

THE UNIVERSITY OF CHICAGO

COHERENCE AND COUPLING OF DRIVEN SPINS IN SILICON CARBIDE

A DISSERTATION SUBMITTED TO
THE FACULTY OF THE PRITZKER SCHOOL OF MOLECULAR ENGINEERING
IN CANDIDACY FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY

BY
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ABSTRACT

Selectively interfacing solid-state defect electron spins to desired control mechanisms and quantum systems is an ongoing challenge in quantum science, particularly in the face of ubiquitous environmental noise. We demonstrate a selection of emergent properties in a remarkably coherent solid-state spin system, the silicon carbide divacancy, when subjected to periodic drives. These properties give rise to new interaction channels and substantially increased protection from environmental fluctuations. In particular, we establish electrically driven coherent quantum interference by applying microwave-frequency electric fields to drive the divacancy’s highly responsive excited-state orbitals and induce Landau-Zener-Stückelberg interference fringes. When combined with the divacancy’s highly coherent spin and optical subsystems, this interaction can serve as the basis for a spin-to-photon interface in the microwave regime. Then, by applying a continuous microwave-frequency drive resonant with two of the three ground-state spin levels of the divacancy, we construct a robust qubit embedded in a decoherence-protected subspace. The qubit is protected from magnetic, electric, and temperature fluctuations, which account for nearly all relevant decoherence channels in the solid state. This culminates in an increase of the qubit’s inhomogeneous dephasing time by over four orders of magnitude (to >22 milliseconds), while its Hahn-echo coherence time approaches 64 milliseconds. Both of these developments elevate the silicon carbide divacancy defect above its already promising status as a platform for quantum information processing, and open the door to real-world applications requiring robust and controllable quantum properties in a technologically mature solid-state host.

CHAPTER 1

INTRODUCTION

1.1 Motivation for solid-state quantum systems

Over the past two decades, the rapid development of miniaturized high-speed electronics, coupled with broad improvements in cryogenic technology and material quality, has brought controllable quantum mechanical systems to the tabletops of many research and industrial laboratories. Since the late 1990s, we have witnessed the development of a vast range of architectures for implementing quantum bits (qubits) and the subsequent scaling to large, complex systems. In 1999, the first demonstration of a controllable quantized system in superconducting circuits was carried out in a Cooper-pair box [1]. Just twenty years later, arrays of superconducting qubits have been used to showcase a quantum advantage over classical computing systems [2]. Quantum dots, which were first proposed as a basis for quantum computation [3] in 1997, have undergone extensive development to yield a highly promising candidate system with excellent readout [4], manipulation [5, 6], and coherence properties [7]. Furthermore, multiple interacting trapped ions have been leveraged to create many-body quantum systems [8, 9] viable for quantum computation purposes.

Yet another class of systems focuses on naturally occurring quantum states in crystal hosts. These solid-state systems, typically found as lattice defects in semiconducting materials, have been studied for a large part of the twentieth century with gemological and classical electronics applications in mind. Only recently have these defect centers been isolated to perform quantum information processing. The prototypical example of solid-state defect centers, the nitrogen vacancy center in diamond, was first demonstrated in 1997 to have the necessary combination of properties to make it viable as a qubit system [10]. Since then, numerous defect systems in diverse hosts [11–21] have been shown to have similar collections of properties required for robust quantum states, but each with variations afforded by the unique nature of each defect configuration and host material.

With so many competing qubit modalities, it is worth asking why all the research attention has not been diverted to just one or two candidates. After all, for mainstream electronic devices, silicon reigns supreme over other materials, such as gallium arsenide or silicon germanium, despite the many advantages offered by these other materials. If the argument is that pre-existing infrastructure or research in silicon gives it an insurmountable advantage as a classical information processing platform, then a corollary could be expected to exist for quantum information processing. One way to address this inconsistency is to consider the two primary thrusts in quantum science on the path to large-scale, low-error systems: long quantum coherence lifetimes of the individual qubits in the system, and fast, yet controllable, interactions between two or more qubits. For a quantum technology to be viable, the constituent qubits typically need to have robustness along these two axes [3]. However, these two parameters cannot be maximized simultaneously in a trivial manner. Abstractly, to have a long-lived quantum system typically indicates that the system is not sensitive to interactions, regardless of whether the source is from a control mechanism or from undesired environmental noise [22]. Such a system would also be more difficult to interface with other quantum systems, as the resultant coupling strengths would be lower. Conversely, a system with qubits that can readily couple to each other will typically have shorter coherence lifetimes. This tradeoff, which is inherent to quantum systems and is not a large concern in classical systems, is a strong driver behind the diversity of quantum architectures. Some qubit modalities may be better suited to one application or another, depending on the need for long lifetimes or better coupling.

Superficially, the class of solid-state defect systems appears to have many shortcomings when compared to systems maximizing one of the two aforementioned parameters. For example, the electron spins in these defect systems are not as robust as isolated nuclear spins, with coherence times many orders of magnitude shorter [23]. This is generally attributed to the electron spins higher gyromagnetic ratio when compared to nuclear spins. Yet despite this high gyromagnetic ratio, it is relatively difficult to interface single electron spins with

each other when considering the required densities to achieve the necessary dipole-dipole interaction strengths [24]. However, these limitations are balanced and potentially outweighed by a slew of advantages afforded by the defect systems atomic nature and solid-state host. In particular, the crystal lattice confers opportunities to fabricate lithographically patterned structures around the quantum state, allowing for a variety of fine control mechanisms to be engineered and implemented [25–30]. The electronic band gap of the semiconducting hosts provide the isolation necessary for quantum phenomena to remain undisturbed, even potentially at room temperature or higher [11, 31–33]. The optical interface of these solid-state defects allow for the encoding of quantum states onto photons, which serve as the basis for long-distance quantum communication protocols [34, 35]. Finally, the natural origin of many of these defects means that their presence is typically intrinsic to many crystals, or can be introduced in a straightforward, controlled, and localized manner through implantation or irradiation techniques [36, 37]. These advantages stand out in particular when we consider the difficulty of implementing optical interfaces and room temperature operation in superconducting qubits and quantum dot systems.

Given these advantages, it is typically beneficial to work within a framework that leverages these intrinsic properties while addressing the shortcomings through engineering principles. For example, short inhomogeneous spin dephasing times can be extended to a significantly longer spin coherence time by utilizing control pulses to implement dynamical decoupling. Isotopic purification can be used to remove noise sources in the host material to further improve spin coherence properties [37–39]. Spin systems have been coupled to superconducting resonators, paving the way for potential scalable quantum information processing techniques [40, 41]. While these results still trail the state-of-the-art developments in other systems with better intrinsic properties for quantum coherence or qubit-qubit coupling, they demonstrate that improvements to these weaknesses are not limited by any law of physics, but instead by the resolution and finesse of the underlying material purity and engineering techniques.

In this thesis, we showcase silicon carbide, as both an excellent material in its own right and as a host to controllable quantum systems embedded within its lattice defects. In particular, we focus on the divacancy defect, which has been the subject of extensive research following the trail blazed by diamond nitrogen vacancy centers. The silicon carbide divacancy enjoys many of the excellent properties of diamond nitrogen vacancy centers such as optical addressing and a robust spin system. Furthermore, the silicon carbide host bestows many unique properties not realizable in diamond-based systems. However, just like many other solid-state defect systems, the same shortcomings apply – coupling mechanisms are still weak compared to other qubit modalities, and spin dephasing times are short in the presence of nuclear spin noise and paramagnetic impurities. We present two demonstrations outlining methods to potentially overcome these shortcomings. The first leverages the divacancy spins higher sensitivity to electric fields in the excited state to coherently couple to external drives, allowing for a quantum electro-optical interface to both reconfigure the optical structure of the divacancy and implement a pathway to electrically mediated couplings. The second work makes use of a symmetry-breaking property in silicon carbide divacancies to implement a coherence-enhancing protocol that greatly improves the spin dephasing time by many orders of magnitude, allowing it to be competitive with nuclear spin systems while retaining a high degree of responsiveness to control mechanisms. These proof-of-principle demonstrations indicate that non-ideal systems, when combined with proper engineering techniques, can be significantly upgraded to overcome their weaknesses and bolster their strengths.

1.2 Thesis overview

This thesis describes the advancement of single quantum systems for robust real-world applications. A particular focus is given to the divacancy defect in silicon carbide, which in recent years has risen to prominence as a candidate model system with excellent versatility, intrinsic resilience to environmental noise, and a multitude of engineerable interactions.

To start, the underlying principles of quantum information are presented in Chapter 2,

beginning with the basic building block of quantum information: the qubit. Then, a selection of methods to assess the equations of motions that govern the evolution of quantum systems under experimental control are described, while an overview of the various loss mechanisms that afflict all physical realizations of qubits are shown in parallel. In Chapter 3, a focus is brought on the application of these basic principles to a particular experimental realization of a qubit, the divacancy defect in silicon carbide. The properties that are shared among many spin-based quantum systems are enumerated, while the attributes that are more particular to the silicon carbide divacancy defect are highlighted. In Chapter 4, the experimental apparatus required to investigate these quantum properties are itemized, covering optical, electronic, and cryogenic components and techniques. In Chapter 5, the underpinnings of many of the results presented in this thesis, in particular the spin and optical properties of the basally oriented divacancy, are expanded upon. This is combined with the demonstration of coherent electrical control of electron orbitals, resulting in Landau-Zener-Stückelberg interference fringes. In Chapter 6, periodic driving of the ground-state spin in a basal divacancy is leveraged to massively enhance its robustness to external perturbations, providing a clear path forward for the divacancy electron spin to be used as a quantum memory as well as a component in a hybrid quantum system. The generalizability of this coherence enhancement technique to other quantum systems is also discussed. Lastly, Appendices A and B detail the critical experimental details regarding the works presented in Chapters 5 and 6, respectively.

This thesis draws from the following published works:

1. **K. C. Miao**, J. P. Blanton, C. P. Anderson, A. Bourassa, A. L. Crook, H. Abe, T. Ohshima & D. D. Awschalom “Universal coherence protection in a solid-state spin qubit” *arXiv preprint*, 2005.06082 (2020)
2. **K. C. Miao**, A. Bourassa, C. P. Anderson, S. J. Whiteley, A. L. Crook, S. L. Bayliss, G. Wolfowicz, G. Thiering, P. Udvarhelyi, V. Ivády, H. Abe, T. Ohshima, Á. Gali & D. D. Awschalom “Electrically driven optical interferometry with spins in silicon carbide”

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3. A. Bourassa, C. P. Anderson, **K. C. Miao**, M. O. Onizhuk, H. Ma, A. L. Crook, J. U. Hassan, H. Abe, T. Ohshima, N. T. Son, G. Galli & D. D. Awschalom “Entanglement and control of single quantum memories in silicon carbide” *arXiv preprint*, 2005.07602 (2020)
 4. N. T. Son, C. P. Anderson, A. Bourassa, **K. C. Miao**, C. Babin, M. Widmann, M. Niethammer, J. U. Hassan, N. Morioka, I. G. Ivanov, F. Kaiser, J. Wrachtrup & D. D. Awschalom “Developing silicon carbide for quantum spintronics” *Appl. Phys. Lett.* (2020)
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 9. H. Seo, A. L. Falk, P. V. Klimov, **K. C. Miao**, G. Galli & D. D. Awschalom “Quantum

decoherence dynamics of divacancy spins in silicon carbide” *Nat. Commun.* **7**, 1-9
(2016)

CHAPTER 2

QUANTUM INFORMATION PROCESSING IN SOLID-STATE SPINS

2.1 Quantum bits

Before diving into how *quantum* bits work, it is particularly useful to formulate a comparison to its familiar classical counterpart, the bit. The bit (or *binary digit*) is a unit of information that can take on exactly one of two distinct basis states. These states are typically represented as “0” and “1”, “on” and “off”, or any other form that is convenient to the application on hand, as long as there are exactly two states in the representation. For the purposes here, we will use the convention of 0 and 1 to denote the two states of a bit. Also, we will not discuss the various physical encodings of bits, and the methods of manipulation associated with those encodings. Instead, we will consider the bit purely as an abstract concept that will allow us to develop the understanding for its quantum counterpart.

A single bit, once prepared into one of the two states, is expected to remain in that state indefinitely until manipulated by some external force. Manipulation is accomplished by applying computational gates to the state. We can demonstrate this by writing the bit in a vector form,

$$0 = \begin{bmatrix} 1 \\ 0 \end{bmatrix} \qquad 1 = \begin{bmatrix} 0 \\ 1 \end{bmatrix}. \qquad (2.1)$$

Then, operations on a bit, such as a NOT gate, which transforms 0 to 1 and 1 to 0, can be represented by the following matrix,

$$\text{NOT} = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}. \qquad (2.2)$$

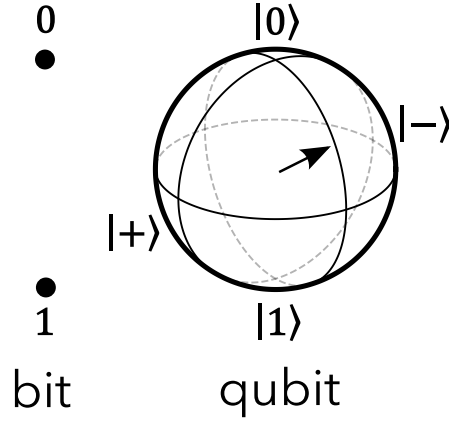


Figure 2.1: **Classical and quantum bits.** Classical bits (left) are represented by two distinct states, and a particular instance can only occupy one of those two states at any given time. Quantum bits (qubits, right) can be represented by a vector in spherical coordinates, where the two states define the poles of the sphere, but superposition states are represented by intermediate vector directions not along either pole. For example, here, $|\pm\rangle = \frac{1}{\sqrt{2}}(|0\rangle \pm |1\rangle)$ are superposition states of the basis vectors.

When applied to the vector representation of 0 or 1, it is clear that the NOT gate yields the other state. This then allows for manipulation of a single bit. Readout of this state then simply involves determining the vector components of the state. Furthermore, upon readout, the state does not change; if the state were 0 prior to readout, it remains 0 until further manipulation.

From this simple picture, the distinction of quantum bits (or qubits) can be readily drawn. First, we will use Dirac notation to represent the basis states for a qubit. Instead of 0 and 1, we now notate these states as $|0\rangle$ and $|1\rangle$, which in vector form remain unchanged,

$$|0\rangle = \begin{bmatrix} 1 \\ 0 \end{bmatrix} \qquad |1\rangle = \begin{bmatrix} 0 \\ 1 \end{bmatrix}. \quad (2.3)$$

One of the most important distinctions from classical bits is the concept of *superposition*, which states that if $|0\rangle$ and $|1\rangle$ are valid states for a particular system, then any linear

combination of $|0\rangle$ and $|1\rangle$ are also valid states. This is typically represented by the expression

$$|\psi\rangle = \alpha |0\rangle + \beta |1\rangle \quad (2.4)$$

where $|\psi\rangle$ is any valid state of the system, and α, β are complex coefficients satisfying $|\alpha|^2 + |\beta|^2 = 1$ for a pure state. Since there are then three degrees of freedom (two real components for each complex coefficient, minus one degree of freedom for an unobservable global phase), a common representation of a single qubit state is a Bloch sphere, where the two basis states are mapped to the poles of the sphere, and any intermediate region is a superposition state.

Furthermore, in many physical systems, pure states encoded as $|\psi\rangle$ may not be realizable over the course of an experiment. For example, many experiments consist of statistical ensembles of quantum systems, which would be unwieldy to handle with a combination of pure states. Instead, by using the density matrix formalism, a density matrix ρ can be constructed,

$$\rho = \sum_n p_n |\psi_n\rangle\langle\psi_n|, \quad (2.5)$$

where p_n is the probability that the system is in the pure state $|\psi_n\rangle$.

We have described how quantum states are represented, but the question remains on how they evolve over time. Just like how classical systems obey Newton's laws of motion and complex trajectories can be modeled from a few simple rules, quantum systems also obey equations of motion to describe evolution of the state as a function of time. These equations are the Schrödinger equation for pure states,

$$\frac{\partial}{\partial t} |\psi(t)\rangle = -\frac{i}{\hbar} H(t) |\psi(t)\rangle, \quad (2.6)$$

where $H(t)$ is the Hamiltonian operator of the quantum system defining the total energy of

the system, as well as the von Neumann equation for mixed states,

$$\frac{\partial \rho(t)}{\partial t} = -\frac{i}{\hbar} [H(t), \rho(t)], \quad (2.7)$$

where $[\cdot, \cdot]$ is the matrix commutator. The von Neumann equation can be further augmented with quantum jump operators to produce the Lindblad master equation, which captures phenomenological decay effects and is used in Chapter 5 to simulate optical two-level systems with rapid decay rates. Overall, these equations of motion are ubiquitous in quantum science and allow for many powerful predictions of the behavior of quantum systems.

Another distinction between qubits and their classical counterparts is the question of measurement. Physically, a quantum system can be in a superposition of an arbitrary number of basis states. The experimentalist can choose an observable to measure (provided an appropriate apparatus sensitive to that observable), which is represented by a Hermitian operator in a finite-dimensional system. Once the measurement is made, the state collapses to one of the eigenstates of the observable, returning a measured value which is the eigenvalue corresponding to the eigenstate. Importantly, the amplitudes α and β determine the probability of the state collapsing into one of the eigenstates. In the case of measuring the projection of a qubit in the $\{|0\rangle, |1\rangle\}$ basis, the probabilities for measuring $|0\rangle$ and $|1\rangle$ are calculated as the squared norm of the amplitudes, $|\alpha|^2$ and $|\beta|^2$, respectively.

Although straightforward in their definitions, these distinctions give rise to a wide variety of complex phenomena that are particular to quantum systems. Throughout this thesis, we make use of these fundamental distinctions to build up a toolbox of measurement techniques to observe and understand the properties of single qubits, and how they respond to the various external control fields we can experimentally apply.

2.2 Time evolution

We are interested in the evolution of quantum states under the application of static and dynamic controls. This typically involves solving the Schrödinger equation,

$$\frac{\partial}{\partial t} |\psi(t)\rangle = -iH(t) |\psi(t)\rangle, \quad (2.8)$$

where we have set $\hbar = 1$. For a general time-dependent Hamiltonian $H(t)$, the expression for the state $|\psi(t)\rangle$ at some time t given an initial state $|\psi(t_0)\rangle$ is

$$|\psi(t)\rangle = \mathcal{T} \exp \left(-i \int_{t_0}^t H(t') dt' \right) |\psi(t_0)\rangle, \quad (2.9)$$

where $\mathcal{T}$ is the time-ordering operator. In some cases it may also be useful to consider the time-evolution of the unitary propagator $U(t)$ acting on the time-independent initial quantum state $|\psi(t_0)\rangle$, allowing us to derive equivalent equations,

$$\begin{aligned} \frac{\partial}{\partial t} (U(t) |\psi(t_0)\rangle) &= -iH(t) (U(t) |\psi(t_0)\rangle) \\ \frac{\partial}{\partial t} U(t) &= -iH(t)U(t), \end{aligned} \quad (2.10)$$

and

$$U(t) = \mathcal{T} \exp \left(-i \int_{t_0}^t H(t') dt' \right). \quad (2.11)$$

For a time-independent Hamiltonian, we can diagonalize and extract energy eigenvalues and eigenvectors,

$$H(t) = \sum_n E_n |u_n\rangle\langle u_n|. \quad (2.12)$$

Eq. 2.9 can then be simplified to remove the time-ordering operator,

$$\begin{aligned}
|\psi(t)\rangle &= \exp\left(-i \int_{t_0}^t H dt'\right) |\psi(t_0)\rangle \\
&= \sum_n |u_n\rangle \underbrace{\langle u_n | \psi(t_0) \rangle}_{c_n} \exp(-i E_n t) \\
&= \sum_n c_n |u_n\rangle \exp(-i E_n t),
\end{aligned} \tag{2.13}$$

where we used the fact that the matrix exponential of a diagonal matrix is also a diagonal matrix. The result is a representation of the time evolution as a linear combination of energy eigenstates, with phase factors dependent on the energy eigenvalues. This is a powerful and intuitive representation of the time-evolution of a quantum state, as the phase relations of the individual energy eigenstates in a superposition can be readily identified.

However, with a time-independent Hamiltonian, it can be seen that the only time-evolution of a quantum state is the acquisition of phase between energy eigenvectors. Notably, the complex coefficients c_n are invariant with time, and population cannot be transferred between the various states in a superposition. In order to effect arbitrary quantum control, time-varying perturbations of the Hamiltonian are necessary. Typically, these perturbations consist of periodic drives corresponding to oscillatory electromagnetic interactions between the quantum system and an external control mechanism. Thus, many Hamiltonians relevant to quantum information processing take the form

$$H(t) = H_0 + H_1(t), \tag{2.14}$$

where H_0 is the static, time-independent Hamiltonian, and H_1 is a time-dependent Hamiltonian, usually influenced by a controllable interaction. Many of the systems studied in this thesis possess a unifying motif of a periodic modulation of some parameter within the

Hamiltonian. Thus, for these systems, the Hamiltonian $H(t)$ obeys the constraint

$$H(t + T) = H(t),$$

where T is the period of the periodic modulation. Although simple in its formulation, this constraint is applicable for Rabi oscillations using a periodic drive, parametric driving of energy levels, as well as the generation of dressed states. Throughout this thesis, Rabi oscillations are used to transfer population between various levels of a quantum system. In Chapter 5, we show that periodic modulation of the energy levels of an optical two-level system yields interferometric signatures. In Chapter 6, we show that dressed states gain massively improved coherence properties under a periodic drive. Thus, methods to efficiently treat these periodic quantum systems are convenient and facilitate analysis of these near-ubiquitous systems. As we will show in the next two sections, these time-dependent systems can be transformed into approximately equivalent time-independent systems, where the dynamics can be greatly simplified.

2.3 Rotating wave approximation

For many quantum systems interacting with an external oscillating control pulse, the Hamiltonian can be constructed like Eq. 2.14. Here, we use the Pauli operators σ_i , which allow for concise formulation of operators on two-level systems. For example, a two-level system with an energy splitting ε_0 along σ_z ,

$$H_0 = \frac{\varepsilon_0}{2} \sigma_z, \tag{2.15}$$

can interact with a time-varying field with amplitude Ω , angular frequency ω , and phase ϕ ,

$$H_1(t) = \Omega \cos(\omega t + \phi) \sigma_x, \tag{2.16}$$

so that the full time-dependent Hamiltonian is

$$H(t) = \frac{\varepsilon_0}{2} \sigma_z + \Omega \cos(\omega t + \phi) \sigma_x. \quad (2.17)$$

A well-known procedure to obtain an excellent approximation of this system is known as the rotating wave approximation (RWA). Near resonance, $\omega \approx \epsilon_0$, the system can be reformulated so that states precessing at an angular frequency ω appear static. First, this entails separating $H(t)$ into H_0 and $H_1(t)$, and then applying a time-dependent unitary operator

$$U = \exp\left(-\frac{i\omega t}{2} \sigma_z\right) \quad (2.18)$$

to a state $|\psi\rangle$ which is a solution of the Schrödinger equation, allowing us to enter the interaction picture. We then obtain $|\tilde{\psi}\rangle = U |\psi\rangle$. In effect, we are performing a transformation to a reference frame that is rotating at angular frequency ω , which makes precessional dynamics at ω appear static. The Schrödinger equation in the interaction picture is

$$\begin{aligned} H|\tilde{\psi}\rangle &= i\frac{\partial}{\partial t}|\tilde{\psi}\rangle \\ HU|\psi\rangle &= i\frac{\partial}{\partial t}(U|\psi\rangle) \\ U^\dagger HU|\psi\rangle &= iU^\dagger \frac{\partial}{\partial t}(U|\psi\rangle) \\ U^\dagger HU|\psi\rangle &= iU^\dagger \left(\frac{\partial U}{\partial t} |\psi\rangle + U \frac{\partial}{\partial t} |\psi\rangle \right) \\ -iU^\dagger \frac{\partial U}{\partial t} |\psi\rangle + U^\dagger HU|\psi\rangle &= i\frac{\partial}{\partial t} |\psi\rangle \\ \tilde{H}|\psi\rangle &= i\frac{\partial}{\partial t} |\psi\rangle \end{aligned} \quad (2.19)$$

The LHS of Eq. 2.19 shows that the interaction picture can be reformulated in the Schrödinger picture, namely with a modified time-dependent Hamiltonian $\tilde{H}$,

$$\tilde{H} = -iU^\dagger \frac{\partial U}{\partial t} + U^\dagger HU. \quad (2.20)$$

First, we consider the leading term in $\tilde{H}$,

$$\begin{aligned} -iU^\dagger \frac{\partial U}{\partial t} &= -U^\dagger U \frac{\omega}{2} \sigma_z \\ &= -\frac{\omega}{2} \sigma_z. \end{aligned} \quad (2.21)$$

Next, we consider the second term in $\tilde{H}$,

$$\begin{aligned} U^\dagger H U &= U^\dagger H_0 U + U^\dagger H_1 U \\ &= H_0 + \Omega \cos(\omega t + \phi) [\cos(\omega t) \sigma_x + \sin(\omega t) \sigma_y] \\ &= H_0 + \Omega \left(\frac{\exp(i\omega t + \phi) + \exp(-i\omega t - \phi)}{2} \right) [\exp(-i\omega t) \sigma_+ + \exp(i\omega t) \sigma_-] \\ &= H_0 + H_{\text{int}}. \end{aligned} \quad (2.22)$$

The interaction Hamiltonian H_{int} can be further expanded,

$$\begin{aligned} H_{\text{int}} &= \frac{\Omega}{2} \left\{ e^{i(\omega - \omega + \phi)t} \sigma_+ + e^{-i(\omega - \omega + \phi)t} \sigma_- + e^{-i(\omega + \omega + \phi)t} \sigma_+ + e^{i(\omega + \omega + \phi)t} \sigma_- \right\} \\ &= \frac{\Omega}{2} [\cos(\phi) \sigma_x - \sin(\phi) \sigma_y + e^{-i(2\omega + \phi)t} \sigma_+ + e^{i(2\omega - \phi)t} \sigma_-] \end{aligned} \quad (2.23)$$

at which point, we invoke the RWA, which dictates that terms with “fast” precession frequency 2ω contribute negligibly to the overall evolution of the system, and can be removed.

The result is then

$$H_{\text{int}} = \frac{\Omega}{2} [\cos(\phi) \sigma_x - \sin(\phi) \sigma_y]. \quad (2.24)$$

The rotating frame Hamiltonian $\tilde{H}$ then is

$$\tilde{H} = \left(\frac{\varepsilon_0}{2} - \frac{\omega}{2} \right) \sigma_z + \frac{\Omega}{2} [\cos(\phi) \sigma_x - \sin(\phi) \sigma_y]. \quad (2.25)$$

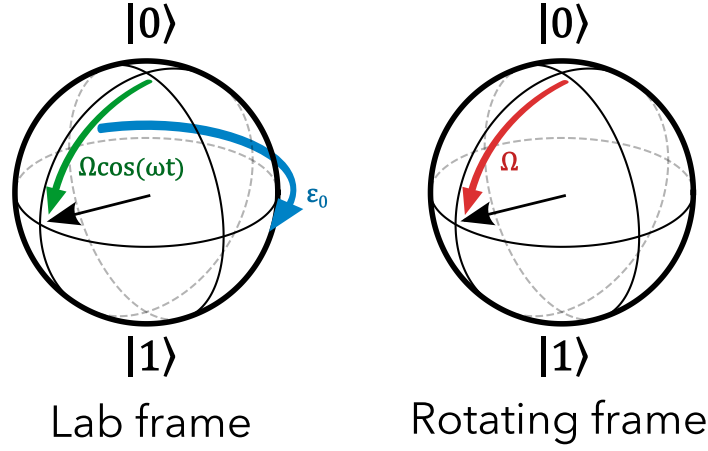


Figure 2.2: **Lab and rotating frame pictures of a qubit.** In the lab frame, the two basis states precess relative to each other at a rate defined by the energy difference. In order to produce a coherent spin rotation, a periodic drive at that precession frequency can be applied in the plane orthogonal to the quantization axis. In a frame rotating at that precession frequency, the two states appear static relative to each other and have the same energy in the absence of a drive. An applied drive also loses its time-dependence in this frame, and lifts the degeneracy of the system.

Furthermore, by defining $\Delta = \varepsilon_0 - \omega$ as the detuning between the applied excitation and the natural resonance, we obtain

$$\tilde{H} = \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2} [\cos(\phi)\sigma_x - \sin(\phi)\sigma_y]. \quad (2.26)$$

Remarkably, this system now has a time-independent Hamiltonian in the rotating frame, which allows for significant simplification of the dynamics of the system. On resonance, $\Delta = 0$, the Hamiltonian is simply

$$\tilde{H}_{\text{res}} = \frac{\Omega}{2} [\cos(\phi)\sigma_x - \sin(\phi)\sigma_y], \quad (2.27)$$

which for an initial state prepared in a σ_z basis state produces an oscillatory transfer of population between one state and the other – otherwise known as Rabi oscillations. Furthermore, depending on ϕ of the applied drive, the phase of the traversed oscillation can be

tuned. This then allows for the creation of any arbitrary single-qubit state.

2.4 Coherent control and Rabi oscillations

The time-independent system shown in Eq. 2.26 can be readily diagonalized to yield energy eigenstates and eigenvalues. We observe that the two states have energies

$$E_{\pm} = \pm \frac{\sqrt{\Omega^2 + \Delta^2}}{2}, \quad (2.28)$$

which allows for evaluation of eigenenergies as a function of drive strength and detuning.

We can also determine the corresponding eigenvectors of these eigenvalues. By defining $\theta = \arctan\left(\frac{\Omega}{\Delta}\right)$, the corresponding eigenvectors can be written as

$$\begin{aligned} |+\rangle &= \sin \frac{\theta}{2} |0\rangle + e^{i\phi} \cos \frac{\theta}{2} |1\rangle \\ |-\rangle &= \cos \frac{\theta}{2} |0\rangle - e^{i\phi} \sin \frac{\theta}{2} |1\rangle. \end{aligned} \quad (2.29)$$

We will set $\phi = 0$ for simplicity in this following example. In this system, for an initial state $|0\rangle$, which is not an eigenstate, we can use Eq. 2.9 to obtain its time evolution

$$\begin{aligned} |\psi(t)\rangle &= \sum_{u=+,-} |u\rangle \langle u|0\rangle \exp(-iE_u t) \\ &= \sin \frac{\theta}{2} |+\rangle \exp(-iE_+ t) + \cos \frac{\theta}{2} |-\rangle \exp(-iE_- t). \end{aligned} \quad (2.30)$$

The population of $|1\rangle$ at any time in free evolution can be written as

$$\begin{aligned}
|\langle 1|\psi(t)\rangle|^2 &= \left| \cos \frac{\theta}{2} \sin \frac{\theta}{2} \exp(-iE_+t) - \sin \frac{\theta}{2} \cos \frac{\theta}{2} \exp(-iE_-t) \right|^2 \\
&= \left| \cos \frac{\theta}{2} \sin \frac{\theta}{2} [\exp(-iE_+t) - \exp(-iE_-t)] \right|^2 \\
&= \frac{\sin^2 \theta}{4} |\exp(-iE_+t) - \exp(-iE_-t)|^2 \\
&= \frac{\sin^2 \theta}{4} [2 - e^{i(E_- - E_+)t} - e^{-i(E_- - E_+)t}] \\
&= \frac{\sin^2 \theta}{4} [2 - 2 \cos[(E_- - E_+)t]] \\
&= \sin^2 \theta \sin^2 \left[\frac{(E_- - E_+)t}{2} \right] \\
&= \left(\frac{\Omega^2}{\Omega^2 + \Delta^2} \right) \sin^2 \left[\frac{\sqrt{\Omega^2 + \Delta^2}t}{2} \right]. \tag{2.31}
\end{aligned}$$

Eq. 2.31 is central to manipulating quantum systems and understanding their response to applied control interactions. First, this expression shows how Rabi oscillations are expected to occur for a particular drive strength and detuning. In particular, when on resonance, $\Delta = 0$, this expression simplifies to

$$|\langle 1|\psi(t)\rangle|^2 = \sin^2 \left(\frac{\Omega t}{2} \right), \tag{2.32}$$

which shows the sinusoidal behavior of a two-level system evolving between the two unperturbed basis states. For non-zero Δ , the evolution trajectory does not complete a full population inversion as the prefactor asymptotically approaches 0 for increasing Δ . Furthermore, the effective rate of oscillation $\Omega' = \sqrt{\Omega^2 + \Delta^2}$ increases.

Next, for a combination of Ω and t so that full population inversion is obtained, $\Theta = \Omega t = \pi$, we can obtain the resonance spectrum as a function of detuning,

$$P(\Delta) = \left[\frac{1}{1 + (\Delta/\Omega)^2} \right] \sin^2 \left[\frac{\sqrt{1 + (\Delta/\Omega)^2}\pi}{2} \right], \tag{2.33}$$

which, when plotted, bears a strong resemblance to a sinc^2 function. However, the profile and relative intensity of sidelobes in Eq. 2.33 differ from a sinc^2 function and generally cannot be interchanged. As we will see later, understanding how this equation behaves at various values of Θ , Ω , and Δ will allow us to evaluate many parameters of a quantum system under study, and is a core tool in bootstrapping coherent control mechanisms in the absence of known parameter values.

2.5 Floquet formalism

Another approach for extracting solutions to the Schrödinger equation for periodic Hamiltonians is to apply Floquet's theorem and reduce the time-dependent system into an equivalent system that shunts the time-dependence into periodic wavefunctions, transforming the system into one that is equivalent to one with a time-independent Hamiltonian. In a sense, the RWA accomplishes the same transformation, but Floquet's theorem is tailored specifically to time-periodic systems, and can be easily scaled up to handle interactions at many oscillation frequencies without significant modifications.

Consider the Schrödinger equation for a time-dependent Hamiltonian $H(t)$ that possesses a periodic time-translation symmetry,

$$H(t) = H(t + T), \quad (2.34)$$

for some period T ,

$$\left(H(t) - i \frac{\partial}{\partial t} \right) |\psi(t)\rangle = 0. \quad (2.35)$$

Floquet's theorem asserts that solutions for $|\psi(t)\rangle$ take the form

$$|\psi(t)\rangle = \sum_n c_n |u_n(t)\rangle \exp(-i\varepsilon_n t), \quad (2.36)$$

where $|u_n(t)\rangle = |u_n(t + T)\rangle$ is the Floquet state obeying the same periodic time-translation

symmetry as the Hamiltonian and ε_n is the quasienergy, which can be considered as an analog of energy in a periodic system. This can be seen by

$$\begin{aligned}
\left(H(t) - i\frac{\partial}{\partial t}\right) |\psi(t)\rangle &= \left(H(t) - i\frac{\partial}{\partial t}\right) |u_n(t)\rangle \exp(-i\varepsilon_n t) \\
&= H(t) |u_n(t)\rangle \exp(-i\varepsilon_n t) - i\frac{\partial}{\partial t} [|u_n(t)\rangle \exp(-i\varepsilon_n t)] \\
&= H(t) |u_n(t)\rangle \exp(-i\varepsilon_n t) - i \exp(-i\varepsilon_n t) \left[\frac{\partial}{\partial t} |u_n(t)\rangle - i\varepsilon_n |u_n(t)\rangle \right] \\
&= \exp(-i\varepsilon_n t) \left(H(t) - i\frac{\partial}{\partial t} - \varepsilon_n \right) |u_n(t)\rangle.
\end{aligned} \tag{2.37}$$

Thus the LHS of Eq. 2.35 can be written as

$$\left(H(t) - i\frac{\partial}{\partial t} - \varepsilon_n \right) |u_n(t)\rangle = 0, \tag{2.38}$$

which can then be rearranged to produce

$$\underbrace{\left(H(t) - i\frac{\partial}{\partial t} \right)}_{\mathcal{H}(t)} |u_n(t)\rangle = \varepsilon_n |u_n(t)\rangle. \tag{2.39}$$

Eq. 2.39 takes on a form similar to the time-independent Schrödinger equation, with ε_n taking the place of the energy eigenvalues. Time-independent assumptions can then be applied here, such as diagonalization to obtain resonance spectra. An invocation of Floquet's theorem becomes useful when it is more straightforward to obtain solutions for Eq. 2.39 instead of Eq. 2.35. It can be shown that the Floquet Hamiltonian $\mathcal{H}(t)$ can be obtained [42] by performing a Fourier decomposition of the original Hamiltonian $H(t)$,

$$H(t) = \sum_n H_n(t) e^{in\omega t}, \tag{2.40}$$

where $\omega = 2\pi/T$. Then, $\mathcal{H}(t)$ is constructed by assembling the Fourier components of $H(t)$

in a block matrix embedded in an infinite-dimension Hilbert space,

$$\mathcal{H}(t) = \begin{bmatrix} \ddots & & \ddots & & \ddots & & \\ \ddots & H_0 + \omega\mathbb{I} & H_{+1} & H_{+2} & & & \\ \ddots & H_{-1} & H_0 & H_{+1} & \ddots & & \\ & H_{-2} & H_{-1} & H_0 - \omega\mathbb{I} & \ddots & & \\ & & \ddots & \ddots & \ddots & & \end{bmatrix} \begin{matrix} |m = +1\rangle \\ |m = 0\rangle \\ |m = -1\rangle \end{matrix}, \quad (2.41)$$

where m is the Floquet mode index. Although $\mathcal{H}(t)$ is infinite-dimensional, it can be truncated to be numerically diagonalized to obtain quasienergies. Furthermore, as shown in Appendix B, the rotating wave approximation can be implemented by neglecting fast rotating terms and reducing $\mathcal{H}(t)$ to a block diagonal matrix [43], which can then be further simplified by isolating the terms in the first Floquet zone (in this work, also termed “upper” and “lower” matrices corresponding to Autler-Townes branches) to yield a finite-dimensional matrix.

2.6 Decoherence

In the previous sections, we outlined the construction and operation of ideal qubits that are entirely isolated from the external environment. In any physical realization of a qubit, however, environmental interactions exist. Much like how error can be introduced into classical bits through high-energy perturbations such as cosmic ray events, error can enter a qubit system in various ways, with the end result that the quantum information encoded within the qubit becomes unusable after a certain timescale.

2.6.1 Energy relaxation (T_1)

A closed quantum system, where the quantum system under investigation cannot interact with any other systems, obeys energy conservation and when prepared in particular state,

cannot evolve to another state with more or less total energy. An isolated qubit prepared in a pure state $|0\rangle$, for example, cannot evolve to $|1\rangle$. It becomes very clear that while fully isolated quantum systems can be useful to understand many fundamental concepts of quantum information, they are neither realizable in nature, nor particularly useful in practical applications. Thus, open quantum systems, which interact with other quantum systems and perturbative fields in the environment, are the subjects of extensive theoretical and experimental research. In fact, all quantum systems experimentally studied in laboratories are open quantum systems. In some cases, the quantum system is sufficiently well-isolated so that a closed quantum system can be used as an approximation for its evolution. Typically speaking, an open quantum system is coupled to its environment through many degrees of freedom, allowing it to exchange energy via various controlled and uncontrolled channels. Controlled channels encompass coherent control mechanisms, which we discuss later as the primary method to effect desired time-evolution of the state. Uncontrolled mechanisms are typically undesired interactions with nearby fluctuating quantum systems and classical noise sources that degrade the quality of the quantum information stored within the qubit.

One of the environmental couplings universally encountered in experimental quantum systems is energy relaxation. Just like a classical bit system, a quantum bit can interact with its environment through energy exchange, where quantum systems in the environment can accept or donate energy at a scale close to the energy splitting of the qubit. We must determine if the environment is more likely to accept or donate energy to the qubit, and at what rate this can occur.

In the density matrix formalism, the basis states $\{|0\rangle, |1\rangle\}$ can be represented as

$$\rho_0 = |0\rangle\langle 0|, \quad \rho_1 = |1\rangle\langle 1|. \quad (2.42)$$

Over time, as the quantum system interacts with external fields and other quantum systems that can donate or accept energy at the energy splitting between $|0\rangle$ and $|1\rangle$, it tends towards

a thermal state

$$\rho_{\text{th}} = \frac{1}{Q} \left(e^{-\varepsilon_0/kT} |0\rangle\langle 0| + e^{-\varepsilon_1/kT} |1\rangle\langle 1| \right), \quad (2.43)$$

where $Q = \sum_i e^{-\varepsilon_i/kT}$, ε_i is the energy of the i th level, k is Boltzmann's constant, and T is the thermodynamic temperature of the environment. It can be seen at temperatures $kT \gg |\varepsilon_0 - \varepsilon_1|$, the thermal state will be approximately

$$\rho_{\text{th}} = \frac{1}{2} (|0\rangle\langle 0| + |1\rangle\langle 1|) = \frac{\mathbb{I}}{2}, \quad (2.44)$$

where $\mathbb{I}$ is the identity. Then, from the von Neumann equation of motion, it can be readily shown that no further useful evolution can be accomplished, since all operators commute with the identity,

$$\frac{\partial \rho_{\text{th}}}{\partial t} = -i [H, \mathbb{I}] = 0. \quad (2.45)$$

Even in the limit where $kT \ll |\varepsilon_0 - \varepsilon_1|$, the result is a thermal state corresponding to the basis state with lower energy. This can be considered as an undesired initialization of the system into one of the basis states, which would still prevent further computation to be performed in the system. Thus, energy relaxation is considered an irreversible process after which quantum information is entirely lost to the surrounding environment, and there are no unitary operations to recover such a state. The rate at which an initial basis state transforms to a thermal state is dependent on processes that are *resonant* with the energy splitting between the basis states, which allows for direct energy transfer between the two basis states and leakage of quantum information out of the system. Furthermore, higher-order interactions involving virtual states and multi-excitation processes can also induce transitions between the basis states.

2.6.2 Inhomogeneous dephasing (T_2^*)

There also exist mechanisms for the quantum system to interact with its environment without energy exchange. Consider a superposition state

$$|\psi\rangle = \frac{1}{\sqrt{2}} (|0\rangle + |1\rangle), \quad (2.46)$$

prepared in a system with a Hamiltonian

$$H = \frac{\varepsilon_0}{2} (|1\rangle\langle 1| - |0\rangle\langle 0|). \quad (2.47)$$

Time evolution of this system using one of the quantum equations of motion yields

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}} \left(|0\rangle + e^{i\varepsilon_0 t} |1\rangle \right), \quad (2.48)$$

where we have removed an unobservable global phase. The result is an accumulation of phase for the $|1\rangle$ state relative to the $|0\rangle$ state. Under ideal conditions, ε_0 is constant and the phase relation between $|1\rangle$ and $|0\rangle$ is known at all times. However, external noise caused by nearby quantum systems can induce fluctuations in the energy splitting,

$$\varepsilon_0 \rightarrow \varepsilon_0 + \delta\varepsilon_0(t), \quad (2.49)$$

where $\delta\varepsilon_0(t)$ is typically a mean-reverting stochastic process, such as an Ornstein-Uhlenbeck process. The resulting state at time t is instead

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}} \left[|0\rangle + \exp \left(i \int_0^t \varepsilon_0 + \delta\varepsilon_0(t') dt' \right) |1\rangle \right]. \quad (2.50)$$

From Eq. 2.50 it can be seen that a superposition is maintained in the face of stochastic noise in ε_0 – measuring the system at any point t will yield $|0\rangle$ and $|1\rangle$ with $p = 0.5$. However, the phase between the qubit states, which is a core component in useful quantum

computation, becomes scrambled. In particular, typically $\delta\varepsilon_0(t)$ has little to no correlation with applied control pulses, and thus many repetitions of the same control pulse sequence will yield different phase relations. When such a process is averaged over multiple repetitions, the result is a characteristic loss of phase information over time. This loss is parameterized by the inhomogeneous dephasing time T_2^* , and can be applied to multiple quantum systems operating in a statistical ensemble, or a time-average of a single quantum system with varying noise over time.

2.6.3 Hahn-echo coherence (T_2)

Suppose we can correct the phase fluctuations that we detailed in the previous section. If the correlation time of $\delta\varepsilon_0(t)$ is sufficiently long, so that the noise term $\delta\varepsilon_0$ can be considered constant in a particular iteration of an experiment (otherwise known as *quasistatic noise*), then we can reformulate the time evolution of the state as

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}} \left(|0\rangle + e^{i(\varepsilon_0 + \delta\varepsilon_0)t} |1\rangle \right), \quad (2.51)$$

where ε_0 is still constant, and $\delta\varepsilon_0$ fluctuates between individual iterations of the experiment but is approximately constant within the iteration. The goal is to remove $\delta\varepsilon_0$ from the evolution dynamics, regardless of its value. Measuring $\delta\varepsilon_0$ may be difficult, and its value may have changed after measurement, so a feedback procedure based on the value of $\delta\varepsilon_0$ can prove to be formidable to implement. Instead, if we are interested in maintaining the equality $|\psi(0)\rangle = |\psi(t)\rangle$ as closely as possible, then we can add a single rotation halfway through the free evolution, at $t = \tau/2$. Specifically, this rotation is designed so that $|0\rangle \rightarrow |1\rangle$ and $|1\rangle \rightarrow |0\rangle$, otherwise known as a quantum NOT gate. The evolution of the state up to $t = \tau/2$, right before this rotation, is

$$|\psi(\tau/2)\rangle = \frac{1}{\sqrt{2}} \left(|0\rangle + e^{i(\varepsilon_0 + \delta\varepsilon_0)\tau/2} |1\rangle \right). \quad (2.52)$$

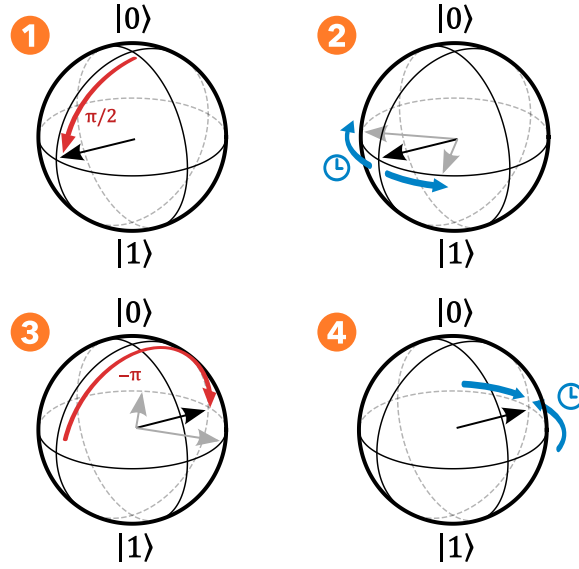


Figure 2.3: **Inhomogeneous dephasing and Hahn-echo refocusing.** A prepared superposition (1) will dephase over time due to inhomogeneities in the energy splitting. Equivalently, an ensemble of identical systems will each experience slightly different energy splittings due to spatial inhomogeneities. Thus, over time (2), each instance of the qubit will accumulate phase at slightly different rates (gray vectors). Without any other controls, this will lead to inhomogeneous dephasing and a loss of phase coherence. A Hahn-echo sequence calls for an additional refocusing pulse (3), which flips all vectors to the opposite side of the Bloch sphere. If the inhomogeneity is quasistatic, which is a good first-order approximation for many systems, then much of the noise can be cancelled out by allowing the system to evolve again for the same amount of time (4). At the end, the state returns to a phase-coherent superposition, which can be further used for quantum information processing.

The applied rotation then swaps the basis states,

$$|\psi(\tau/2)\rangle = \frac{1}{\sqrt{2}} \left(e^{i(\varepsilon_0 + \delta\varepsilon_0)\tau/2} |0\rangle + |1\rangle \right). \quad (2.53)$$

Free evolution for another duration of $\tau/2$ then causes $|1\rangle$ to accumulate phase again,

$$|\psi(\tau)\rangle = \frac{1}{\sqrt{2}} \left(e^{i(\varepsilon_0 + \delta\varepsilon_0)\tau/2} |0\rangle + e^{i(\varepsilon_0 + \delta\varepsilon_0)\tau/2} |1\rangle \right) \quad (2.54)$$

$$= \frac{1}{\sqrt{2}} e^{i(\varepsilon_0 + \delta\varepsilon_0)\tau/2} (|0\rangle + |1\rangle), \quad (2.55)$$

which is equivalent to $|\psi(0)\rangle$ up to a global phase. This sequence is known as a Hahn echo, and the rotation applied halfway through the free evolution of the system is known as a refocusing pulse.

While it appears that a Hahn-echo sequence can perfectly cancel out constant noise, we had made the earlier assumption that noise in the system is quasistatic. However, in reality, no noise source is truly static during the evolution of the system, and there is always a time-dependence of $\delta\varepsilon_0$. It can easily be seen that time variation of $\delta\varepsilon_0$ between the first and second half of the free evolution will cause imperfect refocusing of the superposition state. Generally speaking, a state undergoing a Hahn-echo sequence will still experience phase noise at non-zero frequency, and cause a damping of coherence elements in the density matrix. The resulting decay of the coherence produces a $1/e$ decay time characterized by T_2 , the Hahn-echo coherence time.

CHAPTER 3

DIVACANCY DEFECTS IN SILICON CARBIDE

3.1 Introduction

This section outlines the physical properties of silicon carbide, and how those properties support the existence of robust and controllable quantum systems within the crystal. We provide an overview of silicon carbide, and then move on to discussing the divacancy defect, which hosts an optically active electron spin system. The quantum mechanical properties of the electron spin system are described, as well as how a qubit can be implemented using the spin sublevels. Lastly, the optical interface of the divacancy defect is introduced, as well as how light can be used to initialize and read out the spin.

3.2 Silicon carbide host

Silicon carbide (SiC) is a binary crystal composed of silicon and carbon. SiC has found widespread applications in industrial efforts as an excellent semiconductor material with a wide bandgap, versatility in doping, lattice matching with many other materials, and excellent electronic, optical, and thermal properties. Furthermore, the availability of its constituent elements and long history of material growth research provides many opportunities to secure supply of high quality, wafer-scale material for various research and industrial applications.

Naturally occurring SiC crystals are found in the form of moissanite, named after Henri Moissan. For nearly all commercial, industrial, and research applications, SiC is produced synthetically, through the Acheson process, the Lely method, or chemical vapor deposition. For all of the studies included in this thesis, we work with SiC samples that are grown through high-quality chemical vapor deposition epitaxy. This ensures that the samples have the lowest possible densities of lattice imperfections and environmental impurities that can potentially

adversely affect the quality of desired quantum systems within the crystal. Even then, there is a wide spectrum of growth quality from epitaxially grown substrates [11, 44, 45], and careful selection of a substrate manufacturer can significantly impact the quality of quantum phenomena.

SiC is notable for its large bandgap (as high as 3.33 eV), which places it as a more insulating material when compared to silicon (1.1 eV), but less insulating than diamond (5.5 eV). Although the bandgap is indirect, limiting SiC’s application as a light-emitting material, the wide bandgap allows for the electronic properties to persist at higher temperatures, applied voltages, and operating frequencies. In particular, for quantum information purposes, the wide bandgap can host many isolated levels corresponding to defect sites. As shown in this thesis, the neutral divacancy is one of those defect sites which reside deep in the bandgap, and the wideness of the bandgap relative to the operating temperature suppresses thermally induced fluctuations that would otherwise excite electrons from the valence band into the bound states, or from the bound states to the conduction band [46].

Furthermore, SiC’s high thermal conductivity (as high as 490 W / m·K at 300 K), high melting point (about 3000 K), high Debye temperature ($T_D \approx 1300$ K), bestow excellent robustness in real-world applications where thermal dissipation is ubiquitous. When considered from the perspective of investigating isolated quantum systems, the high thermal conductivity allows for rapid dissipation of applied optical and microwave energy, while the high Debye temperature suggests that phonon modes within the crystal are less likely to interfere with the quantum degrees of freedom of isolated systems.

The high refractive index of SiC ($n \approx 2.6$ at 1 μm) can be exploited to serve as photonic confinement as a waveguide or cavity structure for visible to near-infrared spectra [47–52]. When fabricated into nanostructures, the higher refractive index of SiC compared to surrounding mediums can induce total internal reflection, allowing for high-fidelity light confinement and the creation of a nanophotonic devices. For quantum emitters embedded in bulk crystals, this high refractive index and resulting total internal reflection can hinder the

extraction of emitted photons, but solutions such as solid immersion lenses can overcome some of these losses, as shown in studies in diamond [26, 53], which has a similarly high refractive index.

A particularly attractive property of SiC is its polytypism [44], where the SiC crystal structure can differ along one crystal axis. Although in theory an infinite number of polytypes of SiC can be constructed, only a handful have been physically realized and even fewer are available at commercial scale. The most common polytypes of SiC are the hexagonal 4H and 6H forms, as well as the cubic 3C form. Many material properties vary significantly between the various polytypes, namely lattice constants, electronic bandgap (3.23 eV, 3.05 eV, and 2.36 eV for 4H, 6H, and 3C, respectively) and thermal conductivity at 300 K (370 W / m·K, 490 W / m·K, 360 W / m·K for 4H, 6H, and 3C, respectively). The work presented in this thesis will focus exclusively on 4H-SiC as the semiconductor host for quantum systems, but this does not preclude similar or better systems compared to the ones presented here to exist in other polytypes.

The large bandgap in 4H-SiC (second out of all SiC polytypes only to the rarer 2H-SiC) potentially allows for a wider variety of stable deep levels to exist in the bandgap, and greater electronic stability of those defects under thermal and photo-induced excitation. 4H-SiC has a lattice stacking periodicity of four, giving rise to two distinct local lattice geometries at any particular lattice site. These two distinct local geometries are termed quasicubic (k) sites, as a twist in the lattice produces cubic-like symmetry, and hexagonal (h) sites, where the absence of a twist maintains the hexagonal symmetry of the SiC monolayer. As will be presented later, the local geometry of these lattice sites affect the properties of lattice defects at particular sites, as well as defect complexes comprised of multiple lattice defects.

3.3 Divacancies in 4H-SiC

The divacancy defect complex is comprised of a carbon vacancy adjacent to a silicon vacancy in 4H-SiC. Since each vacancy site can have two local geometries, there are 2^2 possible

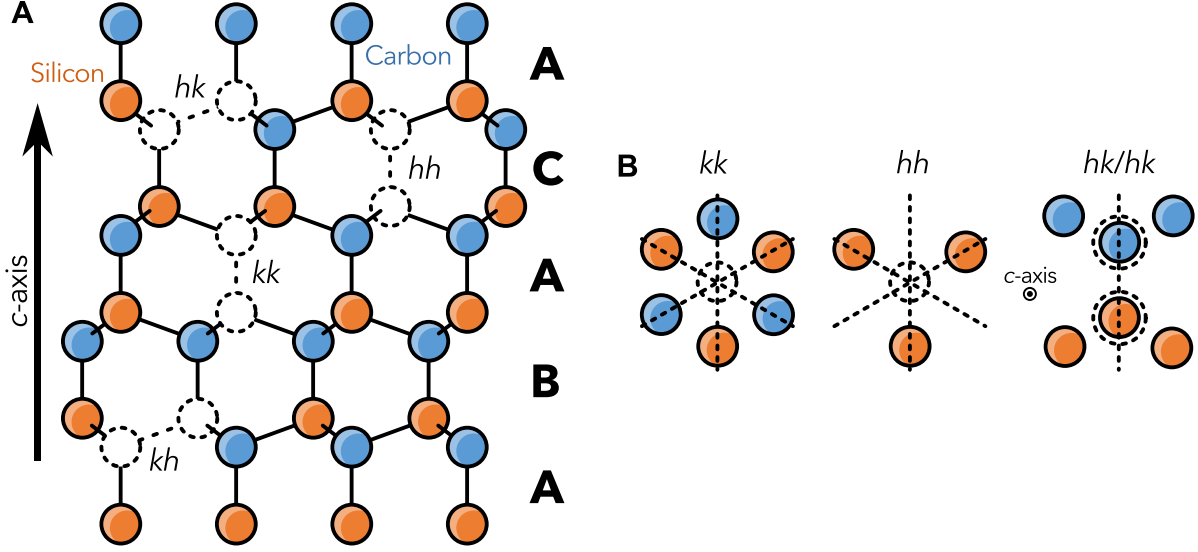


Figure 3.1: **Divacancy defects in 4H-SiC.** (A) 4H-SiC lattice with the four possible divacancy configurations. The stacking periodicity of 4 is shown by the stacking layer labels on the right. The 60° twist on the C layer provides a distinction between the hexagonal (h) and quasicubic (k) sites. (B) The four divacancy configurations viewed along the c -axis. The three-fold rotational and mirror-plane symmetry of the c -axis divacancies (hh and kk configurations) are visible. The basal divacancies (hk and kh configurations) have a lower symmetry, with only a single mirror-plane symmetry and no rotational symmetries. The divacancy axis is also not along the c -axis, which affects the electromagnetic dipole orientations. The reduced symmetry manifests in both the spin and optical properties of basal divacancies.

configurations of divacancies in 4H-SiC. In the notation used in this thesis, the local geometry of the carbon vacancy followed by the local geometry of the silicon vacancy is used to unambiguously label the defect configuration. For example, a kh divacancy refers to a carbon vacancy at a quasicubic site adjacent to a silicon vacancy at a hexagonal site. In this way, all four configurations can be labeled, namely as hh , kk , hk , and kh divacancies. Although natural divacancies exist in all SiC polytypes to varying densities, many applications in quantum science require finer control of divacancy densities. In many studies of divacancy ensembles [11, 32, 44, 54, 55], naturally occurring divacancies exist at high enough densities to allow for off-the-shelf experiments to be performed without any further processing. However, for single defect studies, the densities of these naturally occurring divacancies are too high, prohibiting single defect isolation. Thus, custom SiC wafers grown by research groups or

commercial vendors with extremely low defect densities are chosen as the initial substrate for defect studies. Electron irradiation or ion implantation at carefully controlled doses and particle energies, followed by an annealing procedure, can then form desired defect densities, ranging from sparse single defects to dense ensembles.

The foundational properties of divacancies were determined in ensemble studies [11, 44]. Mechanical and electrical tuning of the spin transitions have been accomplished in divacancy ensembles [54]. Electrical spin driving of the double quantum spin transition has been also shown [27]. Optical polarization of strongly coupled nuclear spins have been shown using the ground- and excited-state spin level anticrossings [56], which has led to demonstrations of two-qubit entanglement in divacancy ensembles at room temperature [32].

Elucidation of some divacancy properties have been accomplished as a result of single defect isolation [57]. With the ability to resonantly excite the divacancy’s optical system and leverage spin-selective optical transitions [45], many exciting avenues for using the divacancy as a quantum information processing platform have been unlocked. Electrical tuning of these single emitters has been demonstrated in intrinsic substrates [58], which has been expanded on recently in intrinsic [29] and doped [30] commercially obtained 4H-SiC. Nuclear spin control has been demonstrated in isotopically purified substrates [39]. Also, photonic emission has been enhanced by integrating the divacancy defect into nanophotonic structures [47], leading to the possibility of using the divacancy as a basis for long-distance quantum network applications.

3.4 Spin-1 systems

In the neutral charge state, the divacancy hosts six electrons from the six total dangling bonds from the three nearest-neighbor carbon atoms and three nearest-neighbor silicon atoms. Four of these six electrons are paired, having no magnetic moment and typically are not involved in ground-state spin dynamics. However, the remaining two electrons occupy two distinct orbitals, allowing for both paired and unpaired configurations. Due to the antisymmetric

configuration of these orbitals minimizing the spin-spin interaction energy, a spin triplet configuration where one electron occupies each of the two orbitals is the stable ground-state configuration. The singlet configuration exists in the divacancy system but is not the lowest energy configuration and is metastable – however, it plays a crucial role in initializing the spin triplet configuration by effectively “erasing” the quantum state encoded in the triplet spin configuration when the spin system undergoes the intersystem crossing.

Starting from a single electron $|s, m_s\rangle$ denoted by its spin quantum number s and its corresponding z -axis projection m_s , the goal is to understand what the implications for angular momentum are when two electrons are combined. In a two-electron system, where each electron has $s = \frac{1}{2}$, the possible spin configurations can be determined by the Clebsch-Gordan coefficients. First, we can obtain the two-electron states $|S, m\rangle$ with total angular momentum S . For total angular momentum z -axis projections $m = \pm 1$,

$$|1, \pm 1\rangle = |1/2, \pm 1/2\rangle \otimes |1/2, \pm 1/2\rangle, \quad (3.1)$$

while $m = 0$ has the following coupled configurations,

$$|1, 0\rangle = \frac{1}{\sqrt{2}} (|1/2, +1/2\rangle \otimes |1/2, -1/2\rangle + |1/2, -1/2\rangle \otimes |1/2, +1/2\rangle), \quad (3.2)$$

$$|0, 0\rangle = \frac{1}{\sqrt{2}} (|1/2, +1/2\rangle \otimes |1/2, -1/2\rangle - |1/2, -1/2\rangle \otimes |1/2, +1/2\rangle). \quad (3.3)$$

Three states satisfy the requirement $S = 1$; these form the triplet configuration

$$|+1\rangle = |\uparrow\uparrow\rangle, \quad (3.4)$$

$$|-1\rangle = |\downarrow\downarrow\rangle, \quad (3.5)$$

$$|0\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle), \quad (3.6)$$

where we have simplified the representation $|s, m_s\rangle$ to $\{|\uparrow\rangle, |\downarrow\rangle\}$. The remaining state with

$S = 0$ constitutes the singlet state

$$|0, 0\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle). \quad (3.7)$$

In the triplet configuration, there are distinct values obtained by projecting the state along a selected axis; furthermore, the ground-state is stable in the triplet configuration and transitions between these distinct states are controllable, as shown in Section 4.8.6. The singlet state, on the other hand, does not have measurable projections and cannot be used to encode a qubit. Thus, from this point onwards, we consider the triplet configuration as the electron spin configuration when we refer to ground-state spin control.

These two electrons interact with the SiC lattice and thus develop distinct energy levels depending on these aforementioned spin configurations. The total energy of the system is obtained by considering how the two electrons interact with each other, as well as with externally applied fields and other spin systems. This is formulated as a Hamiltonian operator, which for the ground-state spin-1 system of the divacancy is

$$H = \mathbf{S} \cdot \mathbf{D} \cdot \mathbf{S} + \gamma \mathbf{B} \cdot \mathbf{S} + \sum_i \mathbf{S} \cdot \mathbf{A}_i \cdot \mathbf{I}_i, \quad (3.8)$$

where $\mathbf{S}$ is the vector of spin-1 operators

$$\mathbf{S} = \begin{bmatrix} S_x \\ S_y \\ S_z \end{bmatrix}, \quad (3.9)$$

$\mathbf{D}$ is the zero-field tensor, γ is the electron gyromagnetic ratio, $\mathbf{B}$ is the magnetic field pseudovector, $\mathbf{A}_i$ is the hyperfine tensor of the i th nuclear spin, and $\mathbf{I}$ is the vector of spin- $\frac{1}{2}$ Pauli operators corresponding to the i th nuclear spin (all naturally occurring nuclei in SiC are either $I = 0$ or $I = \frac{1}{2}$).

First, we isolate the zero-field term,

$$H_{\text{ZF}} = \mathbf{S} \cdot \mathbf{D} \cdot \mathbf{S} \quad (3.10)$$

which defines the dipolar spin-spin interactions between the two electron spins contained within the defect system. For some cases, this term can be intuitively thought of as defining the energy cost of having the two electron spins in an aligned triplet state ($|\uparrow\uparrow\rangle$ or $|\downarrow\downarrow\rangle$) compared to the anti-aligned state ($|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle$). $\mathbf{D}$ has the property of being diagonalizable as well as traceless, allowing us to explicitly define it in matrix form

$$\mathbf{D} = \begin{bmatrix} D_{xx} & & \\ & D_{yy} & \\ & & D_{zz} \end{bmatrix} \quad (3.11)$$

where $D_{xx} + D_{yy} + D_{zz} = 0$. By expanding Eq. 3.10 and rearranging, we obtain

$$H_{\text{ZF}} = D \left(S_z^2 - \frac{S(S+1)}{3} \right) + E \left(S_x^2 - S_y^2 \right) \quad (3.12)$$

where $S(S+1)/3 = 2/3$ and can be ignored as an energy offset. $D = \frac{3}{2}D_{zz}$ is then the axial zero-field splitting, while $E = \frac{1}{2}(D_{xx} - D_{yy})$ is the transverse zero-field splitting. By examining the spectrum of S_z^2 , we clearly see that the axial zero-field splitting lifts the degeneracy between the $|\pm 1\rangle$ states from the $|0\rangle$ state. This is very much in line with our previously described intuitive understanding of the effect of the zero-field tensor. In many experiments using spin-1 systems, including nitrogen-vacancy centers in diamond and c -axis divacancies, D is assumed to be the only contribution to the zero-field tensor, which allows for the spin eigenstates to remain as eigenstates of S_z as long as the primary external perturbation consists of z -axis magnetic fields, given that $[S_z, S_z^2] = 0$.

The operators attached to the transverse zero-field splitting, $S_x^2 - S_y^2$ serve to hybridize the spin sublevels with non-zero S_z projection, $|\pm 1\rangle$. When there are no other contributions to

the spin Hamiltonian beyond the zero-field tensor with nonzero D and E , the spin eigenbasis becomes eigenstates of $S_z^2 + S_x^2 - S_y^2$ and consists of $\left\{ |0\rangle, |+\rangle = \frac{|+1\rangle + |-1\rangle}{\sqrt{2}}, |-\rangle = \frac{|+1\rangle - |-1\rangle}{\sqrt{2}} \right\}$. If nonzero magnetic fields or hyperfine interactions exist, the eigenbasis is in an intermediate regime between being pure eigenstates of S_z and $S_z^2 + S_x^2 - S_y^2$. On the surface, this may appear to complicate the understanding of the spin system as the eigenbasis now is dependent on the magnitudes of D and E , as well as the components of $\mathbf{B}$ and $\mathbf{A}_i$, if they exist. However, as we will see in Chapters 5 and 6, the presence of a transverse zero-field splitting can be leveraged as an immensely powerful ingredient in protecting the spin from external perturbations, as well as introducing new ways to control the ground-state spin system.

The Zeeman interaction term,

$$H_{\text{Zeeman}} = \gamma \mathbf{B} \cdot \mathbf{S} \quad (3.13)$$

allows for tuning of the electron spin energy levels by large ranges, by virtue of the large magnitude of $\gamma / (2\pi) = 28 \text{ GHz/T}$. Furthermore, when $\mathbf{B}$ is time-dependent and oscillating on resonance with spin transitions, rapid coherent control of the spin can be performed. The interplay between the Zeeman interaction and the zero-field splitting has already been briefly discussed; it should be noted that in most divacancy systems, D is typically the largest energy scale in the ground-state spin system and thus sets the quantization axis along S_z^2 . Operators that do not commute with S_z^2 will not affect the system to first order, hence the spin eigenstates and eigenenergies are insensitive to small quantities of B_x and B_y . With the presence of E , the quantization axis becomes $S_z^2 + S_x^2 - S_y^2$ and all components of $\mathbf{S}$ do not commute with it, thus allowing the spin to become insensitive to *all* axes of magnetic fields in the regime $|\mathbf{B}| \ll E$. This is the underlying mechanism to the improved robustness to magnetic fields near $\mathbf{B} = 0$ shown in Chapters 5 and 6.

Lastly, the hyperfine interaction terms

$$H_{\text{hyp}} = \sum_i \mathbf{S} \cdot \mathbf{A}_i \cdot \mathbf{I}_i \quad (3.14)$$

define the electron spin's coupling to nearby nuclear spins. This interaction has been used to mediate two-qubit gates, and is also the basis for decoherence stemming from nuclear spin fluctuations. In this thesis, we typically discard this term, as the purely quantum mechanical interactions defined by this Hamiltonian can be approximated with a classical magnetic field noise. This is done since we are not utilizing these nuclear spin states as basis states in a quantum information application, and instead considering them as perturbations towards the energy levels of the electronic states.

3.5 Response to electric fields

The ground-state spin of many divacancy configurations have been shown to be responsive to applied ac and dc electric fields. The mechanism through which electric fields couple to the spin is derived from a modified zero-field Hamiltonian

$$H_{\text{elec}} = d_{\parallel} F_{\parallel} S_z^2 + d_{\perp} F_{\perp} (S_x^2 - S_y^2) \quad (3.15)$$

where $d_{\parallel}$ and $d_{\perp}$ are the parallel and perpendicular Stark-coupling parameters of the ground-state spin, and $F_{\parallel}$ and $F_{\perp}$ are the parallel and perpendicular electric field components relative to the z -axis. Thus, the effective action of electric fields is to modify the magnitude of D and E . It has been shown that basal divacancies (with non-negligible E) have increased susceptibility to $F_{\perp}$ given that $d_{\perp}/d_{\parallel} > 10$ in these configurations [54]. This becomes particularly relevant when the spin energy levels are determined by the magnitude of E to first order, such as when operating near $\mathbf{B} = 0$. As we will show in Chapter 6, electric field noise can become non-negligible at these conditions and can even be the limiting factor for

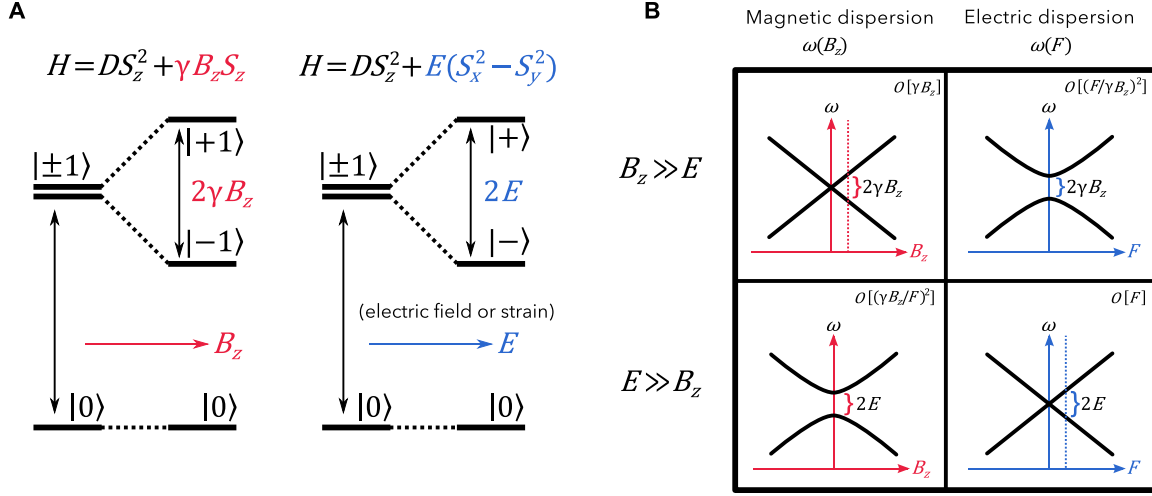


Figure 3.2: **Divacancy ground-state spin energetics.** (A) Ground-state spin energy levels under the application of z -axis magnetic fields B_z and electric field and/or strain F . A transverse zero-field splitting behaves like a static applied strain, lifting the degeneracy of the $|\pm 1\rangle$ states in the absence of any applied fields, and also mixing the spin states into the $|\pm\rangle = \frac{1}{\sqrt{2}}(|+1\rangle \pm |-1\rangle)$ basis. (B) Dispersion relations of applied magnetic field B_z and applied electric field F . In the case of c -axis divacancies, $B_z \gg E$ is typically encountered for most operating conditions. In this scenario, magnetic field fluctuations affect the spin energy to first order, whereas electric field fluctuations are suppressed to second order. For basal divacancies operated at $E \gg B_z$, magnetic fields are suppressed to second order. Instead, the spin is first-order sensitive to electric field perturbations.

spin coherence in some cases. This phenomenon has been described in both nitrogen-vacancy centers in isotopically purified diamond [59] as well as divacancies in isotopically purified SiC [39].

It should also be noted that $S_x^2 - S_y^2$ can be reformulated in terms of the spin raising and lowering operators, $S_{\pm} = S_x \pm iS_y$, as $\frac{1}{2}(S_+^2 + S_-^2)$. In this representation, it becomes clear that time-dependent modulation of E at frequencies resonant with an energy splitting between two spin states separated by two angular momentum quanta, $\Delta m_s = \pm 2$, can potentially induce a transition between those two states. This has been used to actuate magnetically forbidden spin transitions between the $|\pm 1\rangle$ spin states at high magnetic field in c -axis divacancies [27] as well as in driven basal divacancies, as shown in Chapter 6.

3.6 Response to strain and thermal fluctuations

The divacancy's response to applied strain and shear has been characterized using surface acoustic wave devices [28]. Various components of strain and shear can couple to all present terms in the zero-field tensor, and thus $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ mixing can occur, thus allowing for transitions to be realized between all three spin sublevels in the spin-1 system using strain. However, in our system, we simply consider static strain and shear as the relevant perturbative component, which allows us to treat strain and shear in a similar fashion as applied electric fields. Thus, the same modified zero-field Hamiltonian for electric fields can be used to assess the effect of strain and shear, but just with different coupling prefactors.

Thermal effects on the ground-state spin have been studied [44], and the primary contribution is a temperature dependence in D and E . This dependence is more pronounced at higher temperatures, and becomes highly suppressed at cryogenic temperatures [60]. For the purposes of our studies, the temperature dependence of the spin energies can be neglected without affecting the analysis and general understanding.

3.7 Optical fine structure

With the ground-state spin structure described, it is also useful to understand how the spin behaves in the excited state. For neutral divacancy systems, the typical method of optical excitation involves promoting an electron from the upper-most paired orbital to one of the unpaired orbitals forming the ground-state triplet. This energy difference varies between the different divacancy configurations, and can be as high as 277.9 THz for kh divacancies, or as low as 264.9 THz for hh divacancies. Furthermore, the spin manifests in these energy levels as the fine structure, and can be resolved with tunable narrow-line laser.

For c -axis divacancies, diamond nitrogen-vacancy centers, and other defect systems with C_{3v} symmetry, the fine structure is deeply studied and well-understood. The excited state

consists of an orbital-doublet, spin-triplet system resulting in six distinct transition energies, two of which are degenerate under perfect symmetry. External strain and electric field break this symmetry and lift the degeneracy of these two levels, resulting in six resolvable spectral lines. Applied strain and electric fields have been extensively used to tune the excited-state energy levels in a controlled fashion. Furthermore, these resonances have spin-selective character, and can be used for spin-selective initialization and readout, as described in the next section.

Our focus here will be on kh divacancies, where the basally oriented nature of the configuration induces a symmetry-breaking effect in the excited state orbital configurations. This broken symmetry is most easily understood as equivalent to an applied transverse strain, which causes the two excited state orbitals to linearly shift away from each other [61]. In the case of a kh divacancy, this interaction is strong enough to split the two orbitals by over 3 meV, as predicted by density functional theory. In experiments, we see signs of this splitting by only observing three out of the six optical transitions, which is understood to be the fine structure associated with the orbital with lower energy. An optical signature from the higher energy orbital has yet to be definitively reported. Emission spectroscopy of single kh divacancies show only a single spectral line corresponding to the lower energy orbital, with an second line notably absent. This can be explained by invoking Kasha's rule, which states that photoluminescence is expected to occur from the lowest excited state and all excited states with higher energy will rapidly relax to the lowest excited state. Internal conversion involving a rapid non-radiative transition from the upper orbital to the lower orbital is believed to be the relaxation mechanism. However, Kasha's rule is only applicable to divacancies with regards to radiative emission. Absorption of incident laser light is expected to yield spectral resonances corresponding to both the upper and lower orbitals. Thus, the full excited-state structure of the kh divacancy, as well as basal divacancies in general, is still an open question and merits further study.

The lower excited-state orbital, termed A' , hosts a spin triplet configuration. Just like

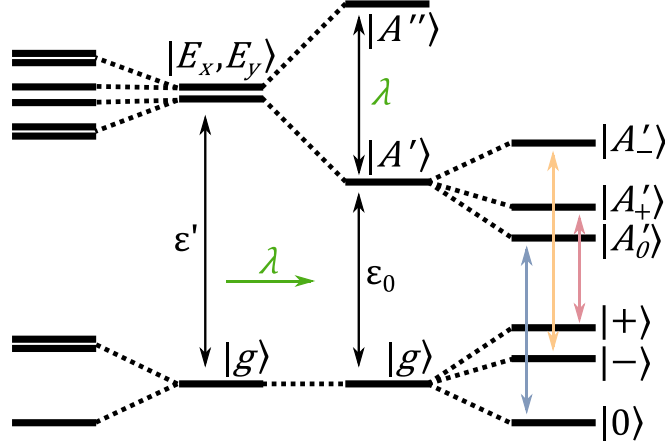


Figure 3.3: **Optical structure of basal divacancies.** The first excited state is an orbital doublet, spin triplet. In the high-symmetry c -axis divacancy configurations, the excited-state orbitals are labeled $|E_x\rangle$ and $|E_y\rangle$. The resulting six states are close in energy at low strain. For basal divacancies, the low symmetry configuration manifests in the excited state as an effective strain, λ . This splits the excited-state orbitals and produces two widely separated spin triplet manifolds. For the kh divacancy, we observe spin-selective optical transitions between the ground-state spin manifold and the first excited state. ε_0 , the energy splitting between the ground-state and excited-state orbitals in kh divacancies, is approximately 277.9 THz.

in the ground state, the presence of both an excited-state axial and transverse zero-field splittings, D_{ES} and E_{ES} , induces a mixed eigenbasis at low applied field. In this case, the excited-state spin Hamiltonian can be expressed as an analog of its ground-state counterpart,

$$H = \mathbf{S} \cdot \mathbf{D}_{\text{ES}} \cdot \mathbf{S} + \gamma \mathbf{B} \cdot \mathbf{S} + \sum_i \mathbf{S} \cdot \mathbf{A}_{i,\text{ES}} \cdot \mathbf{I}_i, \quad (3.16)$$

where $\mathbf{D}_{\text{ES}}$ is the excited-state zero-field tensor and $\mathbf{A}_{i,\text{ES}}$ is the excited-state hyperfine tensor with the i th nuclear spin. In general, since the orbital configuration is different in the excited state, the electron spin wavefunction is also distinctly different in the excited state, so $\mathbf{D}_{\text{ES}} \neq \mathbf{D}$ and $\mathbf{A}_{i,\text{ES}} \neq \mathbf{A}_i$. Thus, non-trivial spin-mixing effects can occur if the magnetic field is applied in a regime where the magnetic field magnitude is non-zero, yet smaller than either excited- or ground-state transverse zero-field splittings. In order to simplify the optical selection rules, it is then typically helpful to operate as close to zero applied magnetic field as possible, ensuring that the spin eigenbases are as close to unchanged

when transitioning from the ground to excited state. This is particularly important when considering the cyclicity of optical transitions. Ideally, optical excitation leaves the spin state unperturbed, and then the optical transition can be continually addressed, yielding many emitted photons to allow for efficient state readout. However, if the eigenbasis between the ground and excited state rapidly fluctuates under optical excitation, it can be expected that spin mixing may introduce adverse perturbations to the spin state, quickly degrading the spin projection and inducing an effective energy relaxation effect in the spin.

For basal divacancies, a major distinction in the optical selection rules must be made when compared to *c*-axis divacancies and diamond NV centers. For those higher symmetry systems at low strain, the optical selection rules dictate that linear polarization excites $|0\rangle$, whereas circular polarization selectively excites $|+1\rangle$ or $|-1\rangle$ to a corresponding excited-state counterpart. With increasing transverse strain, these selection rules are modified so that $|\pm 1\rangle$ are selectively excited using linear polarization [61, 62]. Considering the basal divacancy's lowered symmetry, and its similarity to a *c*-axis divacancy with a large amount of strain, we expect linearly polarized light to excite all three spin sublevels into the first excited-state orbital A' .

The optical structure is also responsive to applied fields and strain. Given the negligible difference in g , the Landé g -factor, of the electron spin between the ground and excited states of the divacancy, magnetic fields do not shift the positions of the optical resonances, to first order. However, the electric dipole is significantly larger in the excited state when compared to the ground state, allowing for a tunable energy difference under applied electric fields. By using the dc Stark effect, the optical resonances of these solid-state spin systems can be continuously adjusted [29, 30, 58, 63, 64]. This phenomenon is utilized in Chapter 5 to implement rapid, gigahertz-frequency shifts of the energy levels in order to produce Landau-Zener-Stückelberg interference. Furthermore, by tuning the optical resonance of two distinct emitters into indistinguishability, remote entanglement protocols can be implemented even in the face of local inhomogeneities [34].

However, uncontrollable effects from electric fields and strain can also couple into the excited state orbital energy levels. Typically, electric fields introduce rapid time-dependent inhomogeneities, while strain is generally more static and induces the aforementioned local inhomogeneities that shift the optical resonances of individual defects from each other. In principle, however, both electric fields and strain can induce similar effects in the excited state. A particular complication that is commonly encountered is that electric field noise is typically generated by photoactive defects nearby the divacancy. These are lattice impurities that can undergo changes in charge state when the control laser excitation is applied. The resulting charge state fluctuations then cause the electric field at the divacancy to become randomly distributed over time. Typically, these fluctuations are unavoidable, as the very nature of probing the divacancy’s optical resonance causes the disturbances to photoactive defects. This has resulted in broadened optical linewidths well above the expected transform limit of approximately 10 MHz [45]. However, recent breakthrough advances in electric field control have shown that statically applied electric fields can induce charge depletion effects, effectively neutralizing the nearby photoactive defects and preventing their charge fluctuation [29, 30]. The result is significantly narrowed optical linewidths, with the best values rapidly approaching the transform limit.

In order to evaluate the limitations of the optical linewidth, an accurate measurement of the radiative lifetime must be performed. This can be accomplished in many ways – one of which is performing coherent control in the optical domain and evaluating the decay from the excited state to the ground state [29, 65, 66]. By optically exciting the solid-state defect with a short pulse of light while histogramming the arrival times of the emitted photons, statistics can be drawn about the excited state population as a function of excitation time as the defect is more likely to emit a photon when it is in the excited state. After the light is disabled, any residual population in the excited state undergoes radiative T_1 relaxation, which manifests as an exponential decay in the number of emitted photons with respect to time after the end of the light pulse.

Experimental demonstrations of the fine structure of basal divacancies and optical coherent control are further detailed in Chapter 5.

3.8 Spin initialization and readout

Quantum information manipulation cannot be practically accomplished without initialization and readout of the quantum state [3]. In the divacancy, as well as many other optically active solid-state defects, the optical interface allows for high-fidelity initialization and readout of the spin. As we will see in later sections, optical initialization is universal to all of our spin manipulation techniques, and readout with high signal-to-noise ratios allows us to make high-quality measurements of the properties of the defect.

We first discuss how spin initialization is accomplished in the divacancy system. As noted in Section 3.7, the ground and excited states of the divacancy have spin triplet character, allowing for spin-preserving resonant optical transitions to be driven. However, the existence of one or more singlet states in the divacancy indicates that there is a pathway for the spin to lose that spin character. The most immediate pathway is the intersystem crossing (ISC), which has been shown to couple the singlet states to the ground- and excited-state triplet manifolds with spin selectivity. By exciting the spin system off-resonantly, all spin states are excited to the excited state with near-equal rates. The ISC preferentially couples the $m_s \neq 0$ spin states relative to the $m_s = 0$ spin state, creating a difference in non-radiative decay rates for the two classes of spin states. Correspondingly, the radiative decay rates are modified, and off-resonant excitation can yield spin-dependent optical contrast of a few percent in divacancy defects [57]. Furthermore, the preferential non-radiative coupling of the $m_s \neq 0$ spin states into a singlet state rapidly “erases” the spin information in those spin states, as the ISC between the singlet states and the ground-state triplet preferentially populates $m_s = 0$. Thus, over many off-resonant optical excitation cycles, the spin is asymptotically initialized into $m_s = 0$ with high fidelity. For nearly all of the experiments described in this thesis, 905 nm and/or 975 nm excitation light is used as an off-resonant excitation source to

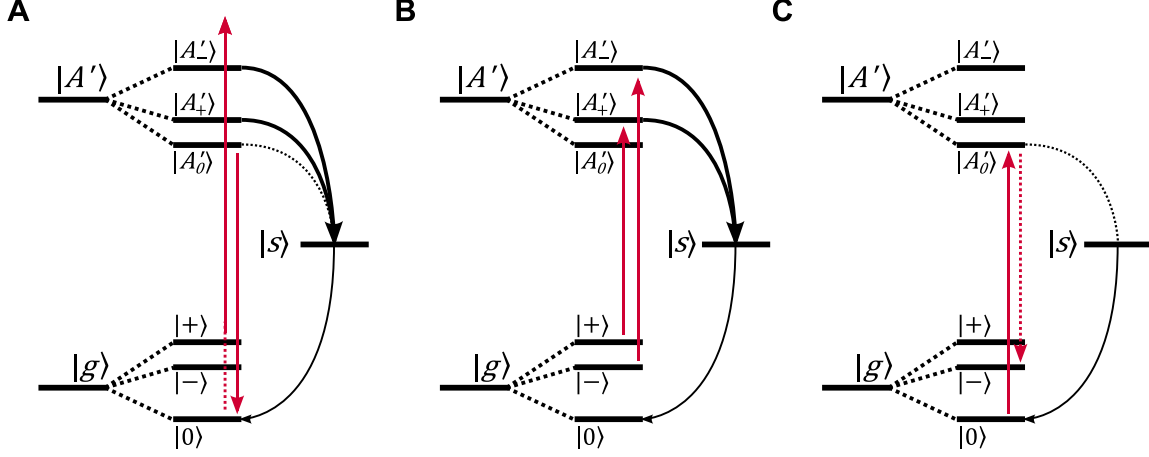


Figure 3.4: **Initialization and readout of basal divacancy spins.** (A) Off-resonant initialization and readout of the spin. By applying laser excitation with higher energy than the energy splitting between the ground and excited states, a transition can occur with the assistance of phonon processes. The states with less $m_s = 0$ character couple stronger to the singlet state $|s\rangle$ compared to the $m_s = 0$ state. The singlet to ground state ISC preferentially populates $m_s = 0$, which initializes the spin. Readout is simultaneously carried out by observing the relatively higher radiative emission from the $m_s = 0$ state. (B) Resonant initialization of the spin. By tuning resonant excitations to directly excite the states orthogonal to $m_s = 0$, rapid depopulation of those states occur through the ISC and excited-state spin mixing. (C) Resonant readout can be accomplished by selectively exciting the spin level corresponding to $m_s = 0$, which permits many optical cycles in general. Excited-state spin mixing causes the spin state to enter into an orthogonal subspace, limiting further readout of the state.

induce spin initialization into $m_s = 0$.

Another avenue of spin initialization is resonant spin pumping, which is a technique commonly used in atomic physics to optically initialize atoms without an intrinsic ISC. Here, a narrow-line tunable laser is applied resonantly with the transitions connecting the states that are not the desired initialized state to the excited state. Once in the excited state, any kind of fluctuation in the eigenbasis may induce spin mixing, which can cause non-zero rates of relaxation back to the ground-state, but with different spin character. By continuously pumping the manifold excluding the initialized state, the eventual desired result is that the spin mixing can occur rapidly enough to selectively transfer population into the

initialized state prior to ground-state relaxation and depolarization. This method has been demonstrated to enhance spin initialization fidelities beyond those reached simply using off-resonant excitation [26]. While this can be used to further improve the spin initialization fidelity in divacancy defects, the already high initialization fidelity ($>94\%$) using solely off-resonant excitation is sufficient for proof-of-principle demonstrations without significant limitations in experimental capabilities.

Once initialized, the spin may be manipulated using any of the previously discussed interactions. It is then typically desirable to be able to measure the resultant evolved spin state with high accuracy and fidelity. We have described how off-resonant excitation can produce spin-dependent photoluminescence. However, the contrast, or the relative difference in photoluminescence between the two spin states, is very low for off-resonant excitation (order 10%). In terms of obtaining good measurement signal-to-noise ratios, this can be detrimental. Furthermore, for particular quantum optics experiments, such as remote entanglement, the emitted photons under off-resonant excitation have less pure polarization, leading to lower entanglement indistinguishability and visibility. To overcome both of these issues, readout through resonant excitation is preferred. By tuning a narrow-line laser to be resonant with the spin-selective optical transition corresponding to exciting the desired “bright” spin state, high contrast spin-selective photoluminescence can be obtained. In principle, excitation of a highly cycling optical transition can yield many photons prior to a disturbance of the spin, whereas spurious excitation of optical transitions corresponding to other spin states is highly suppressed according to the Rabi formula (Eq. 2.31). Experimentally, disturbances to the spin limit the number of photons extracted from the defect system before the spin information is lost. Pivotal demonstrations of single-shot readout of the spin have been achieved by engineering the defect system and overcoming these disturbances and photon collection losses [26]. Furthermore, the resonant nature of the excitation allows for energy splitting between the ground- and excited-state orbitals to be clearly mapped. This is utilized to determine the fine structure of basal divacancies, as well as observe interferometric phenomena

in Chapter 5.

CHAPTER 4

EXPERIMENTAL CONTROL OF QUANTUM SYSTEMS

4.1 Introduction

This section provides an overview of the experimental techniques involved in isolating single divacancies at cryogenic conditions and probing the optical and spin properties of the divacancy at various operating conditions. Confocal microscopy is introduced, as well as the basic optical elements that typically constitute a laser scanning confocal microscope. We then describe how the cryogenic apparatus is assembled and operated in order to ensure reliable operation of single divacancies near 5 K. Then, we outline how defect emission is measured and correlated to optical and spin properties of the divacancy, allowing us to examine the quantum mechanical characteristics of these defect centers. Lastly, we provide a summary of the basic experimental sequences and procedures used to probe the individual quantities of interest, including spin and optical resonance structures as well as coherence lifetimes.

4.2 Confocal microscopy and optical control

At the heart of all our solid-state defect studies is a high-resolution confocal microscope setup. These microscopes are designed so that they are extremely stable with respect to external fluctuations in temperature or motion, while also providing the optical resolution and stability necessary to resolve and track single defects for an extended period of time. Furthermore, the need for versatility and potential rapid reconfiguration to adapt the microscope to various experiments and applications calls for particular design considerations, which are detailed in this section.

Confocal microscopy, in contrast to widefield imaging, is used because of the inherently minute spatial volumes involved in our solid-state defect studies. In particular, single defects

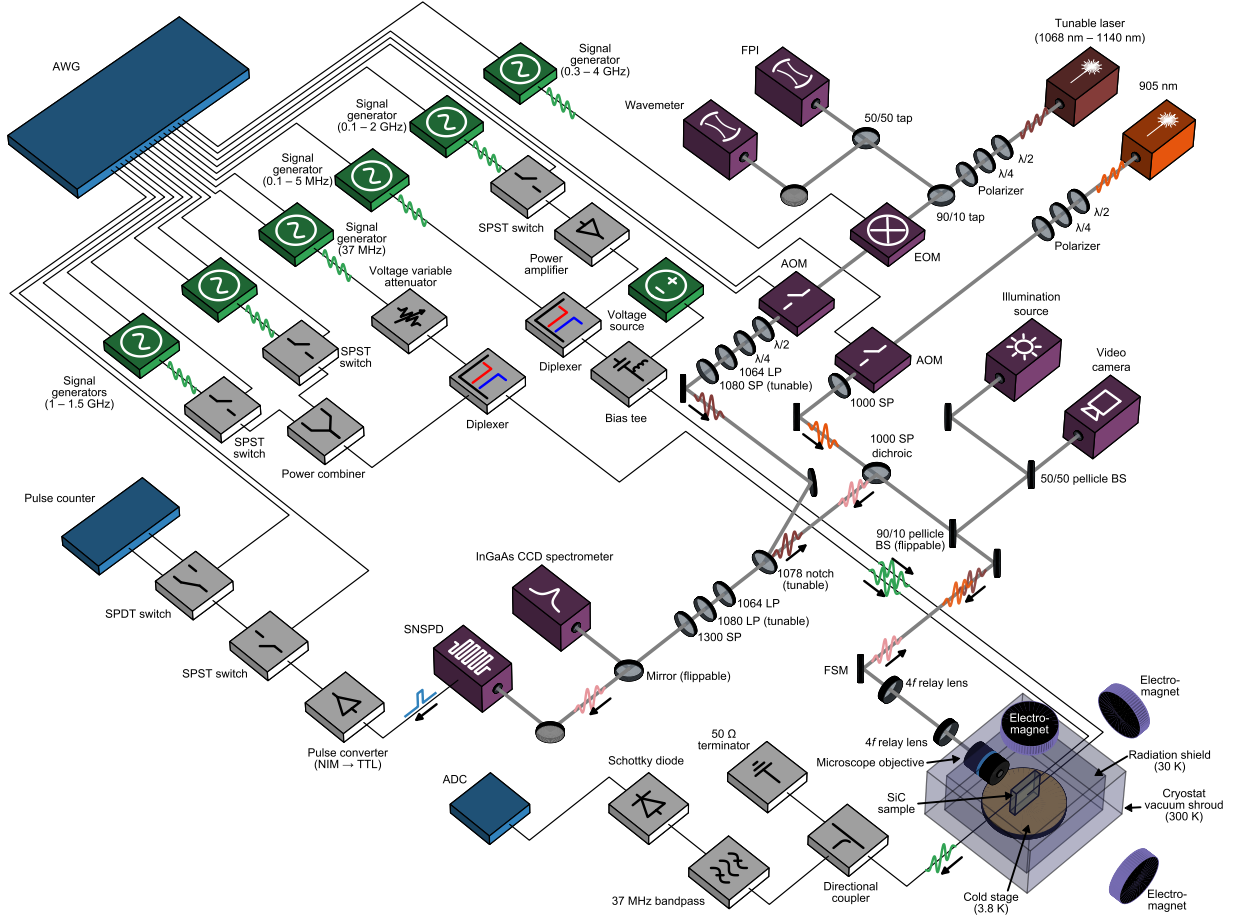


Figure 4.1: **Confocal microscopy setup.** Electrical connections using coaxial cables are indicated by solid black lines. Optical paths are indicated by wide gray lines. Direction of propagation indicated by arrows. Active timing electronics shown as blue modules. Signal generators, oscillators, and voltage sources shown as green modules. Other electronics shown as gray modules. Active optical elements shown as violet modules. Optical elements are mirrors unless otherwise annotated. A microwave line for electrical driving is open-terminated on the sample, while another microwave line for magnetic driving is 50-ohm terminated externally.

are treated as point source emitters, and thus the emission from the region around that point can typically only serve to introduce spurious background into the measurement. In order to optimize the signal from the emitter relative to the background noise, we use confocal microscopy in order to selectively address a narrow focal plane determined by the pinhole size in an optically conjugate plane (in our case, the core size of a single-mode fiber). The spatial resolution within that plane is then determined by the achievable spot size of the

microscope optics, which is effectively diffraction limited at our wavelength range of about $1\text{ }\mu\text{m}$.

The entire microscope setup is assembled on an optical table. The table can be isolated from mechanical disturbances in the laboratory through a pneumatic mechanism that allows the table surface to float on a cushion of nitrogen gas, effectively decoupling it from vibrations in the ground. Throughout these studies, we have not used this decoupling mechanism, as the vibrations in our laboratories were undetectable in the context of our experiments. Furthermore, the large mass of the stainless steel surface significantly dampens mechanical vibrations from human activity and equipment motion, effectively eliminating mechanical fluctuations on the optics.

The confocal microscope setup is shown in Fig. 4.1. We describe the purpose and figures of merit for each component in this following enumeration:

1. **A fixed, 905-nm excitation source:** This off-resonant excitation source is used for off-resonant spin initialization and off-resonant spin readout. It is also used for non-selective excitation of divacancies for initial defect searches. Furthermore, 905-nm excitation light provides charge repumping and stabilization of divacancy defects. The core consideration for this excitation source is the output optical power, which is typically over 200 mW for our diodes. This allows for significant working overhead when the beam passes through passive and active components that have imperfect transmission specifications.
2. **A tunable laser source:** This tunable laser is used for resonant spin initialization and resonant spin readout. Ranging in the near-infrared (1068-1140 nm), the resonant nature of this excitation source means that significantly less power is required for interaction with single defects. However, even then, significant losses are incurred through active and passive components, so high output power at the laser diode is a necessary requirement. Furthermore, spectral stability and finesse is a central requirement for proper addressing of resonant optical transitions. Thus, linewidths much less than the

natural linewidth of the emitter is required (<1 MHz).

3. **Acousto-optic modulators (AOMs):** In order to selectively gate the laser sources, we choose to use AOMs, which are active components consisting of an acousto-optic crystal through which the light is passed. When a resonant surface acoustic wave is applied to the acousto-optic crystal, the acousto-optic effect produces angular deflections in the crystal, with angles dependent on the wavelength of the incident light as well as the wavelength of the surface acoustic mode. We then block the non-deflected light and only allow the first deflected beam to pass, using an optical iris. Radio-frequency excitation is used to generate the surface acoustic waves, which is modulated by external digital logic. The rise and fall times of AOMs are typically on the order of tens of nanoseconds (as seen in Fig. 5.2B), which can be comparable to the excited-state dynamics of divacancy defects. Thus, for applications requiring short rise and fall times, AOMs may not be the ideal device to use, and instead electro-optic modulators should be used. However, the extinction ratios of these devices are typically very good (exceeding 40 dB) and are generally sufficient for use as an optical switch.

4. **Electro-optic modulators (EOMs):** A related optical device to the AOM is the EOM. Specifically, a phase EOM utilizes an electro-optic effect to introduce controllable phase components in the incident light. Time-dependent control of the phase can then allow for near-arbitrary control of the light's amplitude and frequency. Specifically, starting with a plane wave representation of the incident laser,

$$A \exp [i (\omega_0 t + \phi)], \quad (4.1)$$

we can arbitrarily modulate the phase with a time-dependent signal,

$$A \exp [i (\omega_0 t + \phi(t))]. \quad (4.2)$$

If $\phi(t) = B \sin(\omega_1 t)$, then we can use the Jacobi-Anger expansion to obtain

$$A \exp[i(\omega_0 t + B \sin(\omega_1 t))] = A \exp(i\omega_0 t) \sum_{n=-\infty}^{\infty} J_n(B) \exp(in\omega_1 t), \quad (4.3)$$

where $J_n(x)$ is the n th Bessel function of the first kind. For particular values of B and ω_1 , this can be used for the generation of multi-color optical probe signals, as well as rapidly tunable frequency scans of the light incident on the single defect. Furthermore, the bandwidth of EOMs are typically much higher than AOMs, allowing for GHz modulation of the optical light and fast pulsing of a continuous-wave laser.

5. **Fabry-perot interferometer (FPI):** In order to characterize the spectral quality of the light emitted by the tunable laser, a device that can take an unknown light source and return a high-resolution spectrum is required. A high-finesse Fabry-Perot interferometer provides that capability. The cavity sustains a set of modes, each spaced apart by the cavity free spectral range (FSR). The cavity length is tunable by adjusting a built-in piezo's applied voltage, which allows for fine tuning of the spectral position of the cavity mode. When the cavity is scanned over its FSR, the full spectrum of light incident on the cavity can be reconstructed. Absolute information about the spectrum (wavelength/frequency of particular features) cannot be reliably obtained, however, necessitating additional characterization tools, such as a wavemeter.
6. **Wavemeter:** In order to characterize the absolute wavelength of the light emitted by the tunable laser, a device that can compare light with unknown wavelength to a known reference wavelength is needed. Such a device is a wavemeter, which in our microscopy setup is a commercially obtained Fizeau interferometer. The reference wavelength is provided by an internal HeNe laser. The input light must typically be monochromatic, as a multimode input signal can cause the wavemeter to return spurious readings of wavelength.

7. **Linear polarizer:** Although the emission from the various laser sources is polarized, we further ensure that the polarization fidelity of the excitation is optimized by passing the beam through film-based linear polarizers. This can improve the polarization extinction ratio by another five orders of magnitude. This allows for high-fidelity addressing of polarization-selective transitions, potentially increasing the optical absorption efficiency and reducing the back-scattered photons that would otherwise contribute to background noise.
8. **Half- and quarter-waveplates ($\lambda/2$, $\lambda/4$):** The spatial alignment of the electric dipole of single defects indicates that there exists a preferential alignment of the incident excitation for maximal excitation of the optical transition. Thus, these waveplates allow for full control of linear and circular polarization of the resonant excitation, ensuring maximal efficiency when addressing the optical transition. Furthermore, AOMs and EOMs typically have sensitivity to the polarization of incident light, and so arbitrary control of the polarization allows for maximal efficiency in operating these devices.
9. **Combiner dichroic:** In order to direct both the off-resonant and resonant lasers into a single collinear beam, a passive optical combining element is required. Typically, a dichroic filter with a cutoff wavelength situated between the off-resonant and resonant wavelength ranges (between 905 and 1078 nm) is sufficient. We use a 1000 nm shortpass dichroic, which passes the off-resonant laser at 905 nm and almost entirely reflects both the resonant laser and divacancy emission (>1078 nm).
10. **Excitation/collection dichroic:** A problem particular to our excitation scheme is the overlap between the resonant excitation laser and the resulting photoluminescence from the divacancy. When optically excited on resonance, the divacancy emits at the exact same wavelength as the excitation laser through its zero-phonon line, in addition to emitting into a broad phonon-assisted sideband. For many applications demanding

indistinguishable photons, such as remote entanglement and quantum communication protocols, collection of zero-phonon line emission is required. However, in experiments concerned with measuring the spin state or the absorption properties of the optical fine structure, collection of photons emitted into the phonon-assisted sideband is sufficient. In this case, we use a dichroic that reflects light at the zero-phonon line, allowing for resonant laser excitation to be directed to the sample, while transmitting light with wavelengths outside of a 40-nm window around the zero-phonon line. We accomplish this by using a notch filter centered at 1064 nm with a 44 nm full-width at half-maximum reflection window. Although this filter is specific to the kh divacancy emission at 1078 nm, other notch filters can be installed to address other divacancy configurations with comparable effectiveness.

11. **Visible imaging camera:** Prior to using the lasers to find single emitters, we typically use a visible imaging camera setup to view the sample, and position the sample so that the laser is near a region of interest. In tandem, an illumination source is projected onto the sample so that features are visible. Filtering of the illumination source is necessary to mitigate back-scattered light from anti-reflection coatings on lenses from obscuring visible features on the sample.
12. **Fast steering mirror (FSM):** In order to perform laser raster scans for building photoluminescence images of the sample and for rapid spatial feedback and tracking of single defects, a fast steering mirror is introduced into the optical setup prior to the objective. This piece of equipment consists of a mirror mounted on two adjustable axes which are controlled by electromagnetic coils. By inputting control voltages along these two axes, angular motion of the mirror is achieved, allowing for arbitrary deflection angles of a laser beam incident on the sample. In combination with the $4f$ relay pair and microscope objective (discussed next), these deflections, to first order, become translations of the focused beam (as well as the optical interrogation region).

Thus, arbitrary translations of the excitation and collection spots can be obtained by adjusting the angles of the FSM.

13. **$4f$ relay pair:** A $4f$ relay pair (also known as a telescope pair), allows for angular deflections produced by the FSM to manifest as angular deflections at the entrance aperture of the objective. This is accomplished by the pair of lenses in the $4f$ pair, which are spaced from each other at $2f$, where f is the focal length of both of the lenses. The total path length between the FSM and the objective must then sum to $4f$, which allows for the relay pair to be adjusted in position relative to these elements. While one lens produces a focal plane for the collimated beam and converts the angular deflection to a translation, the second lens inverts these operations, yielding a collimated beam and an angular deflection at the end of the relay pair. Special consideration is given to the size of these relay pair lenses, as they must be relatively large (two inch diameter or larger) in order to accommodate the wide range of deflection angles produced by the FSM.
14. **Microscope objective:** We use a microscope objective that is designed for infrared imaging. Here, high numerical aperture (NA) and optical transmission are valued as key specifications, allowing for a large fraction of photons emitted by the defect to be collected and detected. Working distance, or the space between the front of the objective and the focal point of the objective, is also an important parameter to consider, as short working distances can limit the possible types of experiments that can be pursued. In the experiments reported here, $NA = 0.85$ and transmission throughout the divacancy emission spectrum exceeds 70%.

4.3 Cryogenic apparatus

The SiC samples are cooled to cryogenic temperatures in order to activate the high-fidelity optical interface and to suppress phonon processes that scale unfavorably at higher temper-

atures. We accomplish this by using a closed-cycle liquid helium cryostat, which allows for sample temperatures to reach 5 K and below. Special care is taken to develop a system that is particularly robust to vibrations and thermal fluctuations while simultaneously allowing for a wide variety of customization options in the form of optical windows and feedthroughs for microwave coaxial cables and optical fibers.

The operation of closed-cycle cryogenic refrigeration processes is a well-established technique that dates back to the mid-twentieth century. Here we present a brief overview of the closed-cycle process. Our system uses a Gifford-McMahon cryocooler architecture. First, a cryogenic compressor pressurizes helium gas. This pressurized gas is then fed to a cryocooler head, which allows for controlled expansion of that pressurized helium gas. When allowed to expand, the gas does work on its surroundings and extracts thermal energy. The low-pressure helium is then fed back to the cryogenic compressor to complete the cycle. After many cycles of this process, a large amount of thermal energy is extracted from the target system. This continues down to the temperature where helium expansion process equilibrates with its environment and no further work can be done (usually around the boiling point of helium near atmospheric conditions – about 4 K).

A caveat of this process is that the amount of thermal energy that can be extracted per unit time (the cooling power) is relatively small compared to the potential amount of radiative power from the environment. Thus, excellent thermal isolation of the cold stages from the environment is required in order to maintain the thermal stability and the cooling capabilities of the system. This is typically achieved by designing an appropriate vacuum chamber, inside which a material with high thermal conductivity (typically copper) is attached to the cryocooler and serves as the cold stage. The vacuum chamber, once pumped out with a turbomolecular pump, becomes nearly void of any gas molecules, and thus the rate of convective heat transfer between the environment and the cold stage is minimized. Radiative heat transfer is still present, however, and is addressed by the installation of a radiation shield that is highly reflective in the infrared spectrum (typically aluminum or

gold-plated copper). In our system, this radiation shield envelopes the cold stage and sample entirely and ensures that minimal radiative heating occurs.

For our experiments, we also require optical access, as well as a way to feed in microwave signals. These are accomplished through optical windows and microwave feedthroughs, respectively. The optical window is a few-millimeter thick piece of glass that has a custom anti-reflective coating to maximize transmission of laser light and defect photon emission. The microwave feedthroughs consist of hermetically sealed coaxial adapters (typically SMA to SMA) fixed into an aluminum panel that is installed onto the side of the cryostat. This allows for a continuous microwave transmission line to be constructed from the exterior of the cryostat directly to the sample.

In order to optimize thermal properties at the sample, we mount our SiC samples directly to a copper block, which is subsequently mounted to various copper adapter pieces that link it to the cold stage. This is in contrast to mounting the SiC sample onto a printed circuit board (PCB) with exposed ground pours. This change was made after a series of experiments allowed us to determine that the thermal conductivity from one face to the other of the PCB was significantly limited by the weak conductivity of the small copper-plated vias. Thus, incident heat flux from the microscope objective and the radiation shield was not being dissipated rapidly across the PCB to the cold stage, and the resulting sample temperatures were noticeably elevated beyond our expected sample temperature. Once we bypassed the PCB, the sample temperature reduced drastically and became much closer to the cold stage temperature. In order to retain the ease of interfacing with external coaxial connectors, we developed a PCB with a cutout for the sample and allowing for wire bonds to be drawn between the PCB and lithographically patterned designs on the surface of the sample.

4.4 Microwave control of single quantum systems

Recall Eq. 2.27. If we are able to apply electromagnetic fields with frequencies resonant with the energy splitting of two levels, we can typically effect a transition, selection rules

permitting. We accomplish this by routing the outputs of sine wave generators to on-chip, lithographically patterned wires and capacitors. These structures are in close proximity (order $10\text{ }\mu\text{m}$) to the divacancy defects under optical interrogation. This allows for highly efficient delivery of electromagnetic fields, minimizing the amount of dissipated Joule heating in the metal conductors.

Furthermore, many experiments require well-defined timing of these electromagnetic fields, in the form of pulses. In general, pulses are formed through the use of external modulation devices, such as switches and mixers, or by adjusting the inputs and parameters of the devices generating the electromagnetic fields. Modulating the amplitude, frequency, and phase of these electromagnetic fields in a pulsed fashion allows for well-defined rotations between the qubit states, forming the foundations for quantum information manipulation. For example, the calibration of a π rotation in a qubit requires tuning the amplitude and duration of an applied electromagnetic pulse (more details in Section 4.8.6).

We enumerate the core components in the microwave control subsystem of our setup here:

1. **Arbitrary Waveform Generator (AWG):** If the confocal microscope optics can be considered the heart of the setup, then the AWG is the “brain” which allows for powerful operation of the setup. Its core purpose is to properly time and modulate all the events in an experimental sequence, ranging from when the AOMs should be toggled to allow for optical pulses to reach the sample, to when microwave pulses should be applied to the sample to drive spin rotations. It accomplishes this by providing many (upwards of 16) synchronized outputs with very fine timing resolution (bandwidths exceeding 2.4 GHz and sub-nanosecond skew and jitter). Furthermore, as suggested by its name, *arbitrary* operation of these outputs is possible (within the electrical and bandwidth limits of the device). Thus, theoretically any experimental sequence can be implemented as long as the experiment can be operated within the bounds of the device. All of our pulsed sequences with time-resolved pulses of light and microwaves

require an AWG to synchronize the individual operations on the defect. By combining the modulation capabilities within the AWG with internal and external microwave oscillators, arbitrary shaped pulses with amplitude and frequency modulation can be achieved, allowing for exotic qubit operations to be implemented.

2. **Signal generator:** In order to drive many of the spin resonances present in the divacancy system, a periodic signal (specifically, a sine wave) with a tunable frequency is required. For some of these generators, they need to operate around 1 GHz. This order of magnitude is typically defined by the zero-field splitting D , which separates the $m_s = 0$ spin level from the $m_s = \pm 1$ levels. In our system, D is near 1.3 GHz. Thus, many of our signal generators operate from a few hundred MHz up to a few GHz, allowing us to probe the spin levels at low and high magnetic fields. Furthermore, direct modulation of the amplitude, frequency, and phase components of the sine wave can be accomplished through in-phase/quadrature (IQ) modulation. Other signal generators operating at lower frequencies (hundreds of kHz to tens of MHz) and higher frequencies (up to 4 GHz) allow for further control of the spin system using both electrical and magnetic excitation. Additionally, these signal generators supply the local oscillator tones to produce sum and difference frequencies when operating the EOM.

3. **Microwave switch:** A single-pole double-throw (SPDT) switch for two-state modulation of microwave-frequency signals is used for a few purposes.

One of the crucial applications is for additional extinction of microwave drive signals that can potentially induce spurious spin rotations. As an example, without a microwave switch, the typical method of producing a microwave pulse for a spin rotation is to use the IQ modulation on the signal generators. However, the IQ modulation carrier suppression is merely about 40 dBc, which indicates that the extinction (on to off) ratio of power outputted by the signal generator over the full range of IQ modulation only reaches 10000:1. When considering the spin rotation frequency, which is

linearly dependent on the amplitude of the drive, this extinction ratio then becomes only 100:1. Given the fast Rabi frequencies obtained in many of our systems, on the order of 1 MHz, along with the spin's coherence time routinely exceeding 1 millisecond, it is clear that a 100:1 extinction ratio can easily introduce spurious driving that would manifest as errors in an experiment. By inserting a microwave switch between the output of the signal generator and the sample and modulating it on its full scale (yielding an additional 60 dB extinction in power) while simultaneously actuating the signal generator IQ modulation, we obtain total power extinction ratios exceeding 100 dB, which corresponds to a 10^5 :1 Rabi frequency extinction ratio.

Another critical application is the extinction of incoming electrical pulses from the SNSPD corresponding to photon detection events. As described in Section 4.6, proper extinction of photon detection events that have weak or no correlation with the spin state can drastically improve the measurement fidelity of the system.

4. **Power splitter/combiner:** Many different microwave sources are typically combined into one on-chip drive wire or capacitor. This is typically accomplished by using a resistive power combiner (which also doubles as a power splitter when used in reverse). This component combines two microwave inputs into one output, with the caveat that each input signal experiences a 3 dB insertion loss. Furthermore, the isolation between the two input channels can be very low (near 6 dB), which can limit the range of power applied to the inputs.
5. **Diplexer:** When combining two signals with vastly distinct frequency bands, a passive element such as diplexer may be more appropriate than a typical power combiner. At its core, a diplexer is a combined high-pass and low-pass filter which has low insertion loss for in-band signals when applied to the corresponding high-frequency and low-frequency input, but has significantly increased insertion loss for out-of-band signals. This prevents leakage of one input to the other, provided that the frequency specifi-

cations are adhered to. This also allows for low loss combination of the two signals (typically <1 dB in band, compared to the >3 dB for resistive power combiners) and lower dissipation. We use these components to combine driving tones below 100 MHz (for driving spin transitions near zero field or nuclear spin control) with 1 GHz tones for driving the spin transitions primarily separated by D .

6. **Bias tee:** A diplexer with the low-pass filter taken to the extreme is effectively a bias tee. This device allows for efficient combination of dc signals (the bias) with an ac tone. We use this to apply static electric fields in conjunction with ac electric fields for spin and orbital manipulation of the divacancy.
7. **Power amplifier:** For applications where fast pulses are required, a power amplifier is necessary to maintain a sufficiently high pulse amplitude so that the total energy delivered to the quantum system is enough to modulate the spin or orbital levels.
8. **Directional coupler:** A waveguide coupler allows for monitoring of the microwave signal while still allowing the system to experience a broadband $50\ \Omega$ impedance.
9. **$50\ \Omega$ termination block:** By constructing the microwave system in a transmission topology and adding a $50\ \Omega$ termination block at the end, this avoids the presence of standing waves and allows for the dissipation of the majority of the applied microwave power at this termination block, which is designed to be highly efficient in dissipating heat. Without this element, a standing wave antinode could exist at the signal source or at the sample, which could adversely affect the stability of the quantum system.
10. **Bandpass filter:** In applications where the power of a continuous microwave drive must be monitored, a bandpass filter allows for rejection of all other microwave drives not at the continuous drive's frequency. Subsequent electronics can then accurately read the power of the continuous drive tone without spurious power contributions from out-of-band pulses.

11. **Schottky diode:** A Schottky diode can rectify microwave-frequency signals and convert them to dc signals proportional to the amplitude of the ac signal. These are used to monitor the microwave pulse sequence for debugging purposes, as well as a power detector in active feedback stabilization schemes of continuous-wave microwave tones.

4.5 Photon counting

Nearly all of the work presented in this thesis relies on counting the photons emitted from the divacancy defect under certain conditions and correlating those photon statistics with particular experimental configurations. Thus, a precise, high-fidelity method of capturing photons and counting them must be developed. To this end, we use a superconducting nanowire single photon detector (SNSPD) to convert single photons into electric pulses, which can then be manipulated with conventional electronics. A SNSPD is the preferred device for photon counting given its high quantum efficiency (the proportion of photons that are successfully converted to a pulse out of all incident photons) and low timing jitter. While avalanche photodiodes (APDs) are used in many applications to detect single photons, the small semiconductor bandgaps necessary to detect near-infrared photons precludes the use of common silicon-based APDs, requiring the use of less common InGaAs-based devices with significantly increased thermally induced dark count rates and reduced quantum efficiencies. Meanwhile, the SNSPDs used in our experiments have quantum efficiencies exceeding 80% at 1100 nm, with dark count rates below 10 counts per second. The sole drawback of operating a SNSPD is the associated cryogenic apparatus necessary to bring the nanowire detectors to a sufficiently low temperature (<3 K) in order to obtain those aforementioned specifications. However, like the closed-cycle sample cryostat system, a similar closed-cycle helium refrigeration cycle is used in our SNSPD system, allowing for long-term continuous operation without user intervention.

The SNSPD is operated by applying a current bias on the superconducting nanowire structures, thus placing the structure very close to the critical current point where it will

become normal (non-superconducting). An incident photon on the nanowire structure with sufficient energy to break Cooper pairs will reduce the effective critical current at that region, and thus causing that region to become a normal hotspot with non-zero electrical resistance. That change in resistance can then be detected with conventional electronics and can be converted to a voltage pulse. In our SNSPD system, that voltage pulse has an amplitude of about 100 mV, with a duration of tens of nanoseconds. These pulses are then converted by a module which produces TTL pulses with higher amplitudes (up to 5 V), allowing for transmission over long coaxial cables to counter modules.

4.6 Data acquisition

Once we have obtained the electrical pulses corresponding to the emitted photons, we aim to perform statistics on these pulses and correlate these statistics with the experimental parameters of the system. First, we must selectively gate the incoming pulses, as there is typically only a small window in time that we are interested in detecting photon counts, but the SNSPD will detect all photons emitted by the defect. In many of our experiments, this window corresponds exactly with the duration of when the AOM allows the tunable laser to pass through and resonantly excite a spin-dependent transition for spin-selective optical readout. By rejecting all other photon count events (for example, during the weakly-spin-selective off-resonant initialization pulse), we significantly lower the amount of spurious background counts and increase our readout contrast. We accomplish this gating by passing the electrical pulses through a microwave switch, which is actuated by a digital channel on the AWG.

Subsequently, the gated pulses are directed to one of two edge-counting channels on the DAQ by passing the gated pulses through another switch operated in a single-pole double-throw configuration, creating a two-branch logic tree. This allows us to rapidly toggle between the two counter channels at upwards of megahertz rates, enabling differential measurements that are resistant to common-mode noise and fluctuations. Finally, the edge-

counting channels of the DAQ provide a counter value that is incremented whenever an electrical pulse is incident on its input. By measuring the difference of the counter value before and after a known duration, and then normalizing by that time duration, photon count rates are calculated.

4.7 Software control

A central component to having all of the aforementioned components working in harmony is a robust software controller suite. To this end, we have developed SPYRE (Scientific PYthon Research Environment) to allow for rapid iteration and implementation of experimental logic and storage of acquired data. Developed entirely in Python, this environment leverages the mature frameworks of NumPy, SciPy, pandas, and Qt to provide high-level abstraction of data operations. Low-level communication is accomplished using the lantz package, which encapsulates the binary- and string-based messaging between the control computer and individual instruments.

Such a hierarchical design bestows many advantages to the user. Direct control and read-out of the instruments and their individual parameters and statuses can be accomplished by directly utilizing the lantz driver instance, which provides atomic commands to set and read instrument parameters as well as perform actions. This may be useful for one-off procedures or debugging. When scripted experiment logic is required, lantz drivers controlling an ensemble of instruments can be inserted into a Spyrelet (a SPYRE experimental module) and interlaced with loops and conditionals. Boilerplate is minimized, allowing for the user to focus on the details of a particular experiment. Furthermore, the user-friendliness is maintained by providing a straightforward graphical interface for inputting experimental parameter values, as well as real-time plotting of acquired data. These graphical components are decoupled from the experimental logic, allowing for reuse and adaptation to other experiments with little effort. Lastly, the acquired data is managed in a pandas DataFrame, which integrates a comprehensive lineup of built-in data manipulation methods and facilitates data

processing after acquisition and during analysis.

4.8 Experimental procedures

In this section, we provide an overview of the various implemented procedures to perform the various experiments described in this thesis. While not an exhaustive list, these procedures serve as the backbone to the toolbox of methods to probe samples to find single divacancy defects and deeply characterize their quantum properties. In particular, we outline two procedures commonly used to ensure optimal laser operation, as well magnetic field conditions for the best possible quantum coherence properties of the observed divacancy system. We then describe the process to localize a divacancy within the SiC sample, identify its resonant optical spectrum, and then probe its spin properties using the resonant spin-to-photon interface. We restrict our discussion to procedures related to the experiments presented in this thesis, and some methods, such as optically detected magnetic resonance using purely off-resonant excitation, will not be covered. Nonetheless, many of these omitted techniques can see use in certain conditions, such as when the optical resonances are too broad to perform spin-selective operations, which is a potential problem in SiC substrates with lower purity.

4.8.1 *Laser mode calibration*

The tunable laser we use for high-fidelity readout of the divacancy spin is a specially designed unit from Toptica Photonics that emphasizes high output optical power and large tuning ranges in order to address the optical resonances of the various divacancy configurations (typically ranging from 1078 nm to 1132 nm). One particular shortcoming of this external cavity diode laser is the spectral instability over large regions of the tuning range unless additional steps are taken to optimize the laser diode supply current and the voltage applied for piezoelectric fine modulation of the cavity position. More specifically, the supply current and piezo voltage must be adjusted simultaneously in proportion for optimal operation,

and this proportion is given by an adjustable feedforward constant. When the feedforward constant is properly set, any modification of the piezo voltage automatically adjusts the supply current to compensate for the shift in cavity length, allowing for significantly higher mode stability. However, when changing the laser frequency by steps larger than a few tens of gigahertz, the mode supported by the external cavity changes drastically and the laser parameters must be readjusted. However, the optimal new configuration is very difficult to determine, and has very little deterministic behavior based on the laser frequency. Thus, for large scans requiring laser frequency ranges larger than a few tens of gigahertz, a procedure must be developed for unattended operation.

We developed an automated calibration technique to optimize the single-mode nature of tunable laser at arbitrary output wavelengths, based on the manual optimization procedures first described by David Christle from our group. We tap off a small fraction ($<5\%$) of the laser output using a fiber tap and feed it into a characterization Fabry-Perot interferometer, which provides the relative spectral information within its free spectral range $\Delta\nu_{\text{FSR}}$. Under ideal circumstances, the Fabry-Perot interferometer will display n sharp, uniform height peaks as the laser frequency is tuned over a range $\Delta\nu = n \times \Delta\nu_{\text{FSR}}$. However, we often observe the number of detected peaks from the Fabry-Perot interferometer to be much larger or smaller than n when the laser is tuned over $\Delta\nu$, while also having large variations in peak amplitude. For us, this is the clearest sign that the laser is spectrally unstable and is emitting in a multimode manner.

With this in mind, we digitize the output of the Fabry-Perot interferometer by linking its amplified output signal to a data acquisition module input. As the laser frequency is scanned, we obtain a collection of peaks, as previously described. We then perform peak detection on this spectrum and define a loss function $f(\mathbf{x})$ which is proportional to the difference between the observed number of peaks and the expected number of peaks $\bar{n} = \frac{\Delta\nu}{\Delta\nu_{\text{FSR}}}$, as well as the variance in the peak heights. Our search coordinate, $\mathbf{x}$, lies in a 2D space spanned by the laser diode input current and the feedforward constant, which are both freely adjustable

within a predetermined set of boundary values. In general, $f(\mathbf{x})$ is highly nonlinear and possesses many local minima with respect to $\mathbf{x}$. In order to make the optimization routine robust to these conditions, we opted to use a stochastic, derivative-free optimization method in CMA-ES (Covariance Matrix Adaptation Evolution Strategy). We sample the search space using this method and iterate until convergence, returning $\mathbf{x}_{\text{opt}}$. We then perform a check on the loss function to ensure that it is within a particular threshold ϵ , $f(\mathbf{x}_{\text{opt}}) \leq \epsilon$. If this check fails, we restart the optimization algorithm with randomized initial components of $\mathbf{x}$. While in theory this naïve restart condition could result in an infinite loop, in our usage of this procedure we have rarely encountered a need for a restart, and more than one restart has never been encountered under proper operating conditions.

The result of this procedure is a large tuning range around a user-defined center wavelength, throughout which the laser operates in a single-mode manner. This development has directly enabled us to take automated optical absorption spectrum scans with large (>20 GHz) wavelength ranges (for example, Fig. 5.3), which would otherwise be prohibitively difficult to acquire without constant user intervention. Furthermore, this has enabled the acquisition of very fine, megahertz resolution absorption spectra of single divacancies over wavelength ranges of tens of nanometers by iteratively setting the center wavelength, running the spectral calibration procedure described in this section, and then stitching the resulting absorption spectrum sections into one combined spectrum.

4.8.2 *Magnetic field calibration*

Zero-field operation is critical for many of our defect systems, as detailed in Chapters 5 and 6. Experimentally, however, many components inside and outside of the cryostat have slight magnetic character and produce non-zero magnetic fields at the sample. We correct for this by installing a three-axis electromagnet external to the cryostat which can apply a magnetic field in the range of 0.6 mT along each axis. We then perform a procedure to automatically and iteratively zero out the field at the sample, by using our spin system as a calibration

target.

From Chapter 5, we observed that the kh divacancy ground-state spin has a “sweet spot” at $\mathbf{B} = 0$ where spin coherence is maximized due to suppressed inhomogeneity from magnetic field fluctuations. Considering this, we execute a Hahn-echo sequence at an initially unoptimized external field and sweep the free precession time τ to estimate the Hahn-echo coherence time T_2 . We then repeatedly execute the Hahn-echo sequence at $\tau = T_2$ while performing a line search by sweeping the field along each axis of the electromagnet. We iterate this line search over the individual axes until the coherence is improved to a point where no additional improvements can be resolved. This can happen if T_2 has improved significantly and the operating τ is much less than T_2 . At this point, we return back to sweeping τ to obtain a new estimate for T_2 , and repeat the iterative line search with the Hahn-echo sequence using this new τ value. This is then repeated until a convergence criterion is achieved, namely where the measured coherence does not improve within a set margin.

This calibration technique uses the Hahn-echo sequence instead of a Ramsey free precession sequence for first-order robustness to pulse detuning. This is important as changes in the magnetic field during the line searches can induce resolvable energy shifts in the spin levels, potentially producing spurious oscillations in the field sweep and complicating the determination of the optimized field component values. By using a Hahn-echo sequence, which is insensitive to energy detunings to first order, we can unambiguously extract the coherence as a function of magnetic field, and use it as a metric for proximity to $\mathbf{B} = 0$.

4.8.3 *Laser raster scans*

For many of our experiments, we are seeking a localized single divacancy near the surface of the sample. Searching for defects near the surface has many practical advantages, including less optical aberrations encountered from the focused beam of the objective, closer proximity to lithographically patterned structures used for spin and orbital manipulation, as well as

higher purity of substrate material in the epitaxially grown top layer. Thus, we initially search for the surface sample using visible imaging while enabling a highly attenuated laser spot. By moving the piezo stages in the z -axis, we can bring the sample closer to the objective. When at the top surface, both the features on the sample surface as well as the laser spot will become clearly in focus. Lateral movement of the sample is then accomplished using the x - and y -axis piezo actuators in order to find regions of interest, e.g., near a wire, capacitive structure, or optical cavity structure.

At this point, typically the broadest search for single divacancy defects is accomplished by rastering an off-resonant excitation laser (with a wavelength at 905 or 975 nm) while collecting photon emission from the divacancy phonon sideband. High powers (>5 mW) of off-resonant laser light at the objective is typically used to ensure optical saturation of emitters. A significant downside of using off-resonant excitation is its non-selective nature of exciting particular divacancy configurations (all divacancy configurations will absorb off-resonant light at comparable rates), as well as the possibility of exciting other photoluminescing features that are not related to divacancies, such as other color centers and debris on the sample surface. Thus, careful filtering of the collected photoluminescence to within the known divacancy emission spectrum is necessary to reduce spurious background contributions. Furthermore, additional checks, such as ensuring the emitter does not photobleach, as well as examining the spectrum of the emission, are typically necessary to strengthen the evidence that the emission is from a quantum system, such as a divacancy defect.

In a sample with known stable divacancies, another approach is to apply resonant excitation (at high powers to induce power broadening) with a wavelength near the typical optical absorption energies. A raster scan with this laser may then excite the optical transitions of a few divacancies on- or near-resonance, allowing for quick identification of the location and optical transition frequency of these divacancies. This technique is typically worthwhile to pursue at elevated temperatures, where the optical resonance broadens significantly due to phonon-induced processes, or at high divacancy densities where the optical resonance inho-

mogeneity can be neglected and many emitters are expected to be on resonance with the incident laser.

4.8.4 Photoluminescence excitation (PLE) spectra

Clean PLE spectra are critical to high-fidelity operation of single divacancies. However, an outstanding challenge is to reliably determine the optical resonance frequencies in the presence of inhomogeneous shifts from strain and local electric fields. Typically, we overcome this by performing coarse PLE scans by using maximal resonant laser power ($>100\text{ }\mu\text{W}$) to ensure sufficient power broadening, so that large frequency steps can be taken while also resolving resonance features. Once a resonance feature is identified, the laser power is reduced significantly ($<1\text{ }\mu\text{W}$) and a finer scan is performed over the resonance to resolve the fine optical structure. Furthermore, once a bright, cycling optical transition is identified, a resonant laser tuned into resonance with that transition can be used in a laser raster scan to more finely optimize the position of the laser spot with respect to the defect.

The shape of the resonance takes on a Lorentzian lineshape (also known as a Cauchy distribution or Breit-Wigner distribution) in the absence of inhomogeneous broadening contributions. When considering the effects from inhomogeneous broadening, a Voigt profile, or a convolution between a Lorentzian and Gaussian distribution, is more appropriate to describe the lineshape. In the limit of very large inhomogeneous effects, the lineshape becomes Gaussian.

4.8.5 Pulsed optically detected magnetic resonance (ODMR) spectra

After localization of a defect and identifying its optical spectrum, the next steps are typically concerned with understanding the spin properties of the divacancy. In many cases, the optical resonance frequency will already provide sufficient likelihood of successfully identifying the divacancy configuration. However, the spin resonance spectrum can allow for unambiguous identification of the divacancy, as the zero-field splitting parameters of the ground-state spin

are only weakly sensitive to strain-induced inhomogeneity.

While continuous-wave microwave excitation in simultaneity with resonant optical excitation (continuous-wave ODMR) can produce spin-dependent signal, the possibility of electrically induced Landau-Zener-Stückelberg interference fringes [29] can cause changes in the optical absorption spectrum independent of spin dynamics and thus complicate the interpretation of the obtained spin spectrum. Thus, we typically opt to perform pulsed ODMR, where the microwaves applied to induce a spin rotation are temporally separated from the initialization and readout optical pulses. The spin-dependent resonant excitation of the divacancy then probes purely the spin population associated with the resonant optical transition.

In order to perform pulsed ODMR, an appropriate spin rotation angle must be chosen so that when the microwaves are on resonance with the spin transition, a π rotation is ideally performed. However, in the absence of exact information regarding the spin's coupling to an applied microwave-frequency magnetic field, an arbitrarily chosen rotation angle can provide an initial resonance feature in the ODMR spectrum. If we revisit Eq. 2.31, we can obtain the spectrum for particular rotation angles $\Theta = \Omega t$. On resonance, $\Delta = 0$, such a condition produces a full inversion of the spin, and simply finding this local extremum is sufficient to determine the resonance frequency. However, there are two cases that can occur if we apply a rotation with an angle that is not approximately π . If we under-rotate the spin, the resulting spectrum as a function of Δ still yields a local extremum at $\Delta = 0$, indicating the resonance frequency. However, the depth of the resonance may be significantly reduced, as $\sin^2 [\Theta] \ll 1$ for under-rotated pulses, and may make it difficult to obtain the true resonance frequency with high visibility. By then calibrating the π rotation using Rabi oscillations where Ω or t is swept, a proper ODMR spectrum can be subsequently obtained.

In the other case, over-rotation of the spin can result in an ODMR spectrum with local extrema that are *not* the location of the true resonance. This can be particularly confounding if there are multiple resonances in close proximity (at low applied field, for example).

Consider Eq. 2.31 in the case of $\Theta > \pi$. For example, if $\Theta = 2\pi$, then Eq. 2.31 becomes

$$P(\Delta) = \left[\frac{1}{1 + (\Delta/\Omega)^2} \right] \sin^2 \left[\frac{\sqrt{1 + (\Delta/\Omega)^2} (2\pi)}{2} \right]. \quad (4.4)$$

On resonance, $\Delta = 0$, no population transfer is expected. Furthermore, the extrema in the spectrum with respect to Δ are expected when the argument of $\sin^2$ is $(2n + 1)\pi/2$ for all integers n ,

$$\begin{aligned} \sqrt{1 + (\Delta/\Omega)^2} &= (2n + 1)/2 \\ \Delta &= \Omega \sqrt{(2n + 1)^2 / 4 - 1}. \end{aligned} \quad (4.5)$$

The first extrema are encountered at $\Delta = \pm \frac{\sqrt{5}}{2} \Omega$. Thus, simply using the local extremum as the true resonance is no longer suitable for $\Theta > \pi$. An experimentally obtained ODMR spectrum can be fit to the Rabi formula to yield the central resonance frequency, or the symmetry point of Δ can be used to estimate the central resonance frequency, at which point Rabi oscillations can be performed to further optimize Ω and/or t .

4.8.6 *Spin Rabi oscillations*

After the identification of a resonance frequency, Rabi oscillations can be performed by varying the parameter Ω or t in Eq. 2.31 holding $\Delta = 0$. In an experiment, varying the parameter Ω typically involves adjusting the modulation amplitude of a signal generator, and varying the parameter t involves adjusting the pulse duration during which a signal generator is active. As a practical consideration, varying Ω usually allows for finer resolution of Rabi oscillations, as many AWGs have excellent vertical resolution (2^{16} steps for 16 bit resolution, for example), whereas sweeping t is limited by the maximum sampling rate of the device, which for typical AWGs is around 1-2 GHz. Performing Rabi oscillations at MHz Rabi frequencies thus only affords order 1000 time steps. It should be noted that even with the

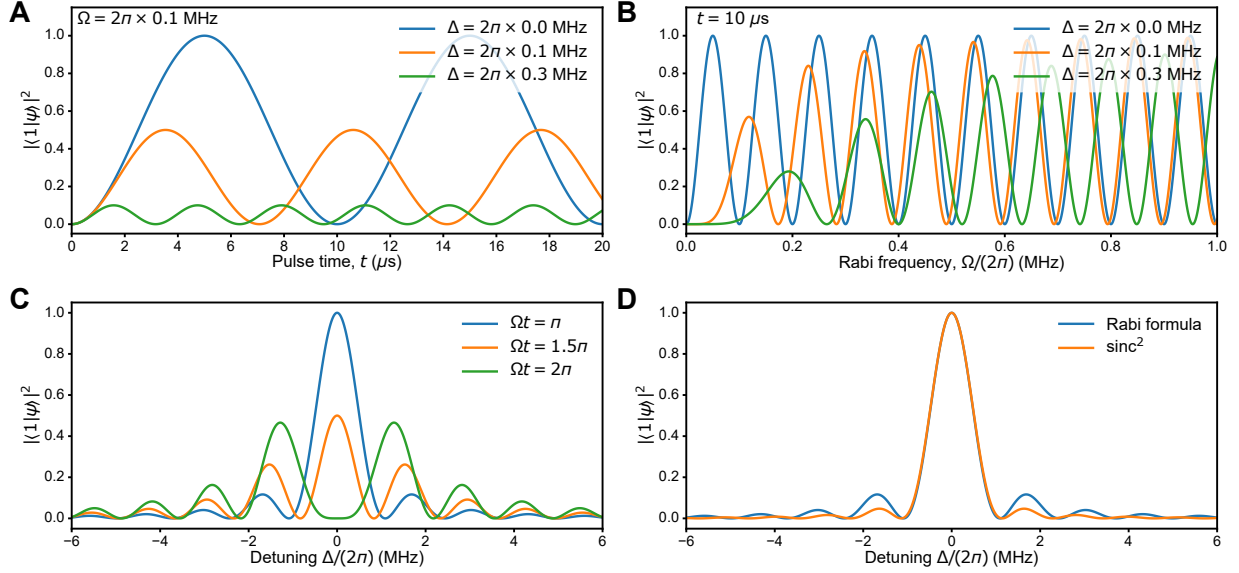


Figure 4.2: **Rabi oscillations and pulsed ODMR.** (A) Rabi oscillations as a function of pulse time for various detunings. It can be seen that the amplitude of oscillations for a given detuning stays constant. As detuning increases, the effective rate of oscillations increases. (B) Rabi oscillations as a function of applied Rabi frequency for various detunings. Here, it can be seen that the amplitude of oscillations increases with higher Rabi frequency, eventually asymptotically saturating at full amplitude. (C) For a fixed product of Rabi frequency and pulse time, a sweep of detuning Δ can yield a pulsed ODMR spectrum. If $\Omega t \neq \pi$, then the resonance frequency may not coincide with an extremum in the spectrum. (D) A non-linear least squares fit of the Rabi formula at $\Omega t = \pi$ with a sinc^2 function. The amplitudes and positions of the sidelobes do not match up and demonstrate that a proper fit of two-level system dynamics requires the Rabi formula.

practical advantages of sweeping Ω instead of t , the two parameters can generate noticeably different Rabi oscillation signals when swept under non-ideal conditions. For an ideal two-level system exactly on resonance, we can use Eq. 2.31 with $\Delta = 0$ and the effect of sweeping Ω or t are indistinguishable. But, if there are other nearby resonances with non-negligible detuning (for example, in the presence of hyperfine interactions from strongly interacting nuclear spins) or other forms of inhomogeneity which cause Δ to deviate from being ideally on resonance, then sweeps of Ω can yield non-sinusoidal behavior as the prefactor $\frac{\Omega^2}{\Omega^2 + \Delta^2}$ non-linearly asymptotes to 1 as Ω exceeds Δ . Meanwhile, sweeps of t are more robust to this concern, as the prefactor is not dependent on pulse time. To avoid the potential non-sinusoidal behavior of sweeping Ω , Ω can be increased so that $\Omega \gg \Delta$ for all sweep points. t

should be reduced proportionally so that the Rabi oscillation period does not change. This is the concept behind applying a “hard” pulse, where the pulse addresses all spin resonances almost equally within a large range of Δ . In contrast, selective pulses, or “soft” pulses, operate in the regime where Ω is smaller than any perturbative inhomogeneity, allowing for fluctuations in Δ to be resolved.

4.8.7 *Ramsey free precession*

Next, with the spin resonance frequency determined and rotation angles calibrated, a measurement of the spin’s inhomogeneous dephasing time can be made using a Ramsey free precession sequence. Recall the procedure outlined in Section 2.6.2. Now, let the basis states be two of the spin-1 levels, such as $|0\rangle$ and $|+1\rangle$. We can prepare a superposition state using a $+\pi/2_x$ pulse with an ideal detuning of zero. Without noise, at some later time t' we can use a $+\pi/2_x$ or $-\pi/2_x$ pulse along the same axis to rotate the state to either $|0\rangle$ or $|+1\rangle$. This differential measurement is known as phase cycling, and results in two separate measurements of two maximally distinguishable basis states. However, consider the stochastic phase accumulation during the free evolution time according to Eq. 2.50. For non-zero noise, the phase trajectory of the spin evolution will become increasingly broad over time, leading to potential phase shifts of the prepared superposition. If a phase of $\pi/2_z$ is accumulated, then the aforementioned phase cycled pulse sequence will yield no distinguishability between the final measured states. More generally, averaging over a distribution of phases will also yield no distinguishability between the averaged measured states, preventing useful computation to be carried out using the coherence of the qubit.

4.8.8 *Hahn-echo sequence*

In Section 2.6.3, we demonstrated the ability to refocus inhomogeneous dephasing effects by implementing a spin rotation halfway through the spin precession. This can be accomplished by applying a π pulse along the x - or y -axis. The Hahn-echo sequence can also refocus

intentional detuning of the system, as any spin precession caused by intentional detuning should also be cancelled in the second half of the sequence. This can be used as a robust method of maintaining spin superpositions during sweeps of parameters that slightly shift the energy difference between the superposition levels. Also, the Hahn-echo sequence is particularly sensitive to noise with time-dependent components with a period of τ , as the two halves of the sequence will both accumulate phase of opposite sign, which becomes additive after the application of the refocusing π pulse. This is exploited in ac sensing schemes, where the value of τ is swept to obtain spectral information of $\delta\varepsilon_0$. The characteristic time for the measured coherence to decay by a factor of $1/e$ is denoted the Hahn-echo T_2 coherence time. Given the spin's reduced sensitivity to slow noise fluctuations (which appear as quasi-static contributions in $\delta\varepsilon_0$), T_2 is generally always longer than T_2^* .

4.8.9 Inversion recovery sequence

A measurement of T_1 requires an inversion of the spin population and then waiting to measure how much that spin population has recovered a thermal distribution, hence the sequence is named an “inversion recovery” sequence. After initialing the spin to $|0\rangle$, a π pulse is used to invert the population to either $|+1\rangle$ or $|-1\rangle$. Then, simply waiting for a duration τ and measuring the population in $|0\rangle$ will allow for an evaluation of T_1 , which can be obtained by sweeping τ and fitting to

$$P_{|0\rangle} = 1 - \exp(-\tau/T_1). \quad (4.6)$$

At cryogenic temperatures, T_1 of the spin can exceed seconds. Thus, it is of utmost importance that no spurious processes can induce a shortening of T_1 , such as laser leakage that would unintentionally initialize the spin to $|0\rangle$, or read out the spin and produce a mixed state. Furthermore, improper microwave power extinction can also shorten the measured T_1 . In many applications, simply obtaining a lower bound for T_1 and determining that it is not the limiting timescale when compared to any other relevant computational timescale (T_2 or

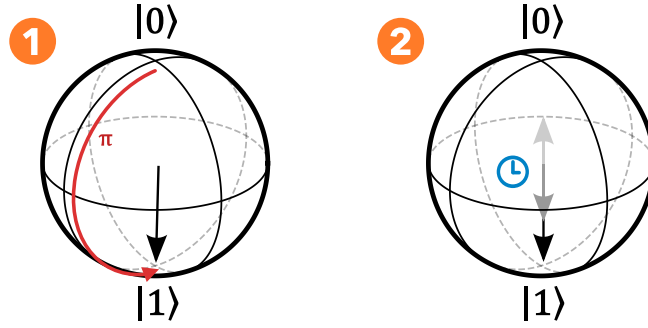


Figure 4.3: **Inversion recovery sequence.** The spin is first initialized and prepared (1) into any other spin sublevel other than $|0\rangle$. By waiting a variable amount of time, the degree of T_1 -induced relaxation can be measured (2) by obtaining the spin projection along the z -axis.

T_2^*) is sufficient to perform useful quantum information processing.

4.8.10 Optical Rabi oscillations

The two levels comprising an optical transition behave quantum mechanically and can be treated as a two-level system. The foundations developed earlier then apply equally, namely the RWA can be applied and coherent control can be accomplished with controlled application of resonant electromagnetic excitation. In the optical domain, transition rates can be higher given the focused nature of optical excitation, higher sensitivity to electromagnetic fields of excited-state orbitals, and significantly larger energy scales, allowing for faster manipulation without exceeding the validity of the RWA.

Unlike measuring Rabi oscillations of the spin, where we perform stroboscopic snapshots of the spin evolution by successively varying the amplitude or length of the applied rotation pulse in between initialization and readout optical pulses, Rabi oscillations in the optical domain can be measured continuously. When the optical two-level system is in the ground state $|g\rangle$, emission of a photon is forbidden, and we can consider that state to be “dark”. Upon absorbing a photon from an applied laser source, the system can be excited to the first excited state $|e\rangle$. Once in $|e\rangle$, interactions with phonons can induce phonon sideband emission, which are then detected with proper filters and detectors. Thus, $|e\rangle$ can be con-

sidered “bright”. As the two-level system interacts with the applied laser pulse, coherent evolution between $|g\rangle$ and $|e\rangle$ is expected, following the RWA treatment. Thus, by observing the emitted photons from the system during the laser pulse, histogramming the arrival times of those photons relative to the start of the pulse, and repeating this experiment to obtain sufficient statistics, the dynamics of optical coherence control can be revealed.

We accomplish this by first initializing the spin to $|g\rangle = |0\rangle$ using an off-resonant laser pulse. Subsequently, a short laser pulse (order 100 ns) is applied resonantly with the transition $|g\rangle \leftrightarrow |e\rangle$. Care is taken to ensure the optical Rabi frequency Ω is sufficiently high relative to decay rates T_1 and T_2 in order to observe oscillatory dynamics. Any emitted photons from the defect that are detected are time-tagged relative to the start time of the resonant laser pulse. After the end of the resonant laser pulse, an additional duration of detecting photons can allow for the observation of the radiative decay rate T_1 . Since very few photons are expected to be emitted during this 100 ns duration, and even less are expected to be collected and detected, this procedure is repeated many times in order to build up accurate statistics for precise extraction of optical parameters.

Pure dephasing (T_2) and radiative decay (T_1) will manifest as damping of the Rabi oscillations, as well as the decay of excited-state population after the end of the resonant laser pulse. Furthermore, spin mixing and relaxation under optical illumination can be extracted, which can quantify the cyclicity of the optical transition under excitation as well as spin mixing and other non-radiative processes in the excited state. These parameters can be extracted through a phenomenological Lindblad master equation model, which is further detailed in Chapter 5.

CHAPTER 5

ELECTRICALLY DRIVEN OPTICAL INTERFEROMETRY

5.1 Introduction

Constructing interfaces between defect-based quantum systems and external control mechanisms is a central focus of solid-state quantum information. Solid-state spins have risen to prominence as qubits with atom-like properties combined with the fabrication advantages offered by the solid-state host. In particular, the neutral divacancy defect in SiC has attracted considerable attention for its near-infrared optical addressability [11], highly coherent spin [67], and adaptability stemming from SiC polytypism [44]. Multiple methods of quantum control [11, 27, 28, 68] have been demonstrated using the divacancy electron spin, while the SiC host material can be doped [30] and fabricated into nanostructures [47–49]. Furthermore, c axis-oriented divacancies have a high-fidelity spin-to-photon interface [45] and exhibit large dc Stark shifts under applied electric fields [30, 58] and strain [54].

While the ground state of solid-state electron spins can be coherently controlled with acoustic [28, 69–71] and electric [27] fields, the excited-state orbitals of these electrons can exhibit substantially larger acoustic [54, 72, 73] and electric [30, 58, 63, 64] couplings, leading to enhanced interactions. In this section, we use the ground- and excited-state electric orbital levels of single, basally oriented kh divacancies in 4H-SiC to implement an optical two-level system (TLS). We demonstrate that this TLS, which has near-lifetime-limited coherence, can couple to electric fields, inducing a controllable σ_z (Pauli-Z) interaction in its Hamiltonian. This enables us to use ac electric fields to modulate the intensity and frequency profile of the kh divacancy resonant absorption spectrum between the levels of the TLS, inducing Landau-Zener-Stückelberg (LZS) interference fringes. Notably, the electric field coupling strength to the excited-state orbital is large enough that we observe classically driven coherent interactions with up to 15 microwave (MW) photons even in the absence of a MW resonator. We show that the coupling can be driven at a wide range of frequencies by

demonstrating the TLS's complex spectral response under two simultaneous electric field drives at distinct frequencies. Last, we show that alongside the coherent optical interface, the kh divacancy's C_{1h} symmetry leads to a ground-state spin with long dephasing times and magnetically drivable spin rotations that span the eigenbasis. These results demonstrate extensive optical and spin control of kh divacancies and pave the way for advanced quantum optics applications demanding high coherence across both photonic and spin subsystems.

5.2 Results

We first outline the kh divacancy's physical configuration and associated orbital and spin energy levels. The kh divacancy defect in 4H-SiC consists of a carbon vacancy V_C adjacent to a silicon vacancy V_{Si} in the basally oriented kh configuration, where k and h refer to quasi-cubic and hexagonal local environments of the V_C and V_{Si} sites, respectively. Dangling bonds from the six neighboring atoms form a localized C_{1h} symmetry system, with a spin-1 orbital singlet ground state and orbital doublet excited state in the SiC electronic bandgap (Fig. 5.1A,B). The resulting defect quantization axis is tilted at an angle of 71° with respect to the crystal c axis. At 5 K, where divacancy photoluminescence is typically the strongest, we take scanning confocal images of a 4H-SiC sample using 905-nm excitation (see Section A.1), revealing isolated emitters in the 10- μm wide gap of an on-chip capacitor (Fig. 5.1C). The emission spectrum of these isolated emitters shows about 10% of the emission in a zero-phonon line near 1079 nm, along with a red-shifted phonon sideband extending out to 1200 nm (Fig. 5.1A, inset), confirming the optical signature as kh divacancies [11, 44].

The C_{1h} symmetry manifests in the kh divacancy ground-state spin sublevels as axial and transverse zero-field splittings D_{GS} and E_{GS} , producing spin eigenstates

$$\left\{ |+\rangle = \frac{|+1\rangle + |-1\rangle}{\sqrt{2}}, |0\rangle, |-\rangle = \frac{|+1\rangle - |-1\rangle}{\sqrt{2}} \right\} \quad (5.1)$$

as represented in the $S_z = \{|+1\rangle, |0\rangle, |-1\rangle\}$ basis at low magnetic field (see Section A.2).

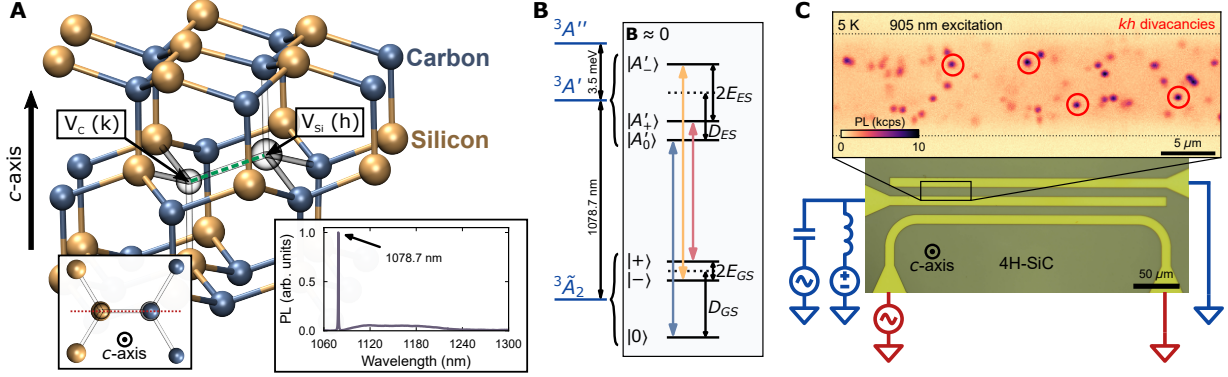


Figure 5.1: **Single kh divacancies in commercially available 4H-SiC.** (A) Lattice configuration of kh divacancies in 4H-SiC. The defect axis is indicated by the green dashed line. Inset (left): Local atomic configuration around the kh divacancy showing C_{1h} symmetry. Inset (right): Emission spectrum of a single kh divacancy. (B) Energy diagram of the kh divacancy. The spin sublevels mix due to the effect of a transverse zero-field splitting $E_{GS/ES}$, causing $|\pm 1\rangle$ to become $|\pm\rangle$ near zero external magnetic field. Spin-selective optical transitions (blue, yellow, and red arrows) enable spin-state readout. (C) Optical image of the 4H-SiC sample with a lithographically patterned capacitor and wire. Inset: Scanning confocal image of the marked region between the coplanar capacitor plates using 905-nm excitation. Highlighted emitters are single kh divacancies.

We label this reduced symmetry spin triplet as ${}^3\tilde{A}_2$. Zero first-order Zeeman (ZEFOZ) transitions emerge (see Section A.2), suppressing magnetic field noise. In addition, magnetically driven spin transitions arise between $|0\rangle \leftrightarrow |\pm\rangle$ and $|+\rangle \leftrightarrow |-\rangle$, which we describe in detail later. In the excited state the C_{1h} symmetry presents itself as a transverse crystal field, producing two widely separated ${}^3A'$ and ${}^3A''$ orbitals (Fig. 5.1B). We do not observe emission from ${}^3A''$ at 5 K, suggesting the presence of internal conversion causing fast relaxation from ${}^3A''$ to ${}^3A'$. Each orbital has three spin sublevels, and the fine structure of each orbital is also subject to longitudinal and transverse zero-field splittings D_{ES} and E_{ES} , which produce spin mixing similar to that in the ground state. Thus, we label the spin sublevels of ${}^3A'$ as $|A'_0\rangle$, $|A'_+\rangle$, $|A'_-\rangle$, analogous to $|0\rangle$, $|+\rangle$, $|-\rangle$ of ${}^3\tilde{A}_2$, respectively.

To map the fine structure of ${}^3A'$, we perform spin-dependent photoluminescence excitation (PLE) spectroscopy (see Section A.1) on a single kh divacancy in a charge-depleted environment, in which photoinduced charge dynamics of impurity sites are suppressed and spectral diffusion is reduced. We identify a central resonance corresponding to the $|0\rangle \leftrightarrow |A'_0\rangle$

transition, as well as two weaker detuned resonances primarily corresponding to $|+\rangle \leftrightarrow |A'_+\rangle$ and $|-\rangle \leftrightarrow |A'_-\rangle$. From these resonance locations, we estimate the excited-state longitudinal and transverse excited-state zero-field splittings, D_{ES} and E_{ES} , to be +970 and -483 MHz, respectively. The notably large magnitude of E_{ES} suggests a significant change in electronic wavefunction distribution between the ground and excited state (see Section A.6). Furthermore, the spin selective nature of these transitions can be used for high-fidelity readout of the ground-state spin. While residual spectral diffusion of the optical transition frequency is observed (see Section A.7), we can sum over individual scans with the wandering compensated, allowing us to extract a 21.0(2)-MHz full width at half maximum of the Lorentzian line shape (Fig. 5.2A, inset).

We probe the excited-state dynamics of single $\hbar\hbar$ divacancies by performing time-correlated fluorescence measurements (see Section A.1). Optical excitation of $|0\rangle \leftrightarrow |A'_0\rangle$ and $|+\rangle \leftrightarrow |A'_+\rangle$ reveals coherent control of both transitions (Fig. 5.2B). A fit of the data to an integrated Bloch equation model reveals longitudinal and transverse relaxation times, as well as the spin relaxation rate under illumination (T_1 , T_2 , and Γ , respectively) (see Section A.4). On the time scale of the experiment, minimal spin relaxation under illumination for $|0\rangle \leftrightarrow |A'_0\rangle$ indicates that it may be cycling, while $|+\rangle \leftrightarrow |A'_+\rangle$ is noncycling and can be used for efficient spin pumping. For both transitions, the optical coherence T_2 approaches the lifetime limit of $2T_1$. We ascribe this to an exponential suppression of orbital mixing, given the large crystal field splitting, since the energy difference between the orbitals is expected to be larger than $k_B T$ (where k_B is the Boltzmann constant). The near-lifetime-limited coherence of both optical transitions suggests that the spectral linewidth obtained through PLE spectroscopy may still be broadened by optical driving [65]. Furthermore, the fidelity of coherent phenomena, such as interferometry and interaction gates in hybrid systems, benefits from long coherence times. In particular, the profile of the LZS interference spectrum depends on optical coherence time, which we show in detail later.

With the optical properties characterized, we can isolate a TLS in the energy structure

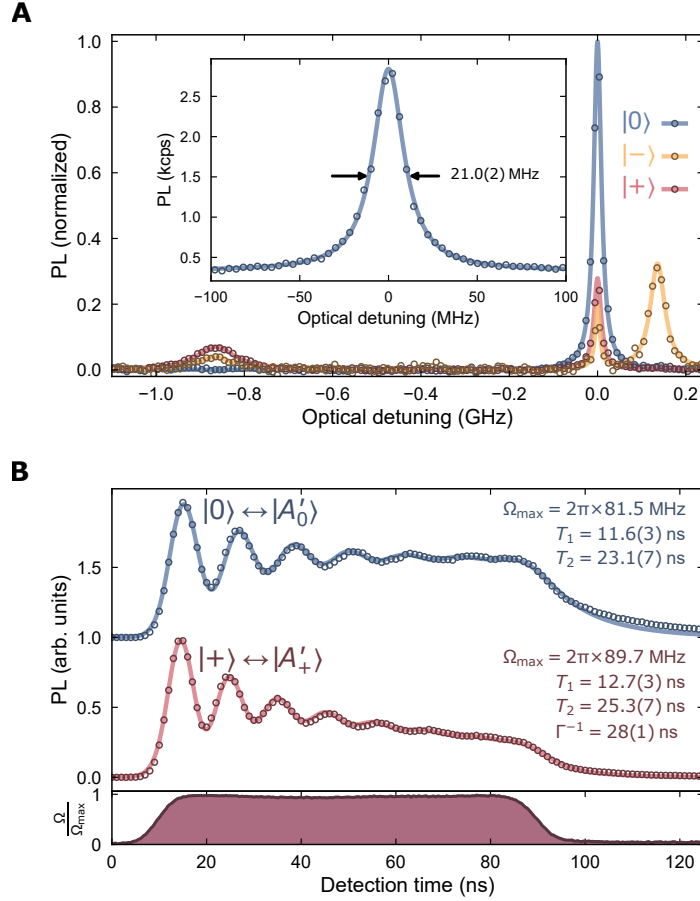


Figure 5.2: **Optical properties of single kh divacancies.** (A) PLE spectrum of a single kh divacancy prepared into $|0\rangle$ (blue), $|-\rangle$ (yellow), and $|+\rangle$ (red) with dc electric field (1 MV/m) applied and spectral diffusion compensated. Optical detuning measured with respect to 277.9337 THz (1078.647 nm). Inset: $|0\rangle \leftrightarrow |A'_0\rangle$ transition exhibiting a narrow, Lorentzian lineshape with spectral diffusion compensated. (B) Optical coherence of kh divacancies. Optical Rabi oscillations between $|0\rangle \leftrightarrow |A'_0\rangle$ (blue circles) and $|+\rangle \leftrightarrow |A'_+\rangle$ (red circles) at 7.6- μ W resonant excitation. Both transitions exhibit near-lifetime-limited optical coherence ($T_2 \approx 2T_1$). $|0\rangle \leftrightarrow |A'_0\rangle$ exhibits no detectable spin relaxation under illumination in this time scale, whereas excitation of $|+\rangle \leftrightarrow |A'_+\rangle$ rapidly depopulates $|+\rangle$. Bottom: The pulse envelope created by the acousto-optic modulator used to gate the resonant, narrow-line laser.

and demonstrate LZS interference under an ac electric field drive. For our TLS, we use $|0\rangle \leftrightarrow |A'_0\rangle$ for its brightness and all-optical preparation sequence. LZS interference arises when the TLS is repeatedly brought through an avoided crossing non-adiabatically, while a Stückelberg phase is acquired between each crossing [74]. Optical driving of the TLS produces an avoided crossing, while the Stark effect allows for controlled passage through that

crossing. According to the Landau-Zener formula, the diabatic transition probability through an avoided crossing of width Δ at a Landau-Zener velocity v is given by $P_{\text{LZ}} = e^{-\frac{2\pi\Delta}{4v}}$. We show that in our system, $v \gg \Delta$ is attainable, allowing us to operate in the non-adiabatic regime. We couple highly confined electric fields between the on-chip planar capacitor plates (Fig. 5.1C) to single kh divacancies, yielding dc Stark shifts of the divacancy's excited-state orbital levels that are resolvable through PLE spectroscopy (see Section A.8). We then apply a gigahertz-frequency ac electric field drive with zero dc bias concurrently with the resonant excitation. We monitor the PLE spectrum as we vary the amplitude $|\mathbf{F}|$ of a sinusoidal electric field drive with frequency $\omega = 2\pi \times 700$ MHz. At zero amplitude, only the bare resonance of $|0\rangle \leftrightarrow |A'_0\rangle$ is observed at zero laser detuning. With increasing amplitude, we produce multiphoton resonances at laser detunings equal to integer multiples of the drive frequency, culminating in classical interactions with up to 15 MW photons when the laser is detuned by $\delta = 15 \times 2\pi \times 700$ MHz (Fig. 5.3A).

With the proper rotating frame transformations (see Section A.3), our observations can be explained by the Hamiltonian of the TLS in the rotating frame:

$$\frac{H''(t)}{\hbar} = \sum_{n=-\infty}^{\infty} \frac{\Delta_n}{2} \left(e^{-in\omega t} \sigma_+ + e^{in\omega t} \sigma_- \right) + \frac{\delta}{2} \sigma_z, \quad (5.2)$$

with

$$\Delta_n = \Omega J_n \left(\frac{\mathcal{A}}{\omega} \right) \quad (5.3)$$

$$\sigma_{\pm} = \frac{1}{2} (\sigma_x \pm i\sigma_y), \quad (5.4)$$

where δ is the optical detuning, Ω is the optical Rabi frequency, $J_n(x)$ are Bessel functions of the first kind, $\mathcal{A}$ is the amplitude of the induced Stark shift from the ac electric field, and ω is the ac electric field frequency. This Hamiltonian leads to LZS interference and multiphoton interactions where the bare optical resonance absorbs or emits quanta of energy resonant

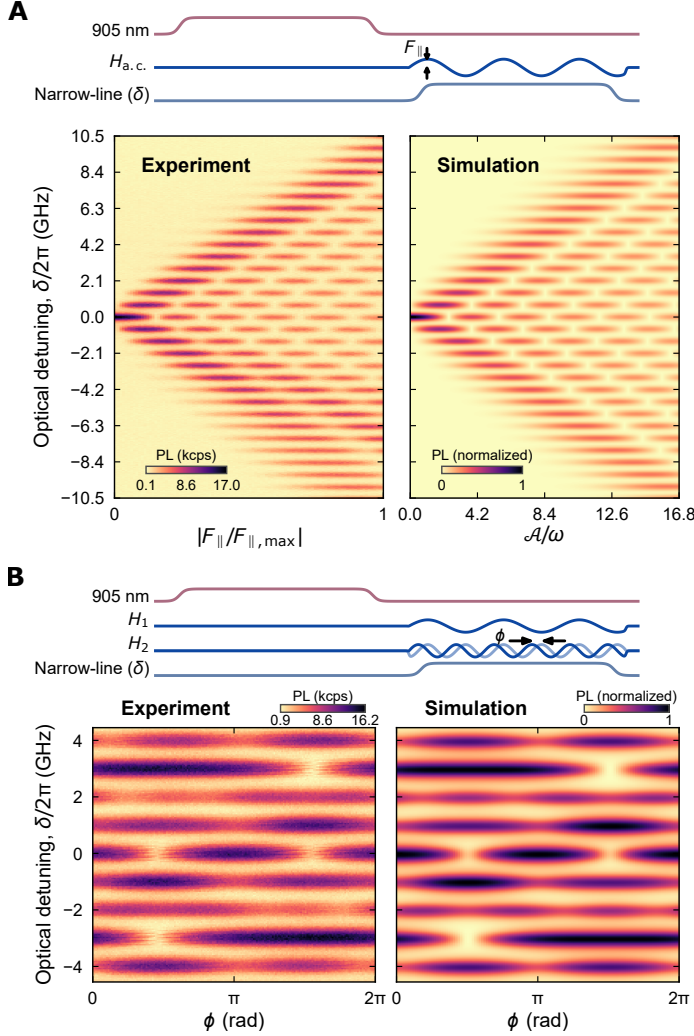


Figure 5.3: **LZS interferometry of kh divacancy absorption spectrum.** (A) Monochromatic LZS interferometry of kh divacancy absorption spectrum. Top: Pulse sequence used to observe LZS interferometry. The interference pattern of $|0\rangle \leftrightarrow |A'_0\rangle$ is measured as a function of $\mathcal{A}$, the induced Stark shift amplitude. Bottom: Interference fringes of $|0\rangle \leftrightarrow |A'_0\rangle$ absorption arise in PLE spectroscopy as electric field magnitude $|\mathbf{F}|$ is increased ($|F_{\parallel, \max}| = 2$ MV/m), proportionally increasing $\mathcal{A}$. Total acquisition time was 19.5 hours. (B) Bichromatic LZS interferometry of kh divacancy absorption spectrum. Top: Pulse sequence used to observe bichromatic LZS interferometry. The interference pattern of $|0\rangle \leftrightarrow |A'_0\rangle$ is measured as a function of the relative phase ϕ of the two drives. Bottom: PLE of a single kh divacancy under two electric field drives ($\omega_1 = 2\pi \times 1$ GHz, $\omega_2 = 2\pi \times 2$ GHz, $\mathcal{A}_1/\omega_1 = \mathcal{A}_2/\omega_2 \approx 2.4048$) as a function of ϕ . Multiphoton resonances arise at $1 \times n$ GHz and $2 \times n$ GHz optical detunings, resulting in fringes from constructive and destructive interference of the two drives. Total acquisition time was 9.1 hours.

with the drive. In our system, Ω approaches $2\pi \times 200$ MHz, while $\mathcal{A}$ is swept up to $2\pi \times 11.76$ GHz. Since $v = \mathcal{A}\omega$ at the avoided crossing, we are in the non-adiabatic regime for nearly all values of $\mathcal{A}$. Integrating the photoluminescence within the carrier resonance shows an approximate $|J_n(\frac{\mathcal{A}}{\omega})|$ dependence, which is expected in the strong optical driving regime, $T_1 T_2 \Omega^2 \gg 4\pi^2$ (see Section A.9). Together, these results demonstrate coherent control of a single kh divacancy's orbital levels with MW frequency electric fields, which can enable a

high-fidelity MW-to-optical interface when driven with single quanta of excitation.

We then turn our attention to bichromatic ac electric field driving enabled by the nonresonant capacitive structure. We apply drives $H_1 = \mathcal{A}_1 \cos(\omega_1 t) \sigma_z$ and $H_2 = \mathcal{A}_2 \cos(\omega_2 t) \sigma_z$, with amplitudes of both drives tuned to the first Bessel zero of the carrier resonance, $\mathcal{A}_1/\omega_1 = \mathcal{A}_2/\omega_2 \approx 2.4048$. We select two arbitrary, commensurable drive frequencies, $\omega_1 = 2\pi \times 1$ GHz and $\omega_2 = 2\pi \times 2$ GHz, both of which couple to the kh divacancy with similar strengths given the broadband nature of the capacitor (see Section A.10). The steady state of the TLS can then be described by higher-dimensional generalized Bessel functions [75]. Specifically, the relative phase of the two drives can be tuned to break time reversal symmetry [76] and produce phase-dependent coherence destruction of tunneling. PLE spectroscopy taken while varying the relative phase ϕ reveals a phase-dependent structure well-matched to simulation (Fig. 5.3B), indicating coherence coupling to electric fields at two frequencies spanning an octave. These results suggest that temporal topological systems [77] can be simulated using polychromatic LZS phenomena in an electrically driven kh divacancy.

Last, we demonstrate the existence of a particularly versatile and coherence ground-state spin system in single kh divacancies. At zero effective magnetic field (see Section A.2), $\mathbf{B}_{\text{eff}} = 0$, the mixed spin basis and resulting vanishing first derivative of the energy dispersion relation enable magnetically driven transitions between all three spin states and long spin dephasing times (Fig. 5.4A). We first show that the electron can spin its eigenbasis by magnetically driving the $|0\rangle \leftrightarrow |+\rangle$ and $|+\rangle \leftrightarrow |-\rangle$ transitions at $D_{\text{GS}} + E_{\text{GS}} = 1.352373$ GHz and $2E_{\text{GS}} = 36.839$ MHz, respectively (see Section A.11). Using resonant spin-selective readout, we observe Rabi oscillations marked by high photoluminescence contrast (Fig. 5.4B,C). We then perform Ramsey interferometry on single kh divacancies prepared into $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |+\rangle)$ at the ZEFOZ point, revealing a ground-state spin dephasing time of $T_2^* = 74$ μs (Fig. 5.4D). Compared to c axis divacancies, which have ground-state spin dephasing time around 2 μs [57], the lengthened spin dephasing time can be attributed to the suppression of first-order magnetic field sensitivity of the ZEFOZ point. Residual second-

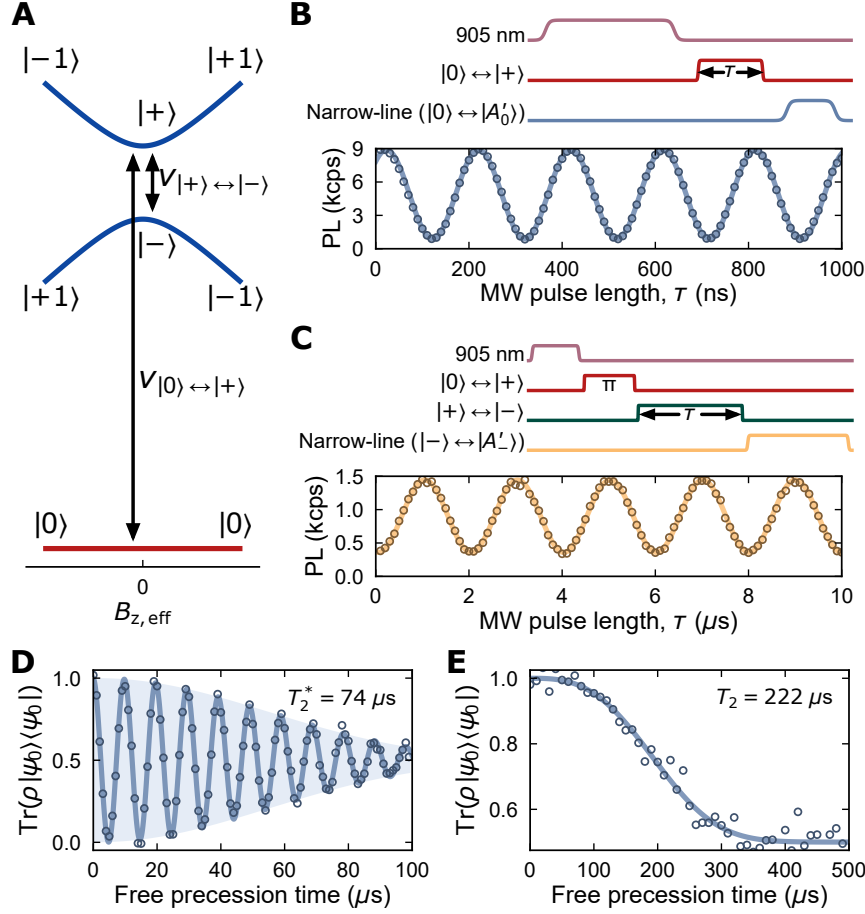


Figure 5.4: **Near-ZEFOZ spin control and dynamics of single kh divacancies.** (A) ZEFOZ transitions near zero effective magnetic field. Energy dispersion with respect to B_z shows the vanishing first derivative of the spin transition energies, $\nu_{|0\rangle \leftrightarrow |\pm\rangle}$ and $\nu_{|+\rangle \leftrightarrow |-\rangle}$, at $B_{z,\text{eff}} = 0$. (B) Top: Pulse sequence used to observe Rabi oscillations between $|0\rangle$ and $|+\rangle$. Bottom: Rabi oscillations of the ground-state spin between $|0\rangle$ and $|+\rangle$. PL measured from $|0\rangle \leftrightarrow |A'_0\rangle$ excitation. (C) Top: Pulse sequence used to observe Rabi oscillations between $|+\rangle$ and $|-\rangle$. Bottom: Rabi oscillations of ground-state spin between $|+\rangle$ and $|-\rangle$. PL measured from $|-\rangle \leftrightarrow |A'_-\rangle$ excitation. The nearby $|0\rangle \leftrightarrow |A'_0\rangle$ transition increases background and reduces the contrast of Rabi oscillations. (D) Ramsey interferometry of a spin superposition prepared in $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |+\rangle)$. Dephasing mechanisms evolve the initial state $\rho(0) = |\psi_0\rangle\langle\psi_0|$ into $\rho(t)$. A MW detuning of +100 kHz is added to increase visibility of the decay envelope. Readout is performed using $|0\rangle \leftrightarrow |A'_0\rangle$ PL. (E) Hahn-echo coherence of $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |+\rangle)$. A Gaussian decay envelope suggests the dominant source of spin coherence is from the fluctuations of the ^{29}Si and ^{13}C nuclear spin bath.

order magnetic field sensitivity and first-order electric field, however, may ultimately limit T_2^* [25, 59] We use a Hahn-echo sequence to suppress higher-order magnetic field noise, al-

lowing us to observe a spin coherence time of $T_2 = 222 \text{ } \mu\text{s}$ at the ZEFOZ point (Fig. 5.4E), comparable to low-field coherence times reported in c axis divacancies [67]. A Gaussian decay envelope suggests that bath ^{29}Si and ^{13}C nuclear spins are a main source of spin decoherence and that these contributions can be reduced by higher-order dynamical decoupling sequences such as XY- n [78].

5.3 Conclusion

Novel interactions between optical and MW photons in a solid-state system can be realized by leveraging the coherent coupling between electric fields and the excited-state orbitals of the kh divacancy. A bevy of MW resonator engineering techniques can allow adaptations to various applications using electric field coupling. For example, recent advances in high kinetic inductance superconducting resonators [79] can be leveraged to enhance zero-point fluctuations of the electric field. Consequently, protocols inspired by both atomic physics and circuit quantum electrodynamics can be explored, including the possibility of qubit-assisted resonator cooling where the optical transitions are driven in the resolved-sideband regime [80] and cavity-enhanced quantum-state readout [81]. Furthermore, the relative ease of confining electric fields allows for independent interrogation of isolated, yet spatially proximal divacancies, enabling a scalable architecture for multi-qubit interactions [82]. Meanwhile, remote entanglement applications requiring high Hong-Ou-Mandel interference visibility will benefit from the lifetime-limited coherence of the optical transitions [83]. Collectively, these properties make kh divacancies a versatile and promising platform to explore spin and optical dynamics coupled to an assortment of quantum control mechanisms.

In particular, the robust and versatile spin properties of kh divacancies have already been shown to be an invaluable asset in extending spin coherence, such as the protocol presented in Chapter 6 as well as in isotopically purified silicon carbide [39]. Some of these later experiments could not be practically executed without the kh divacancy’s excellent spin-to-photon interface, which has also been mapped in this work. Moreover, as a broader paradigm

shift, this work demonstrates that lower-symmetry defects can, somewhat unintuitively, host spin systems that are more coherent than their higher-symmetry cousins. Thus, this work may start a search for low symmetry defects that could be complementary to current efforts to find high symmetry defects (e.g., those with inversion symmetry). Furthermore, the results presented here form a roadmap for operating other basally oriented defects in SiC, including the hk divacancy in 4H-SiC, which has a significantly larger magnitude of E . As shown in Chapter 6, the driven spin coherence properties of a basal divacancy can be dependent on the magnitude of E . The optical properties may be just as favorable for other basal divacancies given the large crystal-field effect in the excited state. Taking into account these advantages, basal divacancies and other lower-symmetry defects of similar form may see a rise in adoption in applications demanding robust spin and optical coherence properties.

CHAPTER 6

UNIVERSAL COHERENCE PROTECTION IN A SOLID-STATE QUBIT

6.1 Introduction

For solid-state spin defects, like the divacancy in SiC, the crystal host plays a significant role in bestowing many favorable properties upon the electron spin, including its spin coherence [57] and optical addressability [26, 45], as well as a variety of engineered interactions for quantum control [28–30, 69, 71, 73]. Since the defect electronic states typically lie deep within the host bandgap, thermal fluctuations cannot disturb the electron. Without these protective properties of the host, many coherent properties would be rapidly quenched through thermal fluctuations. As an example, electron spin resonance experiments performed on unbound electrons typically measure coherence times on the order of nanoseconds [84].

The crystal host, however, can also serve as a limiting factor for many of these coherent phenomena. In particular, naturally abundant SiC has nuclear spin species with non-zero spin, including ^{29}Si and ^{13}C , whose fluctuations generate magnetic noise. Furthermore, all SiC crystals have lattice defects unintentionally incorporated during growth. Some of these defects are paramagnetic and photoactive, and at non-negligible densities can produce large magnitudes of magnetic (in excess of 0.45 mT [32]) and electric fluctuations [29, 30]. When any of these fluctuation sources are nearby our divacancy spin systems, the resulting response of the divacancy to these fields can introduce significant inhomogeneity in the spin and orbital energy levels. For divacancy defects, as well as other defect systems in solid-state hosts, it has been widely demonstrated that magnetic noise from nuclear spin fluctuations severely limit the spin dephasing time to a few microseconds [57, 85], as well as spin Hahn-echo coherence time to about a millisecond [30, 57, 67, 85]. Furthermore, electric noise has been demonstrated to also couple to select spin configurations and can also serve as a limiting factor in spin coherence. Acoustic and strain fluctuations in the crystal can also couple to

the spin and introduce inhomogeneity.

Techniques exist to modify the impurities in the host to lessen their contributions of noise. These include static methods, such as isotopic purification, which lowers the density of fluctuating nuclear spin species, and compensatory doping during growth to suppress the formation of unfavorable defect configurations. There are also dynamic techniques such as spin bath driving and spin bath hyperpolarization, which aim to reconfigure the nuclear spin bath into a less noisy entity. Another class of techniques, pulsed dynamical decoupling, delivers a series of control pulses on the target quantum system to cancel out any spurious, inhomogeneous phase accumulation from these fluctuation sources. While all these techniques have been successfully implemented in a variety of quantum systems, there are many drawbacks associated with each of them. For example, isotopically purified host crystals typically require isotopically purified precursor compounds, which can be rare and expensive, limiting the scale at which substrates can be made available. Pulsed dynamical decoupling can limit the availability of the target quantum system to other desired interactions, such as spin rotations and coupling to other quantum systems.

In this section, we explore another technique to reduce the inhomogeneity of the spin system. Instead of engineering the defect's environment, we opt to reconfigure the spin system's energy levels in a way to make them significantly less responsive to external fluctuations. The basis for this technique lies in the creation of dressed states, which has been explored and implemented in a variety of systems. We show that the creation of these dressed states in the kh divacancy's spin system drastically alters the spin system's response to magnetic, electric, and thermal fluctuations, which account for nearly all the relevant decoherence mechanisms for a solid-state spin system. The resulting spin dephasing time, T_2^* , increases by over four orders of magnitude when compared to dephasing times measured for solid-state spin systems at high magnetic fields (typical for many experiments in order to lift the degeneracy between the spin levels and to exploit particular field configurations for nuclear spin polarization and coupling interactions). Unlike protocols that shuttle quantum information

from the volatile electron spin to a long-lived nuclear spin, these dressed electron spin states retain the electron spin’s highly controllable interfaces while gaining the nuclear spin-like resilience to external perturbations.

6.2 Results

From Chapter 5, we showed that the kh basal divacancy in the 4H polytype of SiC has reduced sensitivity to magnetic noise when operated near zero magnetic field [29]. This robustness primarily arises from the transverse zero-field splitting (ZFS) intrinsic to the spin system’s single mirror plane symmetry. When combined with the spin system’s axial ZFS, clock transitions emerge near $\mathbf{B} = 0$. By operating at this point, the spin has lengthened inhomogeneous dephasing times, among other properties (see section B.4). Here, we demonstrate the implementation of a protocol to further extend the ground-state spin inhomogeneous dephasing time, as well as the Hahn-echo coherence time, of a single kh divacancy in 4H-SiC by simply applying a continuous microwave drive resonant with two of the three ground-state spin-1 sublevels at zero field. This microwave drive induces spin-photon hybridization, producing dressed spin states [69–71, 86–89] that exhibit notably decreased inhomogeneous shifts from nearly all environmental fluctuations at the expense of increased sensitivity to slow fluctuations in the microwave dressing drive. Once these slow fluctuations are controlled for, this protocol leads to exceptionally long spin coherence times in a decoherence-protected subspace. While many dressing protocols decouple the target system from a subset of noise sources, we show that the dressed states in the kh divacancy, by virtue of the system’s large transverse and axial ZFS magnitudes, are protected from magnetic, electric, and temperature fluctuations to high order. Furthermore, there are many practical and experimental advantages of this protocol when compared to typical pulsed dynamical decoupling sequences, not limited to the following:

1. **Arbitrary on-demand manipulation of the spin:** One of the most important

aspects of this protocol that is very difficult to achieve in pulsed dynamical decoupling is the capability to arbitrarily manipulate the spin while under protection. This is typically due to the fact that pulsed dynamical decoupling makes the spin insensitive to *all* phase accumulation, regardless of source or intent. Thus, while pulsed dynamical decoupling is excellent for applications where a static state is protected for a quantum memory purposes, it is more difficult to intersperse control pulses between the dynamical decoupling pulses in a meaningful way that can arbitrarily control the spin. Creative procedures [90, 91] are typically required to recover the capability to interact with the electron spin, as well as produce desired interactions with other quantum systems.

2. **Uninterrupted coherence protection:** During pulsed dynamical decoupling sequences, periodic manipulation of the quantum state is performed using control pulses. Since these control pulses require a non-zero amount of time to execute (a Delta function would necessitate infinite power to drive the spin), combined with the fact that the system Hamiltonian evolves differently when the control pulses are active, uncorrected dephasing can occur during these pulses. Bang-bang dynamical decoupling, where the control pulses are significantly faster than any relevant dephasing process (approaching the Delta function limit), have been investigated theoretically [92, 93] and experimentally [94] as an improvement to avoid these coherence protection interruptions, but carry additional complexities that must be taken into account for successful operation.
3. **Lower peak drive powers:** In order to mitigate the problem of higher sensitivity to noise during control pulses, as mentioned in the previous point, an experimental solution is to shorten the control pulse durations and make them behave more like Delta functions. However, in order to retain the spin rotation angle, the amplitude must be scaled correspondingly so that the integral of the amplitude over the pulse duration remains constant. Since Joule heating typically scales as the square of the

amplitude, this is typically an unfavorable scaling at short pulse durations. In our protocol, a low-amplitude continuous tone is applied, ensuring that there are no sudden dissipative heating events that could induce additional fluctuations in the quantum system.

4. **Fewer non-idealities in implementing the decoupling protocol:** An ideal implementation of pulsed dynamical decoupling typically requires a pulse to rotate the quantum system by an exact angle (usually by $\theta = \pi$ or $\theta = \pi/2$). However, in experimental realizations, pulse errors accumulate and reduce the effectiveness of the dynamical decoupling protocol. Universal dynamical decoupling sequences seek to mitigate this non-ideality [78]. Furthermore, pulses are intrinsically more demanding to generate in experiments, as the rising and falling edges of rectangular pulses typically experience ringing artifacts. Thus, an experimental implementation must carefully consider these aspects to ensure near-ideal operation. A continuous tone, such as the dressing drive in our scheme, does not have a rising or falling edge, and we show that frequency and amplitude errors are either negligible in the context of the driven system, or can be corrected through active measures.
5. **Operation with fewer assumptions about the form of the environmental noise:** In many pulsed dynamical decoupling sequences, certain combinations of parameters can actually enhance the interaction and the rate of entanglement with certain subsets of environmental fluctuation sources. In particular, this is exploited in dynamical decoupling spectroscopy [39, 91, 95] and has been shown as a reliable method for executing two-qubit gates with weakly-coupled quantum systems. However, if the purpose is to operate a target quantum system in a decoherence-protected regime for an arbitrary duration, then pulsed dynamical decoupling sequences can fail to protect the target quantum system at certain times. In our system, there are no times at which these events occur, as the interaction strengths with *all* weakly-coupled quantum sys-

tems are suppressed. Thus, algorithms and applications that require a wide range of execution times during which the quantum system must remain phase coherent may benefit from this scheme.

Importantly, this dressed spin system retains its large magnetic and electric response to resonant microwave control fields, much like the undriven divacancy electron spin. This preserves the capability to rapidly perform quantum operations to coherently manipulate the long-lived quantum state and the ability to prepare arbitrary superpositions in this new basis.

We isolate the kh divacancy used in this study in naturally abundant, commercially available 4H-SiC at 5 K (see Section B.1). The ground-state spin-1 system is initialized and read out through optical pulses, while spin rotations are produced by microwave-frequency magnetic and electric fields from lithographically patterned wires and capacitors, respectively (Fig. 6.1A). We use a three-axis electromagnet for vector control of the external magnetic field, allowing us to access the zero-field regime, as well as produce controlled non-zero fields along the Cartesian axes of the spin system. At $\mathbf{B} = 0$, a magnetically allowed spin transition can be driven between the upper two spin states, $|+\rangle = \frac{1}{\sqrt{2}}(|+1_z\rangle + |-1_z\rangle)$ and $|-\rangle = \frac{1}{\sqrt{2}}(|+1_z\rangle - |-1_z\rangle)$, where $|\pm 1_z\rangle$ denote the $m_s = \pm 1$ sublevels in the S_z basis. This transition is achieved by way of the nonzero Hamiltonian matrix elements $\langle \pm | B_z S_z | \mp \rangle$. We use Ramsey interferometry of the $|+\rangle \leftrightarrow |-\rangle$ transition to determine the transverse ZFS magnitude $E/(2\pi) = 18.353164(4)$ MHz to a high degree of precision and accuracy (see Section B.5), allowing us to apply a continuous microwave-frequency magnetic drive at an angular frequency $\omega = 2E$ resonant with the energy splitting between $|+\rangle$ and $|-\rangle$ (Fig. 6.1B). Under a sufficiently strong magnetic field drive with Rabi frequency Ω , the $|\pm\rangle$ spin levels hybridize with the incident microwave drive and undergo Autler-Townes splitting (see Section B.6) to form dressed spin states $|\pm 1\rangle$ with energy levels offset from that of $|+\rangle$ by $\pm\Omega/2$. These dressed states form the basis for the decoherence-protected subspace of the qubit. We then observe these dressed states by applying a weaker microwave probe pulse and

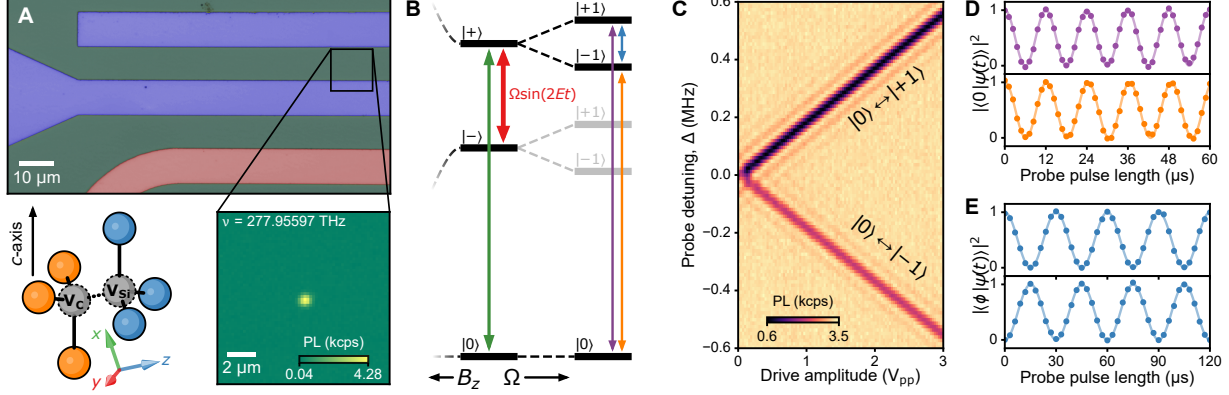


Figure 6.1: **Driven kh divacancy spin system in 4H-SiC.** (A) False-color optical microscope image of the 4H-SiC sample showing lithographically patterned capacitor (blue) and wire (red) which deliver microwave-frequency electric and magnetic fields to the region of the sample between the capacitor plates. Inset: Single kh divacancy under resonant optical excitation, tuned to the $|0\rangle \leftrightarrow |A'_0\rangle$ resonance for this divacancy. Lower left: Lattice diagram of a kh divacancy and nearest-neighbor carbon atoms (blue spheres) and silicon atoms (orange spheres). The defect system's deviation from a tetrahedral symmetry is visible. Spin-1 axes of the kh divacancy shown as green, red, and blue vectors. (B) Energy diagram of the kh divacancy ground-state spin. At $\mathbf{B} = 0$, clock transitions form between the three spin-1 levels. Spin manipulation in this basis consists of resonantly exciting two magnetically allowed transitions $|0\rangle \leftrightarrow |+\rangle$ (green arrow) and $|+\rangle \leftrightarrow |-\rangle$ (red arrow). A continuous microwave drive resonant with $|+\rangle \leftrightarrow |-\rangle$ induces Autler-Townes splitting and forms a hybridized spin-photon system. Magnetically allowed transitions between $|0\rangle$ and the dressed $|\pm 1\rangle$ spin states are coherently controlled with resonant ac magnetic fields (purple and orange arrows), while the transitions between $|+1\rangle$ and $| -1\rangle$ is controlled with resonant ac electric fields (blue arrow). The lower Autler-Townes doublet (gray) is inaccessible due to suppressed magnetic driving of $|0\rangle \leftrightarrow |-\rangle$ (refer to section B.6). (C) Pulsed optically detected magnetic resonance (ODMR) resolving Autler-Townes splitting of the kh divacancy ground-state spin under a continuous drive resonant with the $|+\rangle \leftrightarrow |-\rangle$ spin transition. Δ is the probe frequency detuning from the $|0\rangle \leftrightarrow |+\rangle$ resonance frequency. (D) Magnetically driven Rabi oscillations between $|0\rangle$ and $|+1\rangle$ (upper) and $|0\rangle$ and $| -1\rangle$ (lower) after applying a microwave pulse resonant with the transitions highlighted by the purple and orange arrows in (B). (E) Electrically driven Rabi oscillations between $|+1\rangle$ and $| -1\rangle$ after applying a microwave pulse resonant with the transition highlighted by the blue arrow in (B). To ensure the spin rotation is in the basis $\{|+1\rangle, | -1\rangle\}$, we vary a spin rotation in the pulse sequence and simultaneously perform readout of $|+1\rangle$ ($\phi = +1$) (upper) and $| -1\rangle$ ($\phi = -1$) (lower).

sweeping its frequency detuning Δ from the $|0\rangle \leftrightarrow |+\rangle$ transition frequency (Fig. 6.1C). For subsequent measurements, we operate at $\Omega/(2\pi) = 350$ kHz to mitigate higher-order energy dispersion components (see Section B.9). We realize coherent control within the dressed spin-1 system by driving $\Delta m_s = \pm 1$ transitions $|0\rangle \leftrightarrow |\pm 1\rangle$ with ac magnetic fields (Fig. 6.1D)

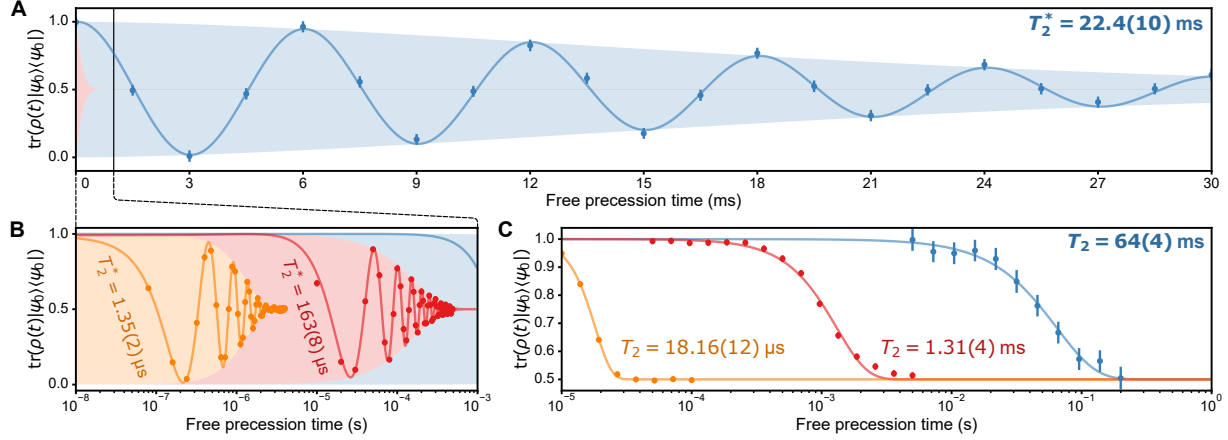


Figure 6.2: **Spin coherence in the decoherence-protected subspace.** (A) Ramsey free precession of the spin in the decoherence-protected subspace. A superposition state $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|+1\rangle + |-1\rangle)$ is prepared in the driven basis at $\mathbf{B} = 0$ mT and then allowed to undergo free precession (blue). A microwave frequency detuning of +166.6 Hz is added to increase visibility of the decay envelope. (B) Ramsey free precession of the spin when encoded outside of the decoherence-protected subspace. Superposition states $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |+\rangle)$ prepared at $\mathbf{B} = 0$ mT (red) and $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |+1_z\rangle)$ at $B_z = 1.2$ mT (orange) exhibit noticeably shortened spin dephasing times. (C) Hahn-echo free precession of the spin when prepared in the decoherence-protected subspace at $\mathbf{B} = 0$ mT (blue, $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|+1\rangle + |-1\rangle)$), and outside of the decoherence-protected subspace at $\mathbf{B} = 0$ mT (red, $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |+\rangle)$) and at $B_z = 1.2$ mT (orange, $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |+1_z\rangle)$). Error bars represent 1 SD.

and a $\Delta m_s = \pm 2$ transition $|+1\rangle \leftrightarrow |-1\rangle$ with ac electric fields [27] (Fig. 6.1E). Readout of the $|\pm 1\rangle$ basis is accomplished by non-adiabatically disabling the dressing drive, rotating into the $\{|0\rangle, |+\rangle\}$ basis, and optically probing the spin population in $|0\rangle$ (see Section B.8).

We quantify the energy inhomogeneity of this driven spin system by preparing a superposition $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|+1\rangle + |-1\rangle)$ in a decoherence-protected subspace and executing a Ramsey free precession sequence. Remarkably, with active feedback procedures in place, we measure a T_2^* spin inhomogeneous dephasing time in this basis to be $22.4(10)$ milliseconds (Fig. 6.2A), which is over four orders of magnitude longer than the value obtained for the same kh divacancy at $B_z = 1.2$ mT without the dressing drive and more than two orders of magnitude longer than the undriven inhomogeneous dephasing time at the $\mathbf{B} = 0$ clock transition (Fig. 6.2B). Moreover, by adding a single refocusing pulse, we extend the lifetime

of the superposition to a T_2 Hahn-echo spin coherence time of 64 (4) milliseconds (Fig. 6.2C). Both of these values are among the longest times measured for an optically addressable electron spin, regardless of isotopic purity of the host. These increases are a direct result of reduced energy inhomogeneity of the spin in the decoherence-protected subspace, combined with active feedback of the spin resonance frequency. We discuss these two components in detail in the following sections.

In order to understand the effect of the individual contributions from magnetic, electric, and temperature fluctuations on spin inhomogeneous dephasing, we investigate the energy dispersion relation between the dressed spin energy levels and each of the fluctuation parameters. We first consider the effect of magnetic fields on the dressed spin levels, as magnetic noise is the typical limiting decoherence channel in many electron spin systems. The dispersion relation of the divacancy ground-state spin becomes non-trivial under a continuous drive resonant with the $|+\rangle \leftrightarrow |-\rangle$ transition, and acquires both quadratic and quartic dependences on magnetic field (see Section B.9). We probe the dispersion relation by applying magnetic fields along the Cartesian x - and z -axes of the spin system while scanning the frequency detuning Δ of a probe drive (Fig. 6.3A,B). Rotational symmetry of the system about the z -axis leads to indistinguishable effects from x - and y -axis fields (see Section B.10). The resulting frequencies of the $|0\rangle \leftrightarrow |\pm 1\rangle$ spin resonances provide us with the driven spin system's spectral response to magnetic fields. To confirm these observations, we develop an analytical model of the driven spin system using Floquet analysis alongside a numerical model through spin-1 master equation simulations (see Section B.14). We find excellent agreement between the experimental data and these models (Fig. 6.3C,D). We then consider the primary source of magnetic noise, the nuclear spin bath, as a simplified isotropic magnetic fluctuator with an estimated fluctuation magnitude at this kh divacancy to be 13 μT (see Section B.11). We perform fine spectral scans of the electrically driven $|+1\rangle \leftrightarrow |-1\rangle$ transition using Ramsey interferometry (Fig. 6.3E,F) and confirm the high degree of insensitivity in this range, illustrating the primary mechanism for the largely increased spin

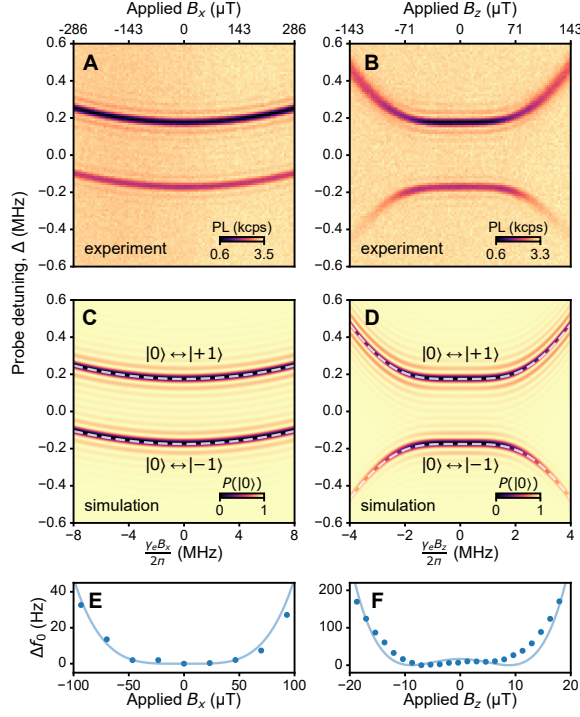


Figure 6.3: **Energy dispersion in the decoherence-protected subspace.** (A,B) Measured spin resonance spectrum of the driven $|0\rangle \leftrightarrow |\pm 1\rangle$ transitions over a range of applied x -axis (A) and z -axis (B) magnetic fields. Unequal photoluminescence contrast between the two resonance branches is a result of the contrast recovery procedure, and is not indicative of unequal transition probabilities between $|0\rangle \leftrightarrow | +1\rangle$ and $|0\rangle \leftrightarrow | -1\rangle$. Δ is the probe frequency detuning from the $|0\rangle \leftrightarrow | +\rangle$ resonance frequency. In (B), inhomogeneous broadening of the resonances is observed at high z -axis fields, where the spin is no longer fully encoded in the decoherence-protected subspace. (C,D) Simulated spin resonance spectrum of the driven $|0\rangle \leftrightarrow |\pm 1\rangle$ transitions over the same range of applied x -axis magnetic field in (A) (C), and applied z -axis magnetic field in (B) (D). Dashed white lines indicate transition energy spectra derived from Floquet analysis. (E,F) Energy difference Δf_0 between the $|+1\rangle$ and $|-1\rangle$ states as a function of applied B_x (E) and applied B_z (F), as measured using Ramsey interferometry of $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|+1\rangle + |-1\rangle)$. Solid lines are the basis state energy differences in the decoherence-protected subspace derived from Floquet analysis with no free parameters (see sections B.9 and B.10). Error bars are smaller than the symbol size.

coherence times. These results, once combined with a phenomenological model of the spin energy inhomogeneity (see Section B.12), allow us to quantitatively confirm that residual inhomogeneity from magnetic fields still plays a role in limiting the spin coherence, even after substantial suppression in the decoherence-protected subspace.

We can then apply the analytical energy dispersion relations to understand the effect of electric noise and temperature fluctuations on spin inhomogeneous dephasing in the decoherence-protected subspace. Electric and temperature fluctuations affect the axial and

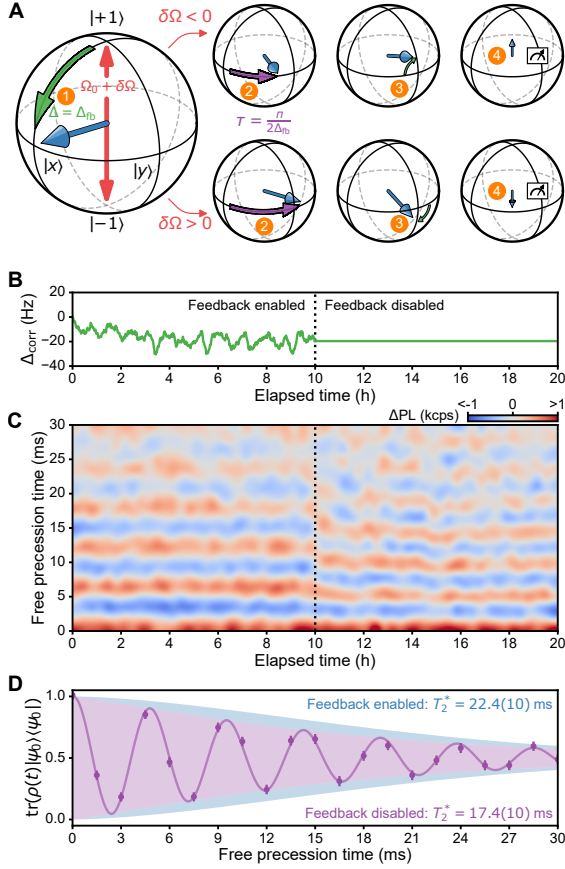


Figure 6.4: **Active spin resonance feedback.** (A) Bloch spheres illustrating the spin resonance feedback protocol. See section B.2 for step-by-step details. (B) Applied corrective detuning to the frequency of the drive resonant with $|+1\rangle \leftrightarrow |-1\rangle$. During hours 0–10, active feedback is applied to the spin and corrective detuning is derived from a proportional factor of the measured error signal. (C) Stabilization of Ramsey free precession of $|\psi_0\rangle = \frac{1}{\sqrt{2}} (|+1\rangle + |-1\rangle)$ with active spin resonance feedback. While active feedback is enabled, the observed Ramsey fringes are stable at the chosen detuning of +166.6 Hz. Once disabled, slow drifts in the dressing drive Rabi frequency shift the effective detuning of the microwave pulse used to prepare the superposition. Interpolation applied to emphasize fringe locations. (D) The Ramsey free precession decay envelope when integrating the individual Ramsey free precession iterations in (C), demonstrating the increased inhomogeneity without the feedback process. Error bars represent 1 SD.

transverse zero-field splitting magnitudes, as well as a corresponding detuning of the dressing drive from the resonance frequency $\omega = 2E$. We use the undriven spin's first-order sensitivity to electric field noise [59] at $\mathbf{B} = 0$ to quantify the magnitude of electric field noise present in the system (see Section B.13). In the decoherence-protected subspace, we gain first-order protection against fluctuations in the zero-field splitting magnitudes, resulting in increased robustness against electric field noise and temperature shifts. This leads to a reduction of electrically induced spin energy inhomogeneity by nearly two orders of magnitude (see Section B.13), and diminishes electric field contributions in limiting the spin coherence in the decoherence-protected subspace. A further reduction of electric field contributions may emerge by applying a dc electric field to deplete fluctuating charges [29, 30], possibly leading to nearly complete elimination of electric field noise in our system.

The energy dispersion relations in the driven basis confirm that the spin’s energy levels depend linearly on the dressing drive Rabi frequency. Hence, amplitude drifts of the dressing drive and corresponding fluctuations in the Rabi frequency $\delta\Omega$ introduce first-order energy inhomogeneity of the dressed spin levels and cause shortening of the measured T_2^* . To this end, we implement active feedback of the dressed spin resonance frequency in order to counteract these slow drifts and reduce inhomogeneity (Fig. 6.4A). By evaluating an error signal derived from Ramsey free precession of the dressed spin (Fig. 6.4B), we measure and correct drifts on the order of 30 Hz in the spin resonance frequency (see Sections B.2 and B.16), consistent with the 100 ppm/°C stability of the dressing drive oscillator. In order to validate the effectiveness of our feedback protocol, we measure Ramsey free precession of $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|+1\rangle + |-1\rangle)$ with and without feedback enabled (Fig. 6.4C). With active feedback enabled, we measure our reported T_2^* of 22.4(10) ms; when disabled, this value shortens to 17.4(10) ms as the dressing drive Rabi frequency inhomogeneity suppresses the spin’s coherence on the timescale of hours (Fig. 6.4D). These results suggest that by incorporating this feedback protocol, we have largely mitigated the effects of dressing drive amplitude fluctuations and that noise sources intrinsic to the host crystal remain as the limiting factor for spin coherence in the decoherence-protected subspace.

6.3 Conclusion

In this section, we presented a considerable increase in inhomogeneous dephasing time by over four orders of magnitude and a thousand-fold increase in Hahn-echo coherence time of a single kh divacancy electric spin under the effect of a single continuous microwave drive tone. Our implementation’s success stems from higher-order protection from magnetic, electric, and temperature fluctuations. This arises from driving a spin system with intrinsic protection from magnetic noise, compared to one with just a linear Zeeman response. The resulting extended spin T_2^* enabled resonant coupling to weakly interacting quantum systems, paving the way to hybrid quantum systems using single divacancy ground-state spins.

Even with modest coupling rates to the spin, unity-cooperativity systems can be realized. Furthermore, the viability of this protocol at zero magnetic field is a substantial advantage, as superconducting systems with low critical fields can be integrated with the spin system. We have shown that, while insensitive to non-resonant magnetic and electric fluctuations, the electron spin is still highly responsive to resonantly applied magnetic and electric control fields. Thus prospective hybrid quantum systems involving the driven kh divacancy spin can leverage any one of magnetic, electric, and acoustic [28, 71] couplings to the spin. Lastly, megahertz-scale tuning of the dressed spin energy levels allows for rapid adjustment of interaction strengths with other quantum systems, enabling the spin to be an efficient quantum bus.

The few requirements for successful implementation of this protocol allow for immediate extension to other quantum systems with similar undriven energy structures. Namely, a non-exhaustive list of these requirements include

1. an n -level quantum system, where $n \geq 3$;
2. an avoided crossing to form between two of those levels, X and Y ;
3. a continuous drive tone that addresses the transitions between X and Y , forming Autler-Townes-split states X' and Y' ;
4. a probe tone that can transfer population from a third level, Z , into X' and/or Y' ;
5. an allowed transition between X' and Y' for state manipulation within the decoherence-protected subspace.

For example, spin-1 systems with larger transverse ZFS values can expect to see improvements from higher dressing drive Rabi frequencies, as well as greater suppression of magnetic field fluctuations in the decoherence-protected subspace. Candidate systems include the basally oriented hk divacancy in 4H-SiC ($E/(2\pi) = 82.0$ MHz [44]) as well as basally oriented nitrogen-vacancy centers in 4H-SiC ($E/(2\pi) = 103$ MHz [20]) and 6H-SiC ($E/(2\pi) = 138$

MHz [18]). Strained nitrogen-vacancy centers in diamond with an appreciable transverse ZFS may also stand to benefit from this protocol [40]. Turning to other systems, donor spins in silicon with robust clock transitions [96, 97], transmon qubits operated at a degeneracy point [98], and molecular spins [99, 100] maybe driven to produce enhanced coherence.

Furthermore, our protocol is compatible with additional forms of noise suppression, including isotopic purification, charge depletion, and pulsed dynamical decoupling. We have shown that our measured spin inhomogeneous dephasing time is consistent with limitations from residual inhomogeneous broadening from nuclear spin bath and charge fluctuations. By reducing the volume density of fluctuating spins in the bath and thus reducing the average dipolar interaction strength between the electron spin and bath nuclear spins [37–39, 101], we expect the magnitude of inhomogeneous electron spin energy shifts to decrease proportionally. Dynamic techniques such as spin bath driving [85] and spin bath hyperpolarization [102] may further reduce the noise contributions from nuclear spin fluctuations. Charge depletion, which has led to the observation of near-transform-limited optical linewidths in divacancies [29, 30], can further suppress electric field contributions. In this section, we demonstrated compatibility with pulsed dynamical decoupling using a Hahn-echo sequence; higher-order dynamical decoupling sequences may allow for spin coherence to rapidly reach the spin-lattice relaxation limit with relatively few control pulses.

Considering these prospective avenues, this dressing protocol represents a major development for potentially improving coherence across a variety of quantum systems, and is a crucial step towards integrating SiC divacancies into robust, real-world quantum technologies. Prior to this work, a significant challenge in interfacing divacancies with other systems (including other divacancies) was the spin’s relatively short T_2^* when compared to the dipole coupling strengths to cavity structures and other quantum systems. This limited many spin coupling works to ensemble couplings [40, 41, 103], which gain in a factor of $\sqrt{N}$, where N is the number of spin systems in the ensemble. From these works, single spin coupling rates are of order $g = 10$ Hz, indicating that single spin coupling to a cavity with a sufficiently

long lifetime can be feasible in the driven basis where T_2^* exceeds tens of milliseconds.

The driven basis offers significantly increased single qubit gate fidelities given the heavily suppressed inhomogeneous dephasing while maintaining the same gyromagnetic ratio. As an estimate of π -pulse fidelities, we can use the expression

$$F_\pi = \exp \left[-\frac{1}{2f\Gamma} \right], \quad (6.1)$$

where F_π is the fidelity of a single π pulse, f is the frequency of driving, and Γ is the decay of Rabi oscillations during drive [104, 105]. For this work, $f = 350$ kHz and Γ can be lower-bounded by T_2^* in the driven basis, which is 22.4 ms. The resulting single π pulse error rate $1 - F_\pi$ is then upper-bounded by approximately 10^{-4} , which would be among the highest single qubit gate fidelities measured in a solid-state system. It is then possible that two-qubit gates that were previously limited by electron spin inhomogeneity can stand to benefit in fidelity, paving the way to scalable qubit technologies using the divacancy ground-state spin. When implemented in other quantum architectures with intrinsically easier routes of multi-qubit coupling, this protocol could serve as the backbone for improving gate fidelities to the point where robust error correction schemes can be executed. In such a favorable scenario, this protocol could expedite the creation of a fault-tolerant quantum computation device.

We described a potential application of the extended T_2^* is the driven spin's use as an on-demand quantum memory. In this context, many remote entanglement schemes [34, 106, 107] require highly coherent stationary qubits while the flying qubits (photons) interact with each remotely. The duration of coherence for the stationary qubits can limit the distance over which the flying qubits can interact. Over a duration of 22 milliseconds, a photon can travel over 6500 kilometers, which effectively allows for traversal across continental length scales. In an actual implementation, photon losses in free space or optical fiber will be the limiting factor for realizable communication distances, and quantum repeaters will be required in

order to successfully create long-distance quantum communication networks. However, the principles presented in this work can potentially be applied to other quantum emitters with more favorable indistinguishable photon emission properties, allowing them to overcome possible shortcomings in the stationary qubit subsystem while also benefiting from improved photonic quantum interference properties.

These examples serve as only a selection in the vast array of possibilities enabled by this coherence protection scheme. Given the few key platform-independent components necessary to realize this protocol, it can be expected to see this protocol open many doors to new experimental regimes in many different quantum architectures.

APPENDIX A

ELECTRICALLY DRIVEN OPTICAL INTERFEROMETRY – DETAILS

A.1 Sample preparation and experimental setup

We created samples from a 4” commercially available wafer where 20 μm of high-purity i-type SiC ($[V_C], [V_{Si}], [N] < 1 \times 10^{15} \text{cm}^{-3}$) was epitaxially grown on a 4° off-axis miscut of the Si face of a high-purity semi-insulating SiC substrate (serial number A3177-14, Norstel AB). We diced 5 mm by 5 mm chips and performed electron irradiation with 2-MeV electrons at a dose of $3 \times 10^{12} \text{e}^-/\text{cm}^2$ to create both V_C and V_{Si} . Subsequent annealing at 850 $^\circ\text{C}$ for 30 min in Ar formed divacancy complexes uniformly throughout the entire sample. Planar capacitors with a 10- μm gap width and wires with 10- μm width made of Ti/Au were then patterned on the sample surface using electron beam lithography. Samples were cooled to 5 K in a closed-cycle cryostat (Cryostation s100, Montana Instruments).

Using a confocal microscopy setup, we performed confocal raster scans using off-resonant excitation at 905 nm (QFLD-905-200S, QPhotonics) or 975 nm (BL975-SAG300, Thorlabs) focused through a microscope objective (LCPLN100XIR, Olympus). We detected emitted photons with $>80\%$ quantum efficiency using a low-jitter, low-dark count superconducting nanowire single-photon detector (SNSPD; Opus One, Quantum Opus). Electrical pulses from the SNSPD were converted transistor-transistor logic and counted using a data acquisition module (PCI-6259, National Instruments). We applied a bandpass filter with a passband of 1064 to 1150 nm to selectively detect emission from kh divacancies while nearly completely rejecting emission from other defects and divacancy orientations. We performed PLE by scanning a narrow-line tunable laser (DL pro, TOPTICA Photonics) and collecting from the phonon sideband of the kh divacancies using a tunable longpass filter (TLP01-1116-25x36, Semrock). We initialized the spin with a pulse of off-resonant light, followed by spin manipulation using microwave and radio-frequency pulses. A pulse of resonant light per-

forms spin-dependent readout. Spin driving with on-chip planar wires was achieved using a signal generator (SG396, Stanford Research Systems) modulated by an arbitrary waveform generator (for $|0\rangle \leftrightarrow |\pm\rangle$ transition; AWG5014C, Tektronix) or an IQ modulator (for $|+\rangle \leftrightarrow |-\rangle$ transition; LTC5598, Analog Devices). LZS interferometry spectra were produced with microwave frequency electric fields applied during the resonant readout pulse. We applied microwaves generated in a similar fashion for spin driving across the on-chip capacitor, producing electric field magnitudes as high as 2 MV/m across the capacitor gap. Time-resolved photon arrival measurements were performed by time-tagging the SNSPD output pulses (Time Tagger 20, Swabian Instruments). We initialized the spin to $|0\rangle$ and performed microwave rotations to prepare $|+\rangle$, followed by a laser pulse resonant with $|0\rangle \leftrightarrow |A'_0\rangle$ or $|+\rangle \leftrightarrow |A'_+\rangle$ for 80 ns. The arrival times of emitted photons in the phonon sideband were time-tagged relative to the start of the resonant laser pulse. Approximately five megacounts were collected over 60 min and histogrammed into 1-ns bins to observe time-resolved dynamics.

A.2 Spin Hamiltonian

The Hamiltonian of the divacancy ground-state spin-1 system is

$$H = D \left(S_z^2 - \frac{S(S+1)}{3} \right) + E \left(S_+^2 + S_-^2 \right) + g\mu_B \mathbf{B} \cdot \mathbf{S} + \sum_i \mathbf{S} \cdot \mathbf{A}_i \cdot \mathbf{I}_i, \quad (\text{A.1})$$

where D and E are the ground-state axial and transeverse zero-field splitting, respectively, μ_B is the Bohr magneton, g is the electron spin g -factor, $\mathbf{B}$ is the applied magnetic field pseudovector, $\mathbf{A}_i$ is the hyperfine tensor of nuclear spin i , and $\mathbf{I}_i$ is the nuclear spin operator vector of nuclear spin i . $\hbar = 1$ is assumed for this derivation. Zeeman interactions of the nuclear spin are neglected.

For simplicity, we first consider a system with one nuclear spin. Assuming B_x and B_y

are negligible, diagonalization of this Hamiltonian yields the following eigenenergies

$$\text{eigval}(H) = \begin{cases} D + \sqrt{C_+^2 + E^2}, & |1\rangle \\ D + \sqrt{C_-^2 + E^2}, & |2\rangle \\ D - \sqrt{C_+^2 + E^2}, & |3\rangle \\ D - \sqrt{C_-^2 + E^2}, & |4\rangle \\ 0, & |5\rangle \\ 0, & |6\rangle \end{cases}, \quad (\text{A.2})$$

and corresponding (unnormalized) eigenstates with a nuclear spin basis $I_z = \{|\uparrow\rangle, |\downarrow\rangle\}$

$$\text{eigvec}(H) = \begin{cases} \frac{E}{C_+ + \sqrt{C_+^2 + E^2}} | +1 \uparrow \rangle + | -1 \uparrow \rangle, & |1\rangle \\ \frac{E}{C_- + \sqrt{C_-^2 + E^2}} | +1 \downarrow \rangle + | -1 \downarrow \rangle, & |2\rangle \\ \frac{E}{C_+ - \sqrt{C_+^2 + E^2}} | +1 \uparrow \rangle + | -1 \uparrow \rangle, & |3\rangle \\ \frac{E}{C_- - \sqrt{C_-^2 + E^2}} | +1 \downarrow \rangle + | -1 \downarrow \rangle, & |4\rangle \\ |0 \uparrow \rangle, & |5\rangle \\ |0 \downarrow \rangle, & |6\rangle \end{cases}, \quad (\text{A.3})$$

where $C_{\pm} = B_z \pm \frac{A_{zz}}{g\mu_B}$. When $C_{\pm} = 0$, the effect of B_z is to negate the hyperfine interaction for a particular nuclear spin orientation. For example, if we consider the system when the nuclear spin is $|\uparrow\rangle$ and $C_+ = 0$, then the electron spin Hamiltonian becomes equivalent to one with no nuclear spin interactions or external magnetic field, which allows us to label the effective external magnetic field as $B_{z,\text{eff}} = B_z + \frac{A_{zz}}{g\mu_B} = 0$. At $C_+ = 0$ ($C_- = 0$), the first derivative of the energy difference ν between $|1\rangle$ ($|2\rangle$) and $|4\rangle$ ($|3\rangle$) as a function of B_z is zero, $\frac{d\nu}{dB_{z,\text{eff}}} = 0$. Similar expressions can be derived for B_x and B_y , showing that $\frac{d\nu}{dB_{x,\text{eff}}} = \frac{d\nu}{dB_{y,\text{eff}}} = 0$ and that the field is $\mathbf{B}_{\text{eff}} = 0$ to fulfill all these conditions. The vanishing first derivative of the energy dispersion, by definition, produces a ZEFOZ

transition.

By operating at the ZEFOZ point, we selected one of the two nuclear spin states. The effective Hamiltonian can be expressed in the $S_z = \{|+1\rangle, |0\rangle, |-1\rangle\}$ basis

$$H_{\text{ZEFOZ}} = \begin{bmatrix} D & 0 & E \\ 0 & 0 & 0 \\ E & 0 & D \end{bmatrix}, \quad (\text{A.4})$$

which can be diagonalized

$$\begin{aligned} H'_{\text{ZEFOZ}} &= U^\dagger H_{\text{ZEFOZ}} U \\ &= \begin{bmatrix} D+E & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & D-E \end{bmatrix}, \end{aligned} \quad (\text{A.5})$$

$$U = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & 0 & 1 \\ 0 & \sqrt{2} & 0 \\ 1 & 0 & -1 \end{bmatrix}, \quad (\text{A.6})$$

where the eigenenergies of the three electron spin states are 0 and $D \pm E$, with corresponding

eigenstates $|0\rangle$ and $\frac{1}{\sqrt{2}}(|+1\rangle \pm |-1\rangle)$. Spin-1 operators in this basis can be expressed as

$$S'_x = \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad (\text{A.7})$$

$$S'_y = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{bmatrix}, \quad (\text{A.8})$$

$$S'_z = \begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix}. \quad (\text{A.9})$$

Thus, spin rotations can be achieved using the nonzero matrix elements (and their Hermitian conjugates) $|\langle 0|S'_x|+\rangle|^2$, $|\langle 0|S'_y|-\rangle|^2$, and $|\langle -|S'_z|+\rangle|^2$. Experimentally, we can realize S'_x and S'_z rotations, while S'_y rotations induce significant off-resonant S'_x rotation, given the defect orientation with respect to the planar drive wire.

A.3 Landau-Zener-Stückelberg interferometry Hamiltonian

The Hamiltonian of the unperturbed optical transition can be considered as a TLS with a ground and excited state $|g\rangle = |0\rangle$ and $|e\rangle = |A'_0\rangle$, respectively. In the $\sigma_z = |e\rangle\langle e| - |g\rangle\langle g|$ basis, this Hamiltonian can be expressed as

$$H(t) = \frac{\Omega \cos(\omega_{\text{opt}} t)}{2} \sigma_x + \frac{\omega_0}{2} \sigma_z, \quad (\text{A.10})$$

where Ω is the optical Rabi frequency, ω_{opt} is the laser frequency, and ω_0 is the optical resonance frequency. In the rotating frame of the TLS, a time-independent Hamiltonian

arises

$$\tilde{H} = \frac{\Omega}{2}\sigma_x + \frac{\delta}{2}\sigma_z, \quad (\text{A.11})$$

where $\delta = \omega_{\text{opt}} - \omega_0$ is the laser detuning from the resonance frequency. This system is analogous to the unperturbed TLS presented by Shevchenko *et al.* [108], with the substitutions $\Delta \rightarrow -\Omega$ and $\epsilon \rightarrow -\delta$.

The addition of a longitudinally coupled ac electric field drive of amplitude $\mathcal{A}$ and frequency ω corresponds to an additional time-dependent Hamiltonian

$$H_{\text{ac}}(t) = \frac{\mathcal{A} \cos(\omega t)}{2} \sigma_z, \quad (\text{A.12})$$

so that

$$\begin{aligned} \tilde{H}'(t) &= \tilde{H} + H_{\text{ac}}(t) \\ &= \frac{\Omega}{2}\sigma_x + \frac{\delta + \mathcal{A} \cos(\omega t)}{2}\sigma_z. \end{aligned} \quad (\text{A.13})$$

Applying a transformation [108] into a frame rotating with $H_{\text{ac}}(t)$ to $\tilde{H}'(t)$ yields

$$\tilde{H}''(t) = \frac{\delta}{2}\sigma_z + \sum_{n=-\infty}^{\infty} \frac{\Delta_n}{2} \left(e^{-in\omega t} \sigma_+ + e^{in\omega t} \sigma_- \right), \quad (\text{A.14})$$

where $\Delta_n = \Omega J_n \left(\frac{\mathcal{A}}{\omega} \right)$ is the effective optical Rabi frequency under ac electrical drive.

A.4 Optical Bloch equation model

We modeled the kh divacancy optical structure as a three-level system in the presence of spin relaxation under illumination. In addition to the ground and excited states $|g\rangle$ and $|e\rangle$, we also considered a trapping state $|s\rangle$ representing noninteracting ground-state spin states. Furthermore, we introduced three relaxation channels in the form of Lindblad operators C_n : radiative decay, $C_1 = \frac{1}{\sqrt{T_1}} |g\rangle\langle e|$; pure dephasing, $C_2 = \frac{1}{\sqrt{T_2^*}} (|e\rangle\langle e| - |g\rangle\langle g|)$; and spin

relaxation under illumination, $C_3 = \sqrt{\Gamma} |s\rangle\langle e|$. Our Hamiltonian takes the form

$$H(t) = \frac{\Omega(t)}{2} (|g\rangle\langle e| + |e\rangle\langle g|) + \delta |e\rangle\langle e|. \quad (\text{A.15})$$

We then used `mesolve` provided by the QuTiP Python package, which solves the Lindblad master equation

$$\dot{\rho}(t) = -i [H(t), \rho(t)] + \frac{1}{2} \sum_n \left(2C_n \rho C_n^\dagger - \{C_n^\dagger C_n, \rho\} \right), \quad (\text{A.16})$$

where ρ is the density matrix, $[A, B] = AB - BA$ and $\{A, B\} = AB + BA$. Fitting to the data allows us to extract T_1 , $T_2 = \left[(2T_1)^{-1} + T_2^{*-1} \right]^{-1}$, and Γ^{-1} .

A.5 Single-photon emission properties of kh divacancies

Single kh divacancies are expected to follow sub-Poissonian photon emission statistics and exhibit antibunching as the optical transition addresses a two-level system. We confirm this by performing correlation spectroscopy on the emission of a single kh divacancy under off-resonant 905-nm excitation. The second order correlation function $g^{(2)}(\tau)$ quantifies the intensity correlations of photon emission events as a function of time τ from another photon emission event. Since a single-photon emitter cannot emit more than one photon at any given moment, we expect the intensity at zero time difference to be zero, $g^{(2)}(0) = 0$.

We detect photons in a Hanbury-Brown-Twiss configuration, where the emission from the defect impinges on the input of a 50/50 beamsplitter and the outputs are routed to individual channels of a superconducting nanowire single-photon detector. By histogramming the photon arrival time differences between the two detector channels, we find a dip at a time difference $\tau = 0$ reaching $g^{(2)}(0) = 0.269$ without background subtractions, indicating single-photon emission but also non-negligible coincidence counts with background fluorescence (Fig. A.1A). We perform a background subtraction procedure using a signal-to-background

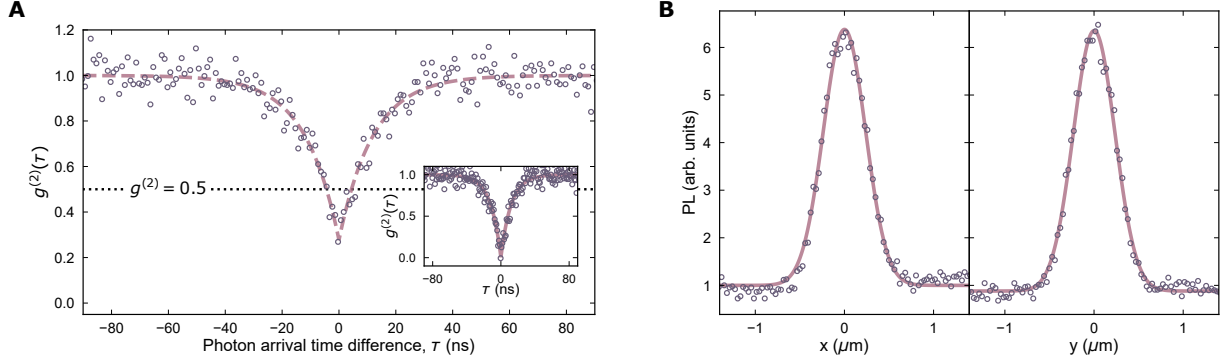


Figure A.1: **Correlation spectroscopy of a single kh divacancy.** (A) Second order correlation function $g^{(2)}$ as a function of photon arrival time difference τ . The dotted line is a best fit to a two-level system model with no background subtraction. $g^{(2)}(0) < 0.5$ indicates single-photon emission. Inset: $g^{(2)}(\tau)$ after background subtraction. (B) Photoluminescence line cuts along the x - and y -axes used to determine signal-to-background ratio used for background subtraction.

ratio of 5.77:1 (Fig. A.1B), producing a background-subtracted value of $g^{(2)}(0) = 0.005$ (Fig. A.1A, inset) and confirming high-fidelity single-photon emission of single kh divacancies.

A.6 Ground- and excited-state spin-spin interactions

The spin-1 nature of the ground- and excited-state spin gives rise to dipolar spin-spin interactions which manifest as axial and transverse zero-field splittings D and E . D and E are determined by ellipsoidicity and rhombicity of the electronic wavefunction distribution, respectively. At $\mathbf{B} \approx 0$, we probe the spin transition frequencies between $|0\rangle$, $|-\rangle$, and $|+\rangle$ and their respective spin-conserving optical transitions in single kh divacancies to establish values for D and E in both the ground and excited state. In the ground state, kh divacancies have a value of D similar to that of other divacancy configurations. The basal orientation of the defect and the resulting neighboring lattice configuration lead to a non-axial electronic wavefunction distribution, with a corresponding non-zero E in the ground state. In the excited state, D is similar to values obtained for c -axis divacancies, while E is significantly larger than its ground-state counterpart. We note that since $|E_{\text{ES}}| > |D_{\text{ES}}/3|$ in the ground-state basis, a rotation can be made to yield D' and E' so that $|E'_{\text{ES}}| < |D'_{\text{ES}}/3|$. By rotating

	D (MHz)	E (MHz)
GS	1333.95	18.42
ES (GS basis)	957	-483
ES (diagonalized)	-1203	237

Table A.1: **Zero-field splitting values for kh divacancies.** D and E are the axial and transverse zero-field splitting components, respectively. Values for D and E in the excited state are determined in the same basis as the ground state, as well as after proper diagonalization.

the zero-field splitting tensor 90° about the y -axis, we set our new coordinate system to be $z' = x$, $y' = y$, and $x' = -z$. In this basis,

$$D' = \frac{3}{2}(E - D/E), \quad (\text{A.17})$$

$$E' = \frac{1}{2}(E + D), \quad (\text{A.18})$$

which correspond to $D'_{\text{ES}} = -1.203$ GHz and $E'_{\text{ES}} = 237$ MHz (see Table A.1). $D'_{\text{ES}} < 0$ in the excited state indicates that the electronic wavefunction distribution is prolate (elongated) along the z' -axis. Although this distortion between the ground- and excited-state electronic wavefunction distribution perturbs the positions of the nearby lattice sites, the spin basis of $\{|0\rangle, |-\rangle, |+\rangle\}$ is invariant under changes of D and E and the spin Hamiltonian at $\mathbf{B} \approx 0$ is undisturbed if changes in hyperfine interactions between the ground and excited state are neglected.

A.7 Spectral diffusion of optical transitions

The presence of impurities other than divacancies in the silicon carbide sample introduces charge trap sites that can either destabilize divacancies into other charge states, or induce spurious Stark shifts as the trap site varies in charge state. Detailed studies in divacancy ensembles and single divacancies have reported excitation wavelength-dependent photoionization of divacancies, converting neutral divacancies to dark charge states. By treating a trap site as a single fluctuating elementary charge at a distance of 100 nm away from the

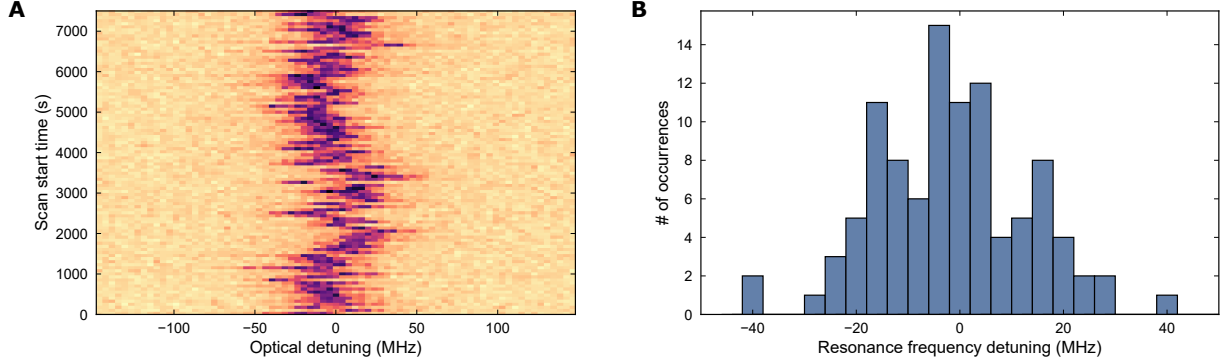


Figure A.2: **Spectral diffusion of a single kh divacancy.** (A) Wandering of the optical resonance central frequency observed in photoluminescence excitation spectra. (B) The distribution of optical resonance central frequencies over the course of 100 scans.

divacancy, we estimate the magnitude of the induced electric field to be $O(100 \text{ kV/m})$, which corresponds to $O(100 \text{ MHz})$ shifts of the excited-state orbital energy for a typical divacancy. We address this challenge by applying dc electric fields, creating a depletion region in a volume around the divacancy. This depletion region is marked by a reduction in fluctuating charge carriers, which decreases the absorption rate of the divacancy with regards to capturing a charge, and also reduces the charge fluctuation rate of nearby trap sites.

We quantify the magnitude of this charge fluctuation by repeatedly acquiring photoluminescence excitation (PLE) spectra on a single kh divacancy under an applied dc electric field. We alternate between a 905-nm initialization pulse and a narrow-line laser pulse that is swept across the resonance, all while a dc electric field is applied. At an applied field of 1 MV/m , we produce a depletion region large enough that fast ($>1 \text{ Hz}$) charge fluctuations of nearby trap sites are diminished, narrowing the optical transition linewidths to about 20 MHz and allowing us to resolve small fluctuations ($<10 \text{ MHz}$) using the narrow optical resonance (Fig. A.2A). We take 100 PLE scans, each taking 75 seconds to complete. In each scan we observe charge fluctuations as shifts of the optical resonance's center frequency. By histogramming each scan's center frequency, we find a broad distribution over 100 scans, which suggests that the charge fluctuations are caused by a small ensemble of trap sites (Fig. A.2B). The residual fluctuations can be attributed to photoionization effects of the

905-nm initialization laser, as well as the finite size of the depletion region created by the applied electric field.

A.8 dc Stark tuning of kh divacancies

In c -axis divacancies, dc Stark tuning has been observed using transverse (orthogonal to the defect axis) and longitudinal (parallel to the defect axis) electric fields. The effect of the transverse field is to split the ${}^3E$ orbital into two separate orbital branches, much like the effect of the transverse crystal field in kh divacancies, while a longitudinal field shifts all levels with equal displacement. In kh divacancies, the large transverse crystal field acts as a large in-built transverse strain, which splits the two orbital branches by order meV. Additional electric fields, either transverse or longitudinal, with magnitudes much smaller than the transverse crystal field serve to linearly perturb the excited-state energy level.

We observe dc Stark shifts by applying a dc electric field across the capacitor. By measuring the resonance frequency of the $|0\rangle \leftrightarrow |A'_0\rangle$ optical transition, we can quantify the kh divacancy's response to dc electric fields (Fig. A.3A,B). Initially, at low applied fields, we observe no dc Stark shifts. This observation can be explained by mobile charge carriers spatially rearranging around the kh divacancy under illumination, producing an equal and opposite electric field vector to the applied field. This phenomenon has also been reported in divacancies in SiC homojunctions, where a characteristic turn-on applied field is required before a complete depletion region is formed and Stark shifts are possible. Starting around 0.4 MV/m, we observe a blueshift of the $|0\rangle \leftrightarrow |A'_0\rangle$ resonance, which continues in a near-linear trend up to 3 MV/m.

We attribute the nonlinear Stark shift behavior near zero applied dc electric field to the creation of a non-uniform depletion region. Only at a critical field does the gap between the capacitor plates become fully depleted, at which point dc Stark shifts are observed. However, we do not expect this nonlinearity to affect high-frequency ac electric fields, given a characteristic Maxwell relaxation time of mobile charge carriers. When the applied electric field

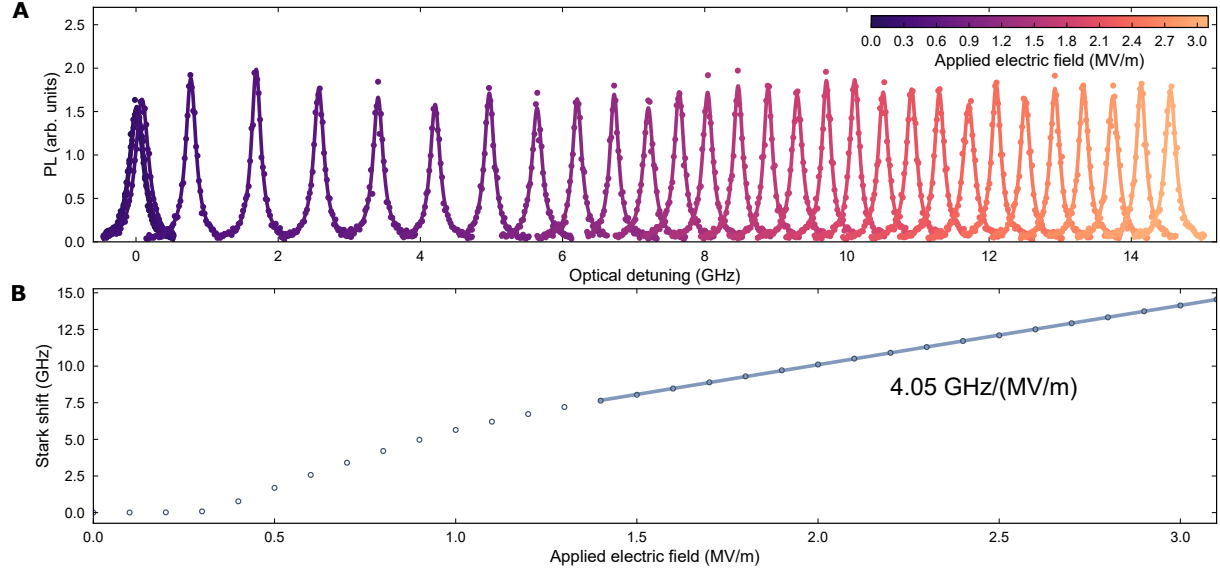


Figure A.3: **dc Stark shifts of single kh divacancies.** (A) Optical resonance $|0\rangle \leftrightarrow |A'_0\rangle$ at various applied voltages from 0 V to 31 V. (B) Center frequencies of the optical resonances in (A) as a function of applied voltage. In the linear regime, a Stark shift sensitivity of 4.05 GHz/(MV/m) is observed.

fluctuates on timescales much longer than the Maxwell relaxation time, the mobile charge carriers can respond to the nonequilibrium electric field by spatially rearranging to cancel out the electric field. At shorter fluctuation timescales corresponding to high-frequency ac electric field drives, the mobile charge carriers do not move significantly in one period of the electric field fluctuations, resulting in negligible field cancellation. If mobile charge carriers produced significant screening, a sinusoidal electric field drive would be distorted at low amplitudes and would yield higher-order harmonics in the Landau-Zener-Stückelberg spectrum. We qualitatively confirm that this is not the case at GHz frequencies in Fig. 5.3B, where the experimental data is well-described by a simulation using a sinusoidal drive and no higher order harmonics are observed at low drive amplitudes.

A.9 Integrated multiphoton resonance lineshape

The steady-state excited state population expression for the n th resonance is

$$P_e(\delta; n) = \frac{\Delta_n^2}{\frac{4\pi^2}{T_1 T_2} + \frac{T_2}{T_1} \delta^2 + \Delta_n^2}, \quad (\text{A.19})$$

$$\Delta_n = \Omega J_n(\mathcal{A}/\omega), \quad (\text{A.20})$$

where Ω is the optical Rabi frequency, $J_n(x)$ are Bessel functions of the first kind, $\mathcal{A}$ is the amplitude of the induced Stark shift from the ac electric field, ω is the ac electric field frequency, T_1 is the radiative decay time, and T_2 is the optical coherence time. The area of the n th resonance is then

$$\begin{aligned} A_e(n) &= \int_{-\infty}^{\infty} P_e(\delta'; n) d\delta' \\ &= \frac{\pi T_1 \Delta_n^2}{\sqrt{4\pi^2 + T_1 T_2 \Delta_n^2}}. \end{aligned} \quad (\text{A.21})$$

We show this by summing the photoluminescence within the baseband resonance of the LZS spectrum as a function of applied ac electric field amplitude, $|F_{\parallel}|$ (Fig. A.4). We expect the intensity $A_e(0)$ to follow a norm-Bessel dependence, $|J_n(x)|$, in the limit of strong driving $T_1 T_2 \Omega_n^2 \gg 4\pi^2$. This can be seen by assessing $A_e(n)$ in that limit

$$A_e(n) \approx \pi \sqrt{\frac{T_1}{T_2}} |\Delta_n|. \quad (\text{A.22})$$

In systems where relaxation mechanisms are more intense than the realizable two-level system Rabi frequency, the expected dependence of the integrated time-normalized excited-state population follows a squared-Bessel function, $J_n(x)^2$. Again, this can be confirmed by assessing $A_e(n)$ in the limit $T_1 T_2 \Omega^2 \ll 4\pi^2$

$$A_e(n) \approx \frac{T_1 \Delta_n^2}{2}, \quad (\text{A.23})$$

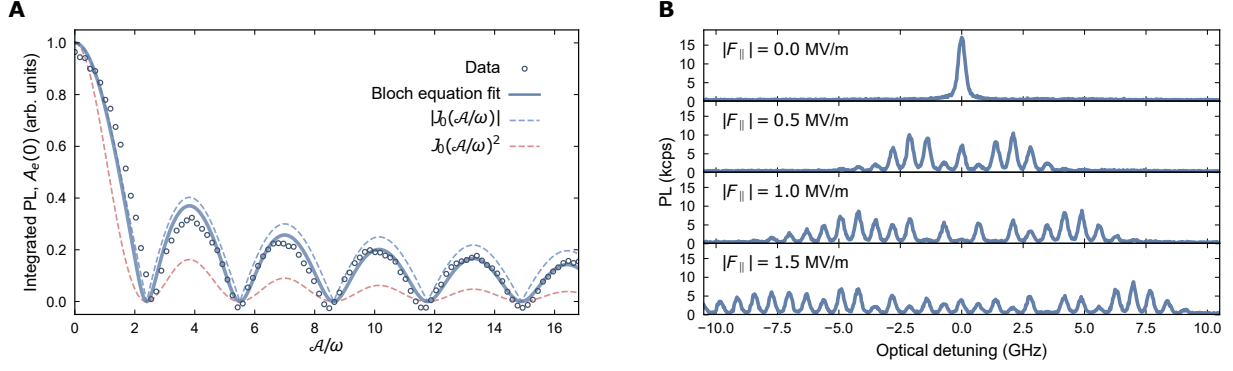


Figure A.4: **Photoluminescence of multiphoton resonances.** (A) Integrated photoluminescence of carrier resonance. Data (open circles) are best fit to a Bloch equation model with weak relaxation compared to optical driving. Dashed lines indicate expected integrated resonance lineshapes in the limit of vanishing relaxation (blue) and vanishing optical drive (red). (B) Line cuts of multiphoton resonances at select electric field amplitudes.

which qualitatively agrees with the behavior near the Bessel local minima. This strong dependence on coherence time has been used in prior works to characterize the intensity of the relaxation mechanisms of a two-level system. Optical linewidths in the zero dc bias regime can reach 440 MHz under strong optical drive, as shown in Fig. A.4B. This confirms that the strong driving regime is achieved and the condition to observe norm-Bessel photoluminescence as a function of applied ac electric field amplitude is satisfied.

A.10 Electrical properties of on-chip planar capacitor

The planar capacitor structure on the sample is comprised of two parallel planar plates of Ti/Au, each of width $10\mu\text{m}$ and length $100\mu\text{m}$, with a gap width of $10\mu\text{m}$. Considering the SiC substrate with a relative permittivity of $\epsilon_0 = 9.66$, we obtain a capacitance of 7.7fF for the capacitor structure. In this geometry, we produce a high-pass filter, where the GHz-frequency range used in our experiments are reflected and yield maximal electric field across the gap. We characterize the behavior of the capacitor by measuring the input voltage port reflection coefficient S_{11} (Fig. A.5A). At low frequencies, we find the structure to be mostly reflective, with decreasing reflectivity at higher frequencies, characteristic of a high-pass filter. While the reflectivity exhibits oscillations characteristic of cable resonances,

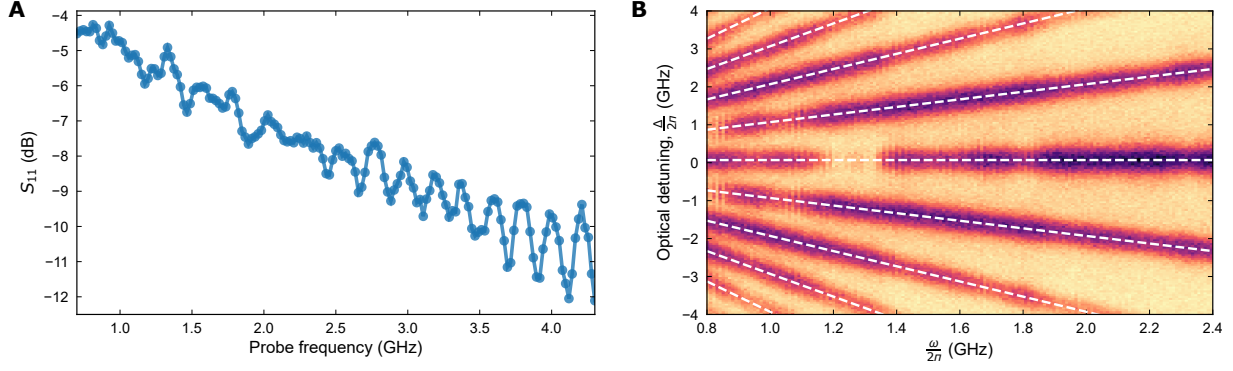


Figure A.5: **Nonresonant device properties.** (A) S_{11} of device showing capacitive behavior at low frequencies. (B) Landau-Zener-Stückelberg interference of a single kh divacancy as a function of electric field modulation frequency. Dashed lines serve as guides to multiphoton resonances, $\Delta = m\omega$, $m = -4, \dots, 4$.

we do not find a strong resonance in the GHz-frequency range. Losses through the cable do not exceed -3.5 dB throughout the probe frequency range and are neglected. The non-resonant structure allows for broadband tuning of the drive frequency. We demonstrate this by observing LZS interference fringes as a function of ac electric drive frequency (Fig. A.5B). With a fixed ac electric field drive amplitude, we vary the frequency of the drive and obtain photoluminescence excitation spectra. We find broad fluctuations in fluorescence as the quantity $x = \mathcal{A}/\omega$ changes, as well as more rapid fluctuations from cable resonances. However, no obvious resonances arising from the capacitor are found. Qualitatively, this confirms that our electric field interaction is not enhanced by any resonant behavior of the capacitor, and that further enhancements can be attained with an intentionally resonant structure.

A.11 Ground-state ZEFOZ spin transitions

At $\mathbf{B} \approx 0$, the effect of E_{GS} is to mix the $S_z = \{|-1\rangle, |0\rangle, |+1\rangle\}$ eigenbasis into $|0\rangle$, $|+\rangle = \frac{1}{\sqrt{2}}(|+1\rangle + |-1\rangle)$, and $|-\rangle = \frac{1}{\sqrt{2}}(|+1\rangle - |-1\rangle)$. An eigenbasis containing $|+\rangle$ and $|-\rangle$ allows for magnetic driving of the $|+\rangle \leftrightarrow |-\rangle$ transition by virtue of $|\langle + | S_z | - \rangle|^2 \neq 0$. Knowing this, we can apply magnetically-driven spin rotations to reach all spin eigenstates

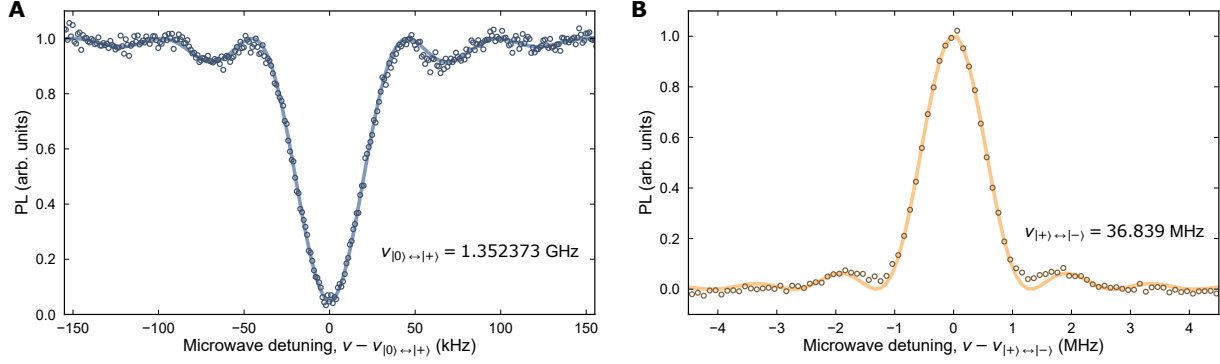


Figure A.6: **Pulsed optically detected magnetic resonance of single kh divacancies.** (A) Spin resonance $|0\rangle \leftrightarrow |+\rangle$ located at $D_{\text{GS}} + E_{\text{GS}} = 1.352373 \text{ GHz}$. Photoluminescence acquired from $|0\rangle \leftrightarrow |A'_0\rangle$ excitation. (B) Spin resonance $|+\rangle \leftrightarrow |-\rangle$ located at $2E_{\text{GS}} = 36.839 \text{ MHz}$. Photoluminescence acquired from $|-\rangle \leftrightarrow |A'_-\rangle$ excitation. Fits are made to a Bloch equation model of a dissipationless two-level system.

in combination with at least one of the spin rotations enabled by $|\langle 0|S_x|+\rangle|^2 \neq 0$ or $|\langle 0|S_y|-\rangle|^2 \neq 0$. We demonstrate the ability to span the eigenbasis by performing ODMR on both the $|0\rangle \leftrightarrow |+\rangle$ and $|+\rangle \leftrightarrow |-\rangle$ transitions. We find these resonances at $D_{\text{GS}} + E_{\text{GS}} = 1.352373 \text{ GHz}$ and $2E_{\text{GS}} = 36.839 \text{ MHz}$, respectively (Fig. A.6). While the $|0\rangle \leftrightarrow |-\rangle$ transition can also be driven, the magnetic dipole of that transition is not well aligned with the magnetic driving field in our experiments. Consequently, the large drive power necessary to perform spin driving of the $|0\rangle \leftrightarrow |-\rangle$ transition produces significant off-resonant driving of the well-coupled $|0\rangle \leftrightarrow |+\rangle$ transition, which obscures ODMR contrast and suppresses spin-rotation fidelity. This limitation can be overcome by introducing a more extreme miscut on the substrate to tilt the magnetic dipole out of plane, or by fabricating transparent drive wires above the defect to produce an in-plane drive field.

A.12 Nuclear spin bath interactions

Naturally abundant silicon carbide consists of non-zero nuclear spin isotopes of Si and C. Specifically, Si is comprised of 4.69% ^{29}Si and C is comprised of 1.07% ^{13}C , both of which are spin- $\frac{1}{2}$ species. As these nuclear spins undergo dipolar interactions with each other, the magnetic field environment of the divacancy fluctuates, resulting in inhomogeneous broaden-

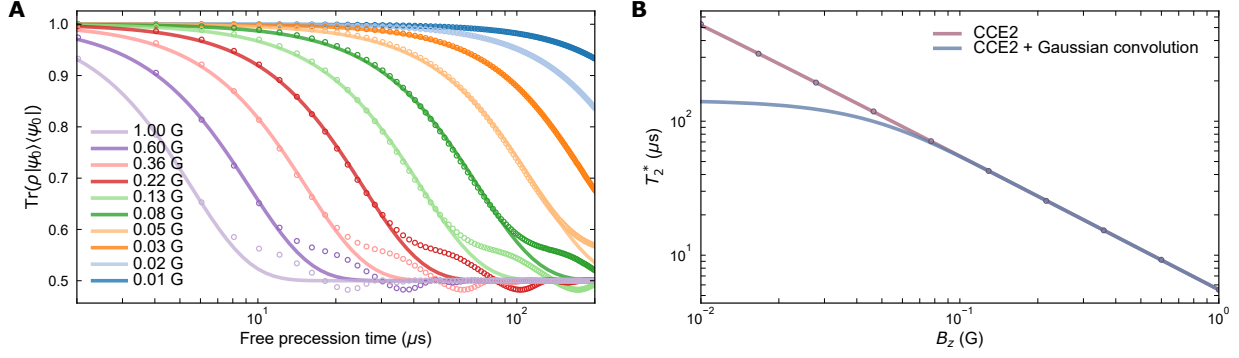


Figure A.7: **Cluster-correlation expansion simulations of kh divacancies.** (A) CCE2 simulations of spin dynamics when the spin is prepared into $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |+\rangle)$ at various z -axis-oriented magnetic fields. (B) T_2^* spin dephasing times derived from the stretched exponential fits of each trace in (A). A Gaussian convolution is incorporated into the model to address the singularity at $B_z = 0$.

ing of divacancy spin resonances. This broadening most readily manifests in the finite spin dephasing times that we observe when the spin is prepared into a superposition state.

By virtue of operating at a ZEFOZ point, the kh divacancy spin is resilient to magnetic field fluctuations, which results in significantly lengthened spin dephasing times compared to c -axis divacancies. However, residual fluctuations in the spin resonance frequency still occur given the non-zero higher-order derivatives of the energy dispersion. We use Cluster-Correlation Expansion (CCE) simulations to quantify these fluctuations and arrive upon estimates for T_2^* for kh divacancies prepared into $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |+\rangle)$ at low external magnetic field. We sample 20 randomly-generated nuclear spin configurations and average the dynamics predicted by CCE2 (second-order CCE) for various fields between 1 μT and 100 μT (Fig. A.7A). We find that as the external magnetic field approaches 0, the spin dephasing time of the superposition $|\psi_0\rangle$ increases (Fig. A.7B). However, we note that our simulations predict a singularity for T_2^* at $B_z = 0$, which arises from the vanishing derivative of the energy dispersion at $\mathbf{B} = 0$ combined with the fact our CCE model ignores nuclear spin-induced backaction on the divacancy electron spin. To address this, we incorporate the effects of an Overhauser field, which manifests as classical, Gaussian-distributed fluctuations of the magnetic field. The magnitude of the Overhauser field can be estimated from c -axis

divacancy spin dephasing times at high field, where the limiting factor for T_2^* is macroscopic magnetic field fluctuations, $\Delta B = \frac{\Delta\nu}{g} = \frac{1}{g\pi T_2^*}$. We then model the low-field behavior of kh divacancies by convolving the spin response obtained through CCE2 simulations over a Gaussian distribution of magnetic fields, the width of which is determined by ΔB . We use a c -axis divacancy high-field T_2^* value of $1\text{ }\mu\text{s}$, which corresponds to $\Delta B \approx 10\text{ }\mu\text{T}$. With decreasing B_z , we find that at $B_z \approx \Delta B$ the spin dephasing time begins saturating towards $105\text{ }\mu\text{s}$. This agrees with the experimental value of $74\text{ }\mu\text{s}$ after considering imperfect field cancellation on the order of $10\text{ }\mu\text{T}$, as well as variations of nuclear spin configurations leading to variations in characteristic T_2^* times.

A.13 DFT calculations of the kh divacancy excited state structure

By considering a C_{1h} symmetry basal divacancy configuration as a distorted C_{3v} symmetry c -axis oriented divacancy configuration, we can obtain the excited state structure of the kh divacancy as a perturbation of c -axis VV structure. The theory of the ${}^3E$ excited state is well established for the C_{3v} symmetry diamond NV-center and c -axis divacancies [61]. At zero strain, the ${}^3E$ excited state includes six substates, $|A_1\rangle$, $|A_2\rangle$, $|E_1\rangle$, $|E_2\rangle$, $|E_x\rangle$, and $|E_y\rangle$, whose relative positions are determined by spin-spin interactions, spin-orbit interactions, and the dynamic Jahn-Teller effect [109]. In the case of kh divacancies, the dominant effect is a crystal field splitting which separates the six ${}^3E$ states into two branches, each with three substates. The branches correspond to the $|X\rangle = |a_1e_x - e_xa_1\rangle$ or $|Y\rangle = |a_1e_y - e_ya_1\rangle$ orbital wavefunctions. In terms of ${}^3E$ states, the states in the A'' branch can be expressed as

$$|A''_+\rangle \approx i(|E_2\rangle + |A_2\rangle) = |Y\rangle \otimes |+\rangle \quad (\text{A.24})$$

$$|A''_-\rangle \approx i(|A_1\rangle - |E_1\rangle) = |Y\rangle \otimes |-\rangle \quad (\text{A.25})$$

$$|A''_0\rangle \approx |E_y\rangle = |Y\rangle \otimes |0\rangle \quad (\text{A.26})$$

and the states in the A' branch as

$$|A'_+\rangle \approx |E_1\rangle + |A_1\rangle = |X\rangle \otimes |+\rangle \quad (\text{A.27})$$

$$|A'_-\rangle \approx |A_2\rangle - |E_2\rangle = |X\rangle \otimes |-\rangle \quad (\text{A.28})$$

$$|A'_0\rangle \approx |E_x\rangle = |X\rangle \otimes |0\rangle \quad (\text{A.29})$$

Ab initio calculations (using the PBE functional) show that the splitting between the branches is 3.5 meV and the A' branch is lower in energy. We note that the upper A'' branch corresponds to a metastable atomic configuration as there exists a relaxation path with no barrier from A'' to A' . The spin-orbit interaction, which is the dominant interaction in the C_{3v} symmetry ${}^3E$ fine structure, is suppressed by the crystal field in kh divacancies. Consequently, there is no observable first-order spin-orbit interaction in both branches.

A.14 DFT calculations of electric field dependent phenomena

In the ground state of the C_{3v} -symmetric NV-center in diamond and c -axis divacancies in SiC, it has been established that the spatial part of the ${}^3A_2$ ground state is stable against strain and electric field variations. Due to mixing of the a'_{a1} and a'_e single electron states in C_{1h} -symmetric basal divacancy configurations, this statement is slightly relaxed. Experimental measurements show, however, that the electric field coupling constant of the spin sublevels remains on the order of 3×10^{-4} GHz/(MV/m), which is negligible compared to the 19 GHz/(MV/m) Stark shift of the GS-ES optical transition energy for basally-oriented divacancies obtained in our *ab initio* simulations, which are detailed later. Therefore, we only focus on the Stark effect and excited-state electric field couplings.

The Stark shift is related to the dipole moment of the transition as

$$H_{\text{el}} = -\mathbf{F} \cdot \mathbf{d}_{\text{GE}} \quad (\text{A.30})$$

where $\mathbf{F}$ is the displacement field and $\mathbf{d}_{\text{GE}}$ is the difference of the point defect's dipole moment between the excited state and the ground state. In order to understand the robustness of the kh VV's optical transition, we first discuss C_{3v} -symmetric configurations. In perfect C_{3v} symmetry, the dipole is aligned along the axis of the defect and thus Eq. A.30 simplifies to $H_{\text{el}} = F_z d_z = F_{\parallel} d_{\parallel}$, which linearly shifts the ${}^3E$ optical transition energy. This behavior can be attributed to the mutual cancellation of perpendicular components. Indeed, the $|X\rangle = |a_1 e_x - e_x a_1\rangle$ and $|Y\rangle = |a_1 e_y - e_y a_1\rangle$ orbital states have non-zero perpendicular dipole components in a way so that $d_{\perp}^X = -d_{\perp}^Y$. The perpendicular dipole moment components of $|X\rangle$ and $|Y\rangle$ cancel out either due to the degeneracy of the zero-strain states or due to the mixed spatial states that even contain $|X\rangle$ and $|Y\rangle$.

In realistic conditions, perfect C_{3v} -symmetric configurations relax due to the presence of local strain and electric fields. For example, a perturbing perpendicular electric field or anisotropic strain can split the ${}^3E$ excited state based on the $|X\rangle$ and $|Y\rangle$ character of the states. As a result, two branches can form, each having either predominantly $|X\rangle$ or $|Y\rangle$ spatial character. At finite temperature, $k_B T$ is expectedly larger than the branch splitting caused by local fields, and thus spin conserving transitions, mediated by low energy acoustic phonon modes, can thermalize the occupation of the branches. Note that thermal fluctuations of the branch occupation give rise to a time-dependent, randomly fluctuating perpendicular dipole moment component, due to the different dipole moments of the branches. In the presence of a perpendicular electric field, the energy of the thermalized ${}^3E$ state will fluctuate and may intensify optical dephasing processes.

In *ab ovo* C_{1h} configurations, like the kh divacancy, the perpendicular dipolar coupling components must be distinguished as an in-plane component d_x and an out-of-plane com-

Defect center	$d_{\perp}$ (d_x) (GHz/(MV/m))	$d_{\parallel}$ (GHz/(MV/m))
Diamond NV center (C_{1h} ES, static Jahn-Teller)	1.79	20.43
hh divacancy (C_{1h} ES, static Jahn-Teller)	1.61	19.07
kh divacancy (<i>ab ovo</i> C_{1h})	0.23	19.17

Table A.2: **Calculated electric dipole for the optical transition in select defect centers.** Parallel (perpendicular) indicates parallel (perpendicular) to the defect axis and x is perpendicular to the defect axis and coplanar with the defect axis and the c -axis.

ponent d_y , where the plane is spanned by the defect axis and the c -axis. Due to symmetry reasons, the d_y dipole component must be zero, thus kh divacancies are insensitive to F_y displacement fields to first order. Furthermore, higher order effects due to F_y that would mix the A' and A'' branches are suppressed by the large crystal field splitting between the branches. Therefore, kh divacancies are decoupled from F_y and only experience F_x and F_z components of the displacement field.

The dipole moment that corresponds to the GS-ES optical transition can be determined from *ab initio* simulations. The calculated dipole moments for the diamond NV-center, hh divacancy, and kh divacancy can be found in Table A.2. Note that in all three configurations (corresponding to a $|X\rangle$ spatial excited state configuration), the parallel component is still the dominant contribution to the electric dipole moment of the defects. As the kh divacancy axis is inclined at an angle of 19.5° with respect to the basal plane, we expect that kh divacancies can couple to in-plane electric fields about 11 times stronger when compared to c -axis divacancy coupling to in-plane electric fields.

APPENDIX B

UNIVERSAL COHERENCE PROTECTION IN A SOLID-STATE QUBIT – DETAILS

B.1 Sample preparation and experimental setup

Our 4H-SiC sample consists of a 20 μm high-purity i-type SiC layer epitaxially grown on a 4° off-axis miscut of the Si face of a high-purity semi-insulating SiC substrate (serial number A3177-14, Norstel AB). Electron irradiation of the sample with 2-MeV electrons at a dose of $3 \times 10^{12} \text{ e}^-/\text{cm}^2$ and subsequent annealing at 850 °C for 30 min in Ar produces neutral divacancies uniformly throughout the epitaxial i-type 4H-SiC. A coplanar capacitor structure with a 10 μm gap width and a wire with 10 μm width made of Ti/Au are then patterned on the sample surface using electron beam lithography. Samples are cooled to 5 K in a closed-cycle cryostat (Cryostation s100, Montana Instruments).

The confocal microscope consists of a 905 nm excitation laser (QFLD-905-200S, QPhotonics) for off-resonant spin initialization, as well as a narrow-line tunable laser (DL pro, TOPTICA Photonics) for resonant spin readout. We focus these excitation beams through a microscope objective (LCPLN100XIR, Olympus). We reject laser scatter and background emission while retaining $\hbar\omega$ divacancy phonon-sideband emission in the range of 1090 nm to 1300 nm by filtering the emission from the sample with a tunable longpass filter (TLP01-1116-25x36, Semrock), a fixed longpass filter (LP02-1064RE-25, Semrock), a fixed shortpass filter (89-676, Edmund Optics), and a notch filter (NF1064-44, Thorlabs). We detect the filtered optical signal with >80% quantum efficiency using a low-jitter, low-dark count superconducting nanowire single-photon detector (SNSPD; Opus One, Quantum Opus). Electrical pulses from the SNSPD are counted using a data acquisition module (PCI-6259, National Instruments).

We drive the spin transition $|0\rangle \leftrightarrow |+\rangle$ and driven single-quantum spin transitions $|0\rangle \leftrightarrow |\pm 1\rangle$ using signal generators (SG396, Stanford Research Systems) modulated by an arbitrary

waveform generator (HDAWG8, Zurich Instruments). We implement the dressing drive on the $|+\rangle \leftrightarrow |-\rangle$ transition using a modulated signal generator (HDAWG8). The outputs of these signal generators are combined using a diplexer (D3005001, Gowanda-BTI) and routed to the on-chip wire, which produces ac magnetic fields. We implement spin rotations on the driven double-quantum spin transition $|+1\rangle \leftrightarrow |-1\rangle$ using a modulated signal generator (HDAWG8), the output of which is routed to the on-chip coplanar capacitor for generation of ac electric fields.

B.2 Feedback protocol details

Noise in the dressing drive manifests as fluctuations $\delta\Omega$ in the dressing drive Rabi frequency Ω_0 . Our feedback protocol, shown in a step-by-step diagram in Fig. 6.4A, is designed to detect these fluctuations. We optically initialize the spin into $|0\rangle$, enable the dressing drive, and rotate to $|+1\rangle$. A spin superposition $|x\rangle = \frac{1}{\sqrt{2}}(|+1\rangle + |-1\rangle)$ is prepared in the dressed basis with a $\pi/2_x$ probe pulse that is detuned by a quantity Δ_{fb} from the desired resonance Ω_0 **(1)** causing the spin to precess at $\Delta_{\text{fb}} - \delta\Omega$. A free precession time τ is chosen so that $\Delta_{\text{fb}}\tau_{\text{fb}} = \pi/2$, which under no fluctuations is a $\pi/2_z$ rotation and causes the spin to evolve to $|y\rangle = \frac{1}{\sqrt{2}}(|+1\rangle + 1|-1\rangle)$. Otherwise the spin precession subtends an angle depending on $\delta\Omega$ **(2)**. A final $-\pi/2_x$ pulse then rotates the spin into the $y-z$ plane **(3)** where it is projected into the $|\pm 1\rangle$ basis and converted into an error signal **(4)**. An ideal, unperturbed resonance frequency corresponds to zero error, while positive or negative $\delta\Omega$ correspond to negative and positive error signals, respectively. A simple proportional feedback based on this error signal is then applied to the spin manipulation frequency at a period of one minute. We choose $\Delta_{\text{fb}}/(2\pi) = 16.66$ Hz and $\tau_{\text{fb}} = 15$ ms to retain sensitivity to 30 Hz fluctuations in $\Omega/(2\pi)$ while maintaining enough signal to efficiently obtain an error signal. We run a ten-second feedback process every minute in the feedback protocol implemented in Chapter 6.

B.3 Spin properties at high magnetic field

At high magnetic field, $\gamma|\mathbf{B}| > E$, where $\gamma/(2\pi) = 28.024$ MHz/mT is the electron spin gyromagnetic ratio, $\mathbf{B}$ is the magnetic field pseudovector, and E is the transverse zero-field splitting, the kh divacancy behaves like a c -axis divacancy system. In this regime, the ground-state spin quantization axis is defined by $DS_z^2 + \gamma\mathbf{B} \cdot \mathbf{S}$, where D is the axial zero-field splitting and $\mathbf{S}$ is the vector of spin-1 operators, instead of primarily by the combined axial and transverse spin-spin interactions, $S_z^2 + S_x^2 - S_y^2$. Under these conditions, the spin energy dispersion relation recovers a linear behavior $\delta\omega = \gamma|\delta B_z|$, where ω is the spin transition energy, and the spin becomes noticeably more susceptible to magnetic fluctuations. In chapter 6, we demonstrate that the kh divacancy investigated for that study has spin coherence properties comparable to c -axis divacancies and nitrogen-vacancy (NV) centers in diamond at high field. We accomplish this by applying 1.2 mT along the z -axis of the divacancy, thus displacing the upper-most level of the ground-state spin by +20 MHz in frequency (Fig. B.1A,B). To evaluate the spin's susceptibility to environmental fluctuations, we then prepare a superposition between the undriven $|0_z\rangle$ and $|+1_z\rangle$ states (Fig. 6.2B), which are approximately energy eigenstates of S_z . The resulting measured values for T_2^* and T_2 are consistent with values reported for c -axis divacancies as well as NV centers in diamond, both of which have similar energy structures at this applied magnetic field. Furthermore, this similarity is supported by the normalized slope of the energy dispersion relation of the spin at $B_z = 1.2$ mT (Fig. B.1C), which is about 0.88 and thus experiences similar degrees of energy inhomogeneity as c -axis divacancies and NV centers in diamond, which have normalized slopes of about 1.

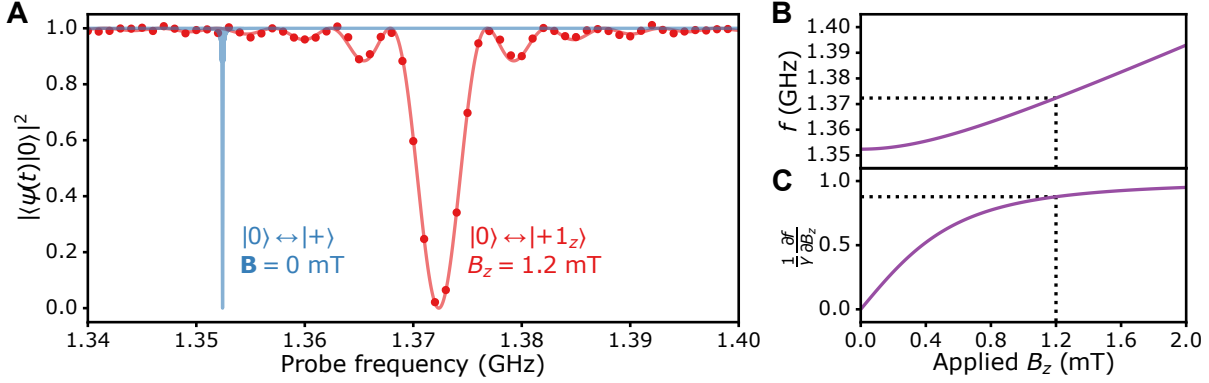


Figure B.1: **kh divacancy spin resonance at non-zero field.** (A) Pulsed optically detected magnetic resonance of the single kh divacancy ground-state spin at $\mathbf{B} = 0$ and $B_z = 1.2$ mT. (B) Energy dispersion relation of the higher-energy spin resonance as a function of applied z -axis magnetic field. (C) The first derivative of the dispersion relation with respect to z -axis field. At $B_z = 1.2$ mT, the kh divacancy behaves like a c -axis divacancy, since the normalized slope of the energy dispersion relation is about 0.88 (the slope of the energy dispersion relation for c -axis divacancies is about 1).

B.4 MW-frequency magnetic excitation of the kh divacancy spin at zero magnetic field

At zero magnetic field, $\mathbf{B} = 0$, the quantization axis of the kh divacancy ground-state spin system is determined by the combined effect of the ground-state axial and transverse zero-field splittings, causing the spin system to hybridize from primarily having eigenstates of S_z to those of $S_z^2 + S_x^2 - S_y^2$. The resulting eigenstates, represented as linear combinations of S_z eigenstates $\{|0_z\rangle, |1_z\rangle, |-1_z\rangle\}$ are

$$|0\rangle = |0_z\rangle, \quad (\text{B.1})$$

$$|+\rangle = \frac{1}{\sqrt{2}} (|1_z\rangle + |-1_z\rangle), \quad (\text{B.2})$$

$$|-\rangle = \frac{1}{\sqrt{2}} (|1_z\rangle - |-1_z\rangle). \quad (\text{B.3})$$

We emphasize that the S_z eigenstates are distinct from the $|\pm 1\rangle$ states in the decoherence-protected subspace presented in chapter 6. Two relevant consequences arise when operating

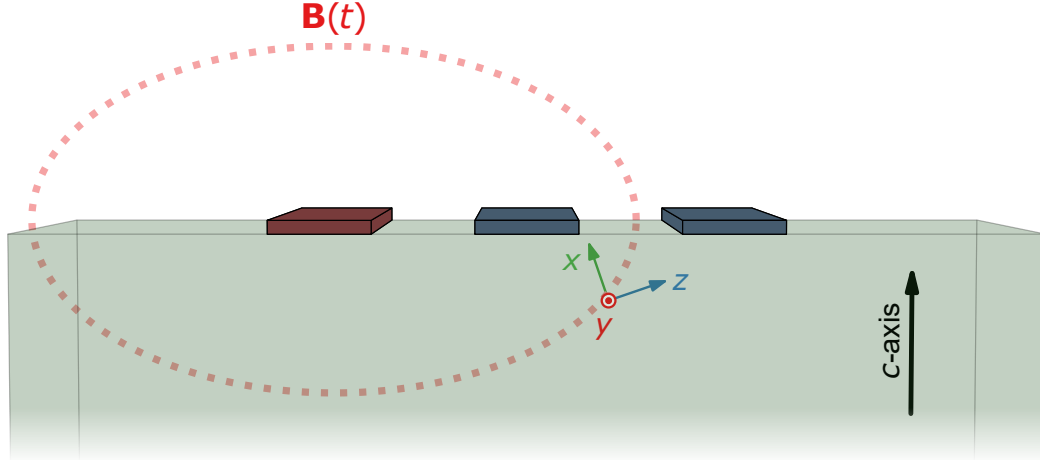


Figure B.2: ***kh* divacancy spin sensitivity to linearly polarized magnetic fields.** Cutaway diagram of the silicon carbide sample corresponding to the optical microscope image shown in Fig. 6.1A. The red wire and blue capacitor plates are at the top of the sample. The green, red, and blue vectors correspond to the axes in the lattice diagram in the lower left of Fig. 6.1A. The *kh* divacancy spin dipoles are tilted into the silicon carbide basal plane. For the *kh* divacancy studied in this work, linearly polarized microwave-frequency magnetic fields (dotted red line), $\mathbf{B}(t)$, nearly always have appreciable parallel components along the *x*- and *z*-axes, but are almost entirely orthogonal to the *y*-axis. Diagram not to scale.

in this eigenbasis. First, the spin transitions between $|0\rangle$ and the other two spin states $|\pm\rangle$ are no longer mediated by circularly polarized magnetic excitation. Instead, linearly polarized magnetic fields aligned along S_x (S_y) selectively drive $|0\rangle \leftrightarrow |+\rangle$ ($|0\rangle \leftrightarrow |-\rangle$). For *kh* divacancies, the S_y magnetic dipole is oriented in the plane of the crystal, nearly entirely orthogonal to the *c*-axis (Fig. B.2). S_x and S_z , on the other hand, have significant vector components parallel to the *c*-axis. This becomes relevant when considering the MW excitation emitted by a lithographically patterned wire on the surface of the sample, which primarily generates magnetic fields parallel to the *c*-axis near the sample surface. This causes the parallel dipole components (S_x and S_z) to be driven efficiently, while the orthogonal dipole component S_y cannot be driven efficiently without significant off-resonant driving of the other spin transitions.

Second, in this eigenbasis, a magnetically allowed spin rotation can be driven between

the upper two spin levels, $|+\rangle \leftrightarrow |-\rangle$. The S_z magnetic dipole mediates this transition, which is efficiently driven by the lithographically patterned wire. This is crucial to our implementation of the dressing drive, allowing us to resonantly address this transition using a low-power, low-noise MW-frequency magnetic excitation that induces minimal dissipative Joule heating and perturbation of the sample and spin system.

B.5 High-fidelity determination of transverse zero-field splitting magnitude

A central component of the protocol presented in chapter 6 is the continuous dressing drive applied on resonance with the $|+\rangle \leftrightarrow |-\rangle$ spin transition. Detuning of the dressing drive frequency from the natural resonance of $|+\rangle \leftrightarrow |-\rangle$ can degrade the dressed spin coherence properties, and thus the resonance frequency of this transition must be determined to high accuracy and precision. We accomplish this by performing Ramsey interferometry at $\mathbf{B} = 0$ on this transition by preparing the superposition $\frac{|+\rangle + |-\rangle}{\sqrt{2}}$ while sweeping the pulse detuning and observing the spin projection after a free precession time of 100 μs (Fig. B.3). The free precession time is chosen to give us sufficient frequency resolution while maintaining enough readout signal. We obtain the true resonance by extracting the pulse detuning at which the spin does not precess in the rotating frame. For the kh divacancy used in this study, we find this resonance to be at $\omega/(2\pi) = 36.706329(7)$ MHz, and establish the magnitude of the transverse zero-field splitting E as half of that value, $E/(2\pi) = 18.353164(4)$ MHz.

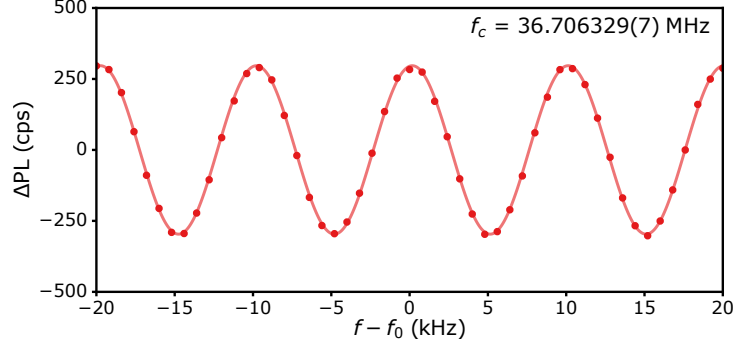


Figure B.3: **Determination of transverse zero-field splitting magnitude.** Ramsey interferometry of the superposition state $|\psi_0\rangle = \frac{1}{\sqrt{2}}(|-\rangle + |+\rangle)$ prepared with a probe frequency f detuned from $f_0 = 36.706157$ MHz. The frequency at which the central fringe photoluminescence is maximized is the frequency where the superposition does not rotate in the rotating frame, simultaneously indicating the frequency of the natural resonance, f_c .

B.6 Floquet treatment of Autler-Townes splitting under a dressing drive

The Hamiltonian of the kh divacancy ground-state spin with an applied ac magnetic drive along the z -axis is

$$H = DS_z^2 + E(S_x^2 - S_y^2) + \gamma \mathbf{B} \cdot \mathbf{S} + \Omega \cos(\omega t) S_z, \quad (\text{B.4})$$

where Ω is the Rabi frequency of the applied driven when on resonance, and ω is the frequency of the applied drive. We set $\hbar = 1$ throughout this derivation for simplicity. Since this Hamiltonian is periodic in time with a period of $T = 2\pi/\omega$, it holds that

$$H(t + T) = H(t). \quad (\text{B.5})$$

We can therefore apply Floquet's theorem to the system and derive the quasienergy spectrum of the ground-state spin, which will serve as stroboscopic energy eigenvalues for our analysis. In order to understand the Autler-Townes splitting spectrum at zero magnetic field, we consider the special case $\mathbf{B} = 0$, but $\Omega \neq 0$. We begin by performing a Fourier decomposition

of H

$$H = \sum_n H_n e^{in\omega t}, \quad (\text{B.6})$$

which yields

$$H_0 = DS_z^2 + E(S_x^2 - S_y^2), \quad (\text{B.7})$$

$$H_{\pm 1} = \frac{\Omega}{2} S_z. \quad (\text{B.8})$$

In the S_z basis, these Fourier-decomposed Hamiltonian components are

$$H_0 = \begin{bmatrix} D & 0 & E \\ 0 & 0 & 0 \\ E & 0 & D \end{bmatrix}, \quad (\text{B.9})$$

$$H_{\pm 1} = \begin{bmatrix} \frac{\Omega}{2} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -\frac{\Omega}{2} \end{bmatrix}. \quad (\text{B.10})$$

We can diagonalize H_0 with a unitary rotation U

$$U = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & 0 & 1 \\ 0 & \sqrt{2} & 0 \\ 1 & 0 & -1 \end{bmatrix}, \quad (\text{B.11})$$

$$\begin{aligned} H'_0 &= U^\dagger H_0 U \\ &= \begin{bmatrix} D+E & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & D-E \end{bmatrix}. \end{aligned} \quad (\text{B.12})$$

In this basis, $H_{\pm 1}$ is

$$H'_{\pm 1} = U^\dagger H_{\pm 1} U = \begin{bmatrix} 0 & 0 & \frac{\Omega}{2} \\ 0 & 0 & 0 \\ \frac{\Omega}{2} & 0 & 0 \end{bmatrix}. \quad (\text{B.13})$$

The Floquet block matrix is then constructed using H'_0 and $H'_{\pm 1}$

$$H_F = \begin{bmatrix} \ddots & & & & \\ & \ddots & & & \\ & & H'_0 + \omega \mathbb{I} & H'_{+1} & \\ & & H'_{-1} & H'_0 & H'_{+1} \\ & & & H'_{-1} & H'_0 - \omega \mathbb{I} & \ddots \\ & & & & \ddots & \ddots \end{bmatrix} \begin{matrix} |m = +1\rangle \\ |m = 0\rangle \\ |m = -1\rangle \end{matrix}, \quad (\text{B.14})$$

where m denotes the Floquet mode index. This yields a Floquet matrix

$$H_F = \begin{bmatrix} \ddots & & & & & & & & \\ & \begin{array}{|ccc|ccc|cc|} \hline D+E+\omega & 0 & 0 & 0 & 0 & \Omega/2 & & & \\ 0 & \omega & 0 & 0 & 0 & 0 & & & \\ 0 & 0 & D-E+\omega & \Omega/2 & 0 & 0 & & & \\ \hline 0 & 0 & \Omega/2 & D+E & 0 & 0 & 0 & 0 & \Omega/2 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \Omega/2 & 0 & 0 & 0 & 0 & D-E & \Omega/2 & 0 & 0 \\ \hline & & & 0 & 0 & \Omega/2 & D+E-\omega & 0 & 0 \\ & & & 0 & 0 & 0 & 0 & -\omega & 0 \\ & & & \Omega/2 & 0 & 0 & 0 & 0 & D-E-\omega \\ \hline \end{array} & & & & & & & & \\ & & & & & & & & \ddots \end{bmatrix} \begin{matrix} |+, m = +1\rangle \\ |0, m = +1\rangle \\ |-, m = +1\rangle \\ |+, m = 0\rangle \\ |0, m = 0\rangle \\ |-, m = 0\rangle \\ |+, m = -1\rangle \\ |0, m = -1\rangle \\ |-, m = -1\rangle \end{matrix}. \quad (\text{B.15})$$

We can facilitate the analytical treatment of this system by applying the rotating wave approximation, which amounts to only retaining the terms necessary to yield a block diagonal

matrix

$$H_{\text{F,RWA}} = \left[\begin{array}{cccccc} \ddots & & & & & \\ & D+E+\omega & & & & \\ & & \omega & & & \\ & & & \begin{array}{cc} D-E+\omega & \Omega/2 \\ \Omega/2 & D+E \end{array} & & \\ & & & & 0 & \\ & & & & & \begin{array}{cc} D-E & \Omega/2 \\ \Omega/2 & D+E-\omega \end{array} \\ & & & & & & -\omega \\ & & & & & & & D-E-\omega \\ & & & & & & & & \ddots \end{array} \right]. \quad (\text{B.16})$$

We can isolate a 3×3 matrix corresponding to the upper Autler-Townes branch

$$H_{\text{F,RWA}}^{\text{U}} = \left[\begin{array}{cccccc} \ddots & & & & & \\ & D+E+\omega & & & & \\ & & \omega & & & \\ & & & \begin{array}{ccc} D-E+\omega & \Omega/2 & 0 \\ \Omega/2 & D+E & 0 \\ 0 & 0 & 0 \end{array} & & \\ & & & & \begin{array}{cc} D-E & \Omega/2 \\ \Omega/2 & D+E-\omega \end{array} & \\ & & & & & -\omega \\ & & & & & & D-E-\omega \\ & & & & & & & \ddots \end{array} \right], \quad (\text{B.17})$$

as well as one for the lower branch

$$H_{\text{F,RWA}}^{\text{L}} = \begin{bmatrix} \ddots & & & & & & & & \\ & D+E+\omega & & & & & & & \\ & & \omega & & & & & & \\ & & & D-E+\omega & \Omega/2 & & & & \\ & & & \Omega/2 & D+E & & & & \\ & & & & & \boxed{\begin{matrix} 0 & 0 & 0 \\ 0 & D-E & \Omega/2 \\ 0 & \Omega/2 & D+E-\omega \end{matrix}} & & & \\ & & & & & & -\omega & & \\ & & & & & & & D-E-\omega & \\ & & & & & & & & \ddots \end{bmatrix}. \quad (\text{B.18})$$

For the analysis of Autler-Townes phenomena, we treat the drive to be on resonance, $\omega = 2E$, so that

$$H_{\text{F,RWA}}^{\text{U}} = \begin{bmatrix} D+E & \frac{\Omega}{2} & 0 \\ \frac{\Omega}{2} & D+E & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad (\text{B.19})$$

$$H_{\text{F,RWA}}^{\text{L}} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & D-E & \frac{\Omega}{2} \\ 0 & \frac{\Omega}{2} & D-E \end{bmatrix}. \quad (\text{B.20})$$

Diagonalizing these systems yields

$$H_{\text{F, RWA}}^{\text{U}} = \begin{bmatrix} D + E + \frac{\Omega}{2} & 0 & 0 \\ 0 & D + E - \frac{\Omega}{2} & 0 \\ 0 & 0 & 0 \end{bmatrix} \begin{matrix} | +1, u \rangle = \frac{| -, m=+1 \rangle + | +, m=0 \rangle}{\sqrt{2}} \\ | -1, u \rangle = \frac{| -, m=+1 \rangle - | +, m=0 \rangle}{\sqrt{2}} \\ | 0, m=0 \rangle \end{matrix}, \quad (\text{B.21})$$

$$H_{\text{F, RWA}}^{\text{L}} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & D - E + \frac{\Omega}{2} & 0 \\ 0 & 0 & D - E - \frac{\Omega}{2} \end{bmatrix} \begin{matrix} | 0, m=0 \rangle \\ | +1, l \rangle = \frac{| -, m=0 \rangle + | +, m=-1 \rangle}{\sqrt{2}} \\ | -1, l \rangle = \frac{| -, m=0 \rangle - | +, m=-1 \rangle}{\sqrt{2}} \end{matrix}, \quad (\text{B.22})$$

where the energy eigenvalues and corresponding eigenstates of the dressed spin system are shown. In order to understand our observation of only the transitions $|0, m=0\rangle \leftrightarrow |\pm 1, u\rangle$ being allowed, we recall that our magnetic drive is primarily aligned along S_x and correspondingly minimized along S_y . The result is $B_x \gg B_y$ for any applied MW-frequency magnetic drive. Hence, we consider S_x as the only practically addressable dipole between $|0\rangle$ and any other state. Combined with the fact that at $\mathbf{B} = 0$ and $\Omega = 0$, H supports spin selection rules with linear polarization, we conclude that the only spin transition that can be practically realized from $|0\rangle$ is $|0\rangle \leftrightarrow |+\rangle$ in our experimental setup.

Furthermore, in the limit of weak driving $\Omega \ll E$ where multiphoton resonances are suppressed, only states with the same Floquet mode can couple to each other,

$$\langle m = m' | m = m'' \rangle \approx \delta_{m', m''}, \quad (\text{B.23})$$

where $\delta_{m, n}$ is the Kronecker delta function. With these considerations, we can determine

that the allowed spin transitions in the dressed basis from $|0, m = 0\rangle$ are

$$|\langle +1, u | S_x | 0, m = 0 \rangle|^2 \neq 0, \quad (\text{B.24})$$

$$|\langle -1, u | S_x | 0, m = 0 \rangle|^2 \neq 0, \quad (\text{B.25})$$

while the transitions to the lower branch vanish,

$$|\langle +1, l | S_x | 0, m = 0 \rangle|^2 = 0, \quad (\text{B.26})$$

$$|\langle -1, l | S_x | 0, m = 0 \rangle|^2 = 0. \quad (\text{B.27})$$

Furthermore, this allows us to confirm that a $\Delta m_s = \pm 2$ spin transition can be actuated by resonant electrical driving,

$$\left| \langle -1, u | (S_x^2 - S_y^2) | +1, u \rangle \right|^2 \neq 0. \quad (\text{B.28})$$

Throughout here and chapter 6, we denote $|\pm 1, u\rangle$ as $|\pm 1\rangle$ and $|0, m = 0\rangle$ as $|0\rangle$ for simplicity.

B.7 Dressing drive non-adiabatic transition

Our readout scheme, detailed in section B.8, relies on disabling the dressing drive non-adiabatically and retaining the state through the transition using the sudden approximation. This allows us to perform a mapping from the dressed basis to the undriven basis in a near-deterministic way, without resorting to time-consuming adiabatic change-of-basis procedures. In order to confirm that the sudden approximation is valid, we assess the probability of a spin state $|\psi\rangle$ changing during the non-adiabatic transition. Such an event is described by a projection operator onto the vector space orthogonal to the initial state

$$Q = 1 - |\psi\rangle\langle\psi|. \quad (\text{B.29})$$

Evaluating the expectation value of Q after the effect of the non-adiabatic transition unitary evolution operator U is equivalent to the probability of an adiabatic transition P_A

$$P_A = \langle \psi | U^\dagger Q U | \psi \rangle. \quad (\text{B.30})$$

It can be shown that P_A can be equivalently formulated as

$$P_A = \tau^2 (\Delta \bar{H})^2, \quad (\text{B.31})$$

where τ is the duration over which U occurs and $\Delta \bar{H}$ is the root mean square (RMS) deviation of the energy observable of the system before and after U . Thus, we must calculate the energy difference between an arbitrary state in the dressed basis and the same one in the bare basis.

Starting with an arbitrary state in the dressed basis

$$|\psi\rangle = \alpha |-1\rangle + \beta |+1\rangle, \quad (\text{B.32})$$

which can also be represented in the bare basis as

$$\begin{aligned} |\psi\rangle &= \frac{\alpha}{\sqrt{2}} (|+\rangle - |-\rangle) + \frac{\beta}{\sqrt{2}} (|+\rangle + |-\rangle) \\ &= \frac{\alpha + \beta}{\sqrt{2}} |+\rangle + \frac{\beta - \alpha}{\sqrt{2}} |-\rangle, \end{aligned} \quad (\text{B.33})$$

we can define $\delta = \frac{\alpha + \beta}{\sqrt{2}}$ and $\gamma = \frac{\beta - \alpha}{\sqrt{2}}$ and obtain

$$|\psi\rangle = \delta |+\rangle + \gamma |-\rangle. \quad (\text{B.34})$$

We then calculate the energy expectation value of the system given an arbitrary state with the dressing drive enabled and disabled, given by the system Hamiltonians H_e and H_d ,

respectively,

$$\langle \psi | H_e | \psi \rangle = \frac{\Omega}{2} (|\beta|^2 - |\alpha|^2) + D, \quad (\text{B.35})$$

$$\begin{aligned} \langle \psi | H_d | \psi \rangle &= |\delta|^2 (D + E) + |\gamma|^2 (D - E) \\ &= D + E (\alpha^* \beta + \beta^* \alpha). \end{aligned} \quad (\text{B.36})$$

The RMS energy deviation over the transition is

$$\begin{aligned} \Delta \bar{H} &= \sqrt{\frac{(\langle \psi | H_e | \psi \rangle - \langle \psi | H_d | \psi \rangle)^2 + (\langle \psi | H_d | \psi \rangle - \langle \psi | H_e | \psi \rangle)^2}{8}} \\ &= \frac{|\langle \psi | H_d | \psi \rangle - \langle \psi | H_e | \psi \rangle|}{2} \\ &= \left| \frac{E}{2} (\alpha^* \beta + \beta^* \alpha) - \frac{\Omega}{4} (|\beta|^2 - |\alpha|^2) \right|. \end{aligned} \quad (\text{B.37})$$

In the limit $\Omega \ll E$, the maximal RMS energy change is obtained for any superposition state where $\alpha^* \beta + \beta^* \alpha = 1$ and $|\beta|^2 - |\alpha|^2 = 0$, resulting in $\Delta \bar{H} \approx E/2$. Considering that the dressing drive modulation bandwidth is limited by the arbitrary waveform generator (AWG) output rise time, which order 1 nanosecond, we arrive on the worst-case probability of an adiabatic transition P_A ,

$$P_A = \tau^2 (\Delta \bar{H})^2 = (1 \text{ ns})^2 (2\pi \times 9 \text{ MHz})^2 \approx 0.003. \quad (\text{B.38})$$

The probability of a non-adiabatic transition P_{NA} is at least the negation of this quantity; $P_{NA} = 1 - P_A \approx 0.997$. Thus, modulation of the dressing drive at nanosecond timescales is non-adiabatic and almost entirely retains the original state vector.

B.8 Readout of dressed qubit basis

We accomplish optical readout of the kh divacancy ground-state spin by exciting its spin-selective optical interface with a tunable narrow-line laser. We use the A' optical branch,

which is about 277.9 THz higher in energy than the ground-state triplet ${}^3A_2$ for low-strain kh divacancies. The fine structure of the A' optical branch consists of three highly spin selective optical transitions, each corresponding to one of the ground-state spin eigenstates at $\mathbf{B} = 0$. Given the large (~ 860 MHz) separation in energy between the $|0\rangle \leftrightarrow |A'_0\rangle$ and $|+\rangle \leftrightarrow |A'_+\rangle$ optical transitions, combined with the ease of manipulating the $|0\rangle \leftrightarrow |+\rangle$ spin transition with linearly polarized MWs generated by the drive wire, we typically choose to work in a ground-state undriven qubit basis consisting of $|0\rangle$ and $|+\rangle$ and perform readout by exciting the cycling transition $|0\rangle \leftrightarrow |A'_0\rangle$ to determine the population in $|0\rangle$.

In the case of readout for a qubit basis consisting of $|0\rangle$ and one of the driven $|\pm 1\rangle$ states, directly exciting $|0\rangle \leftrightarrow |A'_0\rangle$ will also off-resonantly excite $|-\rangle \leftrightarrow |A'_-\rangle$ given the small (~ 130 MHz) energy difference between the two transitions, significantly reducing the contrast between the bright state $|0\rangle$ and the dark states $|\pm 1\rangle$ and potentially suppressing readout signal-to-noise by up to a factor of 3. We resolve this problem by performing a contrast recovery sequence which combines spin rotations with a non-adiabatic shutoff of the dressing drive described in section B.7. We can demonstrate this protocol on an arbitrary state in the dressed $\{|0\rangle, |+\rangle\}$ basis

$$|\psi\rangle = \alpha |0\rangle + \beta |+\rangle. \quad (\text{B.39})$$

By turning off the dressing drive non-adiabatically, the state remains unchanged. In the undriven basis, the state is then expressed as

$$|\psi'\rangle = \alpha |0\rangle + \frac{\beta}{\sqrt{2}} (|+\rangle + |-\rangle). \quad (\text{B.40})$$

A $-\pi/2$ rotation about S_z then produces

$$|\psi'\rangle = \alpha |0\rangle + \beta |+\rangle, \quad (\text{B.41})$$

at which point excitation of $|0\rangle \leftrightarrow |A'_0\rangle$ will yield a high-contrast, low-background measurement of $|\alpha|^2$. A similar sequence can be used when working in the dressed $\{|0\rangle, |-1\rangle\}$ basis, namely by switching to a $+\pi/2$ rotation about S_z after non-adiabatic shutoff of the dressing drive.

When working in the driven $\{|+1\rangle, |-1\rangle\}$ qubit basis, both basis states have components of $|+\rangle$ as well as $|-\rangle$. Since neither $|+\rangle \leftrightarrow |A'_+\rangle$ nor $|-\rangle \leftrightarrow |A'_-\rangle$ are highly cycling transitions, we continue to use $|0\rangle \leftrightarrow |A'_0\rangle$ as the readout optical transition. However, since $|\langle 0|\pm 1\rangle|^2 = 0$ and both states would yield the same photoluminescence, little to no readout contrast will be obtained solely through exciting $|0\rangle \leftrightarrow |A'_0\rangle$. Hence, we can adapt the aforementioned contrast recovery sequence to this basis by considering an initial arbitrary state

$$|\psi\rangle = \alpha |-1\rangle + \beta |+1\rangle. \quad (\text{B.42})$$

We then perform a π rotation using the $|0\rangle \leftrightarrow |-1\rangle$ transition

$$|\psi\rangle = \alpha |0\rangle + \beta |+1\rangle, \quad (\text{B.43})$$

at which point the aforementioned contrast recovery sequence can be performed.

B.9 Energy dispersion from B_z noise contributions

We begin by considering the driven Hamiltonian with only fluctuations along B_z

$$H = DS_z^2 + E(S_x^2 - S_y^2) + \gamma B_z S_z + \Omega \cos(\omega t) S_z. \quad (\text{B.44})$$

We set $\gamma = 1$ for simplicity in this derivation. Following the Floquet analysis procedure outlined in section B.6, we obtain the Fourier components of this Hamiltonian

$$H_0 = \begin{bmatrix} D + B_z & 0 & E \\ 0 & 0 & 0 \\ E & 0 & D - B_z \end{bmatrix}, \quad (\text{B.45})$$

$$H_{\pm 1} = \begin{bmatrix} \Omega/2 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -\Omega/2 \end{bmatrix}. \quad (\text{B.46})$$

We define $G^2 = B_z^2 + E^2$. Then, H_0 can be diagonalized using U

$$U = \begin{bmatrix} \frac{B_z - G}{\sqrt{(B_z - G)^2 + E^2}} & 0 & \frac{B_z + G}{\sqrt{(B_z + G)^2 + E^2}} \\ 0 & 1 & 0 \\ \frac{E}{\sqrt{(B_z - G)^2 + E^2}} & 0 & \frac{E}{\sqrt{(B_z + G)^2 + E^2}} \end{bmatrix}, \quad (\text{B.47})$$

$$\begin{aligned} H'_0 &= U^\dagger H_0 U \\ &= \begin{bmatrix} D + G & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & D - G \end{bmatrix}. \end{aligned} \quad (\text{B.48})$$

$H_{\pm 1}$ can also be rotated into this basis

$$\begin{aligned}
H'_{\pm 1} &= U^\dagger H_{\pm 1} U \\
&= \begin{bmatrix} \frac{\Omega B_z}{2G} & 0 & -\frac{\Omega E}{2G} \\ 0 & 0 & 0 \\ -\frac{\Omega E}{2G} & 0 & -\frac{\Omega B_z}{2G} \end{bmatrix} \\
&\approx \begin{bmatrix} 0 & 0 & -\frac{\Omega E}{2G} \\ 0 & 0 & 0 \\ -\frac{\Omega E}{2G} & 0 & 0 \end{bmatrix}, \tag{B.49}
\end{aligned}$$

where we arrived on the approximation for $H'_{\pm 1}$ by assuming $G \gg B_z$. We then obtain the Floquet matrix

$$H_F = \left[\begin{array}{c} \ddots \\ \begin{array}{|c|c|c|c|c|} \hline \begin{array}{ccc} D+G+\omega & 0 & 0 \\ 0 & \omega & 0 \\ 0 & 0 & D-G+\omega \end{array} & \begin{array}{ccc} 0 & 0 & -\frac{\Omega E}{2G} \\ 0 & 0 & 0 \\ -\frac{\Omega E}{2G} & 0 & 0 \end{array} & & & \\ \hline \begin{array}{ccc} 0 & 0 & -\frac{\Omega E}{2G} \\ 0 & 0 & 0 \\ -\frac{\Omega E}{2G} & 0 & 0 \end{array} & \begin{array}{ccc} D+G & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & D-G \end{array} & \begin{array}{ccc} 0 & 0 & -\frac{\Omega E}{2G} \\ 0 & 0 & 0 \\ -\frac{\Omega E}{2G} & 0 & 0 \end{array} & & \\ \hline & \begin{array}{ccc} 0 & 0 & -\frac{\Omega E}{2G} \\ 0 & 0 & 0 \\ -\frac{\Omega E}{2G} & 0 & 0 \end{array} & \begin{array}{ccc} D+G-\omega & 0 & 0 \\ 0 & -\omega & 0 \\ 0 & 0 & D-G-\omega \end{array} & & \\ \hline \end{array} \right] \begin{array}{l} |+, m=+1\rangle \\ |0, m=+1\rangle \\ |-, m=+1\rangle \\ |+, m=0\rangle \\ |0, m=0\rangle \\ |-, m=0\rangle \\ |+, m=-1\rangle \\ |0, m=-1\rangle \\ |-, m=-1\rangle \end{array} \quad . \tag{B.50}$$

Then, by applying the rotating wave approximation, we construct a block diagonal matrix

$$H_{\text{F,RWA}} = \begin{bmatrix} \ddots & & & & & & \\ & D + G + \omega & & & & & \\ & & \omega & & & & \\ & & & \begin{bmatrix} D - G + \omega & -\frac{\Omega E}{2G} \\ -\frac{\Omega E}{2G} & D + G \end{bmatrix} & & & \\ & & & & 0 & & \\ & & & & & \begin{bmatrix} D - G & -\frac{\Omega E}{2G} \\ -\frac{\Omega E}{2G} & D + G - \omega \end{bmatrix} & \\ & & & & & & -\omega \\ & & & & & & & D - G - \omega \\ & & & & & & & & \ddots \end{bmatrix}. \quad (\text{B.51})$$

allowing us to extract the familiar upper and lower branch matrices

$$H_{\text{F,RWA}}^{\text{U}} = \begin{bmatrix} D - G + \omega & -\frac{\Omega E}{2G} & 0 \\ -\frac{\Omega E}{2G} & D + G & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad (\text{B.52})$$

$$H_{\text{F,RWA}}^{\text{L}} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & D - G & -\frac{\Omega E}{2G} \\ 0 & -\frac{\Omega E}{2G} & D + G - \omega \end{bmatrix}. \quad (\text{B.53})$$

Since we have shown that only the upper Autler-Townes branch is experimentally accessible in section B.6, we will only address the diagonalization of the upper branch matrix

$$\begin{aligned} H_{\text{F,RWA}}^{\text{U}} &= \begin{bmatrix} D - G + \frac{\omega}{2} & -\frac{\Omega E}{2G} & 0 \\ -\frac{\Omega E}{2G} & D + G - \frac{\omega}{2} & 0 \\ 0 & 0 & -\frac{\omega}{2} \end{bmatrix} + \frac{\omega}{2} \mathbb{I} \\ &= \begin{bmatrix} D - \Delta & -\frac{\Omega E}{2G} & 0 \\ -\frac{\Omega E}{2G} & D + \Delta & 0 \\ 0 & 0 & -\frac{\omega}{2} \end{bmatrix} + \frac{\omega}{2} \mathbb{I}, \end{aligned} \quad (\text{B.54})$$

where we have defined $\Delta = G - \frac{\omega}{2}$ as the detuning of the dressing drive from the natural resonance. We can then extract the eigenvalues of this system

$$\omega_1 = -\omega/2, \quad (\text{B.55})$$

$$\omega_2 = D - \sqrt{\left(\frac{\Omega E}{2G}\right)^2 + \Delta^2}, \quad (\text{B.56})$$

$$\omega_3 = D + \sqrt{\left(\frac{\Omega E}{2G}\right)^2 + \Delta^2}. \quad (\text{B.57})$$

For the purposes of understanding the effect of B_z noise contributions, we will assume no drive detuning is present; $\omega = 2E$. The energy difference of the qubit encoded in the decoherence-protected subspace is given by

$$\begin{aligned} |\omega_2 - \omega_3| &= 2\sqrt{\left(\frac{\Omega E}{2G}\right)^2 + \Delta^2} \\ &= 2\sqrt{\left(\frac{\Omega E}{2\sqrt{B_z^2 + E^2}}\right)^2 + \left(\sqrt{B_z^2 + E^2} - E\right)^2} \\ &= 2E \sqrt{\left(\frac{\Omega}{2E\sqrt{\left(\frac{B_z}{E}\right)^2 + 1}}\right)^2 + \left(\sqrt{\left(\frac{B_z}{E}\right)^2 + 1} - 1\right)^2}. \end{aligned} \quad (\text{B.58})$$

A Maclaurin series expansion with respect to B_z/E yields

$$|\omega_2 - \omega_3| \approx \Omega - \frac{\Omega}{2} \left(\frac{B_z}{E}\right)^2 + \left[\frac{3\Omega}{8} + \frac{E^2}{2\Omega}\right] \left(\frac{B_z}{E}\right)^4 + O\left[\left(\frac{B_z}{E}\right)^6\right], \quad (\text{B.59})$$

where we can see that the dispersion relation has quadratic and quartic dependences on B_z/E . From this, we can arrive on a few conclusions:

1. In the limit $\gamma |B_z| < \Omega$, the dispersion relation takes on a primarily quartic dependence.

This provides the highly non-linear suppression of magnetic noise in the regime of small perturbations relative to the dressing drive Rabi frequency.

2. The transverse zero-field splitting magnitude, E , is directly responsible for the reduction of the effect of z -axis magnetic field fluctuations. Thus, greater E is universally beneficial for the efficacy of the decoherence-protected subspace with respect to suppressing magnetic field noise.
3. The dressing drive Rabi frequency, Ω , must be balanced in a way so that the zeroth order energy level separation is large enough, while not overly amplifying the quadratic and quartic contributions of z -axis magnetic field fluctuations. In this case, a larger magnitude of E will also allow for a larger range of Ω to be accessible.
4. The range of fields that the decoherence-protected subspace can efficiently suppress is approximated by the two nonzero roots of the sum of quadratic and quartic terms

$$-\frac{\Omega}{2} \left(\frac{B_z}{E} \right)^2 + \left[\frac{3\Omega}{8} + \frac{E^2}{2\Omega} \right] \left(\frac{B_z}{E} \right)^4 = 0, \quad (\text{B.60})$$

which are approximately of order Ω in the limit $\Omega \ll E$. Thus, the dressing drive Rabi frequency can be tuned to allow the decoherence-protected subspace to suppress a smaller or larger range of magnetic field fluctuations, depending on the environmental parameters.

In our demonstration of this coherence protection protocol, we have chosen $\Omega/(2\pi) = 350 \text{ kHz}$, which is very close to the estimated Zeeman contribution of $\gamma |B_z| / (2\pi) = 364 \text{ kHz}$ derived in section B.11. From our analysis here, larger dressing drive Rabi frequency magnitudes would invariably induce higher spin energy inhomogeneity magnitudes within the range of magnetic field fluctuations, while a smaller a Rabi frequency would fail to protect the system from the complete range of magnetic field fluctuations, creating a more linear dispersion relation at the edges of the magnetic field fluctuation range.

B.10 Energy dispersion from B_x noise contributions

We begin again by considering the driven Hamiltonian, now with only fluctuations along B_x

$$H = DS_z^2 + E(S_x^2 - S_y^2) + \gamma B_x S_x + \Omega \cos(\omega t) S_z, \quad (\text{B.61})$$

We again set $\gamma = 1$ for simplicity in this derivation. Immediately, we see that the system is rotationally invariant along the z -axis, in that a change of basis

$$\begin{aligned} S_x &\rightarrow S_y, \\ S_y &\rightarrow -S_x, \\ B_x &\rightarrow B_y, \end{aligned}$$

leaves the Hamiltonian unaffected up to a sign change. Thus, we will consider the derivation for magnetic field applied along the x -axis to be representative of the effect of magnetic fields applied along any axis orthogonal to the z -axis.

Following the same procedure for determining B_z contributions, we obtain the Fourier components of the Hamiltonian in the basis diagonal to H_0 . We define $F = D + E$ and $G^2 = 2B_x^2 + F^2$,

$$H_0 = \begin{bmatrix} D & B_x/\sqrt{2} & E \\ B_x/\sqrt{2} & 0 & B_x/\sqrt{2} \\ E & B_x/\sqrt{2} & D \end{bmatrix}, \quad (\text{B.62})$$

$$H_{\pm 1} = \begin{bmatrix} \Omega/2 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -\Omega/2 \end{bmatrix}, \quad (\text{B.63})$$

$$U = \begin{bmatrix} \frac{\sqrt{2}B_x}{\sqrt{4B_x^2+(F-G)^2}} & \frac{\sqrt{2}B_x}{\sqrt{4B_x^2+(F+G)^2}} & \frac{1}{\sqrt{2}} \\ -\frac{F-G}{\sqrt{4B_x^2+(F-G)^2}} & -\frac{F+G}{\sqrt{4B_x^2+(F+G)^2}} & 0 \\ \frac{\sqrt{2}B_x}{\sqrt{4B_x^2+(F-G)^2}} & \frac{\sqrt{2}B_x}{\sqrt{4B_x^2+(F+G)^2}} & -\frac{1}{\sqrt{2}} \end{bmatrix}, \quad (\text{B.64})$$

and then apply U to H_0 and $H_{\pm 1}$,

$$H'_0 = \begin{bmatrix} \frac{F+G}{2} & 0 & 0 \\ 0 & \frac{F-G}{2} & 0 \\ 0 & 0 & F-2E \end{bmatrix}, \quad (\text{B.65})$$

$$H'_{\pm 1} = \begin{bmatrix} 0 & 0 & -\frac{B_x\Omega}{\sqrt{4B_x^2-(F-G)^2}} \\ 0 & 0 & -\frac{B_x\Omega}{\sqrt{4B_x^2+(F+G)^2}} \\ -\frac{B_x\Omega}{\sqrt{4B_x^2-(F-G)^2}} & -\frac{B_x\Omega}{\sqrt{4B_x^2+(F+G)^2}} & 0 \end{bmatrix}$$

$$\approx \begin{bmatrix} 0 & 0 & -\frac{B_x\Omega}{\sqrt{4B_x^2-(F-G)^2}} \\ 0 & 0 & 0 \\ -\frac{B_x\Omega}{\sqrt{4B_x^2-(F-G)^2}} & 0 & 0 \end{bmatrix}, \quad (\text{B.66})$$

where in the last line we consider $F+G \gg B_x$ to obtain the approximation for $H'_{\pm 1}$. By

defining $\Omega' (B_x) = -\frac{B_x \Omega}{\sqrt{B_x^2 - (F-G)^2}}$, the full Floquet matrix is then

$$H_F = \left[\begin{array}{c} \ddots \\ \begin{array}{|c|c|c|c|c|c|} \hline \begin{array}{ccc} \frac{F+G}{2} + \omega & 0 & 0 \\ 0 & \frac{F-G}{2} + \omega & 0 \\ 0 & 0 & F - 2E + \omega \end{array} & \begin{array}{ccc} 0 & 0 & \Omega' \\ 0 & 0 & 0 \\ \Omega' & 0 & 0 \end{array} & & & \\ \hline \begin{array}{ccc} 0 & 0 & \Omega' \\ 0 & 0 & 0 \\ \Omega' & 0 & 0 \end{array} & \begin{array}{ccc} \frac{F+G}{2} & 0 & 0 \\ 0 & \frac{F-G}{2} & 0 \\ 0 & 0 & F - 2E \end{array} & \begin{array}{ccc} 0 & 0 & \Omega' \\ 0 & 0 & 0 \\ \Omega' & 0 & 0 \end{array} & & & \\ \hline & \begin{array}{ccc} 0 & 0 & \Omega' \\ 0 & 0 & 0 \\ \Omega' & 0 & 0 \end{array} & \begin{array}{ccc} \frac{F+G}{2} - \omega & 0 & 0 \\ 0 & \frac{F-G}{2} - \omega & 0 \\ 0 & 0 & F - 2E - \omega \end{array} & & & \\ \hline \end{array} \right] \begin{array}{l} |+, m = +1\rangle \\ |0, m = +1\rangle \\ |-, m = +1\rangle \\ |+, m = 0\rangle \\ |0, m = 0\rangle \\ |-, m = 0\rangle \\ |+, m = -1\rangle \\ |0, m = -1\rangle \\ |-, m = -1\rangle \end{array} \quad . \quad (B.67)$$

We implement the rotating wave approximation by reducing H_F down to a block diagonal matrix

$$H_{F,RWA} = \left[\begin{array}{c} \ddots \\ \begin{array}{|c|c|c|c|c|c|} \hline \frac{F+G}{2} + \omega & & & & & \\ \hline & \frac{F-G}{2} + \omega & & & & \\ \hline & & \begin{array}{|c|c|} \hline F - 2E + \omega & \Omega' \\ \hline \Omega' & \frac{F+G}{2} \end{array} & & & \\ \hline & & & \frac{F-G}{2} & & \\ \hline & & & & \begin{array}{|c|c|} \hline F - 2E & \Omega' \\ \hline \Omega' & \frac{F+G}{2} - \omega \end{array} & \\ \hline & & & & & \frac{F-G}{2} - \omega \\ \hline & & & & & & F - 2E - \omega \\ \hline \end{array} \right] \quad . \quad (B.68)$$

We isolate the sub-matrix corresponding to the upper Autler-Townes branch

$$\begin{aligned}
H_{\text{F,RWA}}^{\text{U}} &= \begin{bmatrix} \ddots & & & & & & & & \\ & \frac{F+G}{2} + \omega & & & & & & & \\ & & \frac{F-G}{2} + \omega & & & & & & \\ & & & \boxed{\begin{matrix} F-2E+\omega & \Omega' & 0 \\ \Omega' & \frac{F+G}{2} & 0 \\ 0 & 0 & \frac{F-G}{2} \end{matrix}} & & & & & \\ & & & & F-2E & \Omega' & & & \\ & & & & \Omega' & \frac{F+G}{2} - \omega & & & \\ & & & & & & \frac{F-G}{2} - \omega & & \\ & & & & & & & F-2E - \omega & \\ & & & & & & & & \ddots \end{bmatrix} \\
&= \begin{bmatrix} F-2E + \frac{\omega}{2} & \Omega' & 0 \\ \Omega' & \frac{F+G}{2} - \frac{\omega}{2} & 0 \\ 0 & 0 & \frac{F-G}{2} - \frac{\omega}{2} \end{bmatrix} + \frac{\omega}{2} \mathbb{I}. \tag{B.69}
\end{aligned}$$

Using the resonance condition $\omega = 2E$

$$H_{\text{F,RWA}}^{\text{U}} = \begin{bmatrix} F-E & \Omega' & 0 \\ \Omega' & \frac{F+G}{2} - E & 0 \\ 0 & 0 & \frac{F-G}{2} - E \end{bmatrix} + E \mathbb{I}, \tag{B.70}$$

the eigenvalues of this system are

$$\omega_1 = \frac{F-G}{2}, \tag{B.71}$$

$$\omega_2 = \left(\frac{3F+G}{4} \right) - \sqrt{\frac{(F-G)^2}{16} + \Omega'^2}, \tag{B.72}$$

$$\omega_3 = \left(\frac{3F+G}{4} \right) + \sqrt{\frac{(F-G)^2}{16} + \Omega'^2}. \tag{B.73}$$

We can then determine the energy difference in the decoherence-protected subspace

$$\begin{aligned}
|\omega_3 - \omega_2| &= 2\sqrt{\frac{(F - G)^2}{16} + \Omega'^2} \\
&= 2\sqrt{\frac{(F - G)^2}{16} + \frac{B_x^2 \Omega^2}{4B_x^2 - (F - G)^2}}.
\end{aligned} \tag{B.74}$$

A Maclaurin series expansion with respect to B_x/F yields

$$|\omega_3 - \omega_2| \approx \Omega - \frac{\Omega}{2} \left(\frac{B_x}{F}\right)^2 + \frac{1}{2} \left(\frac{F^2}{\Omega} + \frac{11\Omega}{4}\right) \left(\frac{B_x}{F}\right)^4 + O\left[\left(\frac{B_x}{F}\right)^6\right], \tag{B.75}$$

where we again see similar quadratic and quartic dependences on B_x as we did for the expansion in B_z , but now with a scale factor of F . In our system, F is more than one order of magnitude larger than E , which accounts for the considerable decrease in sensitivity along the x -axis. We can thus conclude

1. Either a large axial or transverse zero-field splitting is necessary for reduction of the effects from transverse magnetic field fluctuations. From this expression, B_x is always suppressed by the effects of the sum of both zero-field splittings.
2. The dressing drive Rabi frequency, Ω , once again must be chosen so that the energy level separation is sufficiently large, while minimizing multiplicative contributions to the quadratic and quartic terms in the expansion.
3. A range of magnetic fields where the inhomogeneity suppression is most effective can be derived through a similar method as detailed in the conclusion of section B.9.

B.11 Estimation of magnetic field fluctuation magnitude from nuclear spin bath

Using a Ramsey free evolution sequence, we measure the undriven T_2^* at $B_z = 1.2$ mT for the kh divacancy used in this study to be $1.35(2)$ μ s. Additionally, we observe no beat patterns in the Ramsey free evolution, allowing us to conclude that the free evolution decay is primarily caused by interactions with a weakly coupled nuclear spin bath. From this assumption, we estimate the upper bound of the magnetic field fluctuations of the bath by relating T_2^* to the magnitude of energy inhomogeneity of the spin sublevels caused by the Zeeman effect. From the uncertainty principle, we first convert T_2^* to an energy fluctuation $\Delta f = \Delta\omega / (2\pi)$ using the following relation

$$\Delta f \approx \frac{1}{2T_2^*}. \quad (\text{B.76})$$

We then obtain the fluctuation intensity of the magnetic field, σ_B , by scaling the energy fluctuation magnitude by the electron gyromagnetic ratio γ :

$$\sigma_B = \frac{\Delta f}{2\gamma} = \frac{1}{4\gamma T_2^*}. \quad (\text{B.77})$$

Using our measured value of T_2^* at $B_z = 1.2$ mT, we estimate the z -axis magnetic field fluctuations to be $2\sigma_B \approx 13$ μ T. At $\mathbf{B} = 0$, where the nuclear spins in the bath are no longer preferentially aligned to a field axis, we assume the magnitude of fluctuations remains the same but becomes an isotropic effect. Thus, for the following sections, we consider 13 μ T as the magnetic fluctuation intensity along the z -axis, as well as in the plane orthogonal to the z -axis.

B.12 Simplified model of spin energy inhomogeneity from nuclear spin bath fluctuations

In the context of the work presented in chapter 6, we approximate the nuclear spin bath as a slowly fluctuating classical magnetic field, with an estimated amplitude derived from the procedure in section B.11. The fluctuation speed is considered to be much slower than any measurement timescale, so the magnetic field is quasi-static during the free evolution of the spin. Effectively, the spin then samples the distribution of magnetic fields from the bath, and inhomogeneous dephasing is the result of integrating all the contributions from the distribution. While this approximation may not hold when the measurement timescale approaches the characteristic timescale of spin bath fluctuations, this simplification allows us to derive a straightforward and predictive model for the protection effects of the decoherence-protected subspace on the spin.

Formally, we model the inhomogeneity of the spin levels by first considering the energy dispersion function of the spin qubit levels with respect to a particular axis i of the magnetic field noise

$$L(B_i) = \omega_1(B_i) - \omega_2(B_i), \quad (\text{B.78})$$

where $\omega_n(B_i)$ is the n th spin energy level's value as a function of magnetic field B_i along axis i . Any inhomogeneity in B_i will also correspond to inhomogeneity in L , leading to a finite dephasing time. To further quantify this, we aim to obtain a response function R relating inhomogeneity in B_i to L . R then provides the instantaneous inhomogeneity of the spin as it is being perturbed by the bath. We obtain R by differentiating L with respect to B_i

$$\begin{aligned} R(B_i) &= \frac{\partial L(B_i)}{\partial B_i} \\ &= \frac{\partial \omega_1(B_i)}{\partial B_i} - \frac{\partial \omega_2(B_i)}{\partial B_i}. \end{aligned} \quad (\text{B.79})$$

For an electron spin primarily under the influence of the Zeeman effect, we obtain a linear

response function $R(B_i) = 1$. For more complex dispersion functions, R is generally dependent on B . We then obtain the Gaussian weighting of R by considering the magnetic field fluctuations as a zero-mean Gaussian distribution

$$\mathcal{N}(B_i|0, \sigma_B) = \frac{1}{\sigma_B \sqrt{2\pi}} \exp \left[-\frac{B_i^2}{2\sigma_B^2} \right]. \quad (\text{B.80})$$

The weighted response function, R' , is then

$$R'(B_i) = R(B_i) \mathcal{N}(B_i|0, \sigma_B) \quad (\text{B.81})$$

By taking the root mean square (RMS) statistic of this quantity, we obtain an approximation of the inhomogeneous broadening from the magnetic field perturbation

$$\delta\omega = \sqrt{\int_{-\infty}^{+\infty} R'(B'_i)^2 dB'_i}. \quad (\text{B.82})$$

In order to apply this model to our system, we must first normalize the inhomogeneity to the electron spin experiencing purely a linear Zeeman effect. This can be done by defining $R(B_i) = 1$ and integrating

$$\begin{aligned} \delta\omega_Z &= \sqrt{\int_{-\infty}^{+\infty} \mathcal{N}(B'_i)^2 dB'_i} \\ &= \frac{1}{\sqrt{2\sigma_B\sqrt{\pi}}}. \end{aligned} \quad (\text{B.83})$$

For the first case, we examine the expected improvements from operating at the undriven clock transition at $\mathbf{B} = 0$. The energy dispersion L between $|0\rangle$ and $|+\rangle$ is

$$L(B_z) = \sqrt{B_z^2 + E^2} + D. \quad (\text{B.84})$$

The response function R is then

$$R(B_z) = \frac{B_z}{\sqrt{B_z^2 + E^2}}. \quad (\text{B.85})$$

The RMS statistic is then

$$\begin{aligned} \delta\omega_{\text{CT}} &= \sqrt{\int_{-\infty}^{+\infty} R'(B'_i)^2 dB'_i} \\ &\approx \frac{\delta\omega_{\text{Z}}}{138}, \end{aligned} \quad (\text{B.86})$$

which predicts that the inhomogeneity at the clock transition is approximately two orders of magnitude lower than at high fields where the Zeeman effect contributes linearly to the energy dispersion. This agrees well with our observation of T_2^* increasing by about two orders of magnitude when changing the operating field from $B_z = 1.2$ mT to $\mathbf{B} = 0$. We note that any discrepancy arising between the predicted and observed ratios may be explained by residual electric field fluctuations that are expected to perturb the spin to first order near zero magnetic field and are not captured by this model.

Next, we address the spin when prepared in the decoherence-protected subspace. From the main text, we expect that magnetic fluctuations along the z -axis are a primary limiting contribution to spin dephasing in the decoherence-protected subspace. Thus, we can derive the response function from the analytical energy dispersion for B_z obtained in section B.9

$$R(B_z) = \frac{B_z \left(4 - \frac{4E}{\sqrt{B_z^2 + E^2}} - \frac{E^2 \Omega^2}{(B_z^2 + E^2)^2} \right)}{2 \sqrt{\left(E - \sqrt{B_z^2 + E^2} \right)^2 + \frac{E^2 \Omega^2}{4(B_z^2 + E^2)}}}. \quad (\text{B.87})$$

The RMS statistic evaluated for the same σ_B is then

$$\begin{aligned}\delta\omega_{\text{DPS}} &= \sqrt{\int_{-\infty}^{+\infty} R' (B'_i)^2 dB'_i} \\ &\approx \frac{\delta\omega_Z}{14200}.\end{aligned}\tag{B.88}$$

Our measured ratio of T_2^* between the electron spin under primarily the Zeeman effect to the spin in the decoherence-protected subspace is about 16600. Thus, this model correctly predicts, to within unity, the expected inhomogeneity improvement and the corresponding increase in T_2^* .

We emphasize that while this model closely predicts our measured ratios of spin inhomogeneity, it should be strictly treated as a phenomenological approach and does not completely capture the dynamics of the nuclear spin bath. In particular, time-correlation of the bath and its inherently quantum nature play a significant role in the dephasing of single quantum states, especially when the spin evolution time approaches the spin-spin coupling timescale of the bath (order 10 to 100 ms). Here, we have instead opted to treat inhomogeneity in time as an inhomogeneity over an effective ensemble of identical quantum systems, each experiencing a distribution of static magnetic fields.

Furthermore, this model is only applicable for quasi-static noise relative to the spin evolution, which plays a major role in determining T_2^* . Spin coherence time under dynamical decoupling T_2 , however, depends on the spin's response to time-dependent noise during free evolution. Since we have neglected time dependence of the noise in this model, we cannot properly predict the expected T_2 coherence times of the spin in the decoherence-protected subspace. For that, a proper treatment would require cluster expansion approaches that consider dynamical interactions between the electron spin and the bath and within the bath itself.

B.13 Contributions from electric and temperature fluctuations

Electric field and temperature fluctuations couple to the divacancy spin system by affecting the axial and transverse zero-field splitting magnitudes. In particular, electric field noise is typically generated whenever optical excitation occurs, as photoactive impurities in the sample undergo charge state fluctuations. Considering this, the model developed in section B.12 is suitable for analysis here, as it can be assumed that minimal electric field fluctuations occur during spin evolution. Thus, we estimate the electric field noise by utilizing a similar argument as one made in section B.11 in order to obtain the magnetic field noise.

At $\mathbf{B} = 0$, the undriven kh divacancy is particularly robust to magnetic field noise but experiences first-order sensitivity to fluctuations in E from electric field noise. Thus we use the measured T_2^* at those conditions as an approximate metric of spin energy inhomogeneity caused by electric field fluctuations

$$\Delta f \approx \frac{1}{2T_2^*}. \quad (\text{B.89})$$

Using the measured value of $T_2^* = 163 \mu\text{s}$, we find the spin energy inhomogeneity stemming from electric field perturbations to be about 3 kHz. By considering the higher sensitivity of E to electric fields when compared to D , we attribute the entirety of this inhomogeneity to fluctuations in E .

We then enter the driven basis to examine the effects of electric noise on the decoherence-protected subspace. We start by obtaining the energy dispersion with respect to drive detuning. We revisit the result calculated in section B.6 without imposing the resonance condition $\omega = 2E$

$$H_{\text{F,RWA}}^{\text{U}} = \begin{bmatrix} D - \Delta & \Omega/2 & 0 \\ \Omega/2 & D + \Delta & 0 \\ 0 & 0 & -\omega/2 \end{bmatrix} + \frac{\omega}{2}\mathbb{I}, \quad (\text{B.90})$$

where we have defined $\Delta = E - \frac{\omega}{2}$ as the detuning of the dressing drive. The resulting

energy levels are

$$\omega_1 = -\frac{\omega}{2}, \quad (\text{B.91})$$

$$\omega_2 = D - \sqrt{\left(\frac{\Omega}{2}\right)^2 + \Delta^2}, \quad (\text{B.92})$$

$$\omega_3 = D + \sqrt{\left(\frac{\Omega}{2}\right)^2 + \Delta^2}. \quad (\text{B.93})$$

The energy difference in the decoherence-protected subspace is again given by the magnitude of the difference of the upper two energy levels

$$|\omega_3 - \omega_2| = 2\sqrt{\left(\frac{\Omega}{2}\right)^2 + \Delta^2}. \quad (\text{B.94})$$

For the purposes of this analysis, we treat the dressing drive as fixed at $\Omega/(2\pi) = 350$ kHz, with no fluctuations. We also treat the frequency noise of the dressing drive as negligible. We immediately see that D does not affect the energy difference in this subspace. The fluctuations of E manifest directly in Δ , $\delta\Delta = \delta E$. Thus, we obtain the response function R , now as a function of Δ

$$R(\Delta) = \frac{2\Delta}{\sqrt{\left(\frac{\Omega}{2}\right)^2 + \Delta^2}}, \quad (\text{B.95})$$

while the electric field fluctuations are still described as a Gaussian distribution, where we use $\sigma_\Delta \approx \Delta f/2 = 1.5$ kHz to describe the electric field fluctuation magnitude

$$\mathcal{N}(\Delta|0, \sigma_\Delta) = \frac{1}{\sigma_\Delta \sqrt{2\pi}} \exp\left[-\frac{\Delta^2}{2\sigma_\Delta^2}\right]. \quad (\text{B.96})$$

Evaluating the relative inhomogeneity yields

$$\begin{aligned}
\frac{\delta\omega_{\text{ZF}}}{\delta\omega_{\text{DPS}}} &= \frac{\sqrt{\int_{-\infty}^{+\infty} \mathcal{N}(\Delta')^2 d\Delta'}}{\sqrt{\int_{-\infty}^{+\infty} R(\Delta')^2 \mathcal{N}(\Delta')^2 dE'}} \\
&= \frac{\sqrt{\int_{-\infty}^{+\infty} \mathcal{N}(E' - E)^2 d\Delta'}}{\sqrt{\int_{-\infty}^{+\infty} R(E' - E)^2 \mathcal{N}(E' - E)^2 dE'}} \\
&\approx 83,
\end{aligned} \tag{B.97}$$

suggesting that the decoherence-protected subspace produces an improvement in protection from electric field noise by almost two orders of magnitude. Further reduction to electric field noise can be achieved by suppressing the source of electric field fluctuations through charge depletion techniques, which has previously been leveraged to observe near-transform-limited linewidths in the electrically sensitive optical spectrum of single divacancies.

Temperature fluctuations are heavily suppressed by the natural insensitivity of D and E in the cryogenic regime, even without the decoherence-protected subspace. However, the improvement in protection from fluctuations in D and E applies equally, regardless of the underlying source of fluctuations. This allows us to conclude that we have achieved a two order of magnitude improvement in thermal protection, on top of the intrinsic suppression at cryogenic temperatures.

B.14 Numerical master equation simulations

We obtain the exact dynamics of the dressed spin system by using a numerical master equation simulation. We implement the system Hamiltonian defined in sections B.9 and B.10, while also adding in a probe field applied along S_x

$$H = DS_z^2 + E(S_x^2 - S_y^2) + \gamma(B_x S_x + B_z S_z) + \Omega \cos(\omega t) S_z + \Omega_p \cos(\omega_p t) S_x, \quad (\text{B.98})$$

where Ω_p is the probe Rabi frequency and ω_p is the probe drive frequency. In order to best approximate experimental conditions, the probe Rabi frequency is chosen as $\Omega_p/(2\pi) = 20$ kHz, while the dressing drive Rabi frequency is chosen to be $\Omega/(2\pi) = 350$ kHz. The system is initialized into $|0\rangle$ and evolved over 25 μs , so that when the probe pulse is on resonance the population in $|0\rangle$ is fully transferred to $|+1\rangle$ or $|-1\rangle$ at the end of the unitary evolution. Both the dressing and probe fields are enabled for the full duration of the simulation. We then sweep the value of B_x (Fig. 6.3C) or B_z (Fig. 6.3D) while varying the detuning $\Delta = \omega - (D + E)$ of the probe field. The remaining population of $|0\rangle$ at the end of the simulation is then recorded.

B.15 Generalizability of the decoherence-protected subspace

Having considered the primarily magnetic fluctuation contributions along all spatial axes coupling through S_x , S_y , S_z , as well as a large subset of fluctuations coupling through product operators (thermal and electric contributions to S_z^2 and $S_x^2 - S_y^2$), we believe it is clear that implementing this decoherence-protected subspace is not reliant on any particular detail or property specific to the $k\hbar$ divacancy or the form of the environmental noise. Instead, its effectiveness is based on the large magnitudes of the matrix elements in the spin Hamiltonian corresponding to S_z^2 and $S_x^2 - S_y^2$. Thus, an arbitrary system that is able to implement these elements in an equivalent fashion, while also capable of driving an equivalent S_z continuous tone, should expect to see coherence protection through a similar mechanism as demonstrated

here. As we have suggested through our analysis of the protection mechanism, the efficacy of the decoherence-protected subspace is strongly dependent on the magnitude of these Hamiltonian matrix elements. Significantly lower values of D and E (or their equivalent in another system) will generally correspond to greater spin inhomogeneity than what has been observed here, as the spin is protected from a smaller range of environmental fluctuation magnitudes. Conversely, we expect this protocol in systems with higher values of E (or its equivalent) to be more effective at suppressing decoherence, up to the T_1 energy relaxation limit.

B.16 Spin resonance feedback

From the quasienergy spectrum of the $|\pm 1\rangle$ basis, we find that Ω , the dressing drive Rabi frequency, provides the quantization energy of the basis states to first order. Thus we can expect fluctuations in the dressing drive Rabi frequency to affect the resonance frequency of the $|+1\rangle \leftrightarrow |-1\rangle$ transition, which also directly affects the spin energy inhomogeneity in this basis. Experimentally, these fluctuations are generated by thermal fluctuations of the AWG oscillator and amplifier stages, which for the AWG model used in the work described in chapter 6 (Zurich Instruments HDAWG8) is specified to be approximately 100 ppm/ $^{\circ}\text{C}$. Our laboratory long-term temperature stability is within 0.5 $^{\circ}\text{C}$, leading to approximately 50 ppm fluctuations of Ω . At $\Omega/(2\pi) = 350$ kHz, this corresponds to fluctuations in the Rabi frequency of $\delta\Omega/(2\pi)$. We have established the spin energy inhomogeneity under magnetic and electric field fluctuations to be within this same range, which indicates that the dressing drive amplitude inhomogeneity may also play a role in limiting the observed spin inhomogeneity, if left unchecked.

By observing the fluctuations of the feedback error signal detailed in sections B.2 and B.16, we conclude that the dressing drive amplitude drifts on the timescale of many minutes. This allows us to effectively implement a feedback protocol given the quasi-static nature of the drift relative to the coherence time of the spin. In the main text, we choose to use a

feedback period of one minute as a compromise between frequently evaluating the error signal and maximizing the duty cycle of experimental acquisition. Depending on the application, the error signal may need to be evaluated more or less frequently than this period in order to properly mitigate the effect of quasi-static drifts. Improvements to photon collection efficiency will also reduce the integration time necessary to reliably evaluate the error signal.

We note that this feedback mechanism, while primarily purposed to counteract drifts from the dressing drive amplitude, also reduces the effect of slow inhomogeneous drifts from local variations in electric and magnetic field. Noise on timescales much shorter than the error signal evaluation time (10 s in this work) but longer than the spin coherence time will average to zero and cannot be corrected for. However, slower fluctuations, possibly from optically induced charge migration and drift in the applied magnetic field, can manifest as a change in the error signal. Since the action of our feedback scheme involves applying corrections to the spin rotation drive detuning, our implementation is also robust to other detectable sources of inhomogeneity.

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