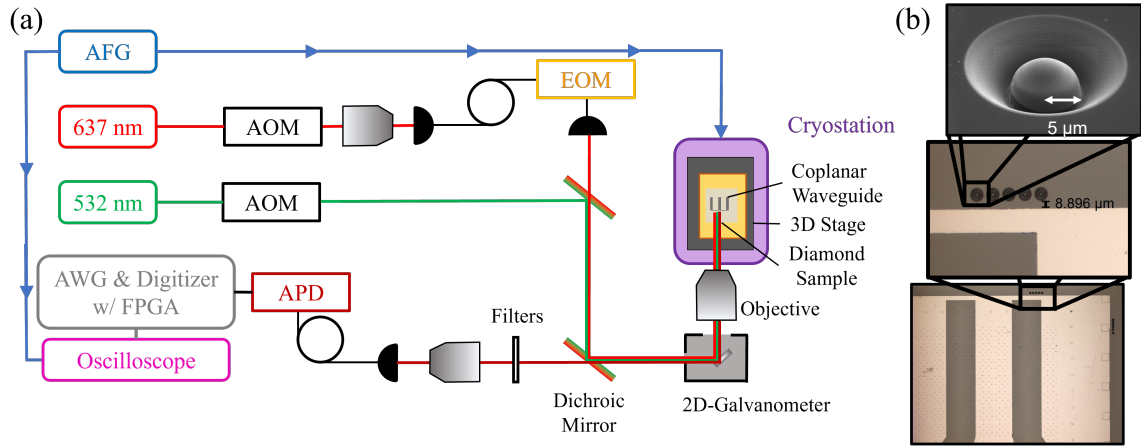


Figure 7.5. (a) Schematic of the experimental setup. Fluorescence from the NV (maroon) propagates backthrough the excitation path, transmitting through a dichroic mirror before being sent to an avalanchephotodiode (APD) detector. The 532 nm (green) laser is used for the initialization of the NV. The 637 nm (red) laser is used for coherent population trapping. The acousto-optic modulators (AOMs) generate the suitable pulse sequence. The estimations are carried out in a field programmable gate array (FPGA) in a Keysight M3302A card, which also contains a digitizer and an arbitrary waveform generator (AWG). An arbitrary function generator (AFG) is used for the generation of an external magnetic field. b) Optical images of the diamond sample with the deposited aluminum coplanar waveguide and focused-ion-beam milled solidimmersion lens (SIL) (top). The SIL is 10 μm in diameter.

7.3.2 Pulse Sequence and Effects of NV Ionization

Continuous resonant excitation of a NV can ionize the NV to its neutral charge state, NV^0 [Robledo et al. \(2011\)](#). To minimize the ionization, we interrupt the sensing process periodically with a 10 μs green laser pulse, with the APD gated off during this initialization or reinitialization process. [Figure 7.6a](#) shows the pulse sequence used for the experimental studies presented in the main text, for which the duration between the two successive green laser pulses is set to 100 μs . Acousto-optic modulators (AOMs) are used for the generation of the desired laser pulse sequences, as shown in [Figure 7.5a](#).

The NV^0 center not only makes no contribution to the sensing process, but also distorts the time series of the photon counts used for the estimation. The effects of the NV^0 can be seen clearly from the photon number probability distributions observed during the sensing process, as shown in [Figure 7.6b](#) and [c](#), for which the duration between the two successive initialization pulses is set to 100 and 500 μs , respectively. For these photon number probability distributions, we time-tag the detected photons for 10 μs and bin the photon counts in 10 ms bins. The solid red line in [Figure 7.6b](#) and [c](#) are the corresponding Poisson distribution, for which the average counting rate is set by the experimentally observed value. As evident from these figures, the photon number distribution in [Figure 7.6c](#) exhibits greater deviation from the Poisson distribution as well as lower average counting rate than that in [Figure 7.6b](#), indicating increasing effects of NV ionization with the increasing separation between successive initialization pulses.

7.3.3 Fidelity of Charge Initialization

To determine the fidelity of charge initialization or reinitialization, we apply a repeating pulse sequence consisting of a 10 μs , 532 nm initialization pulse

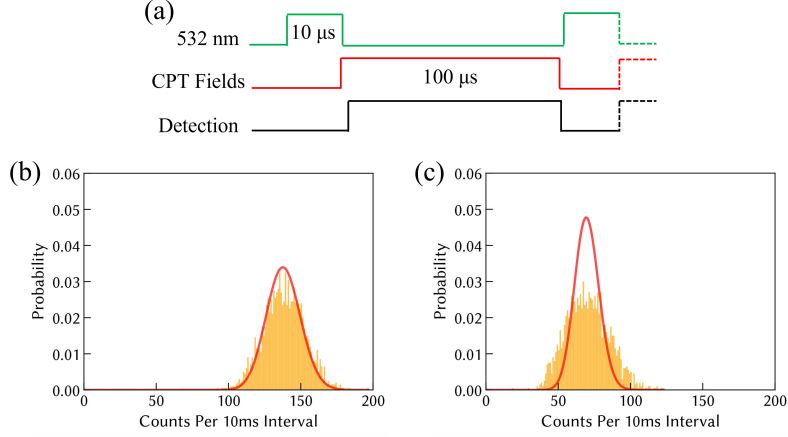


Figure 7.6. (a) Schematic of the pulse sequence used for the real-time sensing, illustrating that the separation between two successive initialization green pulses is $100 \mu\text{s}$. (b) and (c) Photon number probability distribution obtained from time-tagged photon counts, for which the separation between two successive green pulses is 100 and $500 \mu\text{s}$, respectively. The red curves are the Poisson distributions with the average photon counts set by the experimentally observed value.

followed by a 1 ms resonant excitation pulse (near 637 nm) for 20s, time-tagging each photon received. We then bin the number of photons detected during each red pulse (which is our readout interval). [Figure 7.7](#) shows the distribution of photon counts per readout interval, which appears to be bimodal. We apply a least-squares fit of the observed distribution to the sum of two Poisson distributions, one for each charge state. An average count rate of 0.65 and 4.45 counts per readout interval are obtained from the numerical fit for the NV^0 and NV^- , respectively. The relative contribution of counts from the NV^- determines the fidelity for charge initialization. The observed fidelity of approximately 75% is consistent with that of near-surface charge-stable NV centers [Bluvstein, Zhang, and Bleszynski Jayich \(2019\)](#).

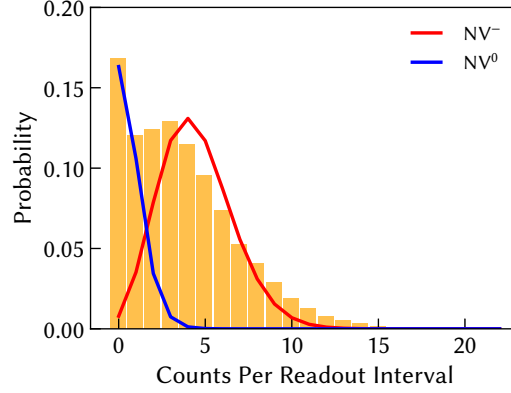


Figure 7.7. Photon number probability distribution, for which the separation between two successive initialization green pulses equals the readout interval (1 ms). The distribution can be described as a sum of two Poisson distributions, with an average count rate of 0.65 and 4.45 counts per readout for the NV^0 (blue curve) and NV^- (red curve) charge states respectively.

7.3.4 External time-varying magnetic field

For the proof-of-principle demonstration, we have applied an external time-varying magnetic field to the NV center. The external field is generated by sending a DC current through a 100 nm thick aluminum coplanar waveguide (CPW) fabricated next to the SIL (see [Figure 7.5a](#)), with the use of an arbitrary function generator (Tektronix AFG30523) or AFG. [Figure 7.8](#) shows an example of the voltage output from the AFG, which follows a simulated Ornstein-Uhlenbeck (OU) process, as will be described in more detail later.

To calibrate the shift in the frequency separation between the $m_s = 0$ and $m_s = +1$ states induced by the external field, we measure the CPT spectral response as a function of the AFG output voltage (see [Figure 7.9a](#)). [Figure 7.9b](#) plots the center frequency derived from the least-square Lorentzian fit to the CPT dip as a function of the corresponding output voltage. The least-square linear fit in [Figure 7.9b](#) yields a frequency-to-voltage slope of 0.66 MHz/V.

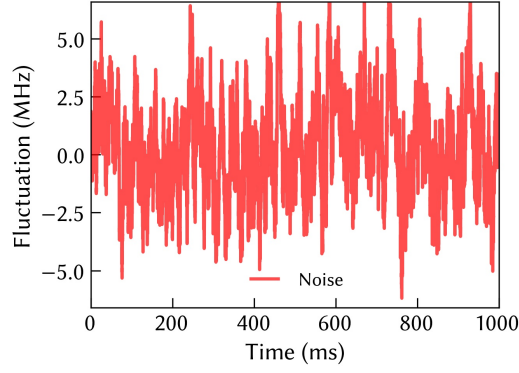


Figure 7.8. An example of an AFG output following a simulated OU-process with a correlation time, $\tau_c = 5\text{ms}$ and standard deviation, $\sigma/2\pi = 2.2\text{ MHz}$.

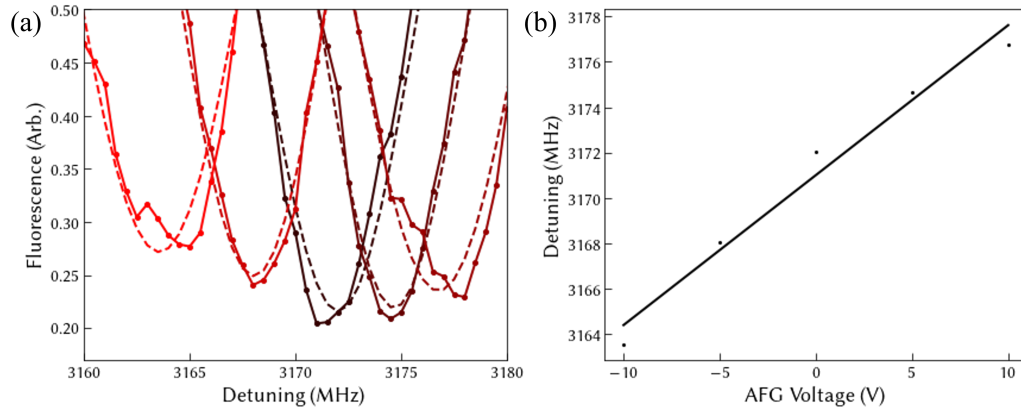


Figure 7.9. (a) Examples of CPT spectra taken at various fixed AFG voltages. The solid lines are least-squares fits to inverted Lorentzians. (b) The detuning at the CPT resonance vs the corresponding AFG voltage. The solid line is the least-square fit.

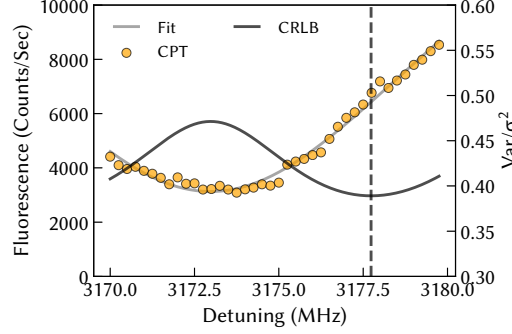


Figure 7.10. An example of a CPT spectral response and the least-square fit to an inverted Lorentzian used in the estimation. The vertical dashed line indicates the position of the fixed detuning, at which the real-time sensing is carried out. The corresponding photon count rate obtained from the CPT spectral response is also the average count rate used in the estimations carried out at this detuning.

7.3.5 Implementation of Estimation

For real-time estimations, we have implemented three different estimators using a FPGA (Xilinx 7k410t) included in the Keysight M3302A card. The FPGA takes as input the time series of detected photon counts, y_n , as well as other relevant parameters, such as the CPT spectral response and the bias or Raman detuning, and then outputs an estimation, $\tilde{x}_n$, of the frequency shift, x_n , induced by the external time-varying magnetic field. Prior to each estimation run, we experimentally measure the CPT spectral response and numerically fit the CPT dip to an inverted Lorentzian. An example is given in [Figure 7.10](#). The numerical fit of the CPT spectral response is then used in the subsequent estimation process.

Here we discuss specifically the implementation of the OU-Bayesian estimator. Similar approaches are also used in the implementation of average count as well as simple Bayesian estimators. For the OU-Bayesian estimator, the arrays, $p_{\bar{y}_n}(y_n|x_n)$ and $p_{OU}(x_n, t_0 + \tau|x_{n-1}, t_0)$, are evaluated for $y_n = 0, 1, 2, 3$ and $-3\sigma \leq x_n \leq 3\sigma$ with a step size of $\sigma/10$. We choose cutoff values of $y_n \leq 3$ and $|x_n| \leq 3\sigma$ since the probabilities for values beyond these ranges are negligible.

These two arrays and the statistical parameters for the OU process including correlation time, τ_c , and standard deviation, σ (in addition to the other parameters discussed earlier) are uploaded to the FPGA as constants to be used during the estimation.

The Keysight M3302A card contains a digitizer and arbitrary waveform generator (AWG) as well as a FPGA (see [Figure 7.11](#)). During the estimation process, photon counts in the form of TTL pulses received by the Keysight M3302A card are digitized by the built-in digitizer. Within the Keysight card, an AND logic block asserts high if the system is gated within the detection pulse shown in [Figure 7.6](#), allowing the signal to proceed to the photon accumulator. For every time interval, $\tau = 10\mu\text{s}$, the photon accumulator block sends the number of photon counts detected in the interval, y_n , to the FPGA. The FPGA then follows the process outlined, with the additional inputs as discussed above, to output an estimated x_n . A voltage proportional to the estimated x_n is then generated by the AWG. We monitor both the estimations and the AFG output voltages (which are proportional to the time-varying external fields) on an oscilloscope (see [Figure 7.11](#)).

Note that the estimators are initially programmed in C++. For the implementation in a FPGA, the C++ programs are converted to the native FPGA language, Verilog, using Xilinx HLS. The processing time for the estimation process from receiving y_n to outputting an estimation is approximately $7\mu\text{s}$.

7.3.6 CPT Experiment

[Figure 7.12a](#) shows the Λ -type three-level system used for the CPT process in the real-time sensing experiment, as also discussed in the main text. It should be

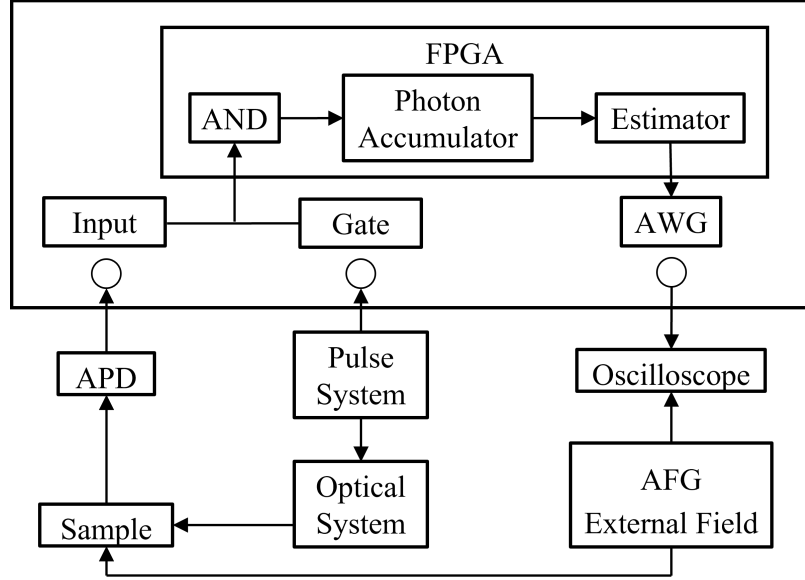


Figure 7.11. A block diagram of the functions of the Keysight card and its relation to the rest of the experimental setup.

noted that the $|+1\rangle$ to $|E_y\rangle$ transition, which is otherwise dipole-forbidden, becomes dipole-allowed due to strain induced state mixing.

To obtain the CPT spectral response, we sweep the frequency of a sideband generated by the EOM in Figure 7.5a across the resonance of the $|0\rangle$ to $|E_y\rangle$ transition, while fixing the frequency of the carrier laser at the resonance of the $|+1\rangle$ to $|E_y\rangle$ transition. Figure 7.12b shows a full CPT spectral response at the resonance of the $|0\rangle$ to $|E_y\rangle$ transition. CPT and the corresponding quenching of the fluorescence occur when the detuning between two laser fields approaches the frequency separation between the two lower states. A close-up of a CPT dip obtained under experimental conditions used for real-time sensing is displayed in Figure 7.12c.

A Lorentzian least-square fit to the CPT dip in Figure 7.12c yields a CPT linewidth approximately 11.6 MHz, which as discussed in the main text limits the sensitivity of the real-time sensing process. Contributions to this linewidth include

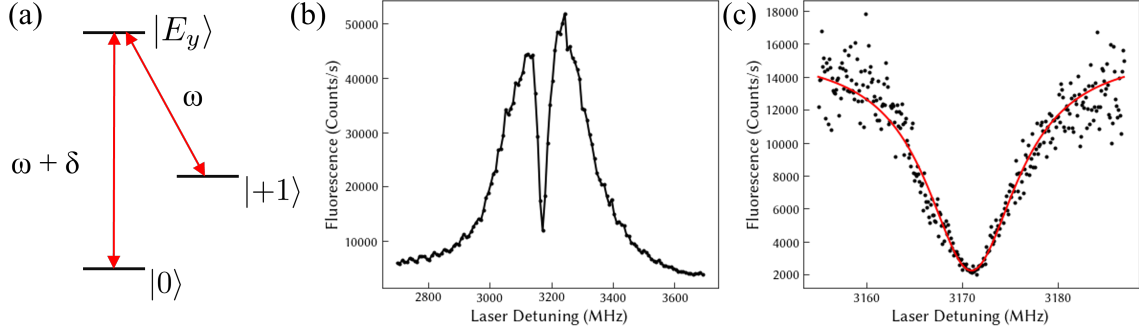


Figure 7.12. (a) The A -type three-level system used in the real-time sensing. (b) A full CPT spectral response occurring at the resonance of the $|0\rangle$ to $|E_y\rangle$ transition. (c) A closer look at the CPT dip obtained under experimental conditions used for real-time sensing. The red line is a least-square fit to an inverted Lorentzian.

the spin dephasing induced by the nuclear spin bath, the hyperfine splitting, and the power broadening. The hyperfine splitting is 2.2 MHz. The linewidth due to the spin dephasing is 0.6 MHz, as determined from a separate optically detected magnetic resonance (ODMR) experiment. To estimate the power broadening, we numerically fit the CPT dip with a sum of three inverted Lorentzians with the same linewidths and with 2.2 MHz hyperfine splitting between adjacent Lorentzians [Smeltzer, McIntyre, and Childress \(2009\)](#). The numerical fit yields a power broadening of 4.7 MHz. Using the standard expression on the power broadening contribution to the CPT resonance, we estimate the Rabi frequency for the optical fields to be of order 5 MHz.

7.3.7 Simulation

To compare the experiment to theoretical predictions we perform a simulation of our experimental procedure. This serves as a verification for our understanding of the system. To do this, we incorporate the ionization process into an adapted theory as a model for simulation. To account for charge initialization fidelity, we label each readout duration, which is $100\mu\text{s}$, as a zero counts readout

with 25% probability, or as a Poissonian readout with 75% probability. This is what we expect from 75% NV^- probability. We then simulate a photon counts series for each time interval $\tau = 10\mu\text{s}$, where counts are generated by the Poisson distribution or a zero count depending on the readout label. The Poisson distribution's average, y_n , along with the conditional probability arrays, $p_{OU}[x_n][x_{n-1}]$ and $p_{\bar{y}_n}[y_n][x_n]$, are obtained from the same measured CPT curve and generated OU signal as during the experiment. We load the arrays to a simulated FPGA in Python, which is then ready for estimation output. Finally, we perform the same data analysis as for the experiment results. As shown in [Figure 7.7](#) in the main text, we can see simulations match the experiment quite well except for minor differences which suggest the existence of extra noise. Also we notice that in [Figure 7.8a](#) in the main text, there are discrepancies between simulations and the experiment, this may be the result of bad fitting around wing of the curve due to the effects of the hyperfine interaction.

7.4 Conclusion

In summary, we have demonstrated real-time magnetometry using dark states in a NV center by estimating the time-varying magnetic field from the corresponding time series of photon counts in a CPT setting. A Bayesian estimator, which takes advantage of the statistical properties of the time-varying field, can effectively update the dynamical information of the field with the detection of a single photon, which is otherwise not feasible with the more conventional average-count estimator that takes no account of the statistical properties. While a NV center has been used as a model system for the proof-of-principle demonstration, the real-time magnetometry can also be extended to other solid-state spin systems [Awschalom et al. \(2018\)](#).

Real-time magnetometry using a single solid-state spin can add a new and powerful tool to quantum sensing. The magnetometry can be used for studies of time-varying magnetic fields in a variety of systems at the nanoscale, for example, nuclear spin baths [Togan et al. \(2011\)](#); [S. H. Wu et al. \(2021\)](#); [Zhao, Hu, Ho, Wan, and Liu \(2011\)](#) and two-dimensional semiconductors [Lovchinsky et al. \(2017\)](#); [Thiel et al. \(2019\)](#). Combining real-time sensing with feedback control also opens new avenues, such as protecting a spin qubit from the fluctuating magnetic environment via feedback control [Shulman et al. \(2014\)](#); [Togan et al. \(2011\)](#).

CHAPTER VIII

CONTINUOUS REAL-TIME MAGNETIC SENSING VIA COHERENT POPULATION TRAPPING AIDED BY FEEDBACK CONTROL

The results presented in this chapter were published in [Turner et al. \(2022b\)](#).

8.1 Introduction

As discussed in Chapter V, using feedback control to compensate environmental noise is an effective way to extend coherence time, provided that we have a real-time estimate of the noise from continuous sensing. For the continuous sensing, BPE can update estimations in real time with the detection of just a single photon [S. H. Wu et al. \(2021\)](#), as demonstrated in a recent magnetometry experiment with a NV center [Turner et al. \(2022a\)](#). An essential requirement for this demonstration is that the statistical properties of the magnetic field fluctuations are known and are taken advantage of in the BPE, which is difficult to meet for many intended applications.

In this chapter we show that this serious limitation can be overcome by combining the continuous BPE with a feedback loop [Stockton, Geremia, Doherty, and Mabuchi \(2004\)](#), followed by a separate verification and optimization process. For the feedback, we apply an additional magnetic field to the NV center to cancel the estimated magnetic field variation. We then verify possible feedback-induced suppression of spin dephasing by using Ramsey interferometry. A key aspect of our approach is to use the spin dephasing rate in the presence of the feedback as an effective cost function for the BPE. We can minimize the cost function and thereby optimize the BPE by systematically varying the statistical parameters used in the

BPE. This verification and optimization process provides us valuable information on the essential statistical properties of the underlying dynamics.

Furthermore, initial cycles of verification and optimization enable the later continuous BPE at the single-photon level regardless of the prior availability of the statistical parameters of the underlying dynamics, which adds a new tool to the quantum sensing toolkit and opens another frontier for exploring quantum dynamics, quantum fluctuations, and feedback control at the nanoscale [Shulman et al. \(2014\)](#); [Zhao et al. \(2011\)](#). This demonstration shows again that quantum control can not only extend the system's coherence time but also, in return, help improve quantum sensing.

8.2 Results

8.2.1 Continuous real-time sensing

We carry out the continuous real-time magnetic field sensing under the setting of coherent population trapping (CPT), for which the $m_s = 0$ and $m_s = +1$ ground spin states of a NV center, with a frequency separation ω_B , are coupled to the E_y excited state with two respective resonant laser fields (see [Figure 8.1](#)) [Golter et al. \(2016\)](#); [Santori et al. \(2006\)](#); [Yale et al. \(2013\)](#). CPT due to destructive quantum interference occurs when the optical coupling is Raman resonant, i.e., when $\delta - \omega_B = 0$, where δ is the detuning between the two laser fields. As illustrated in [Figure 8.1](#), when the average Raman detuning (i.e., the Raman bias), $\Delta = \delta - \langle \omega_B \rangle$, is near the half width of the CPT spectral response, single-photon emissions from the NV center are directly correlated with the magnetic field variations, provided that the variations in ω_B do not exceed the half width. Specifically, $\{\mathbf{y}_n\} = \{y_1, y_2, \dots, y_n, \dots\}$, where y_n is the number of photons detected during the n th time interval with equal duration τ , carries the information

on $\{\mathbf{x}_n\} = \{x_1, x_2, \dots, x_n, \dots\}$, the corresponding change in ω_B . Details of the experimental method will be presented in the next section.

Many dynamical processes of interest can be modeled as an Ornstein-Uhlenbeck (OU) process [de Lange et al. \(2010\)](#); [W. Yang, Ma, and Liu \(2017\)](#); [C. Zhang and Mølmer \(2020\)](#), which is a stationary Gauss-Markov process and features an autocorrelation function,

$$R(t) = \langle x(t_0)x(t_0 + t) \rangle = \sigma^2 e^{-|t|/\tau_c},$$

with σ^2 and τ_c being the variance and the correlation time, respectively. For a proof-of-principle experimental demonstration, we apply to the NV center an external magnetic field fluctuation that follows an OU process with $\langle x(t) \rangle = 0$. To add a feedback loop to the continuous sensing process, we apply to the NV center another magnetic field to effectively subtract the estimated magnetic field from the external fluctuations (see [Figure 8.1](#)).

8.2.2 Bayesian parameter estimations

We use Bayesian inference and the time series of the detected photon counts, $\{\mathbf{y}_n\}$, to generate a time series of estimated frequency changes, $\{\tilde{\mathbf{x}}_n\} = \{\tilde{x}_1, \tilde{x}_2, \dots, \tilde{x}_n, \dots\}$. For the Bayes update,

$$p(x_n|y_n, y_{n-1}, \dots, y_1) \propto p_{\bar{y}}(y_n|x_n) \times p'(x_n|y_{n-1}, \dots, y_1),$$

where $p'(x_n|y_{n-1}, \dots, y_1)$ is the prior probability distribution, $p(x_n|y_n, y_{n-1}, \dots, y_1)$ is the posterior probability distribution, and $p_{\bar{y}}(y_n|x_n)$ is the likelihood of detecting y_n photons in the n th time interval given x_n and follows a Poisson distribution,

$$p_{\bar{y}}(y_n|x_n) = \frac{\bar{y}^{y_n} e^{-\bar{y}}}{y_n!},$$

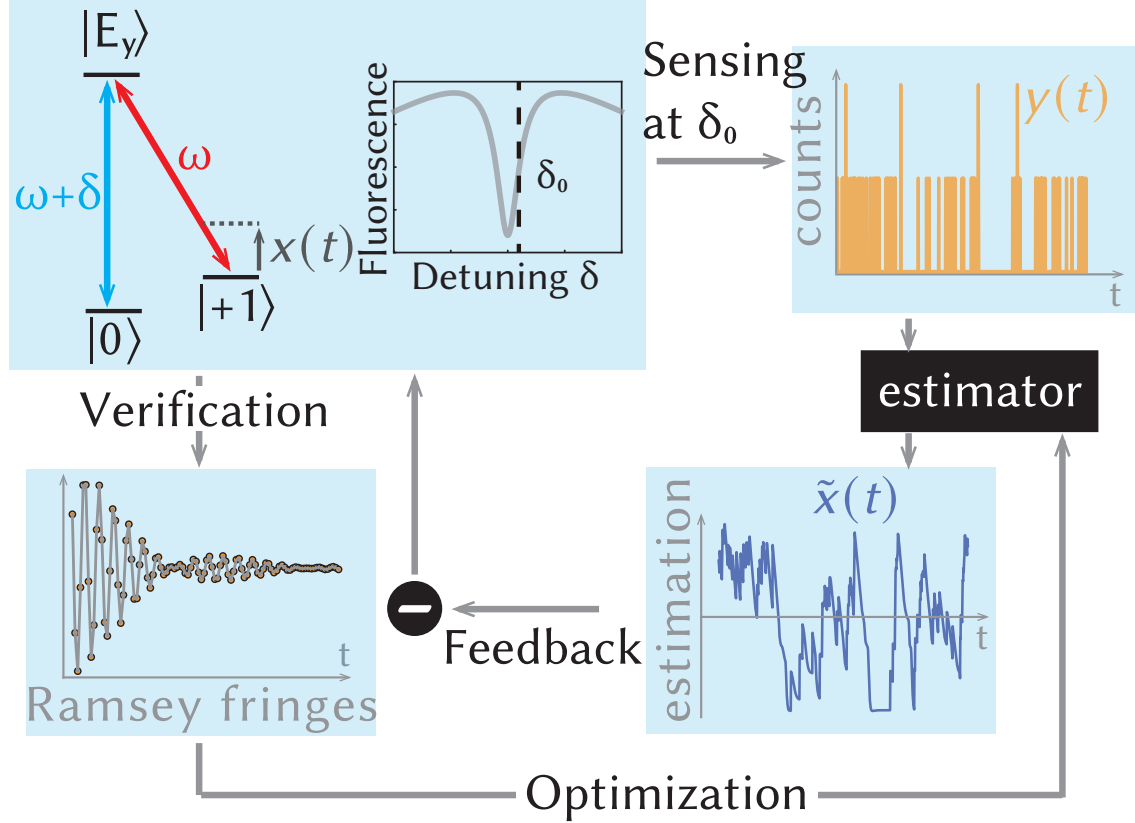


Figure 8.1. Continuous quantum sensing via CPT and with feedback control. The upper-left figure shows the Λ -type three-level system used for the CPT process. With the laser detuning fixed near the half width of the spectral CPT response, a Bayesian estimator can convert in real-time the time series of detected single-photon emissions from the NV center into changes in the frequency separation between the two ground spin states. For the feedback, an additional magnetic field is applied to the NV center to cancel the estimated change. The feedback-induced reduction in the NV spin dephasing rate is verified with Ramsey interferometry. The Bayesian estimator can be optimized by varying the statistical parameters used in the BPE and by minimizing the spin dephasing rate, which serves as the cost function for the BPE.

with $\bar{y}$ being the expected average photon count per updating time interval. The estimation is then given by

$$\tilde{x}(t) = \int p(x, t) x dx.$$

To achieve good time-resolution for the real-time sensing, we have $\bar{y}_n \ll 1$.

Under this condition, Bayesian estimations with a prior that is simply given by the previous estimate quickly converge to the average value of $x(t)$ and provide no information on the dynamics of $x(t)$, as shown in pervious studies [Turner et al. \(2022a\)](#); [S. H. Wu et al. \(2021\)](#).

The dynamical information can be obtained with an improved prior, which takes into consideration the statistical properties of $x(t)$, with

$$\begin{aligned} p'(x_n = x | y_{n-1}, \dots, y_1) &= \int d\omega \cdot p(x_{n-1} = x - \omega | y_{n-1}, \dots, y_1) \\ &\quad \times p_{\text{OU}}(x_n = x_{n-1} + \omega, t + \tau | x_{n-1}, t), \end{aligned}$$

where p_{OU} is the probability of finding x_n at $t + \tau$ given x_{n-1} at t for the OU process,

$$p_{\text{OU}}(x_n, t + \tau | x_{n-1}, t) = \mathcal{N}(x_{n-1}e^{-\tau/\tau_c}, \sigma^2(1 - e^{-2\tau/\tau_c})),$$

with $\mathcal{N}$ denoting a normal distribution [S. H. Wu et al. \(2021\)](#). We refer this Bayesian estimator as the OU-Bayesian estimator. Here the use of the improved prior requires that the statistical parameters of the OU process is known before the sensing process, a requirement that is difficult to meet for many intended applications, as discussed earlier.

[Figure 8.2a](#) shows, as an example, estimations, i.e., $\tilde{x}_n$, obtained with the OU-Bayesian estimator without the feedback as well as the actual fluctuations, x_n . [Figure 8.2b](#) compares the estimations obtained in a relative short time span with the corresponding time series of the detected photon counts. Since $\bar{y}_n \ll 1$,

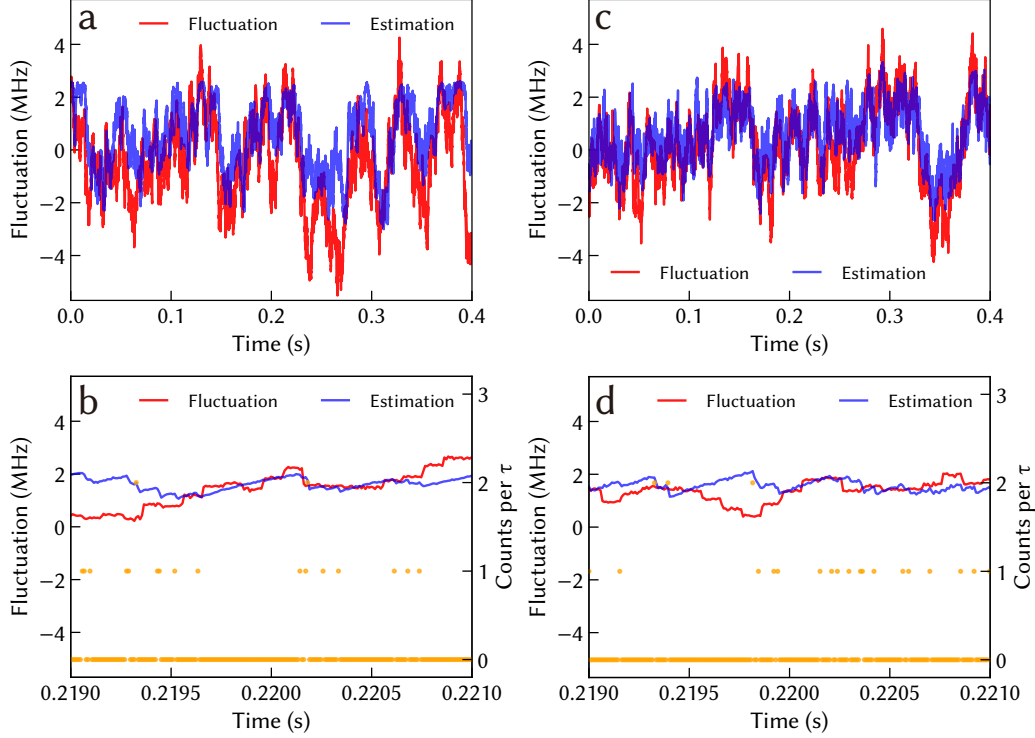


Figure 8.2. Continuous real-time estimations of $x(t)$. a,b, Estimations obtained without feedback. c,d, Estimations obtained with feedback. The red curves are the actual OU-type fluctuations and the blue curves are the corresponding estimations. The orange dots in c and d show the photons counts per updating time interval, τ , which is set to $10 \mu\text{s}$. The correlation between the saw-tooth behavior in the estimations and the detection of a single photon demonstrates the updating of dynamical information with the detection of a single photon. The statistical parameters used for the external OU fluctuations are $\tau_c = 10 \text{ ms}$ and $\sigma/2\pi = 1.9 \text{ MHz}$.

the estimator waits for the arrival of a detected photon to output an estimation, leading to the saw-tooth behavior shown in [Figure 8.2b](#). The correlations between the saw tooth and the photon counts in [Figure 8.2b](#) demonstrate that the continuous real-time estimation effectively updates the dynamical information with the detection of just a single photon [Turner et al. \(2022a\)](#).

[Figure 8.2c](#) shows estimations obtained under the same conditions as those for [Figure 8.2a](#) except that now the estimations are carried out with the feedback loop, which subtracts estimated change, $\tilde{x}_{n-1}$, from x_n such that the actual net change in ω_B is $x_n - \tilde{x}_{n-1}$. In this case, the OU-Bayesian estimations take place with a shifted Raman bias, $\delta - (\langle\omega_B\rangle - \tilde{x}_{n-1})$. In this regard, [Figure 8.2c](#) and [d](#) display the estimations for x_n , not $x_n - \tilde{x}_{n-1}$. Similar to [Figure 8.2b](#), [d](#) shows that the continuous estimation with the feedback control updates the dynamical information with the detection of a single photon.

8.2.3 Estimation verification and parameter optimization

The external magnetic field fluctuations applied to the NV center lead to extra spin dephasing. With the feedback loop, a reduction in the overall magnetic field fluctuations suppresses the extra spin dephasing. We have employed Ramsey interferometry to measure the spin dephasing and have used the suppression of the extra spin dephasing as a verification that the estimation approaches the actual change. [Figure 8.3a](#) shows the pulse sequence used for the feedback and verification.

[Figure 8.3b](#) compares the results of the Ramsey interferometry obtained with and without the feedback loop. We numerically fit the Ramsey fringes with the sum of three sine functions, corresponding to each of the ^{14}N hyperfine spin projections (with $m_I = 0, +1, -1$), multiplied by a Gaussian envelope,

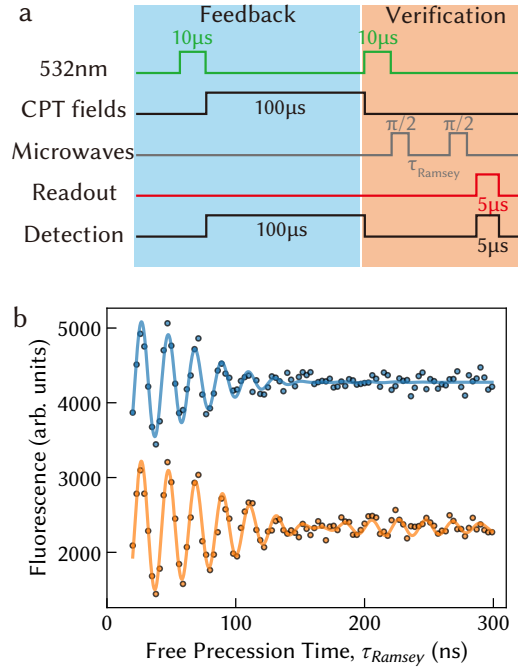


Figure 8.3. Verification using Ramsey interferometry. a, The pulse sequence used for feedback control and Ramsey interferometry, for which the spin readout takes place via the $m_s = 0$ to E_y transition. b, Results of Ramsey interferometry obtained with (bottom curve) and without feedback control (top curve). The statistical parameters used for the external OU fluctuations are $\tau_c = 15$ ms and $\sigma/2\pi = 2.2$ MHz. The feedback leads to an effective reduction in the overall spin dephasing rate. The solid curves show the least-square numerical fit discussed in the text.

$\exp[-(t/T_2^*)^2]$. A spin dephasing time, T_2^* , of 182 ns and 90 ns is derived from the least-square fit of the experimental results with and without the feedback loop, respectively.

The feedback and verification discussed above give us an effective mechanism to obtain information on the statistical parameters of the underlying dynamical process. In this case, we can repeat the feedback and verification cycles shown in [Figure 8.3a](#), while systematically varying the two statistical parameters, σ and τ_c , used for the OU process. [Figure 8.4a](#) plots the heat map of T_2^* obtained from the Ramsey interferometry as a function of σ and τ_c used for the BPE. In this case, $1/T_2^*$ effectively serves as a cost function for the BPE, i.e., the smaller $1/T_2^*$ is, the smaller the estimation error becomes. The maximum improvement of T_2^* by feedback control is plotted in [Figure 8.4b](#) as a function of the memory time of the external magnetic fluctuations.

8.2.4 Discussions

As shown in [Figure 8.4a](#), the maximum T_2^* occurs when τ_c used for the BPE is the same as the corresponding parameter for the actual fluctuations. The repeated cycles of feedback and verification thus correctly optimized parameter τ_c . In comparison, there is a slight deviation between the optimized σ and the corresponding expected value, as can be seen from the heatmap in [Figure 8.4a](#). This deviation is in large part due to experimental uncertainties in the estimation process. In addition, the optimal sensing, i.e., the Cramér-Rao lower bound, for the estimation process can be achieved only if the relevant CPT spectral response is either linear or quadratic, which is not the case for the experiment.

The improvement of T_2^* by feedback control shown in [Figure 8.4b](#) is relatively modest. This is in part because the estimation process is relatively slow.

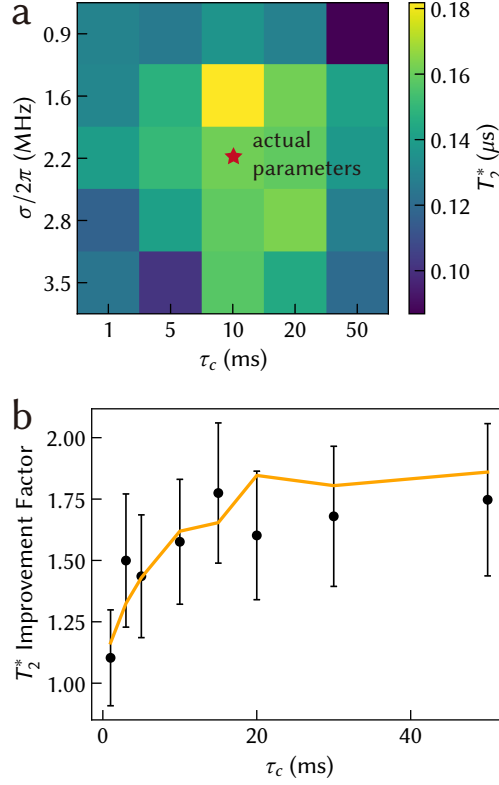


Figure 8.4. Optimization of the statistical parameters used for BPE. a, Spin dephasing time, T_2^* , derived from Ramsey interferometry with feedback control as a function of the memory time and the standard deviation used in the BPE. The actual statistical parameters are $\tau_c = 10$ ms and $\sigma/2\pi = 2.2$ MHz. The maximum T_2^* obtained under feedback loop occurs at $\tau_c = 10$ ms and $\sigma/2\pi = 1.6$ MHz. b, Maximum improvement in T_2^* due to feedback control observed as a function of the memory time of the external magnetic field fluctuations. The error bars in b are derived from the least square numerical fit. The solid orange line is the improvement in T_2^* obtained in a numerical simulation based on the experimental parameters.

Even with updating at the single-photon level, the average time between updates is still about $77 \mu\text{s}$, the inverse of the photon count rate. Both the estimations and the feedback control are thus more effective with relatively slow fluctuations. [Figure 8.4b](#) shows that the improvement observed increases with increasing τ_c for the OU fluctuations, though the improvement saturates when τ_c exceeds 10 ms. At the relatively long τ_c , the improvement becomes limited by the accuracy of the estimations. Overall, the experimentally observed improvement is in good agreement with that obtained from the corresponding numerical simulation, as shown in [Figure 8.4b](#). The improvement will ultimately be limited by the Cramér-Rao lower bound of the estimation process. It should be noted that the modest improvement in T_2^* does not affect the optimization of the statistical parameters used for the BPE, though greater improvement in T_2^* can lead to more robust optimization.

8.3 Methods

8.3.1 Diamond NV sample and setup

An electronic-grade chemical-vapor-deposition grown diamond sample (from Element Six, Inc.) was cooled down to 10 K in an optical cryostat from Montana Instruments, Inc. Optical excitation and collection were carried out with a confocal optical microscope and with a solid immersion lens (SIL) milled into the surface of the diamond. A permanent magnet placed outside of the cryostat was used to split the $m_s = \pm 1$ states by 430 MHz. For the two optical fields used in CPT, a tunable 637-nm diode laser was tuned to the $m_s = +1$ to E_y transition while a sideband generated by an electro-optical modulator (EOM) was tuned to the $m_s = 0$ to E_y transition. The CPT spectral response used for the continuous sensing has a linewidth of 17.9 MHz, including contributions from the hyperfine splitting (2.2

MHz), spin dephasing (0.62 MHz), and power broadening with an estimated Rabi frequency of 10.6 MHz. See the supplementary materials in an earlier study for a similar CPT spectral response as well as a schematic of the experimental setup [Turner et al. \(2022a\)](#).

For the proof-of-principle experiment, an external time-varying magnetic field was applied to the NV center through a coplanar waveguide (CPW) fabricated next to the SIL. The current through the CPW, which follows a simulated OU process, was generated by an arbitrary function generator (AFG). Microwave (MW) pulses used for Ramsey interferometry were also applied via the CPW, with Rabi frequency, $\Omega_{MW}/2\pi = 20$ MHz, and were detuned from the $m_s = 0$ to $m_s = +1$ transition by $\Omega_{MW}/2\pi = 30$ MHz. For feedback control, an additional magnetic field needs to be applied to the NV center to cancel the estimated change in the magnetic field from the OU external magnetic field. Experimentally, this was implemented through the subtraction of the corresponding voltage in the output of the AFG. For BPE, this additional magnetic field can be treated as an effective shift in $\langle\omega_B\rangle$.

8.3.2 Continuous Bayesian parameter estimations

The experiment began with the initialization of the NV to the $m_s = 0$ state using a 10 μs green laser pulse ($\lambda = 532\text{nm}$), followed by two long (100 μs) optical pulses that induce CPT. The fluorescence was continuously collected during the CPT process. BPE were carried out with a field-programmable gate-array (FPGA) in a Keysight M3302A card containing a digitizer and an arbitrary waveform generator (AWG), which accumulates the number of photon counts per update time interval and outputs a voltage corresponding to the estimated frequency fluctuations. See the supplementary materials in an earlier study for a

detailed discussion on the implementation of continuous BPE [Turner et al. \(2022a\)](#). Unless otherwise specified, the Raman bias used is $(\delta_0 - \langle \omega_B \rangle)/2\pi = 3$ MHz and the average photon count rate is near 13000/s.

8.3.3 Verification and optimization

After each 100 μs sensing interval, verification of the feedback process was carried out by Ramsey interferometry. As shown in [Figure 8.3a](#), the pulse sequence for each Ramsey measurement started with a 10 μs green initialization pulse, followed by a MW $\pi/2$ -pulse, which places the NV center in a superposition between the $m_s = 0$ and $m_s = +1$ spin states. The NV center was then allowed to freely precess for a variable duration τ_{Ramsey} . After applying a second MW $\pi/2$ -pulse, the electron population in the $m_s = 0$ spin state was readout through the $m_s = 0$ to E_y transition. For the numerical simulation of the feedback-induced improvement in T_2^* , the experimental parameters, including the experimentally obtained average CPT spectral response, were used. The simulation of the BPE followed the same approach as that in the earlier study [S. H. Wu et al. \(2021\)](#). The finite dynamical range of the FPGA was also accounted for in the simulation.

8.4 Conclusion

The continuous PBE we used assume that the dynamical process involved is an OU process, which is applicable to a wide variety of dynamical phenomena such as nuclear spin fluctuations [de Lange et al. \(2010\)](#); [W. Yang et al. \(2017\)](#). As pointed out in Chapter VI, according to the Bernstein–Widder theorem, OU processes form a basis for expanding any noise with an autocorrelation function $C(t)$ that is completely monotone for $t > 0$. In this regard, the feedback-aided approach introduced in this chapter can be viewed as a spectroscopy of noise in the

OU basis, in contrast to standard dynamical-decoupling spectroscopy, which works in the Fourier basis.

PBE can also be extended to other types of dynamical processes, such as random telegraph noise, which is non-Markovian and non-Gaussian [Song, Chantasri, Tonekaboni, and Wiseman \(2022\)](#). In cases where the statistical model is unknown, machine learning techniques such as neural networks can be used for the estimations. In all these cases, the spin dephasing rate measured in the presence of feedback control can still be used as an effective cost function for the verification and optimization of the relevant estimation process.

CHAPTER IX

SPIN STUDIES OF SiV CENTERS

The results presented in this chapter were published in [S. Wu et al. \(2025\)](#); [S.-H. Wu et al. \(2025\)](#).

9.1 Introduction

In the previous chapters, we developed theory and experiments for CPT-based sensing with NV centers in diamond. However, because the NV center lacks inversion symmetry, its permanent electric dipole makes it sensitive to surface-charge fluctuations, which Stark-shift the optical transitions and cause unstable, broadened lines. This problem is especially severe in nanostructures, limiting sensing sensitivity and hindering cavity-QED studies that would otherwise enhance light–spin interactions for quantum-network building blocks.

Among color centers, the SiV center has inversion symmetry and thereby mitigates these issues. Experiments show that even in nanostructures, down to tens of nanometers the SiV optical linewidth remains within a few times the lifetime limit, making it a strong candidate for cavity QED and nanostructure-embedded studies. To extend CPT-based sensing to SiV centers, we need a clear understanding of their spin states. Because SiV ground-state spins are degenerate in the absence of spin–spin interaction, well-controlled spin transitions require magnetic-field (Zeeman) control with a reliable model. Moreover, CPT and the associated spin dynamics must be characterized specifically for SiV centers. In this chapter, we address these questions through SiV experiments under applied magnetic fields at millikelvin temperatures.

9.2 Experimental determination of energy level structures of diamond silicon vacancy centers in off axis magnetic fields

9.2.1 Introduction

Negatively charged silicon vacancy (SiV) centers in diamond have recently emerged as a promising qubit platform for optical quantum networks and quantum spin mechanics because of their superior optical properties, long spin coherence time at low temperatures, and strain coupling to the orbital degrees of freedom in the ground states [Bradac, Gao, Forneris, Trusheim, and Aharonovich \(2019\)](#); [Meesala et al. \(2018\)](#); [Neu et al. \(2013\)](#); [Rogers, Jahnke, Teraji, et al. \(2014\)](#); [Sipahigil et al. \(2014\)](#); [Sukachev et al. \(2017\)](#). The zero-phonon line of SiV centers contains about 70% of the total fluorescence [Dietrich et al. \(2014\)](#), with nearly lifetime-limited optical linewidth and with the transition robust against surface charge fluctuations. Excellent optical coherence has been observed for SiV centers in diamond nanostructures and in diamond membranes as thin as 100 nm [Evans, Sipahigil, Sukachev, Zibrov, and Lukin \(2016\)](#); [Jantzen et al. \(2016\)](#); [Lekavicius and Wang \(2019\)](#); [Sipahigil et al. \(2016\)](#); [J. L. Zhang et al. \(2017\)](#). Decoherence time exceeding 10 ms, limited by the nuclear spin bath, has been observed at temperatures near 100 mK [Sukachev et al. \(2017\)](#). SiV centers have been incorporated into photonic crystal structures as well as whispering gallery mode (WGM) optical resonators for cavity QED studies and for the development of quantum networks [Chakravarthi et al. \(2023\)](#); [Evans et al. \(2018\)](#); [J. C. Lee, Aharonovich, Magyar, Rol, and Hu \(2012\)](#); [Pauls, Lekavicius, and Wang \(2020\)](#); [Sipahigil et al. \(2016\)](#); [J. L. Zhang et al. \(2018\)](#). In addition, the special energy level structure of SiV centers can enable relatively strong ground-state spin-mechanical interactions [Meesala et al. \(2018\)](#); [Sohn et al. \(2018\)](#), providing a

promising platform for quantum spin-mechanics and the development of mechanical quantum networks [Arrazola, Minoguchi, Lemonde, Sipahigil, and Rabl \(2024\)](#); [Lemonde et al. \(2018\)](#); [Wang and Lekavicius \(2020\)](#). Recent advances in this area include the realization of GHz diamond spin-mechanical resonators with mechanical Q-factors exceeding 106 and with efficient orbital coupling to SiV centers [Joe et al. \(2024\)](#); [X. Z. Li, Lekavicius, Noeckel, and Wang \(2024\)](#).

Microwave and acoustic quantum controls of the spin states of individual SiV centers have been well established [X. Z. Li et al. \(2024\)](#); [Pingault et al. \(2017\)](#). All optical control of SiV spin states have also been demonstrated with coherent population trapping or coherent Raman transitions [Becker et al. \(2018\)](#); [Pingault et al. \(2014\)](#); [Rogers, Jahnke, Metsch, et al. \(2014\)](#); [S. Wu et al. \(2025\)](#). In spite of these experimental advances, a persistent experimental issue is the large and seemingly random variations in the microwave or acoustic transition frequencies or simply magnetic-field induced splitting of the SiV ground spin states, as evidenced in the earlier studies. This is in contrast to diamond nitrogen vacancy (NV) centers, for which the energy levels of the ground states are well characterized and controlled [Doherty et al. \(2013\)](#).

A SiV center is an interstitial point defect consisting of a silicon atom positioned midway between two adjacent vacancies in the diamond lattice. The energy level structure of a SiV depends on the local strain environment, including static strain and the dynamic Jahn-Teller (JT) coupling [Hepp et al. \(2014\)](#). Since the ground states of a SiV center are doubly degenerate, an external magnetic field is needed to induce an energy separation between two relevant spin states. For acoustic or optical spin control, a mixing of the two spin states is also necessary, which can be induced by an off-axis magnetic field, i.e. a field away from the SiV

axis. The ground state energy level structure of a SiV center can thus depend sensitively on the interplay between static strain, dynamic JT effects, and external magnetic fields. An experimental approach for accurate determination of the energy level structure of individual SiV centers in a magnetic field along an arbitrary direction is important for studies such as spin-mechanics, for which matching a spin transition frequency with a mechanical resonance might be needed, and for developing experimental approaches to mitigating the variations in the ground-state energy level separations.

The theoretical model for the SiV energy level structure has been well established [Hepp et al. \(2014\)](#); [Rogers, Jahnke, Doherty, et al. \(2014\)](#). In addition to parameters describing effects of strain and dynamic JT coupling, the model also includes quenching factors that account for the quenching of the orbital gyromagnetic factor due to the dynamic JT interaction. With a large set of experimental data on the dependence of SiV optical transition frequencies on a magnetic field along the (001) direction, an earlier study extracted the relevant parameters by comparing the experimental results with the numerical calculations of the SiV energy level structure and by assuming equal orbital quenching factors for the ground and excited states [Hepp et al. \(2014\)](#). Using this approach to determine SiV energy level structures in a magnetic field along an arbitrary direction, however, is not resource-efficient because minimizing errors in the fitting parameters for a given field direction does not necessarily minimize the errors for other directions. Furthermore, accurate determination of the energy level structure in this case may require unequal orbital quenching factors for the ground and excited states, which is difficult to obtain with the above approach because of the

relatively small contribution from the orbital Zeeman effects as well as the multiple parameters involved in the numerical fitting.

In this chapter, we report the development of an experimental approach that can efficiently determine the energy level structure of individual SiV centers in a magnetic field along an arbitrary direction and extract the key parameters, including unequal orbital quenching factors. Our approach is based on the use of two strain/JT coupling rates (one each for the ground and the excited states) that characterize the combined effects of static strain and dynamic JT coupling and on the fact that orbital Zeeman effects vanish when the field is normal to the SiV axis. We derive the strain/JT coupling rates from the SiV energy level structure in a transverse magnetic field, specifically from an analytical expression for the energy levels, with no complication from orbital magnetic coupling and no numerical fitting. A detailed comparison between theoretical calculations using the obtained strain/JT coupling rates and additional experimental studies on the dependence of the frequencies of the spin-conserved optical transitions on both the amplitude and direction of the magnetic field then extracts unequal orbital quenching factors for the ground and excited states.

9.2.2 Theoretical model

For a negatively charged SiV center, both the ground and excited states are characterized by orbital states, $|e_x\rangle$ and $|e_y\rangle$, and spin states, $|\uparrow\rangle$ and $|\downarrow\rangle$, with the spin-orbit (SO) coupling described by ($\hbar = 1$), $H_{so} = -\lambda_{so}L_zS_z$, where x, y, z are the internal basis of the SiV, with the z-axis along the SiV axis, and so is the spin-orbit coupling rate [Hepp et al. \(2014\)](#). In the Cartesian coordinates, X, Y, Z, of the cubic unit cell, we can define the unit vectors of the SiV basis as $\hat{x} = [1, 1, -2]/\sqrt{6}$, $\hat{y} = [1, -1, 0]/\sqrt{2}$, and $\hat{z} = [1, 1, 1]/\sqrt{3}$. In the absence of external magnetic fields

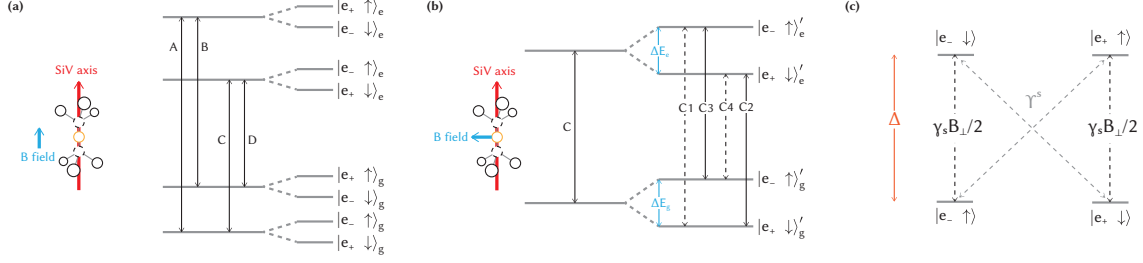


Figure 9.1. (a) SiV energy level structure and optical transitions in the absence of strain and JT coupling. A magnetic field along the SiV axis removes the spin degeneracy, with the optical transitions occurring between the same spin states. (b) Schematic of the energy levels and selection rules for the optical C-transition, including spin-conserved transitions (solid lines) and spin-flip transitions (dashed lines), under a magnetic field normal to the SiV axis. (c) Schematic illustrating that transverse magnetic field induces spin-state mixing (dashed black lines) and strain/JT coupling induces orbital-state mixing (dashed grey lines). Orbital Zeeman effects are not included.

and static strain and in the limit that the dynamic JT effect can be ignored, both ground and excited states of a SiV are characterized by doubly degenerate doublets, with the two upper states being $|e_+, \uparrow\rangle$ and $|e_-, \downarrow\rangle$ and the two lower states being $|e_-, \uparrow\rangle$ and $|e_+, \downarrow\rangle$, where $|e_{\pm}\rangle = (|e_x\rangle \pm i|e_y\rangle)/\sqrt{2}$, as illustrated in Figure 9.1a. The splitting between the upper and lower states in the doublet is λ_{so} , with λ_{so}^g and λ_{so}^e for the ground and excited states, respectively. Optical transitions taking place between the ground- and the excited-state doublets are labeled as A, B, C, D transitions in Figure 9.1a.

The interaction Hamiltonian due to an external magnetic field is given by

$$H_B = f\gamma_L B_z L_z + \gamma_S \mathbf{B} \cdot \mathbf{S}, \quad (9.1)$$

where γ_L and γ_s are the orbital and spin gyromagnetic ratio, respectively, and f is the orbital quenching factor. Because of the small orbital quenching factor (~ 0.1) Hepp et al. (2014), effects of the magnetic field on the orbital states are often ignored. A magnetic field along the z axis removes the spin degeneracy of

all the relevant states, with the optical transitions taking place between states with the same electron spin, as illustrated in [Figure 9.1a](#). In comparison, a transverse magnetic field (i.e., normal to the z-axis) can induce considerable spin-state mixing and thus enable spin-flip transitions (see [Figure 9.1b](#)), as will be discussed in more detail later.

Strain couples only to the orbital states. With symmetry-adjusted linear combination of strain tensor components, α , β , and γ , the strain Hamiltonian is given by (in the $|e_x\rangle$ and $|e_y\rangle$ basis) [Meesala et al. \(2018\)](#)

$$H_{Strain} = \begin{pmatrix} \alpha - \beta & \gamma \\ \gamma & \alpha + \beta \end{pmatrix} \otimes \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \quad (9.2)$$

where α induces an overall energy shift. The JT interaction Hamiltonian is given by in the same basis as

$$H_{JT} = \begin{pmatrix} \Upsilon_x & \Upsilon_y \\ \Upsilon_y & -\Upsilon_x \end{pmatrix} \otimes \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix},$$

where Υ_x and Υ_y are the JT coupling rates along x and y, respectively.

Since strain and dynamic JT coupling have the same symmetry property, it is difficult to experimentally distinguish between these two effects. We thus use a new set of parameters to describe the combined effects of strain and dynamic JT coupling, with $\Upsilon_x^s = -\beta + \Upsilon_x$ and $\Upsilon_y^s = \gamma + \Upsilon_y$. In the basis of the SO eigen states

$\{|e_+, \uparrow\rangle, |e_-, \downarrow\rangle, |e_-, \uparrow\rangle, |e_+, \downarrow\rangle\}$, the total Hamiltonian with $f = 0$ is then given by

$$\begin{aligned}
H_{Total} = & \frac{\lambda_{so}}{2} \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} + \frac{\gamma_s}{2} \begin{pmatrix} B_z & 0 & 0 & B_- \\ 0 & -B_z B_+ & 0 & 0 \\ 0 & B_- & B_z & 0 \\ B_+ & 0 & 0 & -B_z \end{pmatrix} \\
& + \begin{pmatrix} \alpha & 0 & \Upsilon_-^s & 0 \\ 0 & \alpha & 0 & \Upsilon_+^s \\ \Upsilon_+^s & 0 & \alpha & 0 \\ 0 & \Upsilon_-^s & 0 & \alpha \end{pmatrix}, \tag{9.3}
\end{aligned}$$

where $B_{\pm} = (B_x \pm iB_y)$ and $\Upsilon_{\pm}^s = (\Upsilon_x^s \pm \Upsilon_y^s)$. As shown by the Hamiltonian, B_x and B_y mix spin states with the same orbital component, with the mixing ratio scaling with $\gamma_s B_{\perp} / \lambda_{so}$, while Υ_x^s and Υ_y^s mix orbital states with the same spin component, as illustrated in [Figure 9.1c](#).

For a transverse magnetic field, i.e., for $B_z = 0$, we can analytically diagonalize the above Hamiltonian. The eigen energy of the two lower and upper states are respectively given by

$$\begin{aligned}
E_{\pm}^L &= \alpha - \sqrt{(\lambda_{so}/2)^2 + (\gamma_s B_{\perp}/2 \pm \Upsilon^s)^2}, \\
E_{\pm}^U &= \alpha + \sqrt{(\lambda_{so}/2)^2 + (\gamma_s B_{\perp}/2 \pm \Upsilon^s)^2}, \tag{9.4}
\end{aligned}$$

where $B_{\perp} = \sqrt{B_x^2 + B_y^2}$ and $\Upsilon^s = \sqrt{(\Upsilon_x^s)^2 + (\Upsilon_y^s)^2}$, which can be viewed as the effective strain/JT coupling rate. The solution given in [Equation 9.4](#) is valid even if $f = 0$, since the orbital Zeeman effect vanishes when $B_z = 0$. Note that because of the symmetry of the SiV center, the energy level structure depends on Υ^s , instead of Υ_x^s and Υ_y^s . This is also true for a magnetic field in an arbitrary direction.

For the two lower (or upper) eigen states, effects of the transverse magnetic coupling and the strain/JT coupling cancel each other for one state and add together for the other state, as shown by [Equation 9.4](#). This asymmetry lifts the degeneracy of the two states, with the splitting (for both the lower and upper states) given by

$$\Delta E \approx 2\gamma_s B_{\perp} \cdot \Upsilon^s / \lambda_{so},$$

where we assume $\lambda_{so} \gg (\gamma_s B_{\perp}, \Upsilon^s)$. This splitting provides a direct measurement of the effective strain/JT coupling rate, Υ^s . In the limit that the magnetic field is zero, the doublet remains doubly degenerate. In this case, the strain/JT coupling modifies the splitting between the upper and lower states in the doublet according to [Equation 9.4](#). Note that the above theoretical analysis applies to both the ground and excited states of the SiV center.

In the presence of a transverse magnetic field, s_z is no longer a good quantum number. Spin-state mixing induced by the transverse magnetic field enables spin-flip transitions. [Figure 9.1b](#) illustrates the energy level structure and optical selection rules for the C transition. With $\gamma_s B_{\perp} / \lambda_{so} \ll 1$, spin-flip transitions (dashed lines in [Figure 9.1b](#)) are expected to be much weaker than the spin-conserved transitions (solid lines in [Figure 9.1b](#)). As shown in [Figure 9.1b](#), we append a “prime” to the notations for the SO eigen states to denote the new eigen states, which include effects of the spin-state mixing as well as distortions of the orbital states induced by strain/JT coupling. The magnetic field induced splitting for the ground and excited states are

$$\Delta E_g \approx 2\gamma_s B_{\perp} \cdot \Upsilon_g^s / \lambda_{so}^g, \quad (9.5)$$

$$\Delta E_e \approx 2\gamma_s B_{\perp} \cdot \Upsilon_e^s / \lambda_{so}^e, \quad (9.6)$$

respectively, with Υ_g^s and Υ_e^s being the corresponding effective strain/JT coupling rates. The above theoretical analysis suggests a straightforward experimental approach to determining Υ_g^s and Υ_e^s , two key parameters that describe the energy level structure of a SiV. Specifically, ΔE_g and Υ_g^s thus can be determined directly in an experiment. The splitting between the two spin-conserved transitions, given by $\delta E = \Delta E_g - \Delta E_e$, can then be used to derive ΔE_e and thus Υ_e^s .

9.2.3 Experimental results

We have used coherent population trapping (CPT) of the two lower ground states and the frequency splitting between the two spin-conserved C-transitions (i.e., C_2 and C_3 in [Figure 9.1b](#)) in a photoluminescence excitation (PLE) spectrum, when the magnetic field is normal to the SiV axis, to determine the strain/JT coupling rates for the ground and the excited states. We have also carried out detailed studies on dependences of the spin-conserved splitting on the magnetic field amplitude along the Z-axis and on the orientation of the magnetic field in the X-Y plane as well as the z-Z plane. By comparing these experimental results with the theoretical model that uses the obtained strain/JT coupling rates, we then identify contributions from the orbital Zeeman effects, especially effects of unequal quenching factors of the ground and excited states.

SiV centers used in our experiments were implanted approximately 75 nm below the surface of an electronic grade diamond grown by chemical vapor deposition (CVD). The average kinetic energy and dosage of the ^{28}Si used in the implantation are 100 keV and $3 \times 10^9/\text{cm}^2$, respectively. Stepwise thermal annealing up to a temperature of 1200 °C followed by wet chemical oxidation was used for the removal of the damaged surface layer as well as for the formation of SiV centers. The sample was mounted on the mixing chamber of a dilution

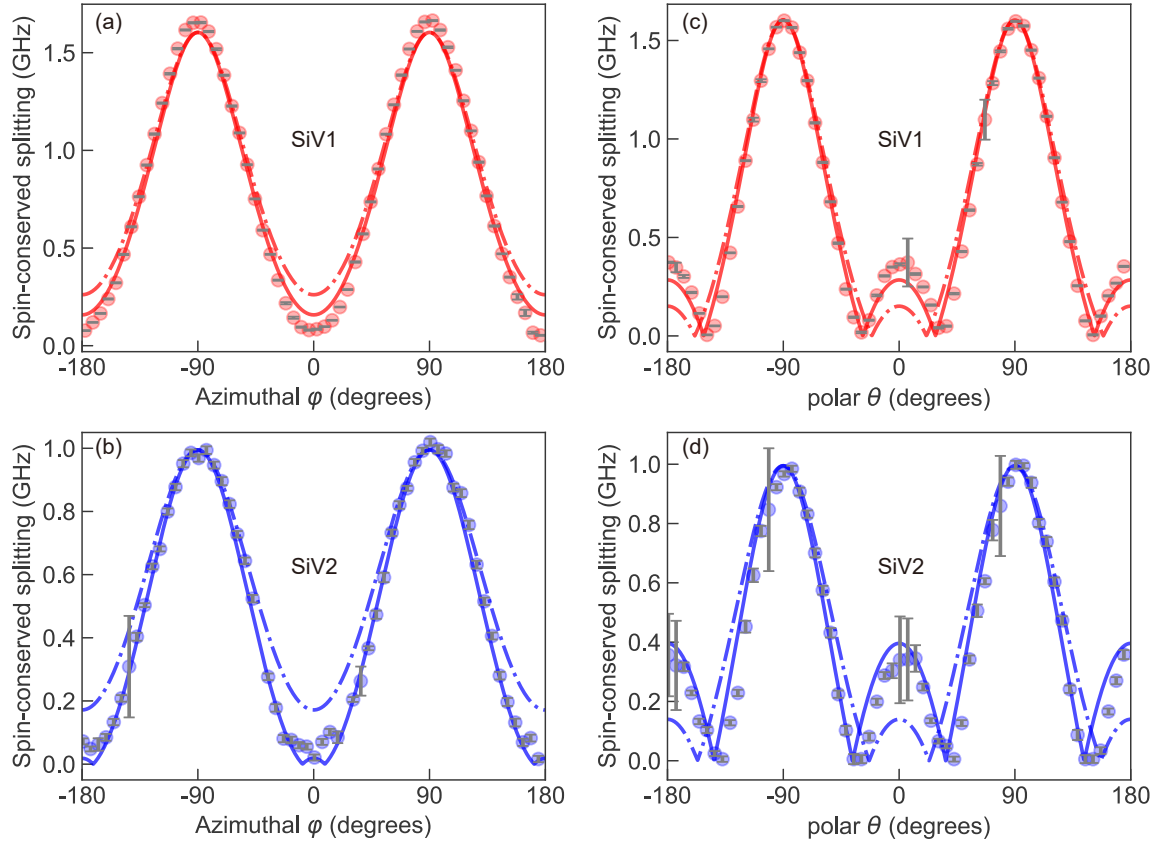


Figure 9.2. (a) and (b) Spin-conserved splitting obtained as a function of the azimuthal angle of the B-field in the X-Y plane. (c) and (d) Spin-conserved splitting obtained as a function of the polar angle (with respect to the z-axis) of the B field in the z-Z plane. The dotted and solid lines represent calculated spin-conserved splitting with equal and unequal orbital quenching factors, respectively, as discussed in the text. Error bars were derived from Lorentzian fits to PLE resonances.

refrigerator (Leiden Cryogenics CFCS81-1000M), which is also equipped with a 3D vector magnet (American Magnetics). The SiV fluorescence was collected with an objective with NA=0.82 integrated into an 8f imaging system inside the refrigerator [Lawrie, Feldman, Marvinney, and Pai \(2021\)](#). SiV fluorescence exiting an optical window at the top of the refrigerator was coupled into a multi-mode fiber with a diameter of $10\mu\text{m}$ and then sent to an avalanche photodiode for photon counting. For PLE spectra, a 532 nm laser pulse was used for the initialization of the SiV center. A red laser with a wavelength near 737 nm (New Focus Velocity TLB-6700) was used for the resonant excitation of the SiV center. An electrooptic modulator (EOM) was used to generate the two optical fields needed for the CPT experiments. One field came from the carrier wave and the other came from the first sideband generated by the EOM. Optical pulses needed were generated with acousto-optic modulators (AOMs). Experimental studies presented in this chapter were performed at a temperature near 4 K and with two SiV centers (SiV1 and SiV2) from two separate samples.

To determine the direction of the SiV axis and especially an axis normal to the SiV axis, we measured the spin-conserved splitting derived from corresponding PLE spectra as a function of the orientation of the magnetic field in the X-Y plane with magnetic field, $B=0.185\text{ T}$, as shown in [Figure 9.2a](#) and b. As the magnetic field rotates in the X-Y plane, the angle between the SiV axis and the field varies from 35.3 degrees to 90 degrees. Theoretically, we expect that the maximum spin-conserved splitting occurs when the magnetic field is normal to the SiV axis. Note that for convenience, we set one of the angular positions, where the spin-conserved splitting reaches the maximal value, as $\varphi = 90^\circ$. From the angular coordinates for the maximum spin-conserved splitting, we can then determine the direction of

the SiV axis. We further confirmed the direction of the SiV axis by showing that the spin-conserved splitting obtained remains nearly unchanged when we rotate the direction of a magnetic field in the plane normal to the SiV axis. In addition, we also measured the spin-conserved splitting as a function of the orientation of a magnetic field in the z - Z plane with $B=0.185$ T, as shown in [Figure 9.2c](#) and [d](#). Note that z is along the SiV axis as discussed earlier.

[Figure 9.3a](#) and [b](#) show the spin-conserved splitting of the C-transition vs the amplitude of the magnetic field when the field is along the Z -axis for SiV1 and SiV 2, respectively. [Figure 9.3c](#) and [d](#) show the spin-conserved splitting as a function of the amplitude of the magnetic field when the field is normal to the SiV axis. [Figure 9.4a](#) shows, as an example, a PLE spectrum of SiV1 obtained when the magnetic field is normal to the SiV axis, with $B=0.13$ T. We will use the spin-conserved splitting shown in [Figure 9.4a](#), along with the CPT resonance position shown in [Figure 9.4b](#), to determine the strain/JT parameter for the ground and excited states.

For the CPT experiment, we used the spin-conserved C_2 transition and the spin-flip C_4 transition in [Figure 9.1b](#) to form a Λ -type three-level system. The spectral position of the CPT resonance directly measures the frequency separation of the two lower energy states in the ground state doublet. In principle, this frequency separation can also be deduced from the corresponding frequency separation between the underlying spin-conserved and spin-flip transitions in the PLE spectrum. Nevertheless, the weak spin-flip transition makes it difficult to accurately determine or even conclusively identify the transition in the PLE spectrum. To search for the CPT resonance, we first fix the optical carrier wave at the spin conserved C_2 transition, while the frequency of the first sideband generated

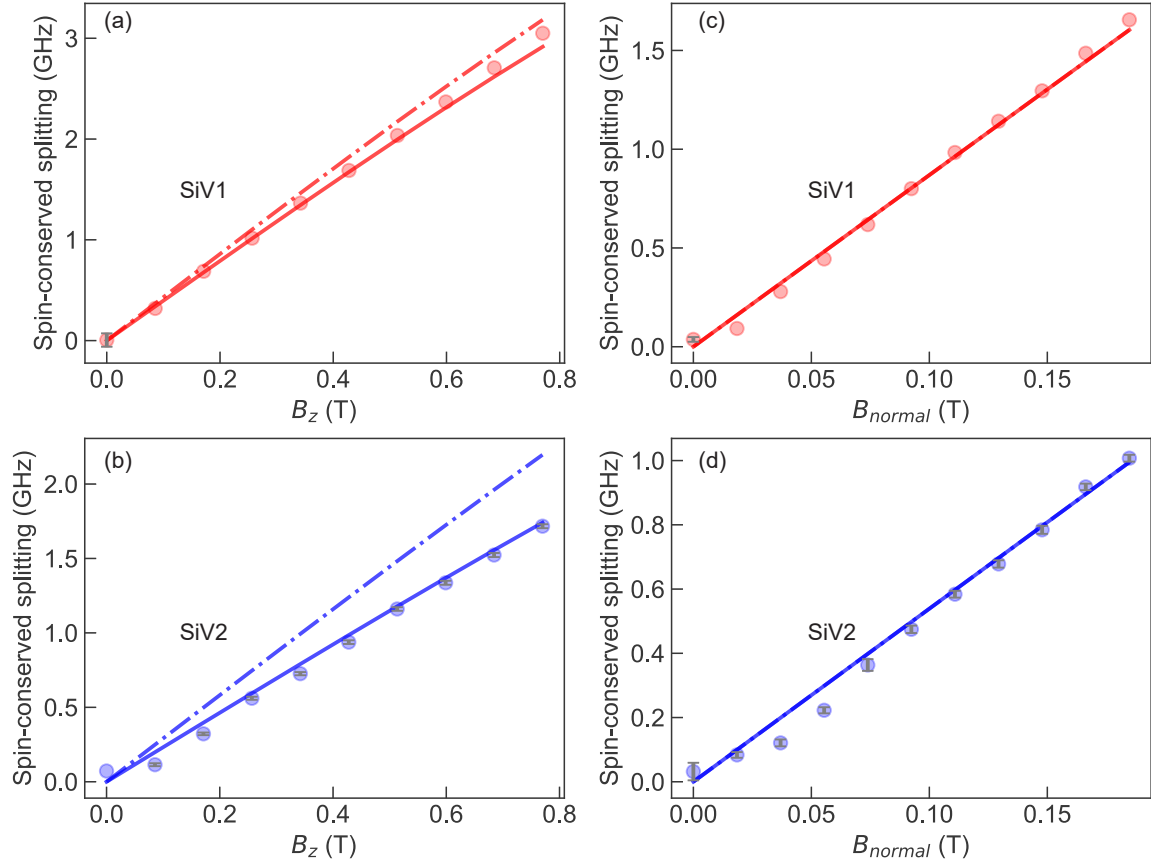


Figure 9.3. (a) and (b) Spin-conserved splitting obtained as a function of the B-field strength and with the B-field along the Z-axis. (c) and (d) Spin-conserved splitting obtained as a function of the B-field strength and with the B-field normal to the SiV axis. Error bars were derived from Lorentzian fits to PLE resonances. The dotted and solid lines represent calculated spin-conserved splittings with equal and unequal orbital quenching factors, respectively, as discussed in the text. The two lines are identical in (c) and (d) because there is no orbital contribution when the field is normal to the SiV axis.

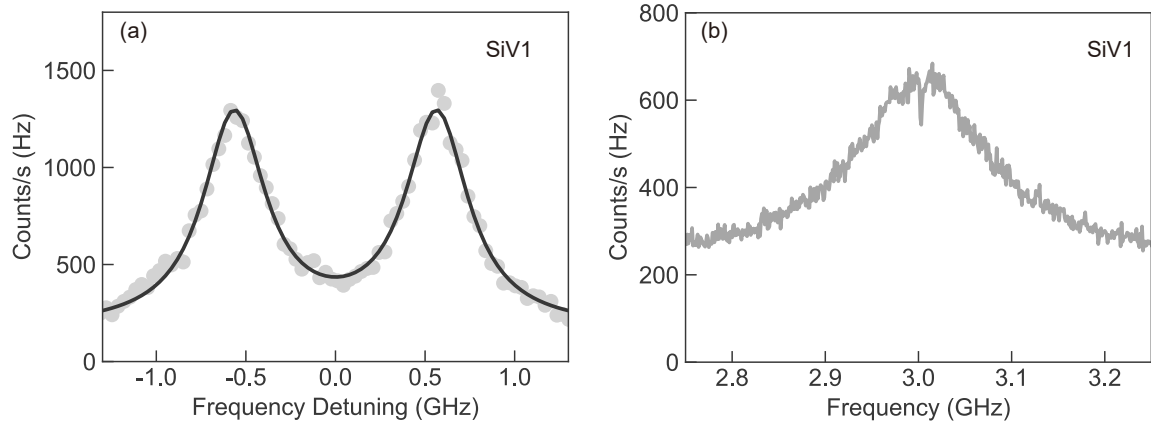


Figure 9.4. (a) PLE spectrum of the SiV C-transition. The two peaks correspond to the C2 and C3 transitions. The solid line is a least-square fit to two Lorentzians. (b) CPT spectral response obtained with an input laser power of $0.25 \mu\text{W}$. All were obtained with $B=0.13 \text{ T}$ along a direction normal to the SiV axis.

by the EOM is scanned over the expected spectral range of the spin-flip transition. After the successful observation of the CPT resonance, we change the experimental setting such that the optical carrier wave is fixed at the weak spin-flip C_4 transition and the first sideband is now near the spin-conserved transition. This is because strong CPT dips occur when the Rabi frequencies for the spin-conserved and spin-flip optical transitions are comparable. [Figure 9.4b](#) shows a CPT spectral response observed for SiV1 in this setting with a magnetic field, $B=0.13 \text{ T}$, normal to the SiV axis.

9.2.4 Analysis and discussions

The spectral position of the CPT resonance and the spin-conserved splitting are $3.01907 (0.00008) \text{ GHz}$ and $1.1300 (0.0045) \text{ GHz}$, respectively, for SiV1 (at 0.13 T) and $3.07956 (0.00025) \text{ GHz}$ and $0.6149(0.0084) \text{ GHz}$, respectively, for SiV2 (at 0.118 T). With [Equation 9.4](#), we obtained $\Upsilon_g^s = 33,46 \text{ GHz}$ and $\Upsilon_e^s = 77,129 \text{ GHz}$, for SiV1 and SiV2, respectively, with an uncertainty less than 2%, where we assumed $\lambda_{so}^g = 45 \text{ GHz}$ and $\lambda_{so}^e = 257 \text{ GHz}$ [Hepp et al. \(2014\)](#). In comparison,

earlier studies using low strain samples have obtained a maximum JT coupling rate of 15 GHz and 76 GHz for the ground and excited states, respectively, based on numerical fittings of a relatively large set of experimental data [Hepp \(2014\)](#). Note that since effects of strain and JT coupling cannot be separated in the experiments, the effects of strain should already be included in the earlier study.

We have used the strain/JT coupling rates obtained above to numerically calculate the spin-conserved splitting under the various magnetic field configurations used in [Figure 9.2](#) and [Figure 9.3](#), with the assumption of equal orbital quenching factor of 0.1 for both the ground and excited states, which has been used in all earlier studies. Note that there is no other fitting parameter used in these calculations. While there are overall good agreements, there are considerable and systematic deviations between the experimental and the calculated values, as shown in [Figure 9.2](#) and [Figure 9.3](#). In particular, the calculations underestimate the spin-conserved splitting when the magnetic field is along the SiV axis (i.e., the z-axis), as shown in [Figure 9.2c](#) and d, and the overestimate the spin-conserved splitting when the magnetic field is along the Z-axis, as shown in [Figure 9.3a](#) and b. These discrepancies can be resolved if we assume unequal quenching factors for the ground and excited states. For this additional numerical fitting, we varied the excited-state quenching factor while fixing all other parameters derived or used in the earlier numerical fitting. A quenching factor of 0.13 and 0.17 for the excited states were obtained for SiV1 and SiV2, respectively, as indicated by the solid lines in [Figure 9.2](#) and [Figure 9.3](#)

[Figure 9.5](#) shows the calculated relative frequencies of the four C-transitions of SiV1 as a function of a magnetic field along the Z-axis using the strain/JT coupling rates obtained above, with equal orbital quenching factor of 0.1 for the

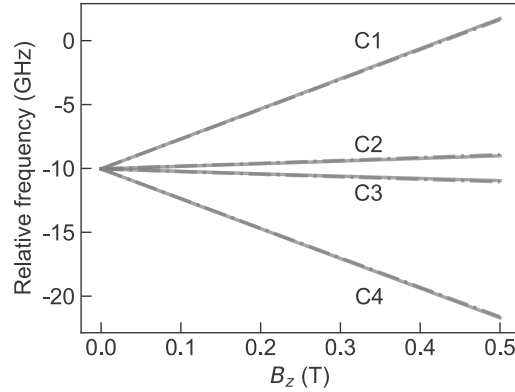


Figure 9.5. Calculated transition frequency for the four C-transitions under a B-field along the Z axis and with the parameters given in the main text. An equal quenching factor of 0.1 was used for the dotted lines. A quenching factor of 0.1 and 0.13 for the ground and excited state, respectively, was used for the solid lines.

ground and excited states (dotted lines) and with a quenching factor of 0.1 and 0.13 for the ground and excited state, respectively (solid lines). At the relatively large frequency scale used for [Figure 9.5](#), the difference in the quenching factors used does not induce discernable differences in the relative frequencies of the optical transitions. This agrees with the earlier study [Hepp et al. \(2014\)](#), which relies on the magnetic field dependence of the energy levels like those shown in [Figure 9.5](#) and assumes equal orbital quenching factors for the ground and excited states. The differences, however, can be clearly observed in [Figure 9.2](#) and [Figure 9.3](#) that feature a relatively small frequency scale, indicating that an accurate description of the SiV energy level structure requires the use of unequal orbital quenching factors. We have been able to achieve the required accuracy, at which relatively small orbital Zeeman effects become important, by first determining the strain/JT coupling rates in a transverse magnetic field, with no complication from the orbital magnetic coupling and with no numerical fitting.

9.2.5 Conclusion

The energy level structure of a SiV center in a magnetic field is characterized by the strain/JT coupling rates and the orbital quenching factors for the ground and excited states. For the accurate experimental determination of these parameters, we have developed an experimental approach, which overcomes the inefficiency and difficulty of the earlier approach. We determine the two strain/JT coupling rates by performing two measurements in a transverse magnetic field, specifically, the splitting between two spin-conserved optical transitions and the CPT resonance of the two lower spin states in the ground states. The orbital quenching factors for the ground and excited states are then extracted from a detailed comparison between the theoretical calculation and the experimental result on the dependence of the spin-conserved splitting on the magnetic field amplitude and direction. The determination of SiV energy level structures in an off-axis magnetic field is important for studies such as acoustic quantum control, cavity QED, and quantum spin-mechanics, and is also essential to the development of experiment approaches that can mitigate the variations of the SiV ground state energy levels due to the local strain environment.

9.3 Coherent population trapping and spin relaxation of a silicon vacancy center in diamond at millikelvin temperatures

9.3.1 Introduction

Color centers in diamond have emerged as a promising qubit platform for quantum information processing [Awschalom et al. \(2018\)](#); [Bradac et al. \(2019\)](#); [Childress, Walsworth, and Lukin \(2014\)](#); [Doherty et al. \(2013\)](#). While negatively charged nitrogen vacancy (NV) centers in diamond feature long spin decoherence times for electron spins even at room temperature [Awschalom et al. \(2018\)](#);

Childress et al. (2014); Doherty et al. (2013), negatively charged SiV centers feature superior optical properties Bradac et al. (2019); Neu et al. (2013); Rogers, Jahnke, Teraji, et al. (2014); Sipahigil et al. (2014). Because of their inversion symmetry, SiV centers feature optical transitions that are robust against charge fluctuations Neu et al. (2013); Rogers, Jahnke, Teraji, et al. (2014); Sipahigil et al. (2014). The zero-phonon line of SiV centers contains about 70% of the total fluorescence, with a nearly lifetime-limited optical linewidth. Excellent optical coherence has been observed for SiV centers in diamond nanostructures and in diamond membranes as thin as 100 nm Evans et al. (2016); Jantzen et al. (2016); Lekavicius and Wang (2019); Sipahigil et al. (2016); J. L. Zhang et al. (2017). All-optical as well as microwave control of SiV spins has been demonstrated Becker et al. (2018); Pingault et al. (2014, 2017); Rogers, Jahnke, Metsch, et al. (2014). In addition, the integral orbital degrees of freedom in the SiV ground spin states make it feasible to control the SiV spin with mechanical vibrations Maity et al. (2020); Meesala et al. (2018), which also suggests the possibility of quantum control of an electron spin at the level of single phonons in a diamond spin-mechanical resonator. Strong orbital interactions with thermal phonons, however, lead to relatively fast spin dephasing and spin relaxation at elevated temperatures.

Spin dephasing, specifically T_2^* , of SiV centers has been investigated with Ramsey interferometry as well as coherent population trapping (CPT) Becker et al. (2018); Pingault et al. (2014, 2017); Rogers, Jahnke, Metsch, et al. (2014), though there have been no reports of CPT studies below 4 K. Phonon-induced spin dephasing can be suppressed with decreasing temperatures. Nearly complete suppression of phonon-induced dephasing has been observed at temperatures near 100 mK, where decoherence times can exceed 10 ms in isotopically enriched

diamond [Sukachev et al. \(2017\)](#). A relatively large ground-state orbital splitting, which can be achieved in group IV color centers such as tin vacancy or induced by an externally applied mechanical strain, can also effectively suppress the effects of thermal phonons [Debroux et al. \(2021\)](#); [Sohn et al. \(2018\)](#). In addition to spin dephasing studies, spin relaxation has also been investigated with spin lifetime measurements at temperatures near 4 K and at 100 mK [Becker et al. \(2018\)](#); [Pingault et al. \(2014, 2017\)](#); [Rogers, Jahnke, Metsch, et al. \(2014\)](#). Note that an earlier study on spin dephasing and spin lifetime in SiV centers between 4 K and 100 mK used HPHT diamond containing a large concentration of substitutional nitrogen impurities [Becker et al. \(2018\)](#), with T_2^* and spin lifetime that differ considerably from that of high purity diamond grown by chemical vapor deposition (CVD). Additional experimental studies between 4 K and 100 mK can thus provide further information on phonon-induced spin dephasing and relaxation processes.

The latest experimental advance on the realization of GHz diamond spin-mechanical resonators with mechanical Q-factors exceeding 10^6 have stimulated considerable efforts in the experimental pursuit of phononic cavity QED with SiV spins [Joe et al. \(2024\)](#); [X. Z. Li et al. \(2024\)](#); [Wang and Lekavicius \(2020\)](#). For these studies, phonon-induced relaxation and CPT related processes at mK temperature are expected to play an important role. Recently, CPT has also been exploited for real-time quantum sensing with a NV center [Turner et al. \(2022a\)](#); [S. H. Wu et al. \(2021\)](#). CPT studies at mK temperatures will be an essential step in the use of CPT-based real-time sensing to suppress spin dephasing in SiV centers induced by the nuclear spin bath.

In this chapter, we report experimental studies of CPT at temperatures as low as 100 mK and spin relaxation between 4 K and 830 mK in a SiV center

subject to a transverse magnetic field. By resolving an issue of power broadening in CPT spectral response, we are able to extract spin linewidth, which is determined by T_2^* , from power dependent CPT linewidth. The CPT studies show that near and below 1 K, phonon-induced spin dephasing becomes negligible compared with that due to the nuclear spin bath of naturally abundant ^{13}C atoms. The temperature dependence of the spin dephasing rates agrees with the theoretical expectation that the phonon-induced spin dephasing arises primarily from orbital relaxation of the SiV ground states induced by the first order electron-phonon interactions. In addition, a nearly 100-fold increase in spin lifetime is observed when the temperature is lowered from 4 K to slightly below 1K. A numerical analysis of the observed temperature dependence of the spin lifetime along with a qualitative consideration of relevant spin relaxation processes in the SiV ground states reveals that two-phonon spin-flip transitions play an essential role in the spin relaxation of the SiV ground states.

9.3.2 Sample and experimental setup

Our experimental studies were carried out with a SiV center implanted about 75 nm below the surface of an electronic grade CVD-grown diamond. The average kinetic energy and dosage of the ^{28}Si used in the implantation are 100keV and $3 \times 10^9/\text{cm}^2$, respectively. Stepwise thermal annealing up to a temperature of 1200 degrees followed by wet chemical oxidation was used for the removal of the damaged surface layer as well as for the formation of SiV centers. The sample was thermally anchored to the mixing chamber of a dilution refrigerator (Leiden Cryogenics CFCS81-1000M), which is fitted with a 3D vector magnet (American Magnetics). The SiV fluorescence was collected with an objective with NA=0.82, followed by an optical collimation system installed in a cold-insertable probe in

the refrigerator [Lawrie et al. \(2021\)](#). SiV fluorescence exiting an optical window at the top of the refrigerator was coupled into a multi-mode fiber with a diameter of 10 μm and then sent to an avalanche photodiode for photon counting. For photoluminescence excitation (PLE) spectra, a 532 nm laser pulse was used for the initialization of the SiV center. A red laser with a wavelength near 737 nm (New Focus Velocity TLB-6700) was used for the resonant excitation of the SiV center. An electro-optic modulator (EOM) was used to generate the two optical fields needed for the CPT experiments. One field came from the carrier wave and the other came from the first sideband generated by the EOM. For the CPT experiments, the laser power given is the power of the 737 nm laser incident on the sample. Optical pulses needed were generated with acousto-optic modulators (AOMs). Time resolved fluorescence measurements were performed with a time tagger (Swabian Instruments).

9.3.3 Results and discussions

For a negatively charged SiV center, both the ground and excited states are characterized by orbital states, $|e_x\rangle$ and $|e_y\rangle$, and spin states, $|\uparrow\rangle$ and $|\downarrow\rangle$. Spin-orbit coupling leads to the formation of doubly degenerate doublets for the ground and excited states, with the two upper states being $|e_+, \uparrow\rangle$ and $|e_-, \downarrow\rangle$ and the two lower states being $|e_-, \uparrow\rangle$ and $|e_+, \downarrow\rangle$, where $|e_{\pm}\rangle = (|e_x\rangle \pm i|e_y\rangle)/\sqrt{2}$, as shown schematically in [Figure 9.6a Hepp et al. \(2014\)](#). The splitting between the upper and lower states is λ_{so} , with ground-state splitting λ_{so}^g near 50 GHz and excited-state splitting λ_{so}^e near 260 GHz. In the absence of an external magnetic field, optical transitions between the ground and excited states are all spin-conserving. An off-axis magnetic field removes the spin degeneracy and induces spin-state mixing of the ground and excited states, as illustrated in [Figure 9.6a](#). The spin-

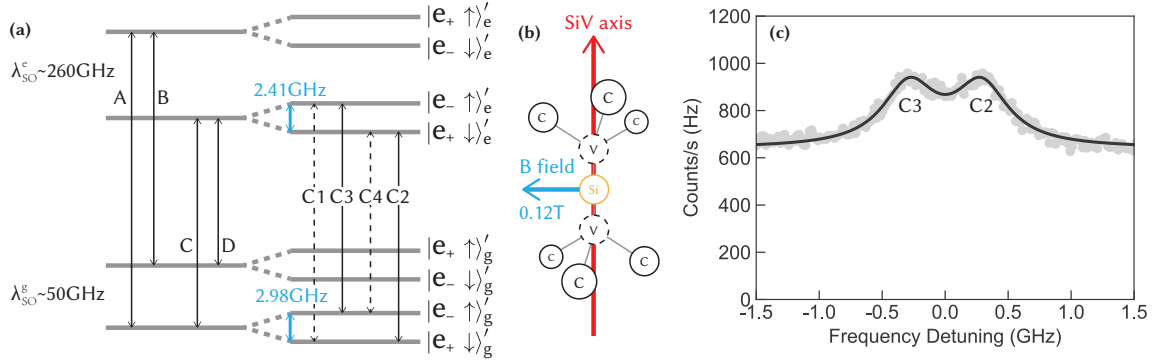


Figure 9.6. (a) Energy level structure of the SiV center in the presence of an off-axis magnetic field. The prime in the state label indicates that the spin states as well as the orbital states are slightly mixed. (b) Schematic of a SiV center subject to a magnetic field normal to the SiV axis. (c) PLE spectrum of the SiV C-transition obtained at 4 K.

state mixing enables spin-flip optical as well as microwave and acoustic transitions. Note that both the spin and orbital states are slightly mixed. The orbital state mixing arises from strain and Jahn-Teller effects [Hepp et al. \(2014\)](#). As illustrated in [Figure 9.6b](#), a magnetic field with a direction normal to the SiV axis and with $B=0.12 \text{ T}$ was used for all the experimental results presented in this chapter, unless otherwise specified, to maximize the spin-state mixing at the given field strength. Our experimental studies were carried out on the C-transition of the SiV center (see [Figure 9.6a](#)). [Figure 9.6c](#) shows the PLE spectrum obtained for the C-transition at 4 K. The two resonances in the PLE spectrum correspond to the spin-conserved C_2 and C_3 transitions.

The spin-conserved C_2 transition and the spin-flip C_4 transition in [Figure 9.6a](#) form a Λ -type three-level system for our CPT experiments. The spectral position of the weak spin-flip transition, however, is difficult to identify directly in the PLE spectrum. We searched for the weak spin-flip transition with two approaches, optical pumping and CPT, but with essentially the same

experimental setting, for which an optical carrier wave is fixed at the spin conserved C_2 transition, while the frequency of the first sideband generated by the EOM is scanned over the expected spectral range of the spin-flip transition. After this set of experiments, we changed the experimental setting such that the optical carrier wave is fixed at the weak spin-flip C_4 transition and the first sideband is now near the spin-conserved transition. This is because strong CPT dips occur when the Rabi frequencies for the spin-conserved and spin-flip optical transitions are comparable. [Figure 9.7a](#) shows, as an example, a CPT spectral response obtained in this setting at 4 K. CPT spectral responses depend strongly on the incident optical powers, as shown in [Figure 9.7b](#). [Figure 9.7c](#) plots the CPT linewidth as a function of the incident optical power obtained at 150 mK. We extract the intrinsic CPT linewidth by fitting the power-broadened CPT linewidth to a linear power dependence. Note that the error bars plotted in [Figure 9.7c](#) and [d](#) include uncertainties in the numerical fits and an additional 10% uncertainty due to polarization fluctuations in the experiment.

[Figure 9.7d](#) plots the spin linewidth or the intrinsic CPT linewidths derived from the linear power dependence of the CPT linewidth as a function of temperature from 4 K to as low as 150 mK. Note that stable temperatures between 4 K and 1 K were difficult to achieve in our current setup. At 4 K, the spin dephasing time, T_2^* , derived from the intrinsic CPT linewidth (2.35 MHz) is in good agreement with that obtained from earlier Ramsey interferometry studies [Pingault et al. \(2017\)](#). Note that CPT linewidths at 4 K reported from earlier studies significantly exceed the intrinsic linewidth obtained in our experiment, which is likely due to power broadening of the CPT spectral response in the earlier experiments. At temperatures near and below 1 K, the intrinsic CPT linewidths are

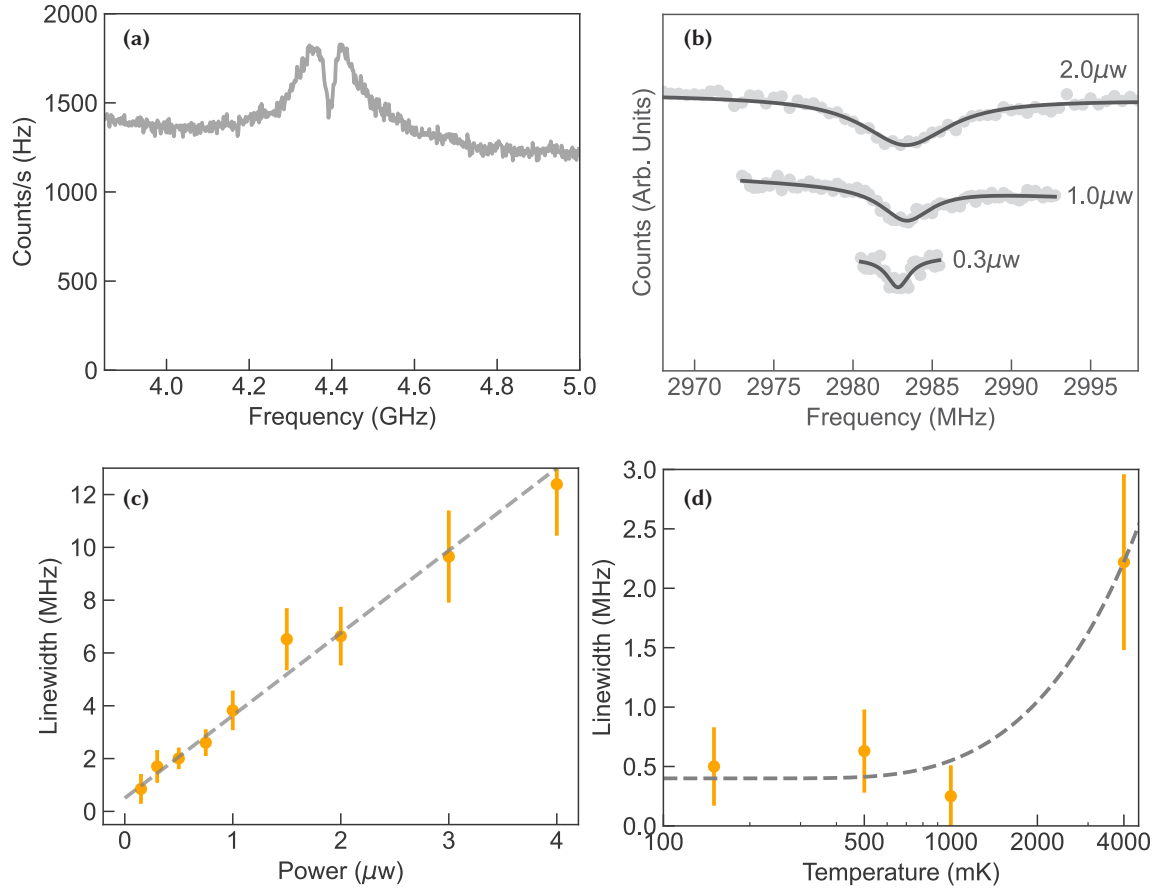


Figure 9.7. (a) CPT spectral response obtained at 4K with an input laser power of 3 μW . (b) Power dependence of the CPT spectral response at 0.15 K. The incident optical powers used are indicated in the figure. (c) Power dependence of the CPT linewidth at 0.15 K. The linear fit yields an intrinsic CPT linewidth of 0.5 $\pm$ 0.33 MHz. (d) The intrinsic CPT linewidth as a function of temperature. The dashed line is a numerical fit, assuming that the phonon-induced dephasing rate is proportional to the thermal occupation of phonons with a frequency of 50 GHz. Except for the CPT spectral response in (a), which was obtained with $B = 0.15$ T along the crystal z-axis, all other data were obtained with $B = 0.12$ T normal to the SiV axis.

nearly independent of temperature, indicating that phonon-induced dephasing is negligible compared with dephasing induced by the nuclear spin bath.

Earlier experimental and theoretical studies have attributed the phonon-induced spin dephasing at temperatures near and above 4 K to the orbital relaxation between the two lower and upper states in the ground state doublet, induced by the first order electron-phonon interaction with phonon frequencies near λ_{so}^g [Jahnke et al. \(2015\)](#). In this case, the phonon-induced spin dephasing rate is expected to be proportional to the thermal phonon occupation, $n(T) = 1/[\exp(h\lambda_{so}^g/k_B T) - 1]$. The numerical fit shown in [Figure 9.7d](#) indicates a good agreement between the experimental result and the theoretical expectation for temperatures below 4 K.

At $T < 1$ K, thermal populations of phonons near 50 GHz are greatly suppressed. Phonon-induced spin dephasing and spin relaxation, however, can still arise from the direct acoustic transition between the two lower energy ground spins states, especially when a transverse magnetic field is used to maximize spin-state mixing. [Figure 9.7d](#), however, indicates that any possible phonon-induced spin dephasing time will have to be much longer than T_2^* induced by the nuclear spin bath. It should be noted that earlier spin dephasing and lifetime studies at 100 mK used a magnetic field along the SiV axis to minimize effects of spin-state mixing [Sukachev et al. \(2017\)](#). In addition, the earlier study using a HPHT diamond sample showed a T_2^* about 29 ns at 40 mK and T_1^{spin} about 9 s at 0.8 K [Becker et al. \(2018\)](#), which differ significantly from the properties of SiV centers in high purity CVD-grown diamond reported in this work as well as earlier experimental studies [Pingault et al. \(2014, 2017\)](#); [Rogers, Jahnke, Metsch, et al. \(2014\)](#).

To probe phonon-induced spin relaxation, we have measured the spin lifetime, T_1^{spin} , using the initialization-readout approach developed in an earlier study [Rogers, Jahnke, Metsch, et al. \(2014\)](#). As illustrated in [Figure 9.8a](#), a laser pulse resonant with the spin-conserved C_2 transition excites the SiV center and initializes it into the spin-up state, $|e_-, \uparrow\rangle$, through an optical pumping process, specifically, decay via the spin-flip C_4 transition. A second laser pulse, again resonant with the C_2 transition, arriving after a waiting period, τ , reads out the population in the spin-down state, $|e_+, \downarrow\rangle$. This population is in large part due to spin relaxation occurring during the waiting period. [Figure 9.8b](#) shows an example of the time resolved fluorescence obtained in an initialization-readout experiment. The initial decay of the fluorescence during the leading edge of the optical pulses reflects the rapid optical pumping process. [Figure 9.8c](#) plots the ratio of the initial peaks of the time-resolved fluorescence observed (the second over the first) as a function of the waiting period. The spin lifetime can be extracted from the exponential recovery of the peak ratio with increasing waiting period.

[Figure 9.8d](#) shows $1/T_1^{\text{spin}}$ extracted from the initialization-readout experiments at temperatures between 4 K and 830 mK. The spin lifetime obtained near 4 K is in good agreement with earlier studies. At $T < 500\text{mK}$, our experiments indicate $T_1^{\text{spin}} > 0.1\text{ ms}$ (not shown in [Figure 9.8d](#)). Note that for the spin lifetime measurement, a time resolution capable of resolving the initial optical pumping peak, which occurs in a 10 ns timescale, is needed. This necessitates a relatively long time for data taking. However, as the waiting period increases, the required data-taking time becomes increasingly challenging to achieve. In particular, during long data-taking periods, the SiV resonance can shift due to spectral diffusion. The charge state may also change due to continuous resonant

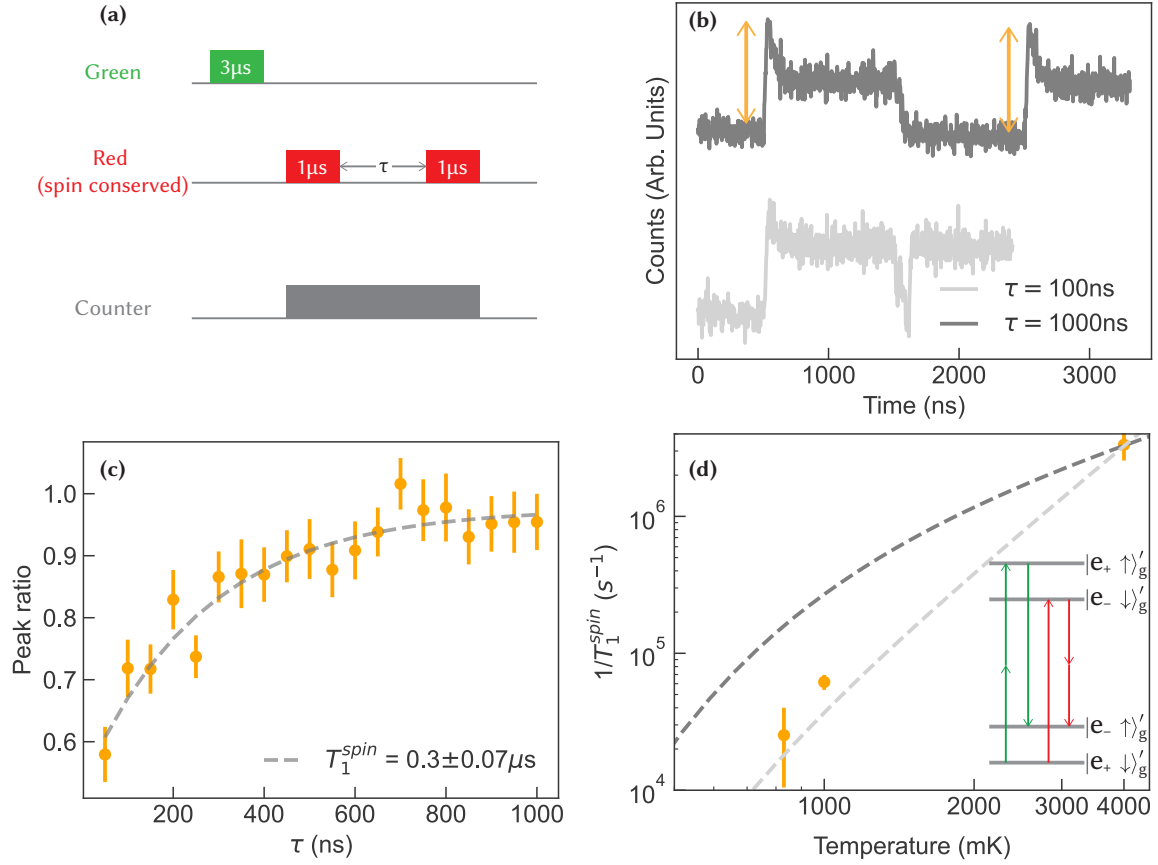


Figure 9.8. (a) Schematic of the pulse sequence used for the spin lifetime measurement. (b) Fluorescence as a function of time showing the initialization and readout of the SiV spin state at 4 K. Orange arrows denote peak values used for numerical fitting. (c) The peak ratio (the second over the first) as a function of the waiting period, τ , at 4 K. The dashed line is an exponential fit to a recovery time of 0.3 μs . (d) Temperature dependence of the spin lifetime. Dark and light dashed lines are numerical fits to temperature dependence of spin relaxation rates that involve single-phonon spin-flip and two-phonon spin-flip transitions, respectively, as discussed in the text. The inset shows schematically two relaxation pathways that involve two-phonon spin-flip transitions.

excitations. These experimental difficulties limited the longest lifetime that can be adequately measured in our current experimental setup to about 0.1 ms.

The nearly 100-fold increase of spin lifetime when the temperature is decreased from 4 K to slightly below 1 K shown in [Figure 9.8d](#) corresponds to strong suppression of phonon-induced spin relaxation. The steep increase of T_1^{spin} also indicates that the spin relaxation results from transitions between the two lower and upper states in the ground state doublet separated by λ_{so}^g . Specifically, the spin relaxation can take place in a dynamical process, for which a SiV in a lower state is excited to an upper state through phonon absorption and then returned to the other lower state through phonon emission. Note that the spin-flip can occur via either phonon absorption or phonon emission. In the limit that the transitions between the lower and upper states in the ground state doublet take place via a single-phonon process, i.e., the first order electron-phonon interactions, the overall spin relaxation rate is expected to scale with $n(T)[1 + n(T)]$. However, as shown by the numerical fit in [Figure 9.8d](#), this temperature dependence can only lead to a 25-fold increase in the spin lifetime when the temperature is decreased from 4 K to 1 K.

The ground-state energy level structure of the SiV center shown in [Figure 9.6a](#) dictates that spin-flip transitions between the lower and upper states in the ground state doublet take place between two states that have the opposite spin orientations but nearly the same orbital components. As a result, the single-phonon spin-flip transition rate depends not only on the spin-state mixing induced by the off-axis magnetic field, but also on the orbital-state mixing induced by strain and JT effects, because the selection rule for acoustic transitions requires that the single-phonon transition changes the orbital states involved in the transition. In

comparison, a two-phonon transition between the lower and upper states does not change the orbital states involved. Because of the relatively small orbital state mixing, the two-phonon spin-flip transition rate can exceed or at least be comparable to the corresponding one-phonon spin-flip transition rate.

The large discrepancy between the observed temperature dependence and the $n(T)[1 + n(T)]$ dependence shown in [Figure 9.8d](#) indicates significant contributions from the two-phonon spin-flip transitions. As shown in the inset of [Figure 9.8d](#), the two-phonon spin-flip transition can take place via either phonon absorption or phonon emission. The corresponding spin relaxation rate is expected to scale with $n_{1/2}^2(T)[1 + n(T)]$ and $n(T)[1 + n_{1/2}(T)]$, respectively, where $n_{1/2}(T)$ is the thermal population for phonons with frequency $=\lambda_{so}^g/2$ (it can be shown that $n_{1/2}^2(T)[1 + n(T)] = n(T)[1 + n_{1/2}(T)]^2$). This temperature dependence can provide a good description of the observed temperature dependence, as shown by the numerical fit in [Figure 9.8d](#). Nevertheless, a detailed theoretical analysis is still needed to determine the relative contributions of the single-phonon and two-phonon spin-flip transitions, which cannot be reliably extracted from the numerical fit.

In addition to the multi-phonon spin relaxation processes discussed above, spin relaxation can also take place through the direct acoustic transition between the two lower energy ground states with a frequency separation near 3 GHz. The spontaneous emission lifetime for the direct acoustic transition is an important parameter for phononic cavity QED studies. At 1 K, the thermal occupation for phonons near 3 GHz is approximately 6. The observed T_1^{spin} near 1 K thus sets a lower limit of about 0.2 ms for the spontaneous emission lifetime for the direct acoustic transition. The actual spontaneous emission lifetime is expected to be

much longer since phonon-induced spin-flip transitions between the lower and upper states of the ground state doublet are expected to play a dominant role near 1 K.

CHAPTER X

SUMMARY AND OUTLOOK

In this thesis, we have developed both the theory and experiments for a CPT-based sensing approach using NV centers in diamond, opening opportunities for sensing applications where continuous measurement is crucial. Our analysis shows that, even under practical experimental conditions, the CRLB can be approached by using an OU Bayesian estimator. A neural-network-based estimator further confirms this optimality, during training the RNN outcomes converges to the predictions of the OU Bayesian model.

We also combine feedback with sensing to probe the spectroscopy of the noise in an OU basis. This makes our CPT-based sensing functional even without a precise priori knowledge of the noise structure. In addition, we report preliminary results on the spin physics and properties of SiV centers under magnetic fields at millikelvin temperatures, a key step toward extending our CPT-based sensing to SiV centers. Beyond these directions, we demonstrate a microsphere-enhanced collection efficiency for confocal experiments, which can benefit quantum sensing applications that favor long-working-distance objectives.

Based on these work, several promising avenues for future exploration are natural:

- (i) Use our OU-basis spectroscopy to probe the actual noise environment, extract its spectrum, and compare the results with standard dynamical-decoupling spectroscopy.
- (ii) With prior knowledge of the real noise, apply feedback techniques to extend the coherence time and explore its limits.

(iii) Improve sensitivity and overall performance using, for example, microsphere-enhanced collection and/or hyperfine-resolved CPT that gives larger slope and higher sensitivity.

(iv) Extend our approach to SiV centers. Owing to their short excited-state lifetime, SiV centers give higher count rate for quantum sensing given excitation leakage is effectively suppressed (e.g., via cross-polarization or ultra-narrow filters), and coupling to cavity QED systems could further boost counts.

APPENDIX A

ENERGY LEVEL STRUCTURES DERIVED FROM GROUP THEORY

A.1 Review of group representation approach

The description in this section is a rephrasing of the more detailed discussion in [Hepp \(2014\)](#). Therefore we are not aiming at a complete and self-contained introduction, but rather at a summary of the basic structure and key concepts that we will need. The basic idea is simple: the symmetry of a quantum system should be reflected in its Hamiltonian and energy levels. Instead of trying to build this symmetry into the Hamiltonian in an implicit, we can describe it systematically with the help of group theory.

A set G of operations with a binary multiplication defined on it is called a group if the following conditions are satisfied: (i) it is closed under the multiplication, (ii) the multiplication is associative, (iii) there exists an identity element and (iv) every element has an inverse. Here the multiplication is defined as a map $G \times G \rightarrow G$ and can be viewed as a generalization of the usual multiplication or addition of numbers. For example, the integers with addition form a group because they satisfy all these requirements.

The structure of a finite group is faithfully encoded in its multiplication table, such as [Table A.1](#), in which E , C_3 , C_2 denote symmetry operations that leave the NV structure in [Figure 2.2](#) unchanged.

In our discussion we are interested in the set of symmetry operations acting on the relevant atomic nuclei of color centers. Under the Born-Oppenheimer approximation we ignore the motion of the nuclei induced by the electrons, and therefore the positions of these nuclei define and dominate the Hamiltonian experienced by the electrons. Due to the lattice symmetry, the set of these nuclear

positions is invariant under certain coordinate transformations, such as rotations or inversions of the coordinate system. These operations acting on the set of nuclear positions form a group, where the multiplication is just the time ordering (composition) of the operations. This group is called the point group.

Although these operations are highly abstract, we can describe them in a more quantitative way by using their matrix form in the standard coordinate basis vectors $\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}}$. By doing so, we are allowed to use matrices as a convenient tool to investigate abstract symmetry operations. A generalization of this idea is called a group representation. If a set of matrices Γ under matrix multiplication reproduces the same multiplication table as a group G that is $\Gamma(a)\Gamma(b) = \Gamma(ab)$, then these matrices form a representation of the group G , and they encode the same structure as the abstract group elements. As an example [Hepp \(2014\)](#), the set of matrices

$$\begin{array}{ccc}
\begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix} & \begin{bmatrix} -\frac{1}{2} & -\frac{1}{2}\sqrt{3} & 0 \\ \frac{1}{2}\sqrt{3} & -\frac{1}{2} & 0 \\ 0 & 0 & 1 \end{bmatrix} & \begin{bmatrix} -\frac{1}{2} & \frac{1}{2}\sqrt{3} & 0 \\ -\frac{1}{2}\sqrt{3} & -\frac{1}{2} & 0 \\ 0 & 0 & 1 \end{bmatrix} \\
\Gamma(E) & \Gamma(C_3^1) & \Gamma(C_3^2) \\
\begin{bmatrix} \frac{1}{2} & \frac{1}{2}\sqrt{3} & 0 \\ \frac{1}{2}\sqrt{3} & -\frac{1}{2} & 0 \\ 0 & 0 & -1 \end{bmatrix} & \begin{bmatrix} \frac{1}{2} & -\frac{1}{2}\sqrt{3} & 0 \\ -\frac{1}{2}\sqrt{3} & -\frac{1}{2} & 0 \\ 0 & 0 & -1 \end{bmatrix} & \begin{bmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{bmatrix} \\
\Gamma(C_2) & \Gamma(C_2') & \Gamma(C_2'')
\end{array}$$

is a valid representation for the point group D_3 described by the multiplication table in [Table A.1](#). We see that all matrices in this representation have a block-diagonal form, and the matrices inside each block also form a representation of the same group. A representation that can be brought into such a block-diagonal form is called a reducible representation, and one that cannot is called an irreducible representation. Any representation can be written as a direct sum of irreducible

representations as $\Gamma = \sum_i a_i \Gamma_{\text{IR},i}$ where a_i are non-negative integers giving the multiplicities of the irreducible representations $\Gamma_{\text{IR},i}$. For a quantum system whose Hamiltonian is defined by a point group, these irreducible representations contain the core information about how the symmetry of the point group is reflected in the corresponding energy structure, as we will describe in the following.

In practice, for a given physical problem we are only interested in a certain set of quantum states. We choose a basis that spans exactly this set of states, for example a dangling-bond basis for the electronic problem or the coordinates of all relevant nuclei when we discuss lattice vibrations and phonons. Any state we care about can be written as a linear combination of these basis states. Once we know how each symmetry operation acts on the basis states, we automatically know how it acts on any state of interest. In this sense, the representation of the group on such a complete basis is sufficient to describe how the symmetry of the point group constrains the problem.

To extract irreducible representations from a given representation and to obtain all possible irreducible representations of a point group, we need an important property of group representations, namely the Schur orthogonality relations. They can be decomposed into several equations. One of them is

$$\sum_{g \in G} \chi^{(\lambda)}(g)^* \times \chi^{(\mu)}(g) = \delta_{\lambda\mu} |G|,$$

where G is a finite group and $\chi^{(\lambda)}(g)$, $\chi^{(\mu)}(g)$ are the traces (characters) of the irreducible representations λ, μ of the group. In particular, for $\lambda = \mu$ and for real characters we obtain $\sum_{g \in G} (\chi^{(i)}(g))^2 = |G|$. Another useful relation is

$$\chi^{(\sigma)}(g) = \sum_i c_i \times \chi_{\text{IR}}^{(i)}(g), c_i = \frac{1}{|G|} \sum_{g \in G} (\chi_{\text{IR}}^{(i)}(g))^* \times \chi^{(\sigma)}(g), \quad (\text{A.1})$$

where $\chi^{(\sigma)}(g)$ is the character of a (possibly reducible) representation, $\chi_{\text{IR}}^{(i)}(g)$ are the characters of the irreducible representations, and c_i are integers. These equations tell us that by projecting the vector consisting of the traces of the current representation for different group elements onto the vectors of irreducible characters, we are able to decompose the representation in a quick and systematic way.

Since the trace of a representation is so important, we often summarize it in the so-called character table of the group, one example of which is shown in [Table A.1](#). In such a table we have reduced the number of columns because elements in the same conjugacy class have the same trace. Indeed, if two elements g and hgh^{-1} are related by conjugation, then their matrices in any representation satisfy $\Gamma(hgh^{-1}) = \Gamma(h)\Gamma(g)\Gamma(h)^{-1}$, so they are similar matrices and therefore have the same trace $\chi(hgh^{-1}) = \chi(g)$ for any representation. Different irreducible representations have different character vectors, and the relation $\sum_{g \in G} (\chi^{(i)}(g))^2 = |G|$ together with the fact that the trace for the identity element equals the dimension of the representation puts strong constraints on the possible dimensions. From these constraints a complete set of irreducible representations can be derived.

Wigner's theorem states the following. The set of all operators that commute with the Hamiltonian of a quantum system form the symmetry group of that Hamiltonian. The commutation relations of the generators determine the algebra of the group. An n -dimensional representation of the symmetry group preserves the multiplication table of the symmetry operations. The possible degeneracies of the Hamiltonian are then given by the dimensionalities of the irreducible representations of the group, and the eigenfunctions belonging to

an n -fold degenerate eigenvalue form a basis for an n -dimensional irreducible representation.

From this theorem we obtain a relatively straightforward path towards analyzing the energy structure. First we identify the point group of the defect center. Then, from the representation of this point group, we find all irreducible representations. From the degeneracies of these irreducible representations we are able to obtain information about the degeneracies of the relevant orbital energy levels. Furthermore, from the electron number in the system and the structure of the orbital energy levels, we can build up the many-electron energy levels.

Additionally, group representation theory can also tell us whether a matrix element corresponding to an interaction term can induce a transition between two states. A matrix element $\langle \psi_i | H | \psi_j \rangle$ is a complex number. If a transition is forbidden by symmetry, this scalar must be zero. If a transition is allowed by symmetry, then a non-zero value of this matrix element must be invariant under all symmetry operations, otherwise the symmetry of the problem would be broken. In other words, a non-zero matrix element that respects the symmetry has to be unchanged under every element of the group. From the group-theoretical point of view, such an invariant scalar transforms according to the totally symmetric one-dimensional representation A_{1g} , which gives 1 for all operations therefore invariant. To find out whether this can happen for given states and a given interaction, we look at how the matrix element transforms when we apply a group operation. One can show that $\Gamma^{(i)} \otimes \Gamma^{(H)} \otimes \Gamma^{(j)}$ is a valid representation that satisfies the multiplication table and is complete for describing the transformation behaviour of $\langle \psi_i | H | \psi_j \rangle$. Therefore, a necessary condition for having a non-zero matrix element is $\Gamma^{(i)} \otimes \Gamma^{(H)} \otimes \Gamma^{(j)} \supset A_{1g}$. Here we have used an important property that the

D_3	E	C_3^1	C_3^2	C_2	C_2'	C_2''		D_3	E	$2C_3$	$3C_2$
E	E	C_3^1	C_3^2	C_2	C_2'	C_2''		A_1	1	1	1
C_3^1	C_3^1	C_3^2	E	C_2''	C_2	C_2'		A_2	1	1	-1
C_3^2	C_3^2	E	C_3^1	C_2'	C_2''	C_2		E	2	-1	0
C_2	C_2	C_2'	C_2''	E	C_3^1	C_3^2					
C_2'	C_2'	C_2''	C_2	C_3^2	E	C_3^1					
C_2''	C_2''	C_2	C_2'	C_3^1	C_3^2	E					

Table A.1. The D_3 multiplication table and character table, where E for unity operation and C_n^m for applying $(2\pi/n)$ -rotation m -times, and prime for C_2 along different axes. And we used Mulliken symbols to provide a shorthand notation for the description of the irreducible representations that the subscripts 1 and 2 refer to the character under C_2 rotations, A, E for one and two dimension representations, and g, u refer to the sign of character under i inversion operation.

tensor product of two representations, $(\Gamma^{(i)}(a) \otimes \Gamma^{(j)}(a))(\Gamma^{(i)}(b) \otimes \Gamma^{(j)}(b)) = (\Gamma^{(i)}(ab) \otimes \Gamma^{(j)}(ab))$, again satisfies the multiplication table and hence serves as a new representation. The character of the tensor product representation is $\text{tr}(\Gamma^{(i)}(g) \otimes \Gamma^{(j)}(g)) = \text{tr}(\Gamma^{(i)}(g))\text{tr}(\Gamma^{(j)}(g))$, so the character of the tensor product is simply the product of the characters of the two factors. This makes it convenient to use character tables to analyze tensor products and to obtain selection rules.

A.2 NV symmetry and energy structure

The negatively charged Nitrogen-Vacancy center (NV^-) in diamond consists of a nitrogen atom and a nearby vacancy. Along with the six carbon atoms which is involved in electron interaction, the total configuration results in a point-group D_3 (see [Figure 2.2](#)), makes it have permanent dipole moment and more susceptible to charge noise near surface. But on the other hand, symmetry and electron number of NV^- end up with two holes in the configuration and makes ground state a spin triplet but with same orbital part. This same orbital part makes phonon interaction decoupled from ground state and forms great spin lifetime and coherence at even room temperature.

$D_3 \otimes D_3$	A_1	A_2	E
A_1	A_1	A_2	E
A_2	A_2	A_1	E
E	E	E	$A_1 \oplus A_2 \oplus E$

Table A.2. The tensor product of two D_3 representations.

The D_3 point group has three types of symmetry operations, one is identity operation E , one is C_3^1 and C_3^2 in which C_n^m denotes apply $(2\pi/n)$ -rotation m -times along the z -axis, and another is C_2, C_2', C_2'' they are π -rotations along a axis in xy plane that is aligned with one of the $C - N$ bond. And D_3 has A_1, A_2, E irreducible representations as shown in [Table A.1](#).

Having the character table, we need to get a complete basis of the system in order to construct representation on it. One complete basis for orbital energy levels can be the orbital of surrounding carbon atoms and nitrogen atom, here we assume the linear combinations of the three carbon orbitals σ_i and one nitrogen orbital $\sigma_4 = \sigma_N$ is a good approximation to the combined system, therefore we can use state vector $\sum_i^4 a_i \times \sigma_i$ to describe and 4×4 matrices $\Gamma^{(\sigma)}(g)$ as representation of all operations in symmetry group. From [Equation A.1](#) and a construction of representation, we end up with decomposition $\Gamma^{(\sigma)} = 2A_1 + E$. If we consider NV^- defect only, then there will be 3+2+1 electrons in the defect and gives excited state and ground state electron configurations, we fill the electron according to Hund rule, and also the DFT verified absolute energy order of these states [Gali and Maze \(2013\)](#). Switch to hole representation then we have as shown in [Figure 2.2](#).

A detailed group theory description of NV center can be found in [Doherty, Manson, Delaney, and Hollenberg \(2011\)](#); [Maze et al. \(2011\)](#), but here we aim to give analysis on ground state only. Because we have two holes in the configuration, the tensor product of two holes gives new representation. In this specific case, each

hole belongs to E , and the tensor product gives $\Gamma^{(E)} \otimes \Gamma^{(E)} = \Gamma^{(A_1)} \oplus \Gamma^{(A_2)} \oplus \Gamma^{(E)}$ as shown in [Table A.2](#), and two holes symmetry

$$A_1 : e_x e_x + e_y e_y, A_2 : e_x e_y - e_y e_x,$$

$$E_x : e_x e_x - e_y e_y, E_y : e_x e_y + e_y e_x.$$

Furthermore, due to the Pauli exclusion principle, the wavefunction must be antisymmetric that means a minus sign must occur upon exchanging two electrons. This restrict the combination between orbital wavefunction and the spin wavefunction, and gives

$$^1A_1 : |e_x e_x + e_y e_y\rangle \otimes |\alpha\beta - \beta\alpha\rangle,$$

$$^1E_{x,y} : |e_x e_x - e_y e_y\rangle \otimes |\alpha\beta - \beta\alpha\rangle, |e_x e_y + e_y e_x\rangle \otimes |\alpha\beta - \beta\alpha\rangle,$$

$$^3A_2 : |e_x e_y - e_y e_x\rangle \otimes \begin{cases} |\beta\beta\rangle \\ |\alpha\beta + \beta\alpha\rangle \\ |\alpha\alpha\rangle \end{cases} \quad ,$$

where we denote spin state by $|\uparrow\rangle = |\alpha\rangle, |\downarrow\rangle = |\beta\rangle$. The Coulomb interaction term between two electrons $V(|r_1 - r_2|)$ will give the energy order of these three states, $^1A_1 > ^1E_{x,y} > ^3A_2$ from the overlap between their orbital parts in which the $|e_x e_y - e_y e_x\rangle$ gives largest orbital separation. Therefore the 3A_2 spin triplet that shares the same orbital wavefunction is the ground state of NV^- as shown in [Figure 2.3](#).

A common intuition from atomic physics is that states with the same orbital part but different S_z components are split by the spin-orbit coupling $H_{SO} = -\lambda \mathbf{L} \cdot \mathbf{S}$, where the minus sign comes from the hole representation. But for the NV^- center ground state 3A_2 , however, the orbital part is a nondegenerate

one-dimensional irrep, and within this orbital subspace all matrix elements of L_α vanish. As a result, H_{SO} does not produce a first-order splitting of the 3A_2 spin triplet. Here we use the rule $\Gamma^{(i)} \otimes \Gamma^{(H)} \otimes \Gamma^{(j)} \supset A_{1g}$ introduced before to analyze non-zero matrix element. Since we have $\Gamma^{(i)} \otimes \Gamma^{(j)} = \Gamma^{(A_2)} \otimes \Gamma^{(A_2)} = A_1$, then the H_{SO} element must also be A_1 to give non-zero transition. But H_{SO} is not a constant number but instead a operator therefore can not couple states within 3A_2 .

The zero-field splitting of the 3A_2 triplet is instead dominated by the spin-spin interaction originating from the magnetic dipole-dipole coupling between the two unpaired electrons,

$$H_{ss} = \frac{\mu_0}{4\pi} \frac{(g_e \mu_B)^2}{r^3} \left[\mathbf{S}_1 \cdot \mathbf{S}_2 - 3 \frac{(\mathbf{S}_1 \cdot \mathbf{r})(\mathbf{S}_2 \cdot \mathbf{r})}{r^2} \right],$$

with $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$. Within the ground 3A_2 triplet, the $\mathbf{r}$ dependence is averaged out as a constant, then spin-spin interaction becomes $H_{ss} = \sum_{\alpha,\beta} D_{\alpha\beta} S_{1\alpha} S_{2\beta}$. Again, we require A_1 symmetry term occurs in H_{ss} to give non zero matrix element. And the only A_1 or constant term for spin are $\mathbf{S}^2 = S_x^2 + S_y^2 + S_z^2$ or S_z^2 terms, since $\mathbf{S}^2 = S(S+1)\hbar^2 = 2\hbar^2$ is the same for all states within the triplet, then we rewrite H_{ss} as

$$H_{ss} = D S_z^2,$$

$$D = \frac{3\mu_0(g_e \mu_B)^2}{16\pi} \int |\psi_{^3A_2}(\mathbf{r}_1, \mathbf{r}_2)|^2 \frac{1 - 3 \cos^2 \theta}{|r_1 - r_2|^3} d\mathbf{r}_1 d\mathbf{r}_2,$$

where θ is the polar angle relative to z axis. This expression gives the spin-spin induced zero-field splitting $D \approx 2.87\text{GHz}$ between $|m_s = \pm 1\rangle$ and $|m_s = 0\rangle$.

A.3 SiV symmetry and energy structure

The negatively charged silicon-vacancy center (SiV⁻) in diamond consists of a silicon atom situated between two adjacent vacancies along a $\langle 111 \rangle$ axis of the diamond lattice. Because the Si-C bond in diamond is longer than the C-

C_i	E	i	C_i	E	i
E	E	i	A_g	1	1
i	i	E	A_u	1	-1

Table A.3. The C_i multiplication table and character table.

C bond, it is energetically favorable for the Si impurity to relax to this bond-centered position between the two opposite vacancies, resulting in a split-vacancy configuration with an inversion center and hence point-group symmetry D_{3d} (see [Figure 2.4](#)). In what follows we fix the nuclear coordinates (Born–Oppenheimer approximation) and, at first, neglect electron–electron and spin–orbit interactions. In this limit the electrons of the SiV^- center experience a Hamiltonian with D_{3d} symmetry.

The D_{3d} group can also be decomposed into $D_3 \times \{E, i\} = D_3 \times C_i \cong D_3 \times C_2$, which is the D_3 group we discussed for NV with an additional operation inversion i that comes from silicon atom situated in the middle. The inversion operation i transforms (x, y, z) vector into $(-x, -y, -z)$. The group C_i multiplication table and character table can be easily derived as [Table A.3](#). Since every operation in D_{3d} is multiplication of one from D_3 and one from C_i . Then we can again use the tensor product of representation of D_3 and C_i as the representation of D_{3d} . And then the character table of D_{3d} group can be obtained from the product of two character table, then we have [Table A.4](#).

Following a similar procedure as NV, but this time the basis is six orbitals σ_i from six carbon atoms, we end up with $\Gamma^{(\sigma)} = A_{1g} + E_g + A_{2u} + E_u$. If we consider SiV^- defect only, then there will be 6+4+1 electrons in the defect and gives excited state and ground state electron configurations, we fill the electron according to Hund rule, and also the DFT verified absolute energy order of these states [Gali and Maze \(2013\)](#). Switch to hole representation then we have as shown in [Figure 2.4](#).

D_{3d}	E	$2C_3$	$3C_2$	i	$2S_6 = 2C_3 \times i$	$3\sigma = 3C_2 \times i$	polynomial
A_{1g}	1	1	1	1	1	1	$x^2 + y^2, z^2$
A_{2g}	1	1	-1	1	1	-1	$R_z = \mathbf{x} \times \mathbf{y}$
E_g	2	-1	0	2	-1	0	$(x^2 - y^2, xy), (xz, yz)$
A_{1u}	1	1	1	-1	-1	-1	
A_{2u}	1	1	-1	-1	-1	1	z
E_u	2	-1	0	-2	1	0	(x, y)

Table A.4. The D_{3d} character table. Polynomial lists the polynomials that transforms as corresponding irreducible representation.

Coulomb and Spin-Orbital interactions can be now introduced as perturbation. We use the rule $\Gamma^{(i)} \otimes \Gamma^{(H)} \otimes \Gamma^{(j)} \supset A_{1g}$ introduced before to give non-zero matrix element. Coulomb interaction is invariant under symmetry operations as it preserves distance, so no extra coupling is introduced. And the SO interaction writes

$$H_{\text{SO}} = -\lambda_{\text{SO}} \mathbf{L} \cdot \mathbf{S}, \quad (\text{A.2})$$

where the minus sign comes from the hole representation.

We can prove that L_x, L_y terms vanish although they are allowed to be non-zero by group theory. The first step is to prove spherical harmonics $Y_l^{\pm m}$ span the $E_{g/u}$ space that $e_{x/y,g/u}$ eigenstates span. A C_3 operation transforms $Y_l^m \mapsto e^{im2\pi/3} Y_l^m$, and C_2 along x -axis gives $Y_l^m \mapsto (-1)^l Y_l^{-m}$, and i operation gives $Y_l^m \mapsto (-1)^l Y_l^m$. Then their representation is equivalent to irreducible representation $E_{g/u}$ given $m = \pm 1 \pmod{3}$ and l is even/odd for $E_{g/u}$. And since Y_l^m is complete in the space of electron orbital, then $e_{x/y,g/u}$ are linear combinations of $Y_l^{\pm m}$ where we ignore the radial part $R_{nl}(r)$. We know that $L_{\pm} = L_x \pm iL_y$ change the m number of Y_l^m , therefore $L_{x/y}$ do not couple a $E_{g/u}$ state to another. Eventually, SO interaction becomes $H_{\text{SO}} = -\lambda_{\text{SO}} L_z S_z$, and corresponding

$\frac{D_{3d}}{\chi^{(3N)}}$	E	$2C_3$	$3C_2$	i	$2S_6 = 2C_3 \times i$	$3\sigma = 3C_2 \times i$
	21	0	-1	-3	0	3

Table A.5. The vibration $3N$ representation.

eigenstates

$$\begin{aligned}
-\frac{\lambda_{SO}}{2} &\leftarrow \begin{cases} |e_- \uparrow\rangle = |e_x - ie_y\rangle \otimes |\uparrow\rangle \\ |e_+ \downarrow\rangle = |e_x + ie_y\rangle \otimes |\downarrow\rangle \end{cases}, \\
+\frac{\lambda_{SO}}{2} &\leftarrow \begin{cases} |e_- \downarrow\rangle = |e_x - ie_y\rangle \otimes |\downarrow\rangle \\ |e_+ \uparrow\rangle = |e_x + ie_y\rangle \otimes |\uparrow\rangle \end{cases}.
\end{aligned} \tag{A.3}$$

This SO interaction mixes the orbital state, lifts the degeneracy of the ground and excited manifold even without the external magnetic field or strain.

A.4 JT, strain and magnetic field

When lattice vibrations (phonons) are present, the nuclei are displaced and the electronic orbitals change. To treat this in a systematic way we again use group theory, this time starting from the $3N$ nuclear coordinates of the six carbon atoms and the silicon atom [Hepp \(2014\)](#), the vibrational representation character table for these seven atoms is [Table A.5](#), and following the same procedure as treating orbital, we get $\Gamma^{(3N)} = 2A_{1g} + A_{2g} + 3E_g + A_{1u} + 3A_{2u} + 4E_u$, and subtract 3 translational and 3 rotational degrees of freedom of the whole defect which does not affect energy structure inside, we end up with $\Gamma^{(3N)} = 2A_{1g} + 2E_g + A_{1u} + 2A_{2u} + 3E_u$. While the strain displaces nuclears, this term does not directly couple to electron, only after we apply the BO approximation and get effective electron Hamiltonian

$$H_{el}(\mathbf{R}) = H_{el}(\mathbf{R}_0) + \sum_i \left(\frac{\partial H_{el}}{\partial Q_{\Gamma i}} \right)_{Q_{\Gamma i}=0} Q_{\Gamma i}, \tag{A.4}$$

where $Q_{\Gamma i}$ are i -th general coordinates of vibration of irreducible representation Γ and $\mathbf{R}_0$ is the nuclear configuration at D_{3d} . And by using $U_{el}(g)H_{el}(\{Q_{\Gamma i}\})U_{el}^\dagger(g) =$

$H_{el}(\{Q'_{\Gamma i}\})$, $U_{\text{nuclear}}(g)\{Q_{\Gamma i}\} = \{Q'_{\Gamma i}\}$ and derivative both sides by $Q_{\Gamma i}$, we can prove $(\partial H_{el}/\partial Q_{\Gamma i})_0$ transforms as $Q_{\Gamma i}$. In order to have vibration couple to orbital state, we again need to have $\langle \psi_i | (\partial H_{el}/\partial Q_{\Gamma i})_0 | \psi_j \rangle \sim \Gamma^{(E_{g/u})} \otimes \Gamma^{(Q)} \otimes \Gamma^{(E_{g/u})} \supset A_{1g}$, therefore we can rule out many irrelevant vibrations except $Q_{\Gamma i}$ of A_{1g}, E_g types. Since $(\partial H_{el}/\partial Q_{\Gamma i})_0$ is a 2×2 matrix with $\{e_x, e_y\}$ basis, and all matrices are linear combinations of $\{I_2, \sigma_x, \sigma_y, \sigma_z\}$, I_2 is A_{1g} and $\sigma_y = i(|e_x\rangle \langle e_y| - |e_y\rangle \langle e_x|)$ flips sign under C_2 operation which swith e_x, e_y then σ_y is A_{2g} , and also can show $\{\sigma_x, \sigma_z\}$ is E_g . Then rewrite the vibration coupling term as

$$\begin{aligned} H_{\text{strain}} &= \sum_i \left(\frac{\partial H_{el}}{\partial Q_{\Gamma i}} \right)_{Q_{\Gamma i}=0} Q_{\Gamma i} \\ &= a_{A_{1g}} Q_{A_{1g}} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + a_{E_g} Q_{E_{gx}} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} + a_{E_g} Q_{E_{gy}} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (\text{A.5}) \end{aligned}$$

in $\{e_x, e_y\}$ basis. One thing suprisingly is that when considering vibronic potential $K(Q_{E_{gx}}^2 + Q_{E_{gy}}^2)/2$, the eigenvalue of such interaction gives ground state energy $-a_{E_g}^2/2K$ at $\sqrt{Q_{E_{gx}}^2 + Q_{E_{gy}}^2} = a_{E_g}/K$ which means ground state shift from origin to a circle centered origin under the coupling to vibration, and this is called Jahn-Teller effect. And when external strain is small, ground state can move along this circle and causing an average effect on azimuthal angle, this will lead to the quenching factor dicussed later, or also called Ham reduction or dynamic Jahn-Teller effect.

In regard of energy structure, strain and JT can not be distinguished. But often time, people prefer to rewrite this strain interaction Hamiltonian in terms of strain tensor,

$$\varepsilon_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right), \quad (\text{A.6})$$

where $\mathbf{u}(\mathbf{x})$ is a displacement field. The result of Hepp (2014); Meesala et al. (2018) are different, here we provide a independent proof to verify. Since ε is a tensor, it transforms as $\varepsilon' = \Gamma \varepsilon \Gamma^{-1}$, and ε_{ij} transforms as $x_i x_j$, where Γ is the symmetry operation/representation under basis (x, y, z) , hence $\Gamma = E_u + A_{2u}$. Therefore $\Gamma \otimes \Gamma$ is representation under basis ε , and also ε is symmetric then $\chi_\varepsilon(g) = (\chi^{(\Gamma)}(g)^2 + \chi^{(\Gamma)}(g^2))/2$, from which we can get $\Gamma_\varepsilon = 2A_{1g} + 2E_g$. The most natural polynomials are $x^2 + y^2, z^2$ for A_{1g} and $(x^2 - y^2, xy), (xz, yz)$ for E_g which gives Meesala et al. (2018) results instantly. So we inherite their notation (with a different σ_z) and

$$\begin{aligned}
H_{\text{strain}} &= \begin{pmatrix} \alpha + \beta & \gamma \\ \gamma & \alpha - \beta \end{pmatrix}, \\
\alpha &= t_\perp(\varepsilon_{xx} + \varepsilon_{yy}) + t_\parallel \varepsilon_{zz}, \\
\beta &= d(\varepsilon_{yy} - \varepsilon_{xx}) - f \varepsilon_{zx}, \\
\gamma &= -2d\varepsilon_{xy} + f \varepsilon_{yz}.
\end{aligned} \tag{A.7}$$

Here we used the local coordinates x, y, z Figure 2.4 of SiV instead of X, Y, Z in lab coordinates Figure 2.1. They can be transformed by expressing x, y, z as $x = (-1, 1, 0)$, $y = (-1, -1, 2)$, and $z = (1, 1, 1)$. Another common external field left is the magnetic field, there are both orbital Zeeman and spin Zeeman terms, but the orbital part is affecting by the dynamic JT effect therefore a quenching factor q is introduced to reduct the standard interaction term, the whole interaction Hamiltonian reads

$$H_{\text{Zeeman}} = q\gamma_L \mathbf{L} \cdot \mathbf{B} + \gamma_S \mathbf{S} \cdot \mathbf{B} = q\gamma_L L_z B_z + \gamma_S \mathbf{S} \cdot \mathbf{B}. \tag{A.8}$$

A complete energy structure is now ready be solved numerically. But the coupling to light is also crucial for experiment, we will briefly talk about the selection rules here. The interaction term is the electric dipole interacts with

electric field of light, again we flip the sign due to hole representation

$$H_{\text{dipole}} = \mathbf{p} \cdot \mathcal{E} = e(x\mathcal{E}_x + y\mathcal{E}_y + z\mathcal{E}_z) = p_0\mathcal{E}_0 + p_-\mathcal{E}_- + p_+\mathcal{E}_+,$$

with $\mathcal{E}$ the electric field of light, and $0, \pm 1$ for linear z -polarization and circular polarizations. By estimating if $\langle \psi_i | H_{\text{dipole}} | \psi_j \rangle \sim A_{1g}$, we are also determining if symmetry operation g changes $\langle \psi_i | H_{\text{dipole}} | \psi_j \rangle$, start from operation C_3 under the basis state with SO coupling

$$\begin{aligned} C_3 (\langle e_{gf} | p_q | e_{ui} \rangle) &= \langle e_{gf} | p_q | e_{ui} \rangle = \langle e_{gf} | C_3^{-1} C_3 p_q C_3^{-1} C_3 | e_{ui} \rangle \\ &= \langle e_{gf} | \omega^f \omega^q \omega^{-i} p_q | e_{ui} \rangle \\ &\rightarrow f = -q + i \end{aligned} \tag{A.9}$$

with $\omega = e^{i2\pi/3}$. We can also confirm that other symmetry operations do not add extra conditions, therefore end up with the selection rules

polarization	$ e_{g+}\rangle$	$ e_{g-}\rangle$
$\langle e_{u+} $	π	σ_-
$\langle e_{u-} $	σ_+	π

, which predicts the polarization of corresponding transitions as shown in [Hepp \(2014\)](#).

APPENDIX B

SOME USEFUL FORMULAS

B.1 Quantum optics

B.1.1 Heisenberg picture and rotating frame

The first principle of quantum mechanics is the Schrödinger equation
(setting $\hbar = 1$)

$$i \frac{d}{dt} |\psi(t)\rangle = H |\psi(t)\rangle,$$

from which we define the density matrix

$$\rho(t) = |\psi(t)\rangle \langle \psi(t)|.$$

Taking the time derivative, we obtain

$$\begin{aligned} \dot{\rho}(t) &= |\dot{\psi}(t)\rangle \langle \psi(t)| + |\psi(t)\rangle \langle \dot{\psi}(t)| \\ &= (-iH |\psi(t)\rangle) \langle \psi(t)| + |\psi(t)\rangle (\langle \psi(t)| iH) \\ &= -i [H, \rho(t)], \end{aligned}$$

which is useful in many cases. This description is called the Schrödinger picture, where the state evolves in time while operators are time independent.

In many situations it is more convenient to work in a picture where the state is kept fixed but operators evolve in time. This leaves all physical observables unchanged, but often simplifies the calculation. This is the Heisenberg picture. It is obtained by applying a unitary transformation $U(t) = e^{iHt}$ to the system such that

$$|\psi\rangle_H = |\psi(0)\rangle = U(t) |\psi(t)\rangle = U(t) |\psi\rangle_S,$$

and

$$O_H(t) = U(t) O_S U^\dagger(t),$$

so that the expectation value of an operator O satisfies

$$\langle \psi |_S O_S |\psi \rangle_S = \langle \psi |_H O_H(t) |\psi \rangle_H.$$

Taking the time derivative of $O_H(t)$, we obtain

$$\begin{aligned} \dot{O}_H(t) &= \dot{U}(t) O_S U^\dagger(t) + U(t) \dot{O}_S(t) U^\dagger(t) + U(t) O_S \dot{U}^\dagger(t) \\ &= i [H, O_H(t)] + U(t) \dot{O}_S(t) U^\dagger(t). \end{aligned}$$

For time-independent Schrödinger-picture operators ($\dot{O}_S = 0$), this reduces to the familiar Heisenberg equation of motion $\dot{O}_H(t) = i[H, O_H(t)]$.

From the equations of motion of operators, we can take the expectation value on both sides. If the equations are linear in the operators, the resulting equations for the corresponding expectation values (complex numbers) are closed and can be solved directly. In this thesis we use ρ_{ij} for a purely atomic system, but when there is coupling between the atom and other systems, it is often convenient to introduce the operators $\sigma_{ji} = |j\rangle \langle i|$, so that $\dot{\rho}_{ij}(t) = \text{Tr}(\dot{\rho}(t) |j\rangle \langle i|) = \langle i | \dot{\rho}(t) | j \rangle$ can be interpreted as the equation of motion for the expectation value of σ_{ji} .

More generally, we can apply a time-dependent unitary transformation $U(t)$ different from e^{iHt} , this is often called going to a rotating frame. For example, consider a time-dependent Hamiltonian (already in the rotating-wave approximation)

$$H(t) = \omega_a a^\dagger a + \Omega (a e^{i\omega_\Omega t} + a^\dagger e^{-i\omega_\Omega t}).$$

It is in general hard to solve the Schrödinger equation directly, since one would need to evaluate

$$|\psi(t)\rangle = \mathcal{T} \exp \left(-i \int_0^t H(t') dt' \right) |\psi(0)\rangle,$$

where $\mathcal{T}$ denotes time ordering.

We now introduce a unitary transformation $U(t) = e^{i\omega_\Omega a^\dagger t}$, and define the transformed state $|\tilde{\psi}(t)\rangle = U(t) |\psi(t)\rangle$. The new state satisfies

$$i \frac{d}{dt} |\tilde{\psi}(t)\rangle = i\dot{U}(t) |\psi(t)\rangle + iU(t) |\dot{\psi}(t)\rangle = \tilde{H}(t) |\tilde{\psi}(t)\rangle, \quad (\text{B.1})$$

the transformed (rotating-frame) Hamiltonian is

$$\tilde{H}(t) = U(t)H(t)U^\dagger(t) + i\dot{U}(t)U^\dagger(t).$$

Using the Baker–Campbell–Hausdorff formula

$$e^A B e^{-A} = \sum_{n=0}^{\infty} \frac{1}{n!} \underbrace{[A, [A, \dots [A, B] \dots]]}_{n \text{ times}},$$

we obtain

$$U(t)aU^\dagger(t) = ae^{-i\omega_\Omega t}, U(t)a^\dagger U^\dagger(t) = a^\dagger e^{i\omega_\Omega t},$$

and

$$i\dot{U}(t)U^\dagger(t) = -\omega_\Omega a^\dagger a.$$

Substituting into the expression for $\tilde{H}(t)$, we obtain an effective time-independent Hamiltonian

$$\tilde{H} = (\omega_a - \omega_\Omega) a^\dagger a + \Omega (a + a^\dagger).$$

The state in the rotating frame differs from the original Schrödinger-picture state only by a unitary phase rotation generated by $a^\dagger a$. For a more general case that $U = e^{iAt}$, we have

$$\tilde{H}(t) = U H U^\dagger - A.$$

B.1.2 Master equation

For a closed quantum system, the time evolution of the density matrix is given by the von Neumann equation

$$\dot{\rho}(t) = -i[H, \rho(t)].$$

For an open quantum system, this can be generalized to the Lindblad equation

$$\dot{\rho} = \mathcal{L}\rho = -i[H, \rho] + \sum_{\alpha} \Gamma_{\alpha} \mathcal{D}(a_{\alpha})\rho,$$

where the Lindblad (dissipator) superoperator is $\mathcal{D}(a)\rho = a\rho a^{\dagger} - \frac{1}{2}(a^{\dagger}a\rho + \rho a^{\dagger}a)$.

We now show how such a Lindblad form arises microscopically under standard approximations. We assume a total Hamiltonian

$$H = H_S + H_B + H_{SB},$$

where H_S is the system Hamiltonian, H_B is the bath (environment) Hamiltonian, and H_{SB} describes the system–bath coupling. The bath Hamiltonian is taken to be

$$H_B = \sum_{|\alpha|} \omega_{\alpha} b_{\alpha}^{\dagger} b_{\alpha} = \sum_{|\alpha|} \omega_{\alpha} b_{-\alpha} b_{\alpha} = \int \omega b_{-\omega} b_{\omega} d\omega,$$

where b_{ω} are bath annihilation operators with $b_{-\omega} = b_{\omega}^{\dagger}$, and we have $b_{\omega} \sim 1/\sqrt{\omega}$ in units.

Very generally, we can write the system Hamiltonian in its eigenbasis as $H_S = \sum_n E_n |n\rangle \langle n|$, so that the natural system operators are the “ladder” operators $S_{mn} = |m_S\rangle \langle n_S|$. We assume a system–bath interaction of the (linear) form

$$H_{SB} = \sum_{m,n} g_{mn} S_{mn} \otimes \int g(\omega) b_{\omega} d\omega,$$

where g_{mn} and $g(\omega)$ are coupling coefficients and we use the notation $b_{-\omega} = b_{\omega}^{\dagger}$.

In the interaction picture with respect to $H_S + H_B$, the operators evolve as

$$S_{mn}(t) = e^{iH_S t} S_{mn} e^{-iH_S t} = S_{mn} e^{-i(E_n - E_m)t},$$

$$b_{\omega}(t) = e^{iH_B t} b_{\omega} e^{-iH_B t} = b_{\omega} e^{-i\omega t}.$$

We reorder terms according to their Bohr frequencies. Defining

$$s_{\alpha} = \sum_{E_n - E_m = \omega_{\alpha}} g_{mn} S_{mn}, B = \int g(\omega) b_{\omega} d\omega,$$

we can write the interaction Hamiltonian as

$$H_{SB} = \sum_{\alpha} s_{\alpha} \otimes B,$$

and in the interaction picture

$$s_{\alpha}(t) = s_{\alpha} e^{-i\omega_{\alpha} t}, B(t) = \int g(\omega) b_{\omega}(t) d\omega.$$

We now invoke the Born approximation, assuming a weak system–bath coupling such that the total density matrix approximately factorizes as $\rho(t) \approx \rho_S(t) \otimes \rho_B$, where $\rho_S(t)$ is the reduced density matrix of the system and ρ_B is the stationary (thermal) state of the environment at equilibrium, $\rho_B = \sum_n p_n |n\rangle \langle n|$. In particular, $\text{Tr}_B(\rho_B b_{\omega}) = \langle b_{\omega} \rangle = 0$, and similarly for products such as $b_{\omega} b_{\omega'}$ that do not conserve excitation number in the thermal equilibrium state. The bath correlation functions are then

$$\text{Tr}_B(\rho_B b_{\omega}(t) b_{\omega'}(t')) = \text{Tr}_B(b_{\omega'}(t') \rho_B b_{\omega}(t)) = \text{Tr}_B(b_{\omega}(t) b_{\omega'}(t') \rho_B),$$

which gives

$$\text{Tr}_B(\rho_B b_{\omega}(t) b_{\omega'}(t')) = \langle b_{\omega}(0) e^{-i\omega t} b_{\omega'}(0) e^{-i\omega' t'} \rangle = \delta(\omega + \omega') \langle b_{\omega}(0) b_{-\omega}(0) \rangle e^{-i\omega(t-t')}.$$

Therefore, for the collective bath operator $B(t)$ we obtain

$$\begin{aligned} \langle B(t) B(t') \rangle &= \int g(\omega) g(\omega') \delta(\omega + \omega') \langle b_{\omega} b_{\omega'} \rangle e^{-i\omega(t-t')} d\omega d\omega' \\ &= \int g(\omega) g(-\omega) \langle b_{\omega} b_{-\omega} \rangle e^{-i\omega(t-t')} d\omega \\ &= \int C_{\omega} e^{-i\omega(t-t')} d\omega, \end{aligned}$$

where we have introduced the bath spectral correlation function C_{ω} . For the system operators we have

$$s_{\alpha}(t) s_{\beta}(t') = s_{\alpha}(0) s_{\beta}(0) e^{-i\omega_{\alpha} t - i\omega_{\beta} t'}.$$

In the interaction picture with respect to $H_S + H_B$, the total density matrix $\rho(t)$ evolves according to

$$\dot{\rho}(t) = -i[H_{SB}(t), \rho(t)].$$

Integrating once,

$$\rho(t) = \rho(0) - i \int_0^t [H_{SB}(t'), \rho(t')] dt'.$$

Substituting this back into the right-hand side gives, to second order in the interaction,

$$\rho(t) = \rho(0) - i \int_0^t [H_{SB}(t'), \rho(0)] dt' - \int_0^t \int_0^{t'} [H_{SB}(t'), [H_{SB}(t''), \rho(t'')]] dt'' dt'.$$

We now trace over the bath degrees of freedom, Tr_B . The first-order term vanishes because $\text{Tr}_B(\rho_B B) = \langle B \rangle = 0$. Thus,

$$\text{Tr}_B(\rho(t) - \rho(0)) = - \int_0^t \int_0^{t'} \text{Tr}_B([H_{SB}(t'), [H_{SB}(t''), \rho(t'')]]) dt'' dt'.$$

Taking the time derivative and using $\frac{d}{dt} \int_0^t f(t') dt' = f(t)$, we obtain an exact equation for the reduced density matrix,

$$\dot{\rho}_S(t) = - \int_0^t \text{Tr}_B([H_{SB}(t), [H_{SB}(t''), \rho(t'')]]) dt''.$$

Using the decomposition $H_{SB}(t) = \sum_{\alpha} s_{\alpha}(t) B(t)$, we can write

$$\begin{aligned} \dot{\rho}_S(t) &= - \int_0^t \text{Tr}_B \left(\left[\sum_{\alpha} s_{\alpha}(t) B(t), \left[\sum_{\beta} s_{\beta}(t'') B(t''), \rho(t'') \right] \right] \right) dt'' \\ &= - \sum_{\alpha, \beta} \int_0^t [(s_{\alpha} s_{\beta} \rho_S(t'') - s_{\beta} \rho_S(t'') s_{\alpha}) \langle B(t) B(t'') \rangle \\ &\quad + (\rho_S(t'') s_{\beta} s_{\alpha} - s_{\alpha} \rho_S(t'') s_{\beta}) \langle B(t'') B(t) \rangle] dt'', \end{aligned}$$

where we have used the Born approximation $\rho(t'') \approx \rho_S(t'') \otimes \rho_B$ and the definition of the bath correlation functions.

Next we make the Markov approximation, assuming that the system dynamics at time t depend only weakly on its history, so we replace $\rho_S(t'')$ by $\rho_S(t)$

inside the time integral. Focusing on the term

$$-\sum_{\alpha,\beta} \int_0^t (s_\alpha(t)s_\beta(t'')\rho_S(t)) \langle B(t)B(t'') \rangle dt'',$$

we substitute

$$s_\alpha(t)s_\beta(t'') = s_\alpha(0)s_\beta(0)e^{-i\omega_\alpha t}e^{-i\omega_\beta t''}, \langle B(t)B(t'') \rangle = \int C_\omega e^{-i\omega(t-t'')}d\omega,$$

to obtain

$$-\sum_{\alpha,\beta} \int_0^t s_\alpha(0)s_\beta(0)e^{-i\omega_\alpha t}\rho_S(t) \int C_\omega e^{-i\omega t}e^{i(\omega-\omega_\beta)t''}d\omega dt''.$$

Because the bath correlations decay on a time scale much shorter than the system dynamics, we can extend the upper limit of the integral over t'' to infinity,

$$\int_0^t e^{i(\omega-\omega_\beta)t''} dt'' \approx \int_0^\infty e^{i(\omega-\omega_\beta)t''} dt'' = \pi\delta(\omega - \omega_\beta) + i\mathcal{P}\frac{1}{\omega - \omega_\beta},$$

where $\mathcal{P}$ denotes the Cauchy principal value. This leads to

$$\begin{aligned} & -\sum_{\alpha,\beta} \int_0^t (s_\alpha(t)s_\beta(t'')\rho_S(t)) \langle B(t)B(t'') \rangle dt'' \\ &= -\sum_{\alpha,\beta} (\pi C_{\omega_\beta} + i\Delta_{\omega_\beta}) s_\alpha(0)s_\beta(0)e^{-i(\omega_\alpha+\omega_\beta)t}\rho_S(t), \end{aligned}$$

where $\Delta_{\omega_\beta} = C_{\omega_\beta} \int \mathcal{P}\frac{1}{\omega-\omega_\beta}d\omega$ is the (frequency-dependent) Lamb-shift contribution.

An analogous expression arises from the term with $\langle B(t'')B(t) \rangle$.

Collecting all terms, we obtain

$$\begin{aligned} \dot{\rho}_S(t) &= -\sum_{\alpha} [(s_\alpha s_{-\alpha}\rho_S(t) - s_{-\alpha}\rho_S(t)s_\alpha) (\pi C_{-\omega_\alpha} + i\Delta_{-\omega_\alpha}) \\ &\quad + (\rho_S(t)s_{-\alpha}s_\alpha - s_\alpha\rho_S(t)s_{-\alpha}) (\pi C_{\omega_\alpha} - i\Delta_{\omega_\alpha})] \end{aligned}$$

where, in the last step, we have applied the rotating-wave approximation and kept only terms with $\beta = -\alpha$, i.e. terms that do not oscillate rapidly as $e^{-i(\omega_\alpha+\omega_\beta)t}$.

Using the relation $s_{-\alpha} = s_\alpha^\dagger$ that follows from the definition of Bohr frequencies,

and rearranging the terms in $\alpha, -\alpha$ pairs, the master equation becomes

$$\dot{\rho}_S(t) = \sum_{\alpha} \left[\pi C_{\omega_{\alpha}} (2s_{\alpha}\rho_S(t)s_{\alpha}^{\dagger} - s_{\alpha}^{\dagger}s_{\alpha}\rho_S(t) - \rho_S(t)s_{\alpha}^{\dagger}s_{\alpha}) - i [\Delta_{\omega_{\alpha}}s_{\alpha}^{\dagger}s_{\alpha}, \rho_S(t)] \right].$$

This is of the standard Lindblad form,

$$\dot{\rho}_S(t) = \sum_{\alpha} \left(\frac{\pi C_{\omega_{\alpha}}}{2} \mathcal{D}(s_{\alpha})\rho_S(t) - i \sum_{\alpha} [\Delta_{\omega_{\alpha}}s_{\alpha}^{\dagger}s_{\alpha}, \rho_S(t)] \right),$$

with a Lamb-shift Hamiltonian contribution that shifts the energy levels. From the derivation we see that each Lindblad operator corresponds to the sum of all ladder operators with a given Bohr energy difference, and different Lindblad operators are associated with different Bohr frequencies.

B.1.3 Solution of a two-level system

In this subsection we derive the steady state solution in a two-level system.

The ground states is denoted by $|g\rangle$ and is coupled to the excited state $|e\rangle$ by a coherent optical fields with Rabi frequencies Ω . In the rotating frame, the Hamiltonian can be written as

$$H = \Delta|e\rangle\langle e| + \frac{\Omega}{2}(|g\rangle\langle e| + |e\rangle\langle g|),$$

where Δ is the detunings between optical field frequency and transition frequency.

The system dynamics are described by a master equation of Lindblad form,

$$\partial_t \rho = -i[H, \rho] + \Gamma \mathcal{D}(|g\rangle\langle e|)\rho + \frac{\gamma}{2} \mathcal{D}(|e\rangle\langle e| - |g\rangle\langle g|)\rho,$$

where Γ is the spontaneous decay rates from $|e\rangle$ to $|g\rangle$, and γ is the pure dephasing.

Explicitly expanding the master equation and using $\dot{\rho}_{ij} = \text{Tr}(\dot{\rho}|j\rangle\langle i|) = \langle i|\dot{\rho}|j\rangle$, we obtain the following equations of motion for the relevant matrix elements:

$$\begin{aligned} \dot{\rho}_{ee} &= i\frac{\Omega}{2}(\rho_{eg} - \rho_{ge}) - \Gamma\rho_{ee}, \\ \dot{\rho}_{gg} &= i\frac{\Omega}{2}(\rho_{ge} - \rho_{eg}) + \Gamma\rho_{ee}, \end{aligned}$$

$$\dot{\rho}_{eg} = -\left(\frac{\Gamma}{2} + \gamma + i\Delta\right)\rho_{eg} + i\frac{\Omega}{2}(\rho_{ee} - \rho_{gg}).$$

We are interested in the steady-state response that gives to the Lorentzian curve, power broadening etc. Therefore we set all time derivatives to zero, $\dot{\rho} = 0$, which leads to

$$\begin{aligned}\rho_{ee} &= i\frac{\Omega}{2\Gamma}(\rho_{eg} - \rho_{ge}), \\ \rho_{eg} &= \frac{i\Omega}{2\left(\frac{\Gamma}{2} + \gamma + i\Delta\right)}(2\rho_{ee} - 1).\end{aligned}$$

Solve these two equations together, we end up with the steady-state solution of excited state population ρ_{ee} that

$$\rho_{ee} = \frac{1}{2 + \frac{\Gamma^2 + 2\Gamma\gamma}{\Omega^2}} \frac{1}{1 + \left(\frac{\Delta}{\sqrt{\left(\frac{1}{4} + \frac{\gamma}{2\Gamma}\right)(2\Omega^2 + \Gamma^2 + 2\Gamma\gamma)}}\right)^2}, \quad (\text{B.2})$$

from which we conclude that in the absence of pure dephasing γ , the transition is a Lorentzian curve with power broadened fullwidth $\sqrt{\Gamma^2 + 2\Omega^2}$ and height $1/(2 + \Gamma^2/\Omega^2)$.

B.1.4 Solution of a three-level system

In this subsection we derive an approximate analytical expression for the CPT dip in a three-level Λ system. The ground states are denoted by $|g\rangle$ and $|s\rangle$ and are coupled to the excited state $|e\rangle$ by two coherent optical fields with Rabi frequencies Ω_1 and Ω_2 , respectively. In the rotating frame, the Hamiltonian can be written as

$$H = \Delta_1|g\rangle\langle g| + \Delta_2|s\rangle\langle s| + \frac{\Omega_1}{2}(|g\rangle\langle e| + |e\rangle\langle g|) + \frac{\Omega_2}{2}(|s\rangle\langle e| + |e\rangle\langle s|),$$

where Δ_1 and Δ_2 are the single-photon detunings of the two optical fields. The system dynamics are described by a master equation of Lindblad form,

$$\partial_t \rho = -i[H, \rho] + \Gamma_1 \mathcal{D}(|g\rangle\langle e|)\rho + \Gamma_2 \mathcal{D}(|s\rangle\langle e|)\rho + \frac{\gamma}{2} \mathcal{D}(|s\rangle\langle s| - |g\rangle\langle g|)\rho,$$

where Γ_1 and Γ_2 are the spontaneous decay rates from $|e\rangle$ to $|g\rangle$ and $|s\rangle$, respectively, and γ is the full width of the spin-state dephasing. Explicitly expanding the master equation yields

$$\begin{aligned}\dot{\rho} = & \left(-i\Delta_1|g\rangle\langle g|\rho - i\Delta_2|s\rangle\langle s|\rho - i\frac{\Omega_1}{2}(|g\rangle\langle e|\rho + |e\rangle\langle g|\rho) - i\frac{\Omega_2}{2}(|s\rangle\langle e|\rho + |e\rangle\langle s|\rho) \right) \\ & + \left(i\Delta_1\rho|g\rangle\langle g| + i\Delta_2\rho|s\rangle\langle s| + i\frac{\Omega_1}{2}(\rho|g\rangle\langle e| + \rho|e\rangle\langle g|) + i\frac{\Omega_2}{2}(\rho|s\rangle\langle e| + \rho|e\rangle\langle s|) \right) \\ & + \Gamma_1 \left(|g\rangle\langle e|\rho|e\rangle\langle g| - \frac{1}{2}|e\rangle\langle e|\rho - \frac{1}{2}\rho|e\rangle\langle e| \right) \\ & + \Gamma_2 \left(|s\rangle\langle e|\rho|e\rangle\langle s| - \frac{1}{2}|e\rangle\langle e|\rho - \frac{1}{2}\rho|e\rangle\langle e| \right) \\ & + \frac{\gamma}{2} \left((|s\rangle\langle s| - |g\rangle\langle g|)\rho(|s\rangle\langle s| - |g\rangle\langle g|) - \frac{1}{2}(|s\rangle\langle s| + |g\rangle\langle g|)\rho - \frac{1}{2}\rho(|s\rangle\langle s| + |g\rangle\langle g|) \right).\end{aligned}$$

Using $\dot{\rho}_{ij} = \text{Tr}(\dot{\rho}|j\rangle\langle i|) = \langle i|\dot{\rho}|j\rangle$, and define $\Delta = \Delta_2 - \Delta_1$, $\Gamma = \Gamma_1 + \Gamma_2$, we obtain the following equations of motion for the relevant matrix elements:

$$\begin{aligned}\dot{\rho}_{ee} &= i\frac{\Omega_1}{2}(\rho_{eg} - \rho_{ge}) + i\frac{\Omega_2}{2}(\rho_{es} - \rho_{se}) - \Gamma\rho_{ee}, \\ \dot{\rho}_{gg} &= i\frac{\Omega_1}{2}(\rho_{ge} - \rho_{eg}) + \Gamma_1\rho_{ee}, \\ \dot{\rho}_{eg} &= -\left(\frac{\Gamma}{2} + \frac{\gamma}{4} - i\Delta_1\right)\rho_{eg} + i\frac{\Omega_1}{2}(\rho_{ee} - \rho_{gg}) - i\frac{\Omega_2}{2}\rho_{sg}, \\ \dot{\rho}_{es} &= -\left(\frac{\Gamma}{2} + \frac{\gamma}{4} - i\Delta_2\right)\rho_{es} + i\frac{\Omega_2}{2}(\rho_{ee} - \rho_{ss}) - i\frac{\Omega_1}{2}\rho_{gs}, \\ \dot{\rho}_{sg} &= -(\gamma + i\Delta)\rho_{sg} - i\frac{\Omega_2}{2}\rho_{eg} + i\frac{\Omega_1}{2}\rho_{se}.\end{aligned}$$

We are interested in the steady-state response that gives rise to the CPT dip in fluorescence. Therefore we set all time derivatives to zero, $\dot{\rho} = 0$, which leads to

$$\begin{aligned}\rho_{eg} &= \frac{i\frac{\Omega_1}{2}(\rho_{ee} - \rho_{gg}) - i\frac{\Omega_2}{2}\rho_{sg}}{\frac{\Gamma}{2} + \frac{\gamma}{4} - i\Delta_1}, \\ \rho_{es} &= \frac{i\frac{\Omega_2}{2}(\rho_{ee} - \rho_{ss}) - i\frac{\Omega_1}{2}\rho_{gs}}{\frac{\Gamma}{2} + \frac{\gamma}{4} - i\Delta_2},\end{aligned}$$

$$\rho_{ee} = \frac{i\frac{\Omega_1}{2}(\rho_{eg} - \rho_{ge}) + i\frac{\Omega_2}{2}(\rho_{es} - \rho_{se})}{\Gamma}, \quad (\text{B.3})$$

$$\rho_{ee} = -\frac{i\frac{\Omega_1}{2}(\rho_{ge} - \rho_{eg})}{\Gamma_1}, \quad (\text{B.4})$$

$$\rho_{ee} = -\frac{i\frac{\Omega_2}{2}(\rho_{se} - \rho_{es})}{\Gamma_2}, \quad (\text{B.5})$$

$$\rho_{sg} = \frac{-i\frac{\Omega_2}{2}\rho_{eg} + i\frac{\Omega_1}{2}\rho_{se}}{\gamma + i\Delta}. \quad (\text{B.6})$$

The inverted Lorentzian CPT dip appears near the two-photon resonance. In this regime we typically have $\Delta_1, \Delta_2, \gamma \ll \Gamma$, so that the optical coherences are mainly limited by the excited-state decay. We therefore approximate $\frac{\Gamma}{2} + \frac{\gamma}{4} - i\Delta_{1/2}$ with $\Gamma/2$ which simplifies to

$$\rho_{eg} = \frac{i\Omega_1(\rho_{ee} - \rho_{gg}) - i\Omega_2\rho_{sg}}{\Gamma}, \quad (\text{B.7})$$

$$\rho_{es} = \frac{i\Omega_2(\rho_{ee} - \rho_{ss}) - i\Omega_1\rho_{gs}}{\Gamma}. \quad (\text{B.8})$$

Combining [Equation B.6](#), [Equation B.7](#), [Equation B.8](#), and use $\rho_{ee} + \rho_{ss} + \rho_{gg} = 1$, assume Ω_1 is complex due to the relative phase but Ω_2 is real, we get

$$\rho_{sg} = \frac{\Omega_1\Omega_2(3\rho_{ee} - 1)}{(\Omega_1^2 + \Omega_2^2) + 2\Gamma(\gamma + i\Delta)}. \quad (\text{B.9})$$

From which we can see that a unstable relative phase between Ω_1, Ω_2 leads to reduced $\langle \rho_{sg} \rangle$ and then contrast. And [Equation B.4](#), [Equation B.5](#), [Equation B.7](#),

Equation B.9 gives

$$\rho_{ee} = \frac{1}{3 + \frac{\Gamma(\Gamma_1\Omega_2^2 + \Gamma_2\Omega_1^2)}{\Omega_1^2\Omega_2^2} + \frac{\Gamma(\Gamma_1\Omega_2^2 + \Gamma_2\Omega_1^2)}{\Omega_1^2\Omega_2^2 \frac{(\Omega_1^2 + \Omega_2^2 + 2\Gamma\gamma)^2 + (2\Gamma\Delta)^2 - (\Omega_1^2 + \Omega_2^2 + 2\Gamma\gamma)(\Omega_1^2 + \Omega_2^2)}{(\Omega_1^2 + \Omega_2^2 + 2\Gamma\gamma)(\Omega_1^2 + \Omega_2^2)}}},$$

compare to a inverted Lorentzian curve

$$D - B \frac{1}{\Delta^2 + C^2} = \frac{1}{\frac{1}{D} + \frac{\frac{B}{D}}{D\Delta^2 + DC^2 - B}},$$

we have

$$\rho_{ee} = \frac{1}{3 + \frac{\Gamma\Gamma_1}{\Omega_1^2} + \frac{\Gamma\Gamma_2}{\Omega_2^2}} \left(1 - \frac{C_{\text{eff}}}{(C_{\text{eff}} + 1)} \frac{(C_{\text{eff}} + 1)^2 \gamma^2}{(C_{\text{eff}} + 1)^2 \gamma^2 + \Delta^2} \right),$$

$$\text{Effective Cooperativity: } C_{\text{eff}} = \frac{(\Omega_1^2 + \Omega_2^2)/2}{\Gamma\gamma},$$

$$\text{Half-width: } C = (C_{\text{eff}} + 1)\gamma = \frac{\Omega_1^2 + \Omega_2^2}{2\Gamma} + \gamma = c \cdot \text{Power} + \gamma, \quad (\text{B.10})$$

$$\text{Dip-contrast: } \frac{B}{DC^2} = \frac{(\Omega_1^2 + \Omega_2^2)}{(\Omega_1^2 + \Omega_2^2 + 2\Gamma\gamma)} = \frac{C_{\text{eff}}}{C_{\text{eff}} + 1}, \quad (\text{B.11})$$

$$\text{Dip-SNR: } \sqrt{\frac{B}{DC^2}} \cdot D = \frac{1}{\sqrt{3 + \frac{\Gamma\Gamma_1}{\Omega_1^2} + \frac{\Gamma\Gamma_2}{\Omega_2^2}}} \frac{C_{\text{eff}}}{C_{\text{eff}} + 1}. \quad (\text{B.12})$$

With a constant power-broadening $\Omega = \Omega_1^2 + \Omega_2^2$, C_{eff} stays constant, and the

Dip-SNR can be maximized by varying ratio $r = \Omega_2^2/\Omega_1^2$ with $\Omega_1^2 = \Omega/(1 + r)$,

$$\text{SNR}(r) \propto \frac{1}{\sqrt{3 + \Gamma\Omega(\Gamma + \Gamma_1 r + \Gamma_2/r)}},$$

$$r_{\text{max}} = \sqrt{\frac{\Gamma_2}{\Gamma_1}}.$$

Considering that $\Omega_{1/2}^2 \propto P_{1/2} \cdot |d_{1/2}|^2 \propto P_{1/2} \cdot \Gamma_{1/2}$, we desire a

$$\frac{P_2}{P_1} = \sqrt{\frac{\Gamma_1}{\Gamma_2}}.$$

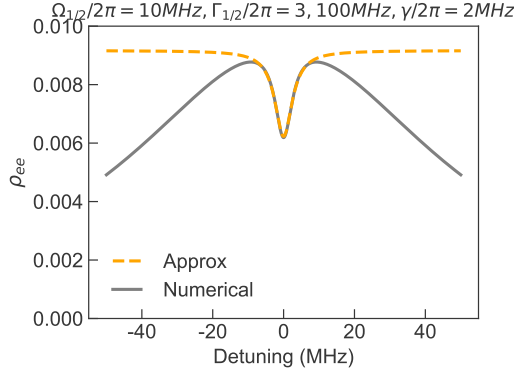


Figure B.1. Comparison between exact numerical results and approximation.

B.2 Wave optics

B.2.1 Helmholtz equation

From Maxwell's equations, we are able to get the vector Helmholtz equation in the absence of current and charge,

$$(\nabla^2 + k^2)\mathbf{E}(\mathbf{r}) = 0, \nabla \cdot \mathbf{E}(\mathbf{r}) = 0,$$

with $k = n\omega/c$, that n, ω are index of refractive and angular frequency of field.

Since a vector differential equation is hard to solve, instead often times a scalar Helmholtz equation is solved that

$$(\nabla^2 + k^2)\psi(\mathbf{r}) = 0,$$

and the solution of vector Helmholtz equation can be generated from Hansen vector wave functions

$$\mathbf{L}(\psi) = \nabla\psi,$$

$$\mathbf{M}(\psi) = \nabla \times (\mathbf{r}\psi),$$

$$\mathbf{N}(\psi) = \frac{1}{k}\nabla \times \mathbf{M}(\psi).$$

They are all solutions of the vector Helmholtz equation and M, N are also divergence-free therefore are solutions under charge free condition.

Since a plane wave $\psi(r) = e^{ikz}$ has no E_z in its corresponding Hansen vector functions, then scalar Helmholtz is sufficient for describing it. And for a more general solution

$$\psi(\mathbf{r}) = \int A(k_x, k_y) e^{i(k_x x + k_y y + \sqrt{k^2 - k_x^2 - k_y^2} z)} dk_x dk_y,$$

we have $|E_z/E| \sim |k_\perp/k|$, then given a small $|k_\perp/k|$ we can ignore the E_z term. In this case,

$$\begin{aligned} \mathbf{E}(\mathbf{r}) &\approx a\mathbf{M}(\psi(x, y)) + b\mathbf{N}(\psi(x, y)) \\ &= \hat{\mathbf{x}}E_x(x, y) + \hat{\mathbf{y}}E_y(x, y) \\ &= \hat{\mathbf{x}} \sum a_i \psi_i(x, y) + \hat{\mathbf{y}} \sum b_i \psi_i(x, y), \end{aligned}$$

this means the solution of vector Helmholtz equation is also linear combinations of orthogonal solution set ψ_i of scalar Helmholtz with different linear polarizations. This condition indicates a almost straight propagation is equivalent to paraxial approximation in free space or weak guiding condition in fiber.

B.2.2 Gaussian beam

Assuming a laser beam propagates along z direction, then we have $\psi(\mathbf{r}) = u(\mathbf{r})e^{ikz}$, corresponding scalar Helmholtz equation is

$$\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2} + 2ik \frac{\partial u}{\partial z} = 0.$$

Here we apply the paraxial approximation that assumes remaining z dependent term in $u(\mathbf{r})$ is small which means $|\partial u/\partial z| \ll |2ku|$ then we have $|\partial^2 u/\partial z^2| \ll |2k\partial u/\partial z|$ therefore

$$\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + 2ik \frac{\partial u}{\partial z} = 0.$$

One orthogonal solution set of this equation is called Hermite-Gaussian modes, and has the form [Wikipedia \(n.d.\)](#)

$$\begin{aligned}\psi_{l,m}(\mathbf{r}) &= \frac{\sqrt{2/\pi}}{\sqrt{2^{l+m}l!m!\omega_0}} \frac{q(0)}{q(z)} \left(-\frac{q^*(z)}{q(z)}\right)^{\frac{(l+m)}{2}} H_l\left(\frac{\sqrt{2}x}{\omega(z)}\right) H_m\left(\frac{\sqrt{2}y}{\omega(z)}\right) \\ &\times \exp\left(-ik\frac{r^2}{2q(z)} - ikz\right),\end{aligned}\quad (\text{B.13})$$

where we defined $q(z) = z + iz_R$, $z_R = \pi\omega_0^2 n/\lambda$, and H_l is Hermite polynomial of order l . For a fundamental HG₀₀ or also called TEM₀₀ mode, we have Gaussian beam

$$\psi_{00}(\mathbf{r}) = E_0 \frac{iz_R}{q(z)} \exp\left(-ik\frac{r^2}{2q(z)} - ikz\right).$$

For a thin spherical lens with curvature $1/R_1$ and $1/R_2$, we know that focal length is given by Lensmaker's equation

$$\frac{1}{f} = (n-1) \left(\frac{1}{R_1} + \frac{1}{R_2} \right).$$

We can estimate the phase change after single side spherical lens at r from the center as $\varphi(r) = -(n-1)(R - \sqrt{R^2 - r^2})$, by applying paraxial approximation and consider both sides, the transmission function is then

$$\begin{aligned}T(r) &= \exp\left(-ik\frac{(n-1)r^2}{2}\left(\frac{1}{R_1} + \frac{1}{R_2}\right)\right) \\ &= \exp\left(-ik\frac{r^2}{2f}\right).\end{aligned}$$

From this we are able to solve Gaussian beam after lens using $\psi_f(\mathbf{r}) = \psi_i(\mathbf{r})T(r)$, the relation follows

$$\frac{1}{q_f(z_f)} - \frac{1}{q_i(z_i)} = \frac{1}{f} = \frac{1}{R_f(z_f)} - \frac{1}{R_i(z_i)} - i\frac{\lambda}{n\pi\omega_f^2(z_f)} + i\frac{\lambda}{n\pi\omega_i^2(z_i)},$$

where we use the convention that $z_{f/i}$ is distance of beam waist to the center of lens, and curvature radius $R(z) = z(1 + (z_R/z)^2)$, beam radius $\omega(z) = \omega_0\sqrt{1 + (z/z_R)^2}$. We then identify that $q(z)$ is just a complex extension of z in

ray optics. The z and z_R after lens are

$$\begin{aligned} z_f &= \frac{z_i f(z_i - f) + f z_{Ri}^2}{(z_i - f)^2 + z_{Ri}^2}, \\ z_{Rf} &= \frac{z_{Ri} f^2}{(z_i - f)^2 + z_{Ri}^2}. \end{aligned} \quad (\text{B.14})$$

In the limit $f \rightarrow 0$, we have $z_f \rightarrow f$. In the following we assume that $\omega_i(z_i)$ is a constant which means $z_{Ri} + z_i^2/z_{Ri} = C = \pi D^2 n / 4\lambda$ that input beam always fill out the full lens diameter D ,

$$z_{Rf} = \frac{f^2}{C} \frac{1}{1 + \frac{f^2 - 2fz_i}{Cz_{Ri}}}, \quad z_i = \pm \sqrt{(C - z_{Ri})z_{Ri}},$$

then it reaches minimal when $(f^2 - 2fz_i)/Cz_{Ri}$ reaches maximal

$$\frac{d \frac{f^2 - 2fz_i}{Cz_{Ri}}}{dz_{Ri}} = 0 = \frac{f}{Cz_{Ri}^2} \left(\frac{Cz_{Ri} - 2z_{Ri}^2}{\sqrt{Cz_{Ri} - z_{Ri}^2}} - f \pm 2\sqrt{Cz_{Ri} - z_{Ri}^2} \right),$$

or when at boundaries $z_{Ri} \rightarrow 0$ or $z_{Ri} = C$. We can observe that $z_{Ri} \rightarrow C$

gives a positive derivative thus as least a local maximal. This corresponding to a collimated input that $z_i = 0$ and produces a

$$z_{Rf} = \frac{Cf^2}{f^2 + C^2} = \frac{f^2 \frac{\pi D^2 n}{4\lambda}}{f^2 + (\frac{\pi D^2 n}{4\lambda})^2}, \quad \omega_{0f} = \frac{\lambda}{\pi \frac{nD}{2f}} \frac{1}{\sqrt{1 + (4f\lambda/\pi D^2 n)^2}}.$$

Since $(f/C)^2 = (4f\lambda/\pi D^2 n)^2$ is much smaller than 1 for a macroscopic lens, and $nD/2f$ is NA under paraxial approximation, then we have the textbook result

$$2\omega_{0f} = \frac{2\lambda}{\pi \text{NA}},$$

although strictly speaking we need a vector Helmholtz equation for high-NA systems but this formula is often accurate enough.

B.2.3 Fiber modes

Here we consider a step-index fiber that $n(r) = n_1$ for core $r \leq a$ and $n(r) = n_2$ for cladding that $r > a$, and weak guiding approximation that $\Delta = (n_1^2 - n_2^2)/2n_1^2 \ll 1$. In these conditions, we can again use the scalar Helmholtz equation and use only a polarization to describe the vector field. We write the solution in

cylindrical coordinate, then

$$\psi(\mathbf{r}) = R(r)\Phi(\phi)e^{i\beta z}.$$

Solving differential equations separately then we end up with

$$\psi(\mathbf{r}) = \begin{cases} AJ_l(\sqrt{k_1^2 - \beta^2}r)e^{il\phi + i\beta_l z} & , r \leq a \\ CK_l(\sqrt{\beta^2 - k_2^2}r)e^{il\phi + i\beta_l z} & , r > a \end{cases},$$

where J_l, K_l are Bessel function of the first kind and Modified Bessel function of the second kind to ensure physical reality while Bessel function of the second kind is unbounded at $r = 0$ and Modified Bessel function of the first kind is non-zero at $r = \infty$.

To satisfy the boundary conditions at $r = a$, we require a characteristic equation

$$\frac{J'_l(\sqrt{k_1^2 - \beta^2}a)}{\sqrt{k_1^2 - \beta^2}J_l(\sqrt{k_1^2 - \beta^2}a)} = \frac{K'_l(\sqrt{\beta^2 - k_2^2}a)}{\sqrt{\beta^2 - k_2^2}K_l(\sqrt{\beta^2 - k_2^2}a)},$$

and a set of solution with different β_{lm} is solved where m denotes the m -th solution. When $\beta \rightarrow k_2$, then solution is no longer bounded outside the fiber core, this is the cutoff of mode. In this limit, and use the recurrence relation of Bessel function, we have

$$\frac{uJ_{l-1}(u)}{J_l(u)} = -\frac{wK_{l-1}(w)}{K_l(w)},$$

where $u = a\sqrt{k_1^2 - \beta^2}$ and $w = a\sqrt{\beta^2 - k_2^2}$, and $K_{-l} = K_l$, $J_{-l} = (-1)^l J_l$. Then from the asymptotic forms, we have $wK_{l-1}(w)/K_l(w) \rightarrow 0$ for $w \rightarrow 0$. This gives

$$\lim_{w \rightarrow 0} \frac{uJ_{l-1}(u)}{J_l(u)} = \frac{VJ_{l-1}(V)}{J_l(V)} = 0,$$

that $V = a\sqrt{k_1^2 - k_2^2}$. Therefore the cutoff is $V = 0$ for $l = 0$ and $V \approx 2.405$ for $l = 1$. This means that a fundamental LP_{01} mode has no cutoff, and other modes start to appear when $V \approx 2.405$ and fiber becomes mutimode.

Under weak guiding condition we have LP_{lm} modes but rigorous solutions are combinations of TE, TM, HE, EH modes. However the fundamental mode is always LP_{01} that has a Gaussian-like shape.

B.2.4 Fiber coupling

For a single mode fiber coupling, the efficiency can be written as the projection of input electric field on the fiber electric field, and has the form

$$\eta = \frac{|\int E_{\text{fiber}}^*(x, y) E_{\text{in}}(x, y) dA|^2}{(\int |E_{\text{fiber}}(x, y)|^2 dA)(\int |E_{\text{in}}(x, y)|^2 dA)}.$$

The estimation of such form gives [Marcuse \(1977\)](#)

$$\eta = \frac{4 \left(4Z^2 + \frac{\omega_1^2}{\omega_2^2} \right)}{\left(4Z^2 + \frac{\omega_2^2 + \omega_1^2}{\omega_2^2} \right)^2 + 4Z^2 \frac{\omega_2^2}{\omega_1^2}}, Z = \frac{D}{n_2 k \omega_1 \omega_2},$$

where D is the longitudinal separation, and assume a graded-index fiber with outer index of refractive n_2 . And similarly the efficiency with only a tilt angle error θ ,

$$\eta = \left(\frac{2\omega_1\omega_2}{\omega_1^2 + \omega_2^2} \right)^2 \exp \left(-\frac{2(\pi n_2 \omega_1 \omega_2 \theta)^2}{(\omega_1^2 + \omega_2^2) \lambda^2} \right),$$

and efficiency with only a offset d ,

$$\eta = \left(\frac{2\omega_1\omega_2}{\omega_1^2 + \omega_2^2} \right)^2 \exp \left(-\frac{2d^2}{\omega_1^2 + \omega_2^2} \right).$$

From those formulas we can tell that multimode fiber is more insensitive to alignment errors and the waist matching error

$$\eta = \left(\frac{2\omega_1\omega_2}{\omega_1^2 + \omega_2^2} \right)^2$$

determines the upper bound of coupling efficiency with perfect alignment. This can be normally optimize by sending laser beam backwards and match inwards and backwards beam waist using the correct objective lens or just rough estimate from the specification.

On the other hand, coupling efficiency of multimode fiber depends on the NA matching that input beam with a large angle can experience loss due to the loose of total internal reflection condition.

B.2.5 Diffracted by AOM

Here we consider a plane wave propagate in $(\cos\theta, -\sin\theta)$ direction and thus has the form $E_{in}(x, y) = E_0 e^{i(k \cos\theta x - k \sin\theta y)}$, an acoustic optical modulator has acoutcis wave propagate in $\hat{\mathbf{y}}$ direction therefore create refractive index modulation $\Delta n(y) = \Delta n_0 \cos(2\pi y/\lambda_a)$, that $\Delta n_0 \propto \sqrt{P_{\text{RF}}/V}$. The corresponding transmission function of such retractive index grating is

$$T(y) = e^{ik\Delta n(y)/\cos\theta} = \sum_m i^m J_m\left(\frac{k\Delta n_0}{\cos\theta}\right) e^{im\frac{2\pi}{\lambda_a}y},$$

where we use the Jacobi-Anger expansion. Then we know that after AOM, the wave has an expression of

$$E_{out}(x, y) = E_{in}(x, y)T(y) = E_0 e^{ik \cos\theta x} \sum_m i^m J_m\left(\frac{k\Delta n_0}{\cos\theta}\right) e^{i(m\frac{2\pi}{\lambda_a} - k \sin\theta)y}.$$

This is the Raman-Nath solution that is in the limit AOM has zero thickness, but this is different from the practical case. For a more general derivation, we consider only the zero and first order diffraction given that driving power of AOM is low and incident angle is near the Bragg angle θ_b that $\sin\theta_b = \lambda/2\lambda_a$

$$E(x, y) = A_0(x) e^{i(k \cos(\theta_b + \delta)x - k \sin(\theta_b + \delta)y)} + A_1(x) e^{i(k \cos(\theta_b)x + k \sin(\theta_b)y)}.$$

Then $E(x + dx, y)$ experiences a $T(y)$ first and then a free evolution from x to $x + dx$, this is

$$\begin{aligned}
E(x, y)T(y) &\approx A_0 e^{i(k \cos(\theta_b + \delta)x - k \sin(\theta_b + \delta)y)} \left(J_0\left(\frac{k\Delta n_0 dx}{\cos(\theta_b)}\right) + iJ_1\left(\frac{k\Delta n_0 dx}{\cos(\theta_b)}\right) e^{i\frac{2\pi}{\lambda_a}y} \right) \\
&+ A_1 e^{i(k \cos(\theta_b)x + k \sin(\theta_b)y)} \left(J_0\left(\frac{k\Delta n_0 dx}{\cos(\theta_b)}\right) - iJ_1\left(\frac{k\Delta n_0 dx}{\cos(\theta_b)}\right) e^{-i\frac{2\pi}{\lambda_a}y} \right) \\
&\approx A_0 e^{i(k \cos(\theta_b + \delta)x - k \sin(\theta_b + \delta)y)} \left(1 + \frac{ik\Delta n_0 dx}{2 \cos \theta_b} e^{ik \cos \theta_b \sin \delta} \right) \\
&+ A_1 e^{i(k \cos(\theta_b)x + k \sin(\theta_b)y)} \left(1 + \frac{ik\Delta n_0 dx}{2 \cos \theta_b} e^{-ik \cos \theta_b \sin \delta} \right).
\end{aligned}$$

With derivative to x

$$E(x + dx, y) - E(x, y) \approx \frac{dE(x, y)}{dx} dx,$$

and after a tedious process, we end up with a coupled equation

$$\begin{aligned}
\frac{dA_0}{dx} &= \frac{ik\Delta n_0}{2 \cos \theta_b} A_1 e^{-ik \sin \theta_b \delta x}, \\
\frac{dA_1}{dx} &= \frac{ik\Delta n_0}{2 \cos \theta_b} A_0 e^{ik \sin \theta_b \delta x}.
\end{aligned}$$

This is the standard coupled equation for two level quantum system, we know the solution is

$$A_1(x) = iA_0(0) \frac{\kappa}{\Omega} e^{i\Delta x/2} \sin(\Omega x),$$

that $\Omega = \sqrt{\kappa^2 + (\Delta/2)^2}$, $\Delta = k \sin \theta_b \delta$, $\kappa = k\Delta n_0/2 \cos \theta_b$. Finally, the first order diffraction efficiency is

$$\left| \frac{A_1(L)}{A_0(0)} \right|^2 = \frac{\kappa^2}{\kappa^2 + \Delta^2/4} \sin^2 \left(L \sqrt{\kappa^2 + \Delta^2/4} \right) = \kappa^2 L^2 \text{sinc}^2 \left(L \sqrt{\kappa^2 + \Delta^2/4} \right).$$

And in the limit of κ or Δ , we are able to recover the standard AOM efficiency formula [Isomet \(n.d.\)](#) that

$$\begin{aligned}
\eta_{0 \rightarrow 1} &\propto \sin^2 \left(\frac{Lk\Delta n_0}{2 \cos \theta_b} \right) \text{sinc}^2 \left(\frac{Lk \sin \theta_b \delta}{2} \right) \\
&\propto \sin^2 \left(\frac{\pi C_0}{\lambda \cos \theta_b} \sqrt{\frac{LP_{\text{RF}}}{H}} \right) \text{sinc}^2 \left(\frac{\pi L \delta}{2 \lambda_a} \right). \tag{B.15}
\end{aligned}$$

APPENDIX C

CAVITY QED WITH SiV CENTERS

C.1 Introduction

The NV center is particularly powerful for quantum sensing because its spin coherence persists to room temperature. However, its geometry lacks inversion symmetry as discussed in chapter II, which gives rise to a permanent electric dipole moment. Consequently, NV optical transitions are intrinsically sensitive to local charge fluctuations. In dielectric systems, such fluctuations are essentially unavoidable because the absence of free carriers, and this problem is severe near diamond surfaces. The associated Stark shifts cause spectral diffusion and undermine transition stability, which complicates cavity coupling and other experiments that require narrow, stable lines.

In contrast, SiV center possesses inversion symmetry. This strongly suppresses first-order sensitivity to electric-field noise and leads to more stable optical transitions even in nanostructures. These properties make the SiV center a natural candidate for cavity quantum electrodynamics.

A cavity enhances light–matter interaction by reducing the optical mode volume and increasing the local field, thus boosting the coupling rate beyond what is feasible in free space. At the single-quantum level, a cavity enables coherent exchange between a “flying” photonic qubit and a stationary SiV qubit, and mediates effective SiV–SiV interactions either within the same resonator or across remote nodes via photonic links. Such interfaces directly address the scalability challenges of solid-state defect platforms and provide key primitives for quantum networking, including quantum repeaters and memory-assisted entanglement distribution [Bhaskar et al. \(2020\)](#); [Knaut et al. \(2024\)](#).

In this chapter, we develop a theoretical framework for SiV–cavity interactions and connect it to measurable observables in typical experimental regimes. We begin with a minimal coupled-oscillator model that gives cooperativity C with simplest model. We then discuss how cavity probing and atom (defect) probing differ operationally. Finally, we briefly introduce some experimental attempts to characterize such a system.

C.2 Cavity QED

The cooperativity C appears in many cavity-QED results. At its core, it arises from the interaction between two damped, coupled oscillators, whose dynamics can be expressed in terms of the dimensionless parameter C . Consider the following Hamiltonian

$$H = \omega_a a^\dagger a + \omega_b b^\dagger b + g(a^\dagger b + ab^\dagger) + \Omega \cos(\omega_\Omega t)(a + a^\dagger),$$

where $\Omega \cos(\omega_\Omega t)(a + a^\dagger) = \sqrt{2}\Omega \cos(\omega_\Omega t)q$ is the driving on the coordinates, this Hamiltonian together with decay channels give equations of motion $\dot{O} = i[H, O]$ under rotating frame,

$$\begin{aligned}\dot{a}(t) &= (-i\Delta_a - \frac{\kappa}{2})a(t) - igb(t) - \Omega, \\ \dot{b}(t) &= (-i\Delta_b - \frac{\gamma}{2})b(t) - iga(t),\end{aligned}$$

where Ω is the external driving under rotating frame. Many times we care about the steady solution of the system as a function of $\Delta_a = \omega_a - \omega_\Omega$, $\Delta_b = \omega_b - \omega_\Omega$, $\omega_a, \omega_b, \omega_\Omega$ are frequencies of oscillator a, b and driving Ω respectively, therefore we

can solve a_{steady} and b_{steady} as

$$\begin{aligned} a_{\text{steady}} &= \frac{\Omega}{-i\Delta_a - \frac{\kappa}{2} + \frac{g^2}{-i\Delta_b - \frac{\gamma}{2}}}, \\ |a_{\text{steady}}|^2 &= \Omega_{\text{eff},a}^2 \frac{1 + \delta_b^2}{(1 + C - \delta_a \delta_b)^2 + (\delta_a + \delta_b)^2}, \\ |b_{\text{steady}}|^2 &= \Omega_{\text{eff},b}^2 \frac{(C/2)^2}{(1 + C - \delta_a \delta_b)^2 + (\delta_a + \delta_b)^2}, \end{aligned}$$

where $\Omega_{\text{eff},a} = 2\Omega/\kappa$, $\Omega_{\text{eff},b} = 2\Omega/g$, $C = 4g^2/\kappa\gamma$, $\delta_a = 2\Delta_a/\kappa$, $\delta_b = 2\Delta_b/\gamma$, therefore in principle the system depends only on these dimensionless parameters. As we will discuss later, when κ or γ is larger than other parameters, the response of the system near the resonance of the slower mode a/b is a function of $\delta_{a/b}$ and C . Here we discuss this solution in three different regime.

In the strong-coupling regime we have $g \gg \kappa, \gamma$. Assuming modes a, b are resonant then $\Delta_a = \Delta_b = \Delta$ and we have

$$\begin{aligned} |a_{\text{steady}}|^2 &= \frac{\Omega^2}{4} \frac{\gamma^2 + 4\Delta^2}{(\Delta^2 - g^2 - \kappa\gamma/4)^2 + (\Delta \frac{\kappa+\gamma}{2})^2}, \\ &\approx \frac{\Omega^2}{4} \left(\frac{1}{(\Delta + g)^2 + (\frac{\kappa+\gamma}{4})^2} + \frac{1}{(\Delta - g)^2 + (\frac{\kappa+\gamma}{4})^2} \right), \end{aligned}$$

where $(\frac{\kappa+\gamma}{4})^2$ is obtained by considering behaviours around $\Delta = \pm g$, and this has the same physics as the vacuum Rabi splitting that we have two Lorentzian peaks with FWHM $(\kappa + \gamma)/2$ separated by $2g$. We can see that $|b_{\text{steady}}|^2$ shares the same profile but with an additional background envelope $1/(1 + \delta_b^2)$.

In bad mode a limit regime we have $\kappa \gg g, \gamma$. Assuming modes a, b are resonant then $\Delta_a = \Delta_b = \Delta$ and we expand the expression near zero δ_b (then δ_a is higher-order infinitesimal),

$$\begin{aligned} |a_{\text{steady}}|^2 &\approx \Omega_{\text{eff},a}^2 \frac{1 + \delta_b^2}{(1 + C)^2 + \delta_b^2} = \Omega_{\text{eff},a}^2 \left(1 - \frac{C(2 + C)}{(1 + C)^2 + \delta_b^2} \right), \\ |b_{\text{steady}}|^2 &\approx \Omega_{\text{eff},b}^2 \frac{(C/2)^2}{(1 + C)^2 + \delta_b^2}, \end{aligned}$$

therefore we see a FWHM $(1 + C)\gamma$ for mode b which is Purcell enhancement that effectively increase the spontaneous emission rate of mode b from γ to $(1 + C)\gamma$. And the background remains $\Omega_{\text{eff},a}^2/(1 + \delta_a^2)$ by taking $\delta_b \rightarrow \infty$, which is the Lorentzian curve of mode a response in the absence of coupling. This result indicates the effect of coupling is a dip of mode b in the middle of mode a .

In bad mode b limit regime we have $\gamma \gg g, \kappa$. Follow a similar procedure, we end up with

$$\begin{aligned} |a_{\text{steady}}|^2 &\approx \Omega_{\text{eff},a}^2 \frac{1}{(1 + C)^2 + \delta_a^2}, \\ |b_{\text{steady}}|^2 &\approx \Omega_{\text{eff},b}^2 \frac{(C/2)^2}{(1 + C)^2 + \delta_a^2}, \end{aligned}$$

which gives the Purcell-enhanced FWHM $(1 + C)\kappa$ for a mode. The mode b response now has the same form as mode a cause the energy exchange is limited by the excitation of mode a . And in this case, the background is zero by taking $\delta_a \rightarrow \infty$ which means that the effect of the coupling is to broaden mode a .

Another approach to understand the results above is as follows,

$$a_{\text{steady}} = \frac{\Omega(-i\Delta - \frac{\gamma}{2})}{(-i\Delta - \frac{\kappa}{2})(-i\Delta - \frac{\gamma}{2}) + g^2} \quad ,$$

we have the identical equation

$$\begin{aligned} \frac{N(\Delta)}{(\Delta - \Delta_-)(\Delta - \Delta_+)} &= \frac{A_-}{\Delta - \Delta_-} + \frac{A_+}{\Delta - \Delta_+}, \\ A_- &= \frac{N(\Delta_-)}{\Delta_- - \Delta_+}, A_+ = \frac{N(\Delta_+)}{\Delta_+ - \Delta_-}, N(\Delta) = \Omega(i\Delta + \frac{\gamma}{2}), \end{aligned}$$

with the solution being

$$\Delta_{\pm} = i(\frac{\kappa + \gamma}{4}) \pm \sqrt{g^2 - (\frac{\kappa - \gamma}{4})^2},$$

where the real part is the frequency resonance and the imaginary part is the decay rate of corresponding resonance. This tells us that the steady state solution of the

system is intrinsically a sum of two Lorentzian peaks with an interference term

$$\begin{aligned} |a_{\text{steady}}|^2 &= \left| \frac{A_-}{\Delta - \Delta_-} + \frac{A_+}{\Delta - \Delta_+} \right|^2 \\ &= \left| \frac{A_-}{\Delta - \Delta_-} \right|^2 + \left| \frac{A_+}{\Delta - \Delta_+} \right|^2 + 2\text{Re} \left(\frac{A_- A_+^*}{(\Delta - \Delta_-)(\Delta - \Delta_+^*)} \right), \end{aligned}$$

for a strong coupling regime this interference term disappears. For a weak coupling $g \ll \kappa, \gamma$ and if one solution has decay rate much faster than the other, we can expand a_{steady} near the resonance of the slower decay solution (say Δ_+) as

$$a_{\text{steady}}(\Delta) \approx \frac{A_+}{\Delta - \Delta_+} + B, B = \frac{A_-}{\Delta_+ - \Delta_-},$$

where we treat the contribution from faster decay solution as static background.

Then we define $\epsilon = -(\Delta - \text{Re}(\Delta_+))/\text{Im}(\Delta_+)$, and

$$a_{\text{steady}}(\Delta) \approx B \frac{\epsilon + q}{\epsilon + i}, q = i - \frac{A_+}{\text{Im}(\Delta_+)B},$$

this gives the standard Fano shape in which a $q \approx 0$ gives Lorentzian dip, $q \rightarrow \infty$ gives Lorentzian peak, and others give asymmetric Fano resonance. This model describes all different behaviours, and the asymmetric Fano resonance is often observed when mode a, b are not resonant and $\Delta_a = \delta + \Delta_b$.

For a more complex system in which an atom interacts with a harmonic oscillator, the above analysis still applies as long as the weak-excitation condition holds, so that the atomic operators $\sigma_{ge} = |g\rangle\langle e|$ can be viewed as oscillator a , or when multiple atoms are present, the collective operator $\sum_i |g\rangle_{ii}\langle e|/\sqrt{N}$, in this case the validity of the weak-excitation approximation can be extended by a factor of $\sqrt{N}$. And from results under different regimes, we can see that it's easier to drive and probe the mode with higher decay rate such that we can observe a dip in the middle which gives better contrast for experimental calibration.

From a quantum information processing perspective, cavity QED – and more generally, systems where an atom is coupled to an cavity mode – are very

appealing for several reasons. First, in the bad-cavity limit, which is often realized in practice (for example, SiV centers or neutral atoms coupled to photonic crystal cavities or Fabry–Pérot cavities), the atomic decay rate into the cavity mode is Purcell-enhanced compared to the no-cavity case. A fraction $C/(1 + C)$ of the emitted photons is collected into the cavity mode. This enhances the collection efficiency, because the emission is both faster and better mode-matched than the cavity mode can be designed to be approximately Gaussian, and therefore couples efficiently to a single-mode optical fiber. More generally, the cavity modifies the local density of optical states (LDOS). When the cavity only affects a subset of the available radiation modes, this leads to Purcell enhancement on top of an essentially unchanged background decay channel. In contrast, when the cavity fully “encloses” the emitter – for example, an atom embedded in a photonic crystal where the free-space modes responsible for spontaneous emission are suppressed – the free-space channel is effectively removed and the cavity determines the total decay rate. If the cavity resonance is engineered to be detuned from the atomic transition, the cavity-induced decay is suppressed and the total decay rate of the atom is reduced.

In the resonant case, the atom induces a narrow dip on top of the cavity resonance with a contrast of order $1/(1 + C)^2$, so by switching the internal state of the atom we can strongly modulate the transmission or reflection of single photons. One can use a first photon to change the atomic internal state such that the dip disappears (or appears) in the cavity response, and thereby selectively reflect or transmit a second photon. This realizes atom-mediated photon–photon entanglement in, for example, a time-bin encoding. Similarly, a shared cavity mode can mediate coherent interactions between spatially separated atoms.

C.3 Probe cavity

As discussed before, in the bad-cavity limit it is experimentally more convenient to probe and drive the cavity mode. In practice, one measures the cavity reflection/transmission rather than directly measuring the intracavity photon occupation. In the following, we provide a more formal derivation using the standard input–output formalism.

C.3.1 Input-Output Formalism

We consider a single cavity mode $a(t)$ coupled to a continuum of external modes $b(\omega, t)$. The interaction plus bath Hamiltonian is

$$H_{\text{int+ext}} = \int \sqrt{\frac{\kappa(\omega)}{2\pi}} (a(t)b^\dagger(\omega, t) + a^\dagger(t)b(\omega, t)) d\omega + \int \omega b^\dagger(\omega, t)b(\omega, t)d\omega,$$

where $b(\omega, t) \sim 1/\sqrt{\omega}$ is the annihilation operator for the continuum mode at frequency ω in the continuous limit, while $a(t) \sim 1$ is the annihilation operator of the discrete cavity mode. In the absence of the cavity, the external field $b(\omega, t)$ evolves freely under the bath Hamiltonian

$$H_{\text{ext}} = \int \omega b^\dagger(\omega, t)b(\omega, t)d\omega.$$

We assume a large bath and Markovian noise, such that the bath correlation time is much shorter than the system time scales. In this limit, the interaction kernel becomes a delta function in the time domain, which corresponds to a flat spectrum $\kappa(\omega) = \kappa$ in the frequency domain. Then we obtain

$$\begin{aligned} \dot{a}(t) &= (\dot{a}(t))_{\text{sys}} - i\sqrt{\frac{\kappa}{2\pi}} \int b(\omega, t)d\omega, \\ \dot{b}(\omega, t) &= -i\omega b(\omega, t) - i\sqrt{\frac{\kappa}{2\pi}} a(t), \end{aligned}$$

where $(\dot{a}(t))_{\text{sys}}$ denotes the contribution from the bare system Hamiltonian (including detuning, drive, nonlinearity, etc.).

Solving the equation for the bath modes, we find

$$b(\omega, t) = e^{-i\omega(t-t_0)}b(\omega, t_0) - i\sqrt{\frac{\kappa}{2\pi}} \int_{t_0}^t e^{-i\omega(t-t')}a(t')dt',$$

where $t_0 \rightarrow -\infty$ is an initial time in the remote past. We now distinguish incoming and outgoing fields via their asymptotic behavior. We define $b_{\text{in}}(\omega, t)$ and $b_{\text{out}}(\omega, t)$ by

$$\lim_{t \rightarrow -\infty} b(\omega, t) = b_{\text{in}}(\omega, t), \quad \lim_{t \rightarrow +\infty} b(\omega, t) = b_{\text{out}}(\omega, t).$$

A natural way to extend $b_{\text{in/out}}(\omega, t)$ to finite times is to let them evolve freely under the bath Hamiltonian H_{ext} , we define

$$b_{\text{in}}(\omega, t) = \lim_{t_0 \rightarrow -\infty} e^{-i\omega(t-t_0)}b(\omega, t_0),$$

$$b_{\text{out}}(\omega, t) = \lim_{t_1 \rightarrow +\infty} e^{-i\omega(t-t_1)}b(\omega, t_1),$$

so that both b_{in} and b_{out} satisfy the free evolution generated by H_{ext} .

We now consider the integrated bath field

$$\int b(\omega, t)d\omega = \int e^{-i\omega(t-t_0)}b(\omega, t_0)d\omega - i\sqrt{\frac{\kappa}{2\pi}} \int_{t_0}^t dt' \int e^{-i\omega(t-t')}a(t')d\omega.$$

Taking $t_0 \rightarrow -\infty$ and using the definition of b_{in} , the first term becomes the freely propagated input. The second term can be evaluated using $\int e^{i\omega(t-t')}d\omega = 2\pi\delta(t-t')$. Since the integration range is $t' \in (-\infty, t)$, we use the standard convention $\int_{-\infty}^t \delta(t-t')f(t')dt' = \frac{1}{2}f(t)$, which accounts for the contribution of the delta function at the integration boundary. We thus obtain

$$\int b(\omega, t)d\omega = b_{\text{in}}(t) - 2\pi i \frac{\sqrt{\kappa/(2\pi)}}{2} a(t).$$

Similarly, by propagating back from $t_1 \rightarrow +\infty$ using $b_{\text{out}}(\omega, t_1)$, one finds

$$\int b(\omega, t)d\omega = b_{\text{out}}(t) + 2\pi i \frac{\sqrt{\kappa/(2\pi)}}{2} a(t).$$

Substituting these expressions into the cavity equation of motion, we obtain

$$\begin{aligned}\dot{a}(t) &= (\dot{a}(t))_{\text{sys}} - i\sqrt{\frac{\kappa}{2\pi}}b_{\text{in}}(t) - \frac{\kappa}{2}a(t), \\ \dot{a}(t) &= (\dot{a}(t))_{\text{sys}} - i\sqrt{\frac{\kappa}{2\pi}}b_{\text{out}}(t) + \frac{\kappa}{2}a(t), \\ a(t) &= \frac{i\sqrt{\kappa/(2\pi)}}{\kappa}(b_{\text{out}}(t) - b_{\text{in}}(t)).\end{aligned}$$

To proceed to the frequency domain, we adopt the Fourier transform conventions

$$\begin{aligned}\mathcal{F}[f(t)](\omega) &= \frac{1}{\sqrt{2\pi}} \int f(t)e^{i\omega t}dt, \\ \mathcal{F}^{-1}[f(\omega)](t) &= \frac{1}{\sqrt{2\pi}} \int f(\omega)e^{-i\omega t}d\omega,\end{aligned}$$

this is a minus sign different from the conventional Fourier transform conventions.

Assuming $a(t)$ vanishes at $t \rightarrow \pm\infty$, we have $\mathcal{F}[\dot{a}(t)](\omega) = -i\omega a(\omega)$. We

further assume that the system contribution in frequency space is linear in $a(\omega)$,

$\mathcal{F}[(\dot{a}(t))_{\text{sys}}](\omega) = Aa(\omega)$, where A is, in general, a complex coefficient determined by the system Hamiltonian (it may contain detuning and damping terms intrinsic to the cavity). One finds after Fourier transformation

$$\frac{b_{\text{out}}(\omega)}{b_{\text{in}}(\omega)} = \frac{-i\omega - A - \frac{\kappa}{2}}{-i\omega - A + \frac{\kappa}{2}} = 1 - \frac{1}{-i\omega - A + \frac{\kappa}{2}}.$$

For a two-level atom case, assume weak excitation $(\rho_{ee} - \rho_{gg}) \approx -1$,

$$H_{\text{sys}} = \omega_a a^\dagger a + \omega_e |e\rangle \langle e| + g(a|e\rangle \langle g| + a^\dagger |g\rangle \langle e|),$$

$$(\dot{a})_{\text{sys}} = (-i\omega_a - \frac{\kappa_0}{2})a - ig\rho_{eg},$$

$$\dot{\rho}_{eg} = (-i\omega_e - \frac{\Gamma}{2})\rho_{eg} + ig a(\rho_{ee} - \rho_{gg}).$$

Then we have

$$A = -i\omega_a - \frac{\kappa_0}{2} - \frac{g^2}{i(-\omega + \omega_e) + \frac{\Gamma}{2}},$$

and at resonant we have

$$\left| \frac{b_{\text{out}}(\omega = \omega_e = \omega_a)}{b_{\text{in}}(\omega = \omega_e = \omega_a)} \right|^2 = \left| 1 - \frac{1}{\frac{2g^2}{\Gamma(\kappa_0 + \kappa)} + 1} \right|^2 = \left| \frac{C}{C + 1} \right|^2.$$

Therefore for a high cooperativity system, the probe experiences the same phase as a large detuning case.

C.4 Probe atom

In the good-cavity limit, as we discussed before, it is easier to probe the atom rather than the cavity, since the cavity shows no dip. A photonic cavity coupled to a two-level system can be viewed as a generalization of two damped coupled oscillators given weak excitation. Here we derive, in more detail, how a phononic cavity interacts with a two-level system. We consider two scenarios: (i) an OMIT-type interaction, in which two laser fields couple two phonon states to the excited state; and (ii) phonon-assisted PLE, which uses only a single laser field. While the former seems preferable, because it does not consume phonons (so no phonon driving is required) and yields a higher base count rate by operating on the zero-phonon transition, it still requires the weak-excitation regime to preserve the dip contrast, otherwise the two-level system saturates and the harmonic-oscillator approximation breaks down. And we will also show that the OMIT-type interaction is indeed coupled oscillators system while we drive and probe the atom mode, and phonon-assisted PLE is the system we drive cavity mode but probe atom mode.

C.4.1 OMIT-system effective Hamiltonian

With phonon in Hamiltonian

$$H = \omega_m b^\dagger b + g_m (b^\dagger + b) |e\rangle \langle e| + \left(\left(\frac{\Omega_1}{2} + \frac{\Omega_2 e^{i\Delta t}}{2} \right) |e\rangle \langle g| + h.c. \right),$$

after Schrieffer-Wolff transformation

$$U = \exp \left(\frac{g_m}{\omega_m} (b^\dagger - b) |e\rangle \langle e| \right),$$

then $\tilde{H} = U H U^\dagger$, and use the Baker–Campbell–Hausdorff formula that $e^A B e^{-A} = \sum_{n=0}^{\infty} \overbrace{[A, \dots [A, [A, B]] \dots]}^n / n!$,

$$\begin{aligned} U (\omega_m b^\dagger b) U^\dagger &= \omega_m b^\dagger b - g_m (b^\dagger + b) |e\rangle\langle e| + \frac{g_m^2}{\omega_m} |e\rangle\langle e|, \\ U (g_m (b^\dagger + b) |e\rangle\langle e|) U^\dagger &= g_m (b^\dagger + b) |e\rangle\langle e| - 2 \frac{g_m^2}{\omega_m} |e\rangle\langle e|, \\ U \left(\frac{\Omega}{2} (|e\rangle\langle g| + |g\rangle\langle e|) \right) U^\dagger &= \frac{\Omega}{2} (e^{\frac{g_m}{\omega_m} (b^\dagger - b)} |e\rangle\langle g| + e^{\frac{g_m}{\omega_m} (b - b^\dagger)} |g\rangle\langle e|), \\ \tilde{H} &= \omega_m b^\dagger b - \frac{g_m^2}{\omega_m} |e\rangle\langle e| + \left(\left(\frac{\Omega_1}{2} + \frac{\Omega_2 e^{i\Delta t}}{2} \right) e^{\frac{g_m}{\omega_m} (b^\dagger - b)} |e\rangle\langle g| + h.c. \right). \end{aligned}$$

We apply a rotating frame to remove $\omega_m b^\dagger b - \frac{g_m^2}{\omega_m} |e\rangle\langle e|$, and expand in Lamb-Dicke regime (ignore multi-phonon process and $\frac{g_m^2}{\omega_m}$ terms and RWA)

$$\begin{aligned} \tilde{H} &= \left(\frac{\Omega_1}{2} + \frac{\Omega_2 e^{i\Delta t}}{2} \right) e^{i \frac{g_m^2}{\omega_m} t} e^{\frac{g_m}{\omega_m} (e^{i\omega_m t} b^\dagger - e^{-i\omega_m t} b)} |e\rangle\langle g| + h.c., \\ \tilde{H} &\approx \frac{\Omega_1}{2} |e\rangle\langle g| + \frac{\Omega_{\text{eff}}}{2} e^{i\delta t} b |e\rangle\langle g| + h.c., \end{aligned}$$

$\delta = \Delta - \omega_m$, and $\Omega_{\text{eff}} = \Omega_2 g_m / \omega_m$. Then transform again to eliminate time dependent term,

$$H = \delta b^\dagger b + \frac{\Omega_1}{2} |e\rangle\langle g| + \frac{\Omega_{\text{eff}}}{2} b |e\rangle\langle g| + \frac{\Omega_1}{2} |g\rangle\langle e| + \frac{\Omega_{\text{eff}}}{2} b^\dagger |g\rangle\langle e|,$$

and

$$\partial_t \rho = -i[H, \rho] + \Gamma \mathcal{D}(|g\rangle\langle e|) \rho + \gamma_u \mathcal{D}(b^\dagger) \rho + \gamma_d \mathcal{D}(b) \rho,$$

where the $\frac{g_m^2}{\omega_m}$ terms in Lindblad operator after rotating frame is also dropped, and $\gamma_u = n_{th} \kappa$, $\gamma_d = (n_{th} + 1) \kappa$.

C.4.2 OMIT-system exact solution

Here we try to solve the system exactly without weak excitation approximation. Often time we convert the equations of motion of operators to the equations of motion of complex numbers (expectation values) by taking expectation

for both sides of equations. This works fine for linear coupled equations, but here due to the nonlinearity there will be terms like $b^\dagger(\rho_{ee} - \rho_{gg})$. However given the coupling between atom and cavity is weak compared to the decay channel, $\Omega_{\text{eff}} \ll (\Gamma + \gamma_u + \gamma_d)$ then we can remove the correlation between systems and $\rho(t) \approx \rho_{\text{atom}}(t) \otimes \rho_{\text{cavity}}(t)$. Then

$$\dot{\rho}_{\text{atom}} = -i\left[\frac{\Omega_1}{2}|e\rangle\langle g| + \frac{\Omega_{\text{eff}}}{2}\langle b|e\rangle\langle g| + \frac{\Omega_1}{2}|g\rangle\langle e| + \frac{\Omega_{\text{eff}}}{2}\langle b^\dagger|g\rangle\langle e|\right]\rho_{\text{atom}} + \Gamma\mathcal{D}(|g\rangle\langle e|)\rho_{\text{atom}},$$

$$\dot{O}_{\text{cavity}} = i\left[\delta b^\dagger b + \frac{\Omega_{\text{eff}}}{2}b\langle\rho_{ge}\rangle + \frac{\Omega_{\text{eff}}}{2}b^\dagger\langle\rho_{eg}\rangle, O_{\text{cavity}}\right] + \gamma_u\mathcal{D}^\dagger(b^\dagger)O_{\text{cavity}} + \gamma_d\mathcal{D}^\dagger(b)O_{\text{cavity}},$$

$$\mathcal{D}^\dagger(c)O = c^\dagger Oc - \{c^\dagger c, O\}/2 \text{ and } \mathcal{D}(c)O = cOc^\dagger - \{c^\dagger c, O\}/2, \langle\rho_{ge}\rangle = \langle|e\rangle\langle g|\rangle = \langle\sigma_{eg}\rangle,$$

$$\langle\dot{b}\rangle = (-i\delta - \frac{\kappa}{2})\langle b\rangle - i\frac{\Omega_{\text{eff}}}{2}\langle\rho_{eg}\rangle,$$

$$\langle\dot{\rho}_{ge}\rangle = -i\left(\frac{\Omega_1}{2} + \frac{\Omega_{\text{eff}}}{2}\langle b^\dagger\rangle\right)(\langle\rho_{ee}\rangle - \langle\rho_{gg}\rangle) - \frac{\Gamma}{2}\langle\rho_{ge}\rangle,$$

$$\langle\dot{\rho}_{ee}\rangle = i\left(\frac{\Omega_1}{2} + \frac{\Omega_{\text{eff}}}{2}\langle b^\dagger\rangle\right)\langle\rho_{eg}\rangle - i\left(\frac{\Omega_1}{2} + \frac{\Omega_{\text{eff}}}{2}\langle b\rangle\right)\langle\rho_{ge}\rangle - \Gamma\langle\rho_{ee}\rangle.$$

Solve for steady state solution, then we have

$$\rho_{ge} = \frac{-i(\frac{\Omega_1}{2} + \frac{\Omega_{\text{eff}}}{2}b^\dagger)(2\rho_{ee} - 1)}{\Gamma/2},$$

$$b = -i\frac{\frac{\Omega_{\text{eff}}}{2}\rho_{eg}}{i\delta + \frac{\kappa}{2}},$$

$$\rho_{ee} = \frac{i(\frac{\Omega_1}{2} + \frac{\Omega_{\text{eff}}}{2}b^\dagger)\rho_{eg} - i(\frac{\Omega_1}{2} + \frac{\Omega_{\text{eff}}}{2}b)\rho_{ge}}{\Gamma}.$$

We have

$$\rho_{ge} = \frac{-i\frac{\Omega_1}{2}(2\rho_{ee} - 1)}{\frac{\Gamma}{2} - \frac{(\frac{\Omega_{\text{eff}}}{2})^2}{\frac{\kappa}{2} - i\delta}(2\rho_{ee} - 1)},$$

$$\rho_{ee} = \frac{i\Omega_1}{2\Gamma}(\rho_{eg} - \rho_{ge}) - \frac{\kappa}{\Gamma} \frac{(\frac{\Omega_{\text{eff}}}{2})^2}{(\frac{\kappa}{2})^2 + \delta^2} |\rho_{ge}|^2.$$

With $a = (\delta/\kappa)^2$, $b = (\Omega_1/\Gamma)^2$, $C = \Omega_{\text{eff}}^2/\kappa\Gamma$, $x = 2\rho_{ee} - 1$, we can solve the system exactly as

$$\begin{aligned}\rho_{ge} &= \frac{-i\Omega_1 x(\kappa - 2i\delta)}{\Gamma\kappa - \Omega_{\text{eff}}^2 x - 2i\delta\Gamma}, \\ \rho_{eg} - \rho_{ge} &= \frac{2i\Omega_1 x(\Gamma\kappa^2 - \kappa\Omega_{\text{eff}}^2 x + 4\delta^2\Gamma)}{(\Gamma\kappa - \Omega_{\text{eff}}^2 x)^2 + 4(\delta\Gamma)^2}, \\ |\rho_{ge}|^2 &= \frac{\Omega_1^2 x^2(\kappa^2 + 4\delta^2)}{(\Gamma\kappa - \Omega_{\text{eff}}^2 x)^2 + 4(\delta\Gamma)^2}.\end{aligned}$$

After simplification,

$$(x+1)((1-Cx)^2 + 4a) = -2bx(4a+1),$$

$$C^2 x^3 + (C^2 - 2C)x^2 + (1 + 4a + 2b + 8ab - 2C)x + (1 + 4a) = 0.$$

According to Implicit function theorem, we can expand x in series of C near $C = 0$,

$$\begin{aligned}x &= -\frac{1}{1+2b} - \frac{4b}{(1+4a)(1+2b)^3}C + O(C^2), \\ \rho_{ee} &\approx \frac{b}{1+2b} - \frac{2b}{(1+4a)(1+2b)^3}C = \frac{b}{1+2b} - \frac{(\frac{\kappa}{2})^2}{(\frac{\kappa}{2})^2 + \delta^2} \frac{2b}{(1+2b)^3}C.\end{aligned}$$

Then the dip/peak contrast is $\sim 2C/(1 + 2(\Omega_1/\Gamma)^2)^2$, and full width $\sim \kappa$. We can see how the excitation Ω_1/Γ reduces the contrast by adding a factor $1/(1 + 2(\Omega_1/\Gamma)^2)^2$.

C.4.3 OMIT-system weak excitation

Another easier way to give the results to by recognize

$$H = \delta b^\dagger b + \frac{\Omega_1}{2}|e\rangle\langle g| + \frac{\Omega_{\text{eff}}}{2}b|e\rangle\langle g| + \frac{\Omega_1}{2}|g\rangle\langle e| + \frac{\Omega_{\text{eff}}}{2}b^\dagger|g\rangle\langle e|,$$

is basically

$$H = \delta b^\dagger b + \frac{\Omega_1}{2}(\sigma + \sigma^\dagger) + \frac{\Omega_{\text{eff}}}{2}(b\sigma^\dagger + b^\dagger\sigma),$$

under the weak excitation approximation that $\sigma_{ge} \approx \sigma$, where $\delta = \Delta - \omega_m$ is the frequency difference between detuning Δ of two driving fields and the mechanical

mode frequency ω_m . This is damped coupled oscillators in good cavity limit which we drive and probe the atom. More generally, there should be a $\Delta_1 |e\rangle \langle e|$ term in H to count the detuning between transition frequency ω_e and driving frequency ω_1 of Ω_1 , but since this term is effectively suppress by the large atom decay Γ therefore we drop this term for a simpler model. During the mode searching, we scan the relative frequency between two driving fields Δ and try to observe the narrow dip of mechanical mode of linewidth $\sim 2\kappa$ when $\delta = \Delta - \omega_m = 0$ requirement is satisfied.

C.4.4 Phonon assisted PLE

Follow a similar procedure as OMIT derivation, and under weak excitation approximation, we have

$$\begin{aligned}\dot{b} &= (-i\delta - \frac{\kappa}{2})b - i\frac{\Omega_{\text{eff}}}{2}\sigma + \Omega_b, \\ \dot{\sigma} &= (-i\Delta_2 - \frac{\Gamma}{2})\sigma - i\frac{\Omega_{\text{eff}}}{2}b,\end{aligned}$$

that an driving term Ω_b for phononic cavity is introduced, $\delta = \omega_m - \omega_\Omega$ is the detuning between mechanical frequency ω_m and driving frequency ω_Ω of Ω_b , and $\Delta_2 = \omega_e - \omega_m - \omega_2$ is the detuning between transition frequency $\omega_e - \omega_m$ and driving frequency ω_2 of Ω_2 , here a different rotating frame is used to make driving term time-independent. And solve for steady state solution, we get

$$\rho_{ee}(\delta) \approx |\sigma|^2 = \frac{16n_{\text{ph}}\Omega_b^2\Omega_{\text{eff}}^2}{\kappa^2\Gamma^2} \frac{1}{(1+C)^2 + (2\delta/\kappa)^2},$$

where $C = n_{\text{ph}}\Omega_{\text{eff}}^2/\kappa\Gamma$, this is the standard cavity QED results with $\sqrt{n_{\text{ph}}}$ enhanced Ω_{eff} as a consequence of average phonon occupancy. The phonon occupation can be estimated by modelling specific system, in our case, it can be evaluated from [X. Li, Lekavicius, Noeckel, and Wang \(2024\)](#). In the experiment, we scan the driving frequency of Ω_b to try to meet the requirement $\delta = \omega_m - \omega_\Omega = 0$ where a peak of

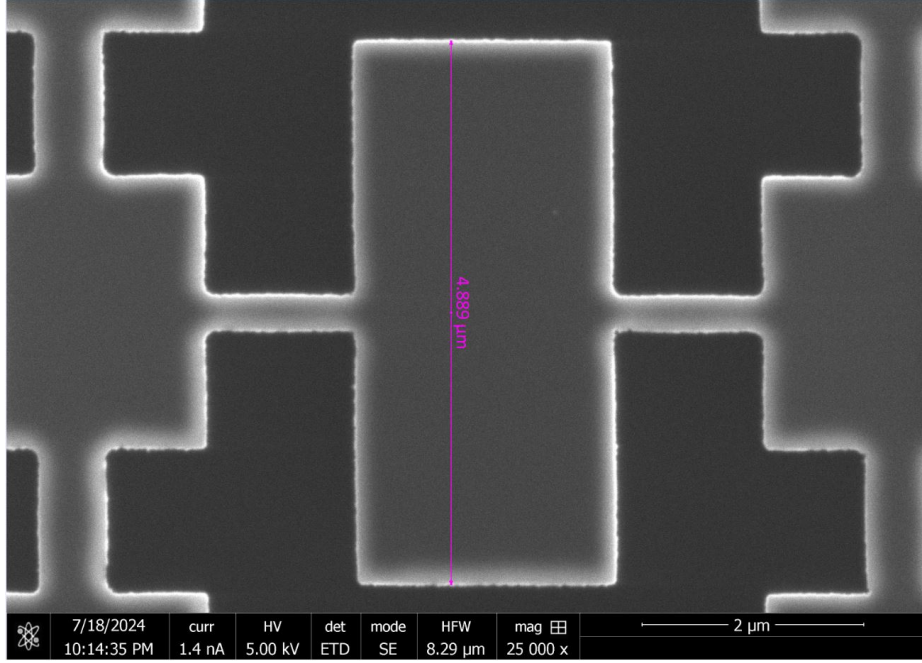


Figure C.1. SEM image of a 2GHz resonator.

linewidth $\sim 2\kappa$ is observed, while a small detuning Δ_2 has negligible effect on the results.

C.5 Phononic cavity with compression mode of membrane

Using the phonon assisted PLE approach discussed before, we are able to search for the mode frequency of phononic resonator. Since the ρ_{ee} is function of phonon occupation, when we scan the phononic driving frequency across the resonance frequency we will observe a sharp peak of ρ_{ee} which has a linewidth given by the κ of resonator, that is at the order of $\sim 100\text{Hz}$.

While we can estimate the length therefore the eigenfrequency of such a resonator using SEM as shown in Figure C.1. It's hard to accurately determine frequency within 5% range, hence we need to scan $\sim 100\text{MHz}$ with $\sim 100\text{Hz}$ step size while keep exposure time sufficient for each point in order to resolve signal

from background. A typical scan will therefore take ~ 10 Hours, and any approaches can reduce overhead is beneficial.

To extract as much as possible information while minimizing exposure overhead, we used the sequential probability ratio test (SPRT) algorithm to implement the theoretical optimal scanning. This algorithm starts with a pair of hypotheses, $H_0 : p = p_0$ and $H_1 : p = p_1$ that state if the parameter being testes is likely be the value presetted. The algorihtm will keep collecting new data and add up information it carries until one of the hypotheses is reached, and the threshold can be varied by assigning different desired type I and type II errors α, β . In our experiment, the parameter being tested is the average photon count λ during duration τ , and photon counts n data follows Poisson distribution with mean λ .

The information from data is log-likelihood ratio

$$\ln \Lambda_i = n_i \ln \frac{\lambda_1}{\lambda_0} - (\lambda_1 - \lambda_0),$$

and we keep collecting for another τ duration until accumulated information is no longer within this range

$$\ln \frac{\beta}{1 - \alpha} < \sum_i \ln \Lambda_i < \ln \frac{1 - \beta}{\alpha}.$$

We set type II error rate β low $\sim 10\%$ because we don't want to miss the real signal, while we set type I error α lower to reduce fake signal as even 1% fake signal from our 10^6 data points can make results unclear. This SPRT is proven to be the best algorithm which satisfies given error rates α, β .

One example of mode obtained with 1GHz resonator is shown in [Figure C.2](#), gives resonant frequency at 976.66MHz. And the triple peaks come from the fact that we do not drive the mechanical mode using a CW laser but instead a pulsed

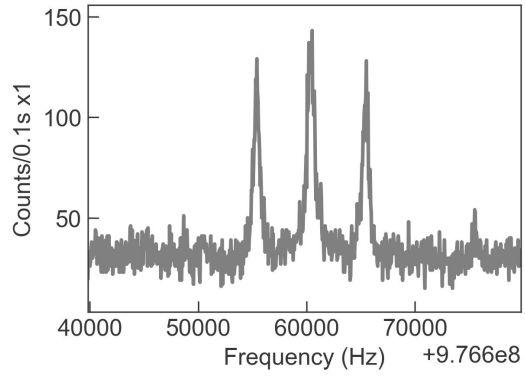


Figure C.2. One example of obtained phononic cavity mode.

pattern to minimize the effect on PLE due to 1550nm. This makes the driving not a single frequency but in principle contains infinite many components.

APPENDIX D

ADIABATIC POPULATION TRANSFER OF DRESSED SPIN STATES WITH QUANTUM OPTIMAL CONTROL

The results presented in this chapter were published in [S.-H. Wu et al. \(2019\)](#).

D.1 Introduction

Stimulated Raman adiabatic passage (STIRAP) has been extensively used for quantum control of three-level systems, in particular, Λ -type systems that feature two lower spin states with robust spin coherence [Bergmann, Theuer, and Shore \(1998\)](#); [Bergmann, Vitanov, and Shore \(2015\)](#); [Vitanov, Rangelov, Shore, and Bergmann \(2017\)](#). In STIRAP, the quantum system evolves adiabatically, following a dark instantaneous eigenstate, which is a coherent superposition of the two lower states controlled by two external driving pulses with a counterintuitive pulse sequence. The fidelity of the STIRAP process is limited by the finite lifetime of the spin coherence and, for many solid-state spin systems, spin dephasing induced by a slowly fluctuating environment. Modifications to STIRAP have been pursued in order to speed up the relatively slow adiabatic passage [Berry \(2009\)](#); [Demirplak and Rice \(2003, 2008\)](#). One approach uses an auxiliary pulse to induce coupling between the two lower states [Chen, Lizuain, Ruschhaupt, Guéry-Odelin, and Muga \(2010\)](#); [Unanyan, Yatsenko, Bergmann, and Shore \(1997\)](#). This counter-diabatic coupling can be designed to cancel precisely nonadiabatic transitions between the instantaneous eigenstates induced by the two driving pulses, leading to a net transitionless driving. Another approach uses specially-designed temporal shapes for the two driving pulses to achieve superadiabatic transitionless driving (SATD). This approach can be implemented in the limit of either zero dipole detuning or

large dipole detuning [Baksic, Ribeiro, and Clerk \(2016\)](#); [Chen and Muga \(2012\)](#); [Du et al. \(2016\)](#). In addition, systematically-corrected control pulses using Magnus expansion techniques have also been proposed for the achievement of transitionless driving [Ribeiro, Baksic, and Clerk \(2017\)](#).

Both conventional STIRAP and SATD STIRAP have been applied to negatively-charged nitrogen vacancy (NV) centers in diamond [Golter and Wang \(2014\)](#); [Zhou et al. \(2017\)](#), a solid state spin system that has recently emerged as a promising qubit system for quantum information processing [Awschalom, Bassett, Dzurak, Hu, and Petta \(2013\)](#); [Awschalom et al. \(2018\)](#); [Childress et al. \(2014\)](#); [Doherty et al. \(2013\)](#). SATD STIRAP has also been applied to a three-level superconducting circuit [Vepsäläinen, Danilin, and Paraoanu \(2019\)](#). In spite of these successful experimental demonstrations, the transfer efficiency observed in the NV experiments are far from ideal. This is by no means surprising. Although spin decoherence time exceeding 1 ms has been observed in isotopically purified diamond samples [Balasubramanian et al. \(2009\)](#), NV centers in diamond samples with natural abundance of ^{13}C are subject to slow fluctuations of the ^{13}C nuclear spins. This nuclear spin bath leads to T_2^* , the effective spin dephasing time, on the order of 1 μs . This spin dephasing process can be suppressed with dynamical decoupling techniques [de Lange et al. \(2010\)](#). The sequence of π -pulses used in dynamical decoupling, however, is incompatible with adiabatic passage. Alternatively, the spin dephasing process can be suppressed with the use of dressed spin states, using microwave fields to dress the electron spin states [Golter et al. \(2014\)](#); [Rabl et al. \(2009\)](#); [Xu et al. \(2012\)](#).

In this chapter, we report theoretical studies of adiabatic population transfer using dressed spin states. To avoid the nonadiabatic excitation of the

upper state in the Λ -type three-level system, we have implemented the adiabatic population transfer with a large optical detuning. The implementation is based on quantum optimal control, specifically, the quantum optimization technique of Chopped Random Basis (CRAB) [Caneva, Calarco, and Montangero \(2011\)](#); [Doria, Calarco, and Montangero \(2011\)](#), a recently developed numerical algorithm for quantum optimal control [Glaser et al. \(2015\)](#); [Werschnik and Gross \(2007\)](#); [Yuan, Koch, Salamon, and Tannor \(2012\)](#). We show that for diamond with natural ^{13}C abundancy, transfer efficiency exceeding 97% can be achieved with the CRAB-optimized temporal pulse lineshapes and with the use of the dressed spin states. Overall, these studies demonstrate that dressed spin states are highly effective in overcoming detrimental effects of spin dephasing in the adiabatic passage process and that the CRAB algorithm can provide a simple and yet powerful technique for quantum optimal control of the adiabatic passage.

D.2 Theoretical results

D.2.1 Λ -type systems and dressed spin states

For a NV center in diamond, either the E_y or E_x excited state can serve as the upper state, $|e\rangle$, and the $m_s = 0$ and the $m_s = -1$ (or $m_s = 1$) states can serve as the two lower spin states of a Λ -type three-level system, as shown schematically in [Figure D.1a](#). The dipole optical transition between E_y (or E_x) state and the $m_s = -1$ state can be enabled by strain induced excited-state mixing or the excited state level anti-crossing, as demonstrated in earlier studies [Santori et al. \(2006\)](#); [Yale et al. \(2013\)](#). Coherent population trapping (CPT) in this Λ -type system has already been demonstrated in a number of earlier experiments [Golter et al. \(2016\)](#); [Santori et al. \(2006\)](#); [Yale et al. \(2013\)](#). To use dressed spin states to suppress effects of spin dephasing, we apply a strong and resonant

microwave (MW) field, with frequency ω_B , between the two lower spin states. The corresponding Hamiltonian in the rotating frame is described by

$$\bar{H}_{mw} = \hbar\delta |-1\rangle \langle -1| + \hbar\frac{\Omega_0}{2}(|0\rangle \langle -1| + |-1\rangle \langle 0|),$$

where δ is the bath-induced energy fluctuation of the $|-1\rangle$ state and Ω_0 is the Rabi frequency for the MW driving field. The eigenstates of this Hamiltonian are the semiclassical dressed states given by

$$\begin{aligned} |+\rangle &= \sin\theta_0 |0\rangle + \cos\theta_0 |-1\rangle, \\ |-\rangle &= \cos\theta_0 |0\rangle - \sin\theta_0 |-1\rangle, \end{aligned}$$

where $\theta_0 = \tan^{-1}[\Omega_0/(\sqrt{\Omega_0^2 + \delta^2} + \delta)]$. The corresponding eigenvalue is $E_{\pm} = \pm\hbar\sqrt{\Omega_0^2 + \delta^2}/2$. The energy of the dressed spin states can be immune to the fluctuations of the nuclear spin bath if $\Omega_0 \gg |\delta|$. For the numerical calculations, we have set $\Omega_0/2\pi = 50$ MHz. These dressed spin states will be used as the two lower states of the new Λ -type system, as illustrated schematically in [Figure D.1b](#).

For adiabatic passage between the two dressed spin states, we start with an initial state,

$$|\psi\rangle_i = (|0\rangle - |-1\rangle)/\sqrt{2}.$$

To prepare the system in this initial state, we first initialize the NV center in the $m_s = 0$ state and then use a $\pi/2$ pulse to drive the NV into $|\psi\rangle_i$, as shown schematically in [Figure D.1c](#). The final state or the target state is given by

$$|\psi(t)\rangle_f = (|0\rangle + e^{-i\omega_B t} |-1\rangle)/\sqrt{2}.$$

Following the adiabatic passage process along with a suitable waiting period, $|\psi(t)\rangle_f$ can be driven to $|-1\rangle$ with a $\pi/2$ pulse for the experimental state detection, as indicated in [Figure D.1c](#).

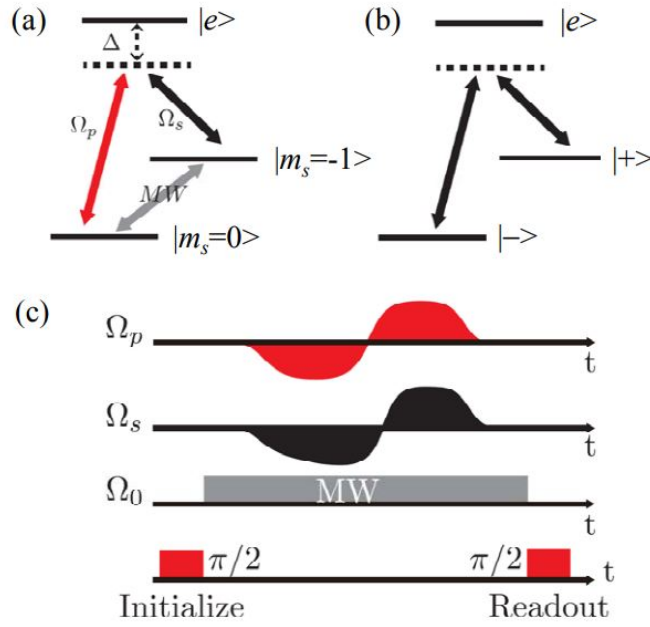


Figure D.1. (a) Schematic of a Λ -type three level system in a NV center, for which the two lower states can also couple to a strong and resonant microwave field. (b) Schematic of the resulting Λ -type three level system with the dressed spin states as the two lower states. (c) Schematic of the pulse sequence for adiabatic population transfer of dressed spin states. The $\pi/2$ microwave pulses are used for the preparation and readout of dressed spin states.

D.2.2 Adiabatic population transfer

For adiabatic population transfer, the overall Hamiltonian is given by $H = H_0 + H_{mw} + V$, where $H_0 = \hbar\omega_e |e\rangle \langle e| + \hbar(\delta + \omega_B) |-1\rangle \langle -1|$ is the Hamiltonian for the three-level system and H_{mw} is the microwave interaction Hamiltonian in the Schrödinger picture. The optical interaction Hamiltonian is given by

$$V = \frac{\hbar\Omega_s(t)}{2}(|-1\rangle \langle e| e^{i\omega_s t} + h.c.) + \frac{\hbar\Omega_p(t)}{2}(|0\rangle \langle e| e^{i\omega_p t} + h.c.),$$

where $\Omega_s(t)$, $\Omega_p(t)$, ω_s , and ω_p are the Rabi frequencies and optical frequencies for the two optical driving fields, respectively. While a set of density matrix equations with the dressed-state basis can be used, these equations, which include unusual decay terms [Berman and Malinovsky \(2011\)](#), are more difficult to use than the much simpler density matrix equations for the bare states. In this regard, we have carried out numerical calculations of the adiabatic passage process using the Lindblad master equation for the bare states, which includes explicitly interactions of both MW and optical fields as well as the population decay rate for $|e\rangle$, Γ , and the spin decoherence rate, γ_s . The Lindblad master equation used is given by

$$\dot{\rho}(t) = -\frac{i}{\hbar}[H, \rho(t)] + \sum_k [L_k \rho(t) L_k^\dagger - \frac{1}{2}\{L_k L_k^\dagger, \rho(t)\}],$$

where $L_0 = \sqrt{\Gamma/2} |0\rangle \langle e|$, $L_1 = \sqrt{\Gamma/2} |-1\rangle \langle e|$ and $L_3 = \sqrt{\gamma_s}(|-1\rangle \langle -1| - |0\rangle \langle 0|)$ are the jump operators. Note that the Raman resonant or two-photon resonant condition (with $\delta = 0$) for the adiabatic passage of the relevant dressed spin states is given by $\omega_p - \omega_s = \omega_B + \Omega_0$, where the formation of the MW-driven dressed states leads to a corresponding shift in the Raman resonant condition.

An important constraint in the adiabatic population transfer between dressed spin states is that Ω_{eff} , the effective Rabi frequency between the two spin states induced by the external optical fields, needs to be small compared with Ω_0 ,

the MW Rabi frequency used for creating the dressed states. This is because power broadening of the effective two-level transition can lead to undesirable excitations of nearby dressed states. In addition, in the limit of large dipole optical detuning, the effective optically-induced decay rate due to the nonadiabatic excitation of the excited state is $(\Omega/2\Delta)^2\Gamma$, where Ω is the relevant optical Rabi frequency and Δ is the dipole detuning, with $\Gamma/2\pi = 14$ MHz for NV centers in diamond [Tamarat et al. \(2006\)](#). Given the above two constraints, we set the maximum optical Rabi frequency, $\Omega_s/2\pi$ and $\Omega_p/2\pi$, to be $\sqrt{2} \times 100$ MHz, along with $\Delta/2\pi > 1.5$ GHz.

D.2.3 CRAB and far off-resonant SATD

For adiabatic population transfer with CRAB-based quantum optimal control, we optimize the transfer efficiency from the initial to the target states by varying the temporal pulse lineshapes of the two external optical fields, with a fixed overall duration for the transfer process and a fixed dipole optical detuning. We also constrain the optimization process on a maximum optical Rabi frequency. The numerical implementation of the CRAB algorithm has been carried out with the use of Quantum Toolbox in Python (QuTiP), an open-source software for simulating the dynamics of open quantum systems.

For comparison, we have also carried out theoretical calculations using the far offresonant SATD scheme. The far off-resonant SATD approximates the Λ -type three-level system as an effective two-level system in the limit of large dipole detuning. For a Λ -type three-level system with $\Delta = \omega_e - \omega_p = \omega_e - \omega_B - \omega_s$ and with $\Omega_0 = \delta = 0$, the effective Rabi frequency, Ω_{eff} , between the two lower states is given by

$$\Omega_{\text{eff}} = \frac{\Omega_s(t)\Omega_p(t)}{2\Delta}.$$

The effective detuning, Δ_{eff} , for the two-level transition is

$$\Delta_{\text{eff}} = \frac{\Omega_p^2(t) - \Omega_s^2(t)}{4\Delta}.$$

Solving the above two equations for Ω_s and Ω_p , we can express the Rabi frequencies of the external optical fields in terms of the effective Rabi frequency and the effective detuning,

$$\begin{aligned}\Omega_p(t) &= [2\Delta(\sqrt{\Omega_{\text{eff}}^2 + \Delta_{\text{eff}}^2} + \Delta_{\text{eff}})]^{1/2}, \\ \Omega_s(t) &= [2\Delta(\sqrt{\Omega_{\text{eff}}^2 + \Delta_{\text{eff}}^2} - \Delta_{\text{eff}})]^{1/2}.\end{aligned}\tag{D.1}$$

A counter diabatic coupling can be applied between the two lower states such that the overall system can remain in the instantaneous dark state, with the Rabi frequency for the counter diabatic coupling given by [Du et al. \(2016\)](#)

$$\Omega_a = \frac{2[\dot{\Omega}_p(t)\Omega_s(t) - \Omega_p(t)\dot{\Omega}_s(t)]}{\Omega_p^2(t) + \Omega_s^2(t)}.$$

For the far off-resonant SATD, the counter diabatic coupling can be implemented by a suitable modification of both Ω_{eff} and Δ_{eff} . As shown in the earlier study [Du et al. \(2016\)](#), the modification is

$$\begin{aligned}\tilde{\Omega}_{\text{eff}}(t) &= \sqrt{\Omega_{\text{eff}}^2(t) + \Omega_a^2(t)}, \\ \tilde{\Delta}_{\text{eff}}(t) &= \Delta_{\text{eff}}(t) + \frac{d}{dt} \tan^{-1}\left(\frac{\Omega_a(t)}{\Omega_{\text{eff}}(t)}\right).\end{aligned}$$

Using [Equation D.1](#), we can then derive the modified Rabi frequencies for the two external optical fields, $\tilde{\Omega}_s(t)$ and $\tilde{\Omega}_p(t)$.

[Figure D.2](#) shows the numerical calculations obtained for bare spin states ($\Omega_0 = 0$), for which we have assumed $\delta = 0$ (no nuclear spin bath), a spin decoherence rate $\gamma_s/2\pi = 1$ kHz, and $\Gamma/2\pi = 14$ MHz. [Figure D.2a](#) plots the temporal pulse lineshapes derived for the far off-resonant SATD. [Figure D.2b](#) shows the temporal pulse lineshapes obtained with quantum optimal control using the

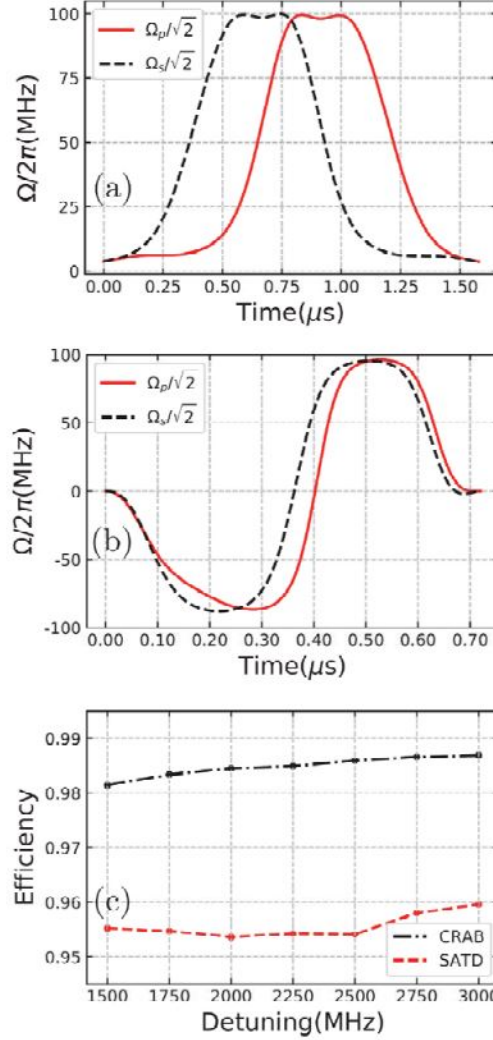


Figure D.2. (a) Temporal pulse lineshapes of the two external optical fields for the faroff-resonant SATD, with dipole detuning $\Delta = 3$ GHz. Ω is the Rabi frequency. (b) Temporal pulse lineshapes of the two external optical fields optimized with the CRAB algorithm, with $\Delta = 3$ GHz. (c) Adiabatic population transfer efficiency of dressed spin states as a function of $\Delta/2\pi$, obtained with the far off-resonant SATD (dashed line) and CRAB (dash-dotted line) schemes. No effects of spin dephasing due to the nuclear spin bath are included.

CRAB algorithm. A striking feature of the CRAB temporal pulse lineshapes is that the overall pulse area for each optical pulse is near zero. [Figure D.2c](#) shows the adiabatic transfer efficiencies obtained with the far off-resonant SATD and those obtained with the CRAB algorithm.

As shown in [Figure D.2c](#), the transfer efficiency increases slightly with increasing detuning, which is expected since a large dipole detuning reduces nonadiabatic excitation of the upper state population. Furthermore, the transfer efficiency obtained with the CRAB algorithm is considerably greater than that obtained with far off-resonant SATD, since the far off-resonant SATD is not optimal for the detuning used for [Figure D.2](#).

D.2.4 Suppressing effects of spin dephasing with dressed spin states

Since the nuclear spin bath fluctuates at a relatively slow timescale, we assume that for the duration of the adiabatic population transfer, E_{-1} , which is the energy level of state $|-1\rangle$, remains unchanged. To describe spin dephasing induced by the nuclear spin bath, we let E_{-1} take a value according to a normal distribution, with

$$E_{-1} = \hbar(\omega_B + \delta) = \hbar[\omega_B + \mathcal{N}(0, \sigma^2)], \quad (\text{D.2})$$

where $\mathcal{N}(0, \sigma^2)$ is a normal distribution with mean=0 and variance= σ^2 . For each σ , we ran a large number of simulations with the relevant detuning adjusted according to [Equation D.2](#). Effects of spin dephasing are investigated with $\sigma/2\pi$ varied from 0 to 2 MHz. Numerical calculations were carried out for the dressed spin states with $\Omega_0/2\pi = 50$ MHz and also for bare states (with $\Omega_0 = 0$), for which we calculate directly the transfer efficiency from state $|0\rangle$ to state $|-1\rangle$.

[Figure D.3a](#) compares the adiabatic transfer efficiencies obtained for bare and dressed spin states as a function of σ and with CRAB-based quantum optimal

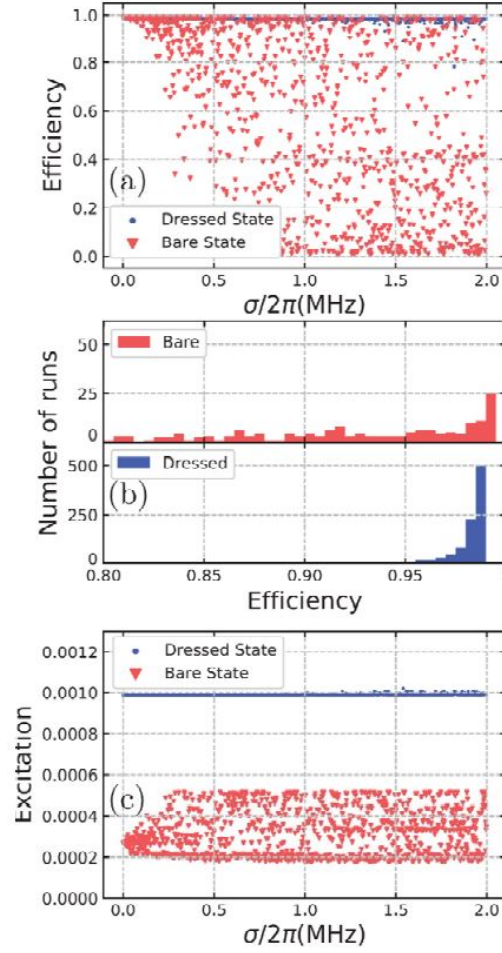


Figure D.3. (a) Adiabatic population transfer efficiency for dressed spin states and bare spin states obtained with the CRAB algorithm and with $\Delta = 3$ GHz, as $\sigma/2\pi$ is varied from 0 to 2 MHz. For each data point, E_{-1} takes a value according to the normal distribution given in Equation D.2. (b) The distribution of the adiabatic population transfer efficiency for dressed spin states and bare spin states (with $\Delta = 3$ GHz). A total number of 1000 simulation runs with $\sigma/2\pi = 2$ MHz are plotted for each distribution. (c) Nonadiabatic excitation of the excited state for the simulation runs shown in (a).

control. Identical optical temporal lineshapes are used for both the dressed state and bare spin state calculations. Similar to [Figure D.2b](#), the duration of the population transfer is set to $0.72 \mu\text{s}$. A maximum Rabi frequency of $\Omega/2\pi = 2 \times 100$ MHz is used for the dressed spin states, while a maximum Rabi frequency of $\Omega/2\pi = 100$ MHz is used for the bare spin states. This is because in order to have the same Ω_{eff} for the bare and the dressed spin states, Ω_s and Ω_p used for the dressed spin states needs to be a factor of $\sqrt{2}$ of those used for the bare spin states.

For the bare spin states, the transfer efficiency displays large fluctuations as σ increases, which is expected because of the corresponding fluctuations in the relevant Raman detuning. In comparison, the transfer efficiency remains nearly 98% for the dressed spin states, demonstrating nearly complete suppression of the effects of spin dephasing on the adiabatic population transfer. [Figure D.3b](#) shows the histogram of a theoretical simulation, for which we took $\sigma/2\pi = 2$ MHz and carried out 1000 calculation runs with E_{-1} taking a value according to the normal distribution given in [Equation D.2](#). As shown in the histogram, the population transfer efficiency for the bare states exhibits a broad distribution ranging from 0.8 to greater than 0.99. In contrast, for the dressed spin states, the distribution of the population transfer efficiency is characterized by a sharp peak centered near 0.98. The width of the peak is less than 1%, which again confirms the nearly complete suppression of the effects of spin dephasing on adiabatic population transfer by the dressed spin states.

It should be noted that the dressed spin states exhibit a slightly greater nonadiabatic excitation of the excited states than the corresponding bare states, as shown in [Figure D.3c](#). This is due to the fact that greater optical Rabi frequencies are used for the dressed spin states. In addition, the excited state population

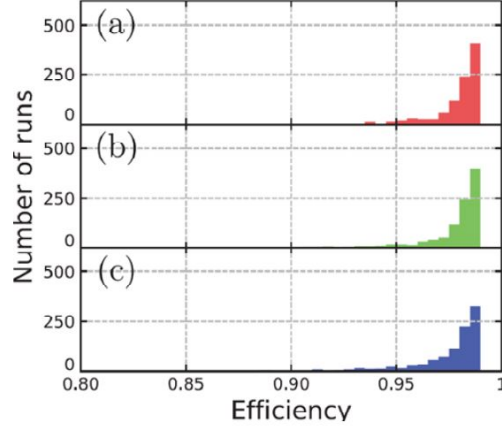


Figure D.4. The distribution of the adiabatic population transfer efficiency for dressed spin states. A total number of 1000 simulation runs with $\sigma/2\pi = 2$ MHz are plotted for each distribution. From the top to bottom, effects of spin dephasing are including in initialization and transfer, in readout and transfer, and in initialization, transfer, and readout, respectively. Other conditions are the same as those used in [Figure D.3b](#).

is nearly independent σ for the dressed states, but shows some variation with increasing σ for the bare states, as shown in [Figure D.3c](#).

The theoretical results shown in [Figure D.3a](#) and [b](#) assume perfect state initialization and readout for the dressed spin states. Both of these steps, however, are also subject to spin dephasing. [Figure D.4](#) shows the histogram of the transfer efficiency obtained under the same condition as that used for [Figure D.3b](#), except that effects of spin dephasing on the state initialization and readout are included in [Figure D.4a](#) and [b](#), respectively. For [Figure D.4c](#), effects of spin dephasing on the entire process including state initialization, transfer, and readout are included. The average efficiency obtained from [Figure D.4a](#), [b](#), and [c](#) are 97.4%, 97.43%, 97.06%, respectively. In comparison, the average efficiency obtained from [Figure D.3b](#) is 97.72%.

D.3 Conclusion

In conclusion, theoretical studies using nitrogen vacancy centers in diamond as a model system demonstrate that dressed spin states can be a highly effective approach for suppressing or circumventing effects of spin dephasing in adiabatic population transfer. The numerical simulations also show that the simple chopped random basis algorithm is a powerful tool for optimal quantum control, enabling efficient and robust adiabatic population transfer. These results should stimulate further experimental efforts on adiabatic population transfer in solid state spin systems.

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