Quantum Control of the Tin-Vacancy Spin Qubit in Diamond

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Group-IV color centers in diamond are a promising light-matter interface for quantum networking devices. The negatively charged tin-vacancy center (SnV) is particularly interesting, as its large spin-orbit coupling offers strong protection against phonon dephasing and robust cyclicality of its optical transitions toward spin-photon-entanglement schemes. Here, we demonstrate multiaxis coherent control of the SnV spin qubit via an all-optical stimulated Raman drive between the ground and excited states. We use coherent population trapping and optically driven electronic spin resonance to confirm coherent access to the qubit at 1.7 K and obtain spin Rabi oscillations at a rate of $\Omega/2\pi = 19.0(1)$ MHz. All-optical Ramsey interferometry reveals a spin dephasing time of $T_2 = 1.3(3)$ $\mu$s, and four-pulse dynamical decoupling already extends the spin-coherence time to $T_2 = 0.30(8)$ ms. Combined with transform-limited photons and integration into photonic nanostructures, our results make the SnV a competitive spin-photon building block for quantum networks.

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I. INTRODUCTION

A light-matter quantum interface combines deterministic and coherent generation of single photons with a long-lived matter qubit [1–4]. This combination constitutes a foundational building block for quantum networking systems that exploit far-field radiation to generate remote entanglement and near-field interactions to realize nonlinear photonic gates [5–7]. Candidate systems include isolated atoms [8–11] and solid-state spins in the optical domain [12–14], as well as superconducting quantum circuits in the microwave regime [15]. An efficient quantum emitter is correspondingly well suited for light-assisted manipulation of its internal degrees of freedom [16–18]. The optical domain offers the critical advantage of wireless control fields which can be confined spatially to an optical wavelength, allowing for selective control of individual systems on that length scale, and high-speed control arising from a high electric field density coupling to typically large electric dipole moments [19–22].

Diamond stands out as a particularly promising solid-state host for scalable fabrication of quantum light-matter interfaces [23], enabling all-optical control [24–27]. Within this material platform, the nitrogen-vacancy center (NV) has been used for pioneering quantum networking tasks owing to its excellent spin coherence [28–32]. Scaling up faces the challenge of improving its optical performance with tailored nanostructures [33], which remains difficult owing to the NV’s sensitivity to nearby surfaces as a result of its permanent electric dipole moment [34,35]. In contrast, the group-IV color centers [36–47] are naturally compatible with photonic nanostructures owing to their inversion symmetry [36,48] in which collection efficiencies exceeding 90% [49] and large cavity-coupling efficiencies of more than 95% have been recently demonstrated [50–52]. Of these, the negatively charged silicon-vacancy center (SiV) is the most studied, with demonstrations of coherent control of its ground state by microwave [53], all-optical [54], and acoustic [55] drive techniques. At millikelvin temperatures, where dephasing due to single-phonon scattering between orbital levels is suppressed, coherence...
times up to 13 ms [56] allow for more mature demonstrations of entanglement [49, 57, 58]. Building on these achievements, the recently reported tin-vacancy center (SnV) [44–46, 59] shares the desirable optical properties of SiV—large Debye-Waller factor of approximately 0.6 [60] with transform-limited photons [46]—and provides the additional advantages of (1) a long spin lifetime of 10 ms at 3.25 K (extrapolated to >1 s at 1.7 K) [46] and (2) optical cyclicity in the presence of an off-axis magnetic field or strain, which can allow for simultaneous high-quality single-shot readout and efficient coupling to nuclear spins. These advantages stem from a large spin-orbit coupling, which suppresses decoherence due to single-phonon scattering in the ground state and establishes a common quantization axis between the ground and excited states, providing robust spin cyclicity. Conversely, the strong spin-orbit coupling also gives rise to orbital-forbidden spin transitions, which has limited microwave-based spin control [46] and may necessitate advanced microwave line engineering to achieve fast, coherent control of the SnV spin qubit.

In this paper, we demonstrate all-optical multiaxis coherent control of the SnV spin qubit by driving its efficient and coherent optical transitions with microwave-modulated laser fields. We demonstrate the flexibility of the all-optical approach by implementing coherent population trapping, optical Rabi driving, Ramsey interferometry, and dynamical decoupling of the SnV spin qubit. We further measure an electron-nuclear hyperfine-coupling strength of 42.6(4) MHz for a spin-active Sn isotope. These results confirm the promise of SnV as a competitive next-generation light-matter quantum interface.

II. COHERENT OPTICAL ACCESS TO THE SnV SPIN QUBIT

The lilac frames in Fig. 1(a) illustrate the formation of the energy-level structure for the negatively charged SnV under the spin-orbit, Zeeman, and hyperfine couplings (see Supplemental Material [61] Sec. 1). The strong spin-orbit coupling results in the ground- and excited-state manifolds having two orbital levels split by 850 and 3000 GHz, respectively [46], with a 484-THz (619-nm) optical transition energy between the manifolds (see Supplemental Material [61] Sec. 1). An external magnetic field lifts the degenerate spin-orbit states via the Zeeman effect. Our qubit is defined as the Zeeman-split electronic spin states in the lower orbital branch $|\downarrow\rangle$ and $|\uparrow\rangle$. Spin-orbit interaction sets spin quantization along the SnV crystallographic symmetry axis for both ground and excited states. However, this axis pinning is weaker in the ground-state

![Image](https://example.com/image.png)

**FIG. 1.** SnV and Raman drive. (a) Lilac panels: energy levels of the SnV split by spin-orbit coupling (left), Zeeman splitting (middle), and hyperfine splitting (right). Blue panel: qubit defined by the $|\downarrow\rangle$ and $|\uparrow\rangle$ electronic levels. An optical lambda scheme is defined by two optical fields $\omega_1$ and $\omega_2$ detuned by $\Delta$ from the excited state $|E\rangle$. The Raman drive frequency $\omega_R = \omega_1 - \omega_2$ is the energy offset between the two fields. (b) $|E\rangle$ population plotted in orange circles as a function of $\omega_R$ for $\Delta = 0$ and 200 mT magnetic field strength applied at 54.7° relative to the SnV symmetry axis. The pulse sequence consists of an approximately 10-µW green stabilization pulse for 50 µs, $B_2$ reset pulse for 30 µs, and Raman drive with $p = 40(4)$ nW for 2 µs. The solid curve is a three-level model describing coherent population trapping. (c) $|\uparrow\rangle$ population plotted in orange circles as a function of $\omega_R$ for $\Delta = 600$ MHz at 204-mT magnetic field strength. After the stabilize and reset pulses, the sequence includes an initialization pulse for 30 µs, a Raman drive with $p = 40(4)$ nW for 1 µs, and a readout pulse on $A_1$ for 30 µs. The solid curve is a two-Lorentzian fit resulting in an average linewidth of 900(200) kHz and spin resonances split by 42.6(4) MHz.
manifold, and its spin quantization axis can be perturbed by strain or magnetic field applied perpendicular to the SnV symmetry axis [37] with negligible impact on the excited-state manifold. This mismatch between the quantization axes of the ground and excited spin states allows for the spin-cycling transitions ($A_1$ and $B_2$) to achieve single-shot optical readout of the qubit, in tandem with the spin-flipping transitions ($A_2$ and $B_1$) to realize an optical lambda scheme (see Supplemental Material [61] Sec. I). The branching ratio between the spin-cycling and the spin-flipping relaxation rates $\eta \approx 80$ measured for the SnV center studied in this work (see Supplemental Material [61] Sec. II) is consistent with a moderate-strain perturbation of 186(3) GHz (see Supplemental Material [61] Sec. I).

The light blue frame in Fig. 1(a) highlights how we leverage the SnV optical transitions to realize an optical lambda scheme between the $|\downarrow\rangle$ and $|\uparrow\rangle$ qubit states ($|\downarrow\rangle$ and $|\uparrow\rangle$, respectively) and the excited state $|\Sigma\rangle$. This is achieved by simultaneously driving the $A_1$ and $A_2$ transitions with lasers at frequencies $\omega_1$ and $\omega_2$, respectively, detuned relative to one another by the Raman frequency $\omega_R = \omega_1 - \omega_2$. The Raman scheme is further detuned from the excited state by the single-photon detuning $\Delta$. For unpolarized light, the Rabi rate at which the spin is driven is then $\Omega = (1/\sqrt{\eta})(p/p_{sat})(\Gamma^2/4\Delta)$, where $p$ is the power in each of the optical fields driving $A_1$ and $A_2$, $p_{sat} \approx 5$ nW is our emitter’s saturation power for the spin-cycling transition, and $\Gamma/2\pi = 35$ MHz is the excited-state relaxation rate [62] (see Supplemental Material [61] Sec. III). Typical experimental values of order $p/p_{sat} = 10$ and $\Delta/2\pi = 300$ MHz, and a measured $\eta = 80(5)$ at 0.2-T magnetic field (see Supplemental Material [61] Sec. II) place the spin Rabi frequency in the MHz range, which comfortably exceeds the inhomogeneous dephasing rate $1/T_2^* \approx 1 \mu$s$^{-1}$ [46], as required for coherent spin control. Driving the spin optically also causes a detuning-dependent excited-state scattering rate $\Gamma_{OS} = (p/p_{sat}) \times (\Gamma^2/8\Delta^2)$ [62], which introduces a spin-relaxation rate $1/T_{1,OS} = \Gamma_{OS}/\eta$ and a spin-dephasing rate $1/T_{2,OS} = \Gamma_{OS}$ (see Supplemental Material [61] Sec. III). Maximizing the fidelity of a $\pi/2$ gate with respect to $\Delta$, we find an optimal balance when $\Gamma_{OS} = 1/T_2$ (see Supplemental Material [61] Sec. III).

Our SnV device consists of a nanopillar array fabricated into an Sn$^+$ ion-implanted diamond (see Supplemental Material [61] Sec. IV) and is cooled to 1.7 K in a magneto-optical cryostat (see Supplemental Material [61] Sec. V) [46]. Our all-optical measurement sequences include stabilization, reset, initialization, Raman drive, and readout pulses. The stabilize pulse uses a 532-nm laser to prevent blinking, which has been proposed to occur as a result of charge-state conversion of the SnV [60]. The initialize (reset) pulse consists of resonantly driving the $A_1$ ($B_2$) transition, which polarizes the SnV spin into a $|\uparrow\rangle$ ($|\downarrow\rangle$) state in a time $\eta/\Gamma$ (approximately equal to 1 $\mu$s) via relaxation through the weakly allowed spin-flipping transition $A_2$ ($B_1$), achieving up to 99% initialization fidelities (see Supplemental Material [61] Sec. II). The reset pulse polarizes the population into the $|\downarrow\rangle$ state immediately before the initialization pulse, such that the fluorescence intensity of the initialization pulse corresponds to approximately 100% of the population. The fluorescence intensity of the readout pulse resonant on $A_1$ and normalized to that of the initialization pulse then provides a direct measurement of the population in the $|\downarrow\rangle$ state. The Raman drive encompasses all coherent pulse combinations we use in this work relying on a stimulated Raman process. For the optical pulses within the Raman drive, $\omega_{1,2}$ are realized using the two sidebands generated by passing a single-frequency laser through a microwave (MW)-modulated electro-optic modulator. The MW-modulation frequency splits the sidebands by $\omega_R$, the MW-modulation amplitude determines the power $p$ in each of the $\omega_{1,2}$ sidebands, and the MW-modulation phase dictates the relative phase between $\omega_{1,2}$ sidebands, the phase of the Raman drive $\phi$.

Figure 1(b) displays a coherent-population-trapping (CPT) measurement as a first step to verify coherent optical access to the SnV spin qubit, where the $\omega_1$ and $\omega_2$ sidebands drive the spin-conserving and spin-flipping transitions, respectively, for $\Delta = 0$. The top panel shows the CPT pulse sequence, in which initialization, drive, and readout pulses are combined into one drive and measure step. The main panel presents the steady-state SnV fluorescence during the drive pulse as a function of $\omega_R$. We observe a broad feature, whose width is comparable to the excited-state linewidth $\Gamma$, accompanied by two narrower dips. A narrow dip in the SnV fluorescence spectrum (see Supplemental Material [61] Sec. VI) corresponds to the generation of a dark coherent superposition of the two ground states, $|1/\sqrt{\eta}\rangle|\downarrow\rangle - |\sqrt{\eta}\rangle|\uparrow\rangle$ (see Supplemental Material [61] Sec. VI) and is obtained when $\omega_R$ matches the spin-splitting frequency. In the presence of a spin-active Sn isotope ($I = 1/2$), the CPT resonance splits into two dips arising from two nuclear-spin-preserving transitions that are separated by the corresponding hyperfine-coupling rate. The CPT spectrum in Fig. 1(b) indicates that the electronic spin qubit of the single SnV color center we measure, confirmed via intensity-correlation measurements (see Supplemental Material [61] Sec. VII) is indeed coupled to a spin-1/2 nuclear spin. Fitting a theoretical model (black curve) to the CPT data, using a Lindbladian master-equation formalism (see Supplemental Material [61] Sec. VI) reveals a hyperfine-coupling strength of approximately 40 MHz commensurate with previous reports on other group-IV
color centers [40,53]. The depth of the two CPT resonances confirms that the coherences of the spin ground states and the optical transitions are sufficient to implement coherent optical drive [40].

Having identified the two spin resonances via CPT, we move to the far-detuned stimulated Raman regime $\Delta \gg \Gamma$ to suppress scattering from the excited state during the coherent drive sequence. The top panel of Fig. 1(c) shows the pulse sequence, where the initialization, drive, and readout pulses are now separate operations. The main panel of Fig. 1(c) presents the population recovery of the $|\downarrow\rangle$ state as a function of $\omega_R$. When $\omega_R$ matches one of the electronic spin resonances, population from the initial $|\uparrow\rangle$ state is transferred to the $|\downarrow\rangle$ state, resulting in a peak in the $|\downarrow\rangle$ population. We fit our hyperfine-split double-peaked spectrum with two independent Lorentzian line shapes (solid curve), thereby allowing for high-precision measurement of the hyperfine constant for this Sn isotope, $A = 42.6(4)$ MHz. In the remainder of this work, we optically address a single-electronic-spin transition $\omega_e$ [defined as the lower energy peak in Fig. 1(c)], which sets the stage for its coherent control.

### III. Multiaxis Coherent Control

We now demonstrate coherent spin control in the stimulated Raman regime. Figure 2(a) shows the Rabi oscillations of the population in the $|\downarrow\rangle$ state as we sweep the drive pulse duration $T$, with $\omega_R = \omega_p$. By fitting the data to a two-level model under a master-equation formalism (solid curve), we extract a Rabi rate of $\Omega/2\pi = 3.6(1)$ MHz (see Supplemental Material [61] Sec. III). Increased optical power straightforwardly yields Rabi rates of up to $\Omega/2\pi = 19.0(1)$ MHz (see Supplemental Material [61] Sec. VIII). This is an improvement of well over 3 orders of magnitude in spin Rabi frequency over direct microwave control realized thus far for SnVs [46] (see Supplemental Material [61] Sec. I). The inset shows the dependence of Rabi rate on power and detuning, with the expected linear dependence $\Omega \propto p/\Delta$. Our model further yields a total damping rate of $5.6(3) \mu s^{-1}$, as the sum of a depolarization ($T_1$ process) rate of $0.28(2) \mu s^{-1}$ and dephasing ($T_2$ process) rate of $5.3(3) \mu s^{-1}$, with the latter in good agreement with the expected scattering rate $\Gamma_{\text{OS}} = 3(1) \mu s^{-1}$ at a detuning of $\Delta/2\pi = 1200$ MHz. Finally, the model allows us to extract a $\pi/2$-gate fidelity of $90.9(7)$% (see Supplemental Material [61] Sec. III C). The same scattering-induced dephasing mechanism sets an upper limit on the fidelity of all subsequent measurements involving more complex pulses. While the pulse fidelity achieved here remains modest, operating at larger detuning with increased laser power places high-fidelity gates within reach (see Supplemental Material [61] Sec. III C).

Full qubit control requires coherent drive about an arbitrary axis. Our approach realizes this via the combination of the two-photon detuning, $\delta = \omega_R - \omega_e$, and $\phi$, the MW-controlled phase between the $\omega_{1,2}$ sidebands. The latter sets the control axis within the equatorial plane of the Bloch sphere, which is particularly relevant for implementing control sequences from the nuclear-magnetic-resonance toolbox [63,64]. We demonstrate this multiaxis control in Fig. 2(b) via a drive sequence comprising two $\pi/2$ pulses, with the first driving the spin about the $x$ axis of the Bloch sphere [40].
sphere ($\phi = 0$) and the second driving the spin about an axis rotated by an angle $\phi$ away from the $x$ axis. The population of the $|\downarrow\rangle$ state depends periodically on $\phi$ over the full $2\pi$ range, where the cumulative drive for the maximum (minimum) $|\downarrow\rangle$ population corresponds to an effective $\pi$ ($0$) pulse. The phase dependence of the population readout confirms our ability to choose the quantum-state rotation axis.

IV. MEASURING SnV SPIN COHERENCE

We use multiaxis coherent control to implement Ramsey interferometry in order to measure the inhomogeneous dephasing time $T_2^*$ of the SnV spin qubit. The top panel of Fig. 3(a) shows the corresponding pulse sequence comprised of two $\pi/2$ pulses separated by a time delay $\tau$. We further impose a periodic recovery of the Ramsey signal by varying the rotation angle $\phi$ for the second $\pi/2$ pulse as a function of $\tau$, such that $\phi = \tau \omega_S$ with the serrodyne frequency $\omega_S/2\pi = 5$ MHz. The main panel of Fig. 3(a) presents the dependence on $\tau$ and $\delta$ of the $|\downarrow\rangle$ population, which oscillates as a function of $\tau$ with a sum frequency given by $\omega_{\text{Ramsey}} = \omega_S + \delta + \Delta_{ac}$, where $\Delta_{ac}$ is the differential ac Stark shift. The latter term originates from the $|\downarrow\rangle$ state’s stronger coupling to the Raman fields, and is only present during the Raman drive, thus acting as an effective detuning between the free precession rate of the spin and that of the drive’s rotating frame (see Supplemental Material [61] Sec. IX). The period of the Ramsey fringes follows the expected $2\pi/\omega_{\text{Ramsey}}$ behavior.

Figure 3(b) is an example line cut of the $|\downarrow\rangle$ population as a function of $\tau$ for a fixed $\delta/2\pi = -1$ MHz. Fitting with the function $e^{-(\tau/T_2^*)^2} \sin(\omega_{\text{Ramsey}}\tau)$ yields $\omega_{\text{Ramsey}}/2\pi = 7.27(3)$ MHz, and hence, $\Delta_{ac}/2\pi = 3.3(5)$ MHz, comparable to the expected value (see Supplemental Material [61] Sec. VIII). We note that gate fidelity reduces the contrast in these measurements, but it does not affect the coherent spin precession between the two $\pi/2$ pulses. The Gaussian envelope $e^{-(\tau/T_2^*)^2}$ provides an estimate of the spin inhomogeneous dephasing time $T_2^*$. By applying our model to the data for each $\delta$ in Fig. 3(a), we extract an average $T_2^* = 1.3(3)$ $\mu$s. This is well within the range of expected inhomogeneous dephasing times limited by the naturally abundant $^{13}$C nuclear spins in diamond [46,56] and indicates that the SnV coherence is not phonon limited at 1.7 K.

V. IMPLEMENTING DYNAMICAL DECOUPLING

To prolong the SnV spin-qubit coherence beyond the timescale set by the low-frequency magnetic noise of $^{13}$C nuclei, we embed dynamical decopling protocols within our optical pulse sequence, as illustrated in Fig. 4(a). We implement two example protocols: Hahn echo [65] comprising a single rephasing $\pi$ pulse about the $x$ axis (orange frame) and CPMG-2, a Carr-Purcell-Meiboom-Gill sequence [66] comprising two rephasing $\pi$ pulses about the $y$ axis (purple frame). Sweeping the phase of the final $\pi/2$ pulse $\phi$ from 0 to $4\pi$ and the decoupling delay time $\tau$ produces the two-dimensional maps of the $|\downarrow\rangle$ population in Fig. 4(a). The phase-dependent modulation of the Hahn echo signal lasts for approximately 30 $\mu$s, while the CPMG-2 signal extends significantly longer. Figure 4(b) presents the extracted visibility for the $\phi$-dependent modulations for both Hahn echo and CPMG-2 protocols as a function of the decoupling delay time $\tau$. Fitting the Hahn echo visibility (orange circles) as a function of $\tau$ with a stretched exponential function $\exp\left[-(\tau/T_2^*)^n\right]$ reveals an extended coherence time $T_2^* = 28.3(6)$ $\mu$s. The exponent $n = 3.6(3)$ is consistent with a noise spectrum from a slowly evolving nuclear-spin bath in diamond [56,67]. Applying the same fit function to the CPMG-2 visibility...
to the function of the total decoupling time and phase shown as a function of the total decoupling time. Our gate fidelities can be prolonged further with straightforward improvements of CPMG-4 coherence time for SiV at 100 mK and could be prolonged further with single-shot readout, and nuclear-spin access all via the optical transitions. Our SnV spin-coherence time and Rabi rate can both be improved with stronger optical fields, which facilitates the suppression of optical scattering. Gate fidelities can be improved with technical refinements, and tailored pulse protocols are expected to result in 99.6% gate fidelity for π rotations [70]. An immediate next step toward realizing an efficient quantum memory is extending our all-optical approach to control the intrinsic Sn nuclear spin [71]. Further, integrating the SnV into photonic nanostructures [44,50,51,68] will increase the photon-collection efficiency, and in parallel can strengthen the optical Rabi drive. Such structures should therefore enable efficient coherent control of an electronic spin coupled to a nuclear quantum memory with single-shot readout, a key building block for quantum networks [5,6]. Finally, going beyond an efficient local-area quantum network necessitates operation in the telecommunication bands. The similarity of the SnV wavelength to that of the NV makes all progress to date on efficient frequency conversion of NV photons to telecommunications bands [72,73] readily applicable for the SnV as well.

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