

Transform-limited photons from a tin-vacancy spin in diamond

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Solid-state quantum emitters that couple coherent optical transitions to long-lived spin states are essential for quantum networks. Here we report on the spin and optical properties of single tin-vacancy (SnV) centers in diamond nanostructures. Through magneto-optical spectroscopy at 4 K, we verify the inversion-symmetric electronic structure of the SnV, identify spin-conserving and spin-flipping transitions, characterize transition linewidths, and measure electron spin lifetimes. We find that the optical transitions are consistent with the radiative lifetime limit and that the spin lifetimes are longer than for other inversion-symmetric color centers under similar conditions. These properties indicate that the SnV is a promising candidate for quantum optics and quantum networking applications.

A central goal of quantum information processing is the development of quantum networks consisting of stationary, long-lived matter qubits coupled to flying photonic qubits [1, 2], with applications in quantum computing, provably secure cryptography, and quantum-enhanced metrology [3]. Among matter qubits, quantum emitters in wide-bandgap semiconductors [4, 5] have emerged as leading systems as their coherent, spin-selective optical transitions act as an interface between quantum information stored in their spin degrees of freedom and emitted photons. While most work has so far focused on the nitrogen-vacancy (NV) center in diamond [6–8], its relatively poor optical properties, including a low percentage of emission into the coherent zero-phonon-line (ZPL) [9] and large spectral diffusion when located near surfaces [10, 11], have fueled the investigation of alternative emitters. These include the group-IV color centers in diamond [12], comprising the silicon-vacancy (SiV) [13–16], germanium-vacancy (GeV) [17, 18], and the recently observed lead-vacancy (PbV) [19] centers. These centers have a large fraction of emission into the ZPL and a crystallographic inversion symmetry that limits spectral diffusion and inhomogeneous broadening [20, 21]. Unlike the NV center, however, the electronic spin coherence of SiV and GeV centers is limited by phonon scattering to an upper-lying ground-state orbital [22, 23], requiring operation at dilution-refrigerator temperatures (~ 100 mK) [24, 25], or controllably induced strain [26] to achieve long coherence times.

The tin-vacancy (SnV) center in diamond [27, 28] is a group-IV color center that promises favorable optical properties and long spin coherence time at readily

achievable temperatures (liquid helium, ~ 4 K). DFT calculations predict that the SnV has the same symmetry as the SiV and GeV [9], while experimental measurement of a large ground-state orbital splitting indicates that single-phonon scattering, the dominant spin dephasing mechanism of SiV and GeV centers at liquid helium temperatures, should be suppressed significantly [27]. In this work, we report spectroscopic measurements that are consistent with the conjectured electronic structure of the SnV, demonstrate that its optical transitions are radiative lifetime-limited, show optical initialization and readout of its electronic spin, and, finally, demonstrate improved spin decay times compared to SiV and GeV centers in similar conditions. These single-emitter measurements indicate the strong potential of the SnV for quantum optics and quantum networking applications.

We perform cryogenic magneto-optical spectroscopy on single SnV centers created through Sn implantation in an ultra-pure diamond grown by chemical vapor deposition [29] to determine the electronic properties of the SnV. The SnV energy spectrum is analogous to those of other group IV centro-symmetric color centers, such as the SiV and GeV centers. Its center of symmetry, located at the impurity atom (Sn, Ge, Si), leads to the color center having no permanent electric dipole. This makes it insensitive to first-order fluctuations in electric field and thus favours spectral stability and emission into the zero-phonon line. This symmetry is also responsible for the presence of doubly degenerate ground and excited orbital states, as depicted in Fig. 1a, which are split by spin-orbit interaction leading to characteristic optical transitions consisting of two doublets. At 4 K, the upper branch of the excited state undergoes fast thermaliza-

tion to the lower branch, leading to only the transitions labeled γ and δ being visible under non-resonant excitation, as shown in Fig. 1b. Figure 1c displays the evolution of the optical spectrum of a single emitter when a magnetic field is applied along the [001] crystallographic direction of the diamond (corresponding to an angle of 54.7° with respect to the high symmetry axis of the SnV along the $\langle 111 \rangle$ directions). The visible transitions γ and δ each split into four lines, analogously to the SiV and GeV centers, which is consistent with the degeneracy of its expected spin $1/2$ being lifted.

Owing to the common symmetry of group-IV color centers, the optical emission spectra can be simulated using the same group-theoretical model developed for SiV [13]. This model includes the Jahn-Teller effect or inherent pre-strain (such effects have similar Hamiltonians), spin-orbit coupling, and the orbital and spin Zeeman effects. By adapting the values of the model parameters to fit the experimental spectra, we obtain the simulated spectra displayed on the right panel of Fig. 1c. From this simulation, we determine that the ~ 850 GHz ground-state splitting for unstrained emitters in the absence of magnetic field is almost entirely due to spin-orbit coupling (99%). Spin-orbit interaction accounts for only 80% of the ~ 3000 GHz excited state splitting, with the rest being due to the Jahn-Teller effect. These couplings impact the electronic spin state associated with energy levels: the spin has an effective quantization axis defined by the combination of the orientation of the external magnetic field and the spin-orbit coupling, which tends to align the spin along the SnV high symmetry axis. As a consequence, in the range of magnetic field strengths applied, the spin quantization axis is dominated by the spin-orbit coupling. However, since the relative strengths of the spin-orbit coupling, Jahn-Teller effect, and magnetic field differ between the ground and excited states, the orientation of the effective quantization axes in the ground and excited states differs slightly. This leads, as shown in Fig. 1c, to optical transitions mostly taking place between states with largely overlapping spin orientations (B2, A1, B4, A3), and weak transitions between states of nearly opposite spin orientations (B1, A2, B3, A4).

Strain, likely induced during implantation, affects the spectrum of the SnV in a similar way to that of the SiV: it alters the orbital components of the states and thus the spin quantization axes via spin-orbit coupling. As a consequence, optical selection rules are relaxed and emission into the weaker transitions is reinforced. We observe this in strained SnV spectra (Fig. 1d), where the observed intensity of the weaker transitions is an order of magnitude greater than in the unstrained case (Fig. 1c). The spectrum can be simulated accurately using the same parameters as for the unstrained case and adding the strain Hamiltonian developed in Ref [29–31]. These findings elucidate the photophysics of the SnV center and con-

firm its similarity with the previously described group-IV centers.

Following the determination of the SnV electronic structure, we investigate the coherent optical properties of individual SnV centers in nanofabricated pillars. Figure 2a shows a confocal scan of SnV centers in pillars with radius $R = 150$ nm at 4 K under non-resonant 532 nm excitation. After identifying a SnV center through non-resonant scans, we perform photoluminescence excitation spectroscopy (PLE) by scanning the frequency of a narrowband laser across the SnV γ optical transition and detecting fluorescence emission in the phonon sideband. Across several single SnV centers, we measure a mean linewidth (Lorentzian FWHM) of 57 ± 17 MHz [29], with some emitters displaying linewidths down to 30 ± 2 MHz (Fig. 2b). As a further demonstration of optical coherence, we measure the second-order photon correlation function $g^{(2)}$ for the emitter presented in Fig. 2b under resonant excitation and an additional continuous low-power non-resonant repump at 532 nm (Fig. 2c) [29]. The characteristic antibunching $g^{(2)}(0) = 0.09 \pm 0.03$ confirms single photon emission under resonant excitation. At a resonant optical power above saturation, we furthermore observe Rabi oscillations in the $g^{(2)}$ with a decay time of 5.9 ± 0.3 ns, indicating coherence between the driven levels on this timescale. Finally, we measure the fluorescence lifetime of this SnV center after short (\sim picosecond pulsed) 532 nm non-resonant excitation (Fig. 2d). After a short Gaussian feature associated with the direct response of our detectors to the initial pump pulse, we observe an exponential decay of fluorescence with a time constant $\tau = 4.5 \pm 0.2$ ns. Together with the transition linewidth measurement, this indicates that the best-case SnV optical transitions are consistent with the lifetime limit of $(2\pi\tau)^{-1} = 35 \pm 5$ MHz. The mean transition linewidth is within a factor of 1.5 of the transform limit. Lifetime-limited optical transitions demonstrate the high optical quality of the SnV and are essential for its use as an optically interfaced spin qubit.

To access and probe the expected spin degree of freedom of the unstrained SnV, we implement spin-selective resonant excitation in the presence of an external magnetic field. We apply a magnetic field of 9 T along the [001] crystallographic direction and excite the transition labeled A1 corresponding to a strong transition between levels of largely overlapping spin orientations, as depicted by an upward red arrow in Fig. 3a. We thus populate the Zeeman sublevel labeled A. The resulting fluorescence is shown in Fig. 3b, where only emission from transition A3 is visible. When exciting resonantly the other strong transition, B2, and thus populating the other Zeeman sublevel B in the same excited state orbital branch, only emission from transition B4 is visible. This dependence shows evidence of selection rules in the thermalization mechanism within the excited state [32] confirming that the SnV possesses a spin $1/2$ and demonstrating its op-

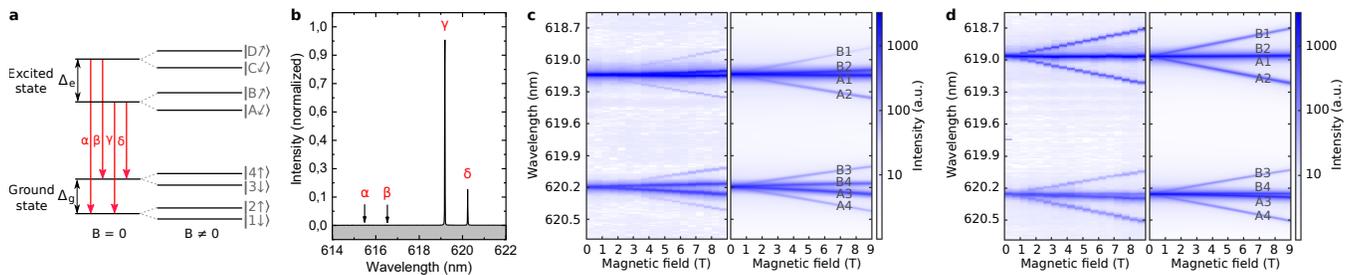


FIG. 1. SnV center electronic structure. (a) The SnV energy levels, in the absence of magnetic field, are composed of doubly-split ground and excited states with respective splittings Δ_g and Δ_e , resulting in four optical transitions (red arrows) labeled from α to δ . A magnetic field splits each level into two. The spin state expected from the SnV model (see main text) is indicated for each level with up and down gray arrows. The tilted arrows in the excited state represent the difference in spin quantization axis compared to the ground state. (b) Fluorescence spectrum of a single SnV center at 4 K without magnetic field. (c) Left panel: Magnetic field-dependent fluorescence spectrum of a single unstrained SnV center. Right panel: simulated magnetic field dependence based on the model developed for the SiV center [13]. The transitions are labeled according to the participating states. The simulated transition intensities were obtained from Fermi's golden rule for dipolar electric transitions and thus depend on the orbital and spin components of the initial and final states. (d) Magnetic field dependence of the fluorescence spectrum of a strained SnV center, both experimental (left panel) and simulated using the strain simulation developed for SiV in ref. [30, 31] (right panel).

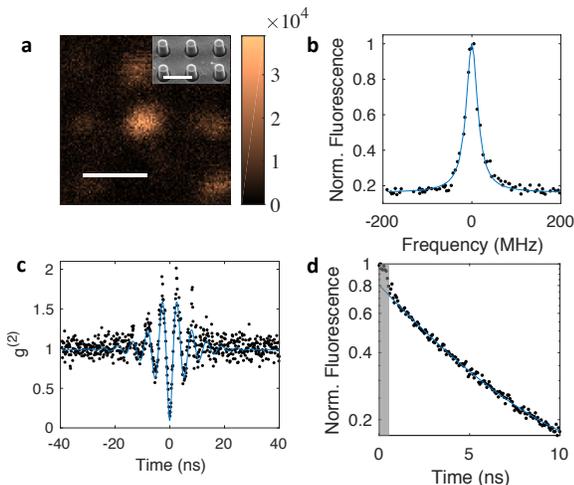


FIG. 2. SnV center optical coherence. (a) Confocal microscope image of SnV centers in nanopillars with radius $r = 150$ nm (inset, scanning electron micrograph). Scale bars are 1 μm . (b) Resonant PLE spectrum of a single SnV optical transition. Each point was integrated for 0.6 seconds. Blue line: Lorentzian fit with FWHM of 30 ± 2 MHz. (c) Second-order correlation function under resonant driving. (d) Fluorescence decay after non-resonant pulsed excitation at $t = 0$. The gray shaded area between 0 and 0.5 ns indicates the instrument response.

tical accessibility. As a second step, we tune the laser frequency into resonance with the weak transitions B1 and A2 between levels of differing spin states, as depicted in Fig. 3c. Figure 3d shows the fluorescence from transitions B4 and A3. The capacity to excite such transitions confirms relaxation of the optical selection rules

due to the different effective spin quantization axes between ground and excited states. This observation of spin selection rules also confirms the assignment of the experimentally measured transitions to the energy levels of the model.

With the spin selection rules determined, we measure optical spin polarization through time-resolved fluorescence. A PLE spectrum of a SnV center with ~ 0.2 T of applied field along the [001] axis is shown in Fig. 4a, where the lines correspond to the spin-conserving A1 and B2 transitions as labeled in Fig 3c. When applying a resonant pulse on transition A1, the observed fluorescence decays with time as shown in Figure 4b. This intensity decay $I(t) = \alpha P_1(t) + \beta P_2(t)$ indicates spin polarization as population P is pumped from state 1 (bright) to state 2 (dark). Taking $\beta/\alpha \sim 0$ and an initial thermal population $P_1(0) = 1 - P_2(0) = 0.5$, the observed contrast after a time $t_{\text{pol}} > 100 \mu\text{s}$ is $C = (I(0) - I(t_{\text{pol}}))/I(0) = 0.55$ which corresponds to a polarization of the spin with $P_2(t_{\text{pol}}) = (C + 1)/2 = 0.78$. Having polarized the spin, we measure its population decay time $T_1 = 57 \pm 12 \mu\text{s}$ from the ratio of fluorescence at the leading edge of the initial polarizing pulse $I(0)$ to that of a second readout pulse delayed by a dark interval of length τ . This decay time is about an order of magnitude longer than that of the SiV and GeV centers considering similar magnetic field orientation and temperature [17, 23, 25, 29, 33], highlighting the promise of the SnV as a spin qubit. Figure 4d shows that T_1 decreases with temperature, similarly to the SiV. This decrease follows the Bose-Einstein population of the phonon mode resonant with the orbital splitting of the SnV [29], indicating that single phonon-mediated excitation to the upper ground state orbital branch remains the dominant spin decay mecha-

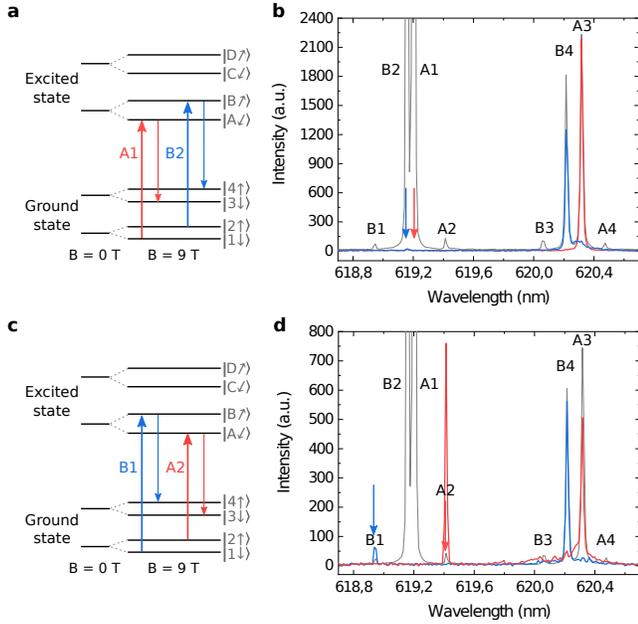


FIG. 3. Spin-selective transitions. (a) Energy levels of the SnV center under external magnetic field depicting resonant excitation (upward arrows) and subsequent fluorescence (downward arrows) for transitions A1 (red) and B2 (blue) between levels of nearly overlapping spin states. (b) Experimental spectra at 9 T for non-resonant excitation (light gray curve) and resonant excitation of A1 (red curve) and B2 (blue curve). The excitation wavelengths are indicated by arrows of the corresponding colors. (c) Energy levels of the SnV center illustrating resonant excitation (upward arrows) and subsequent fluorescence (downward arrows) for transitions B1 (blue) and A2 (red) between levels of nearly opposite spin states. (d) Experimental spectra at 9 T for non-resonant excitation (light gray curve) and resonant excitation of B1 (blue curve) and A2 (red). The excitation wavelengths are indicated by arrows of the corresponding colors. Leakage from the laser is visible in both cases at the excitation wavelength.

nism [22, 23].

These results show a straightforward path for improvement. Spin polarization can be increased by addressing a "spin-flipping" transition which has a significantly higher branching ratio to the opposite spin state in the ground state. As for the spin decay time, it has been shown for SiV and GeV centers that aligning the magnetic field along the color center axis improves spin T_1 by several orders of magnitude [17, 24, 33]. A similarly aligned magnetic field is expected to improve T_1 for the SnV center towards the millisecond scale at 4 K. Furthermore, theoretical predictions based on experiments performed with SiV indicate that cooling SnV to 2 K is equivalent to cooling SiV to 100 mK in terms on phonon absorption [27, 29]. Since the spin coherence time is dominated by the same single-phonon absorption mechanism for group IV centers [17, 23], a similar improvement is expected for SnV compared to SiV [27]. Alternatively, an

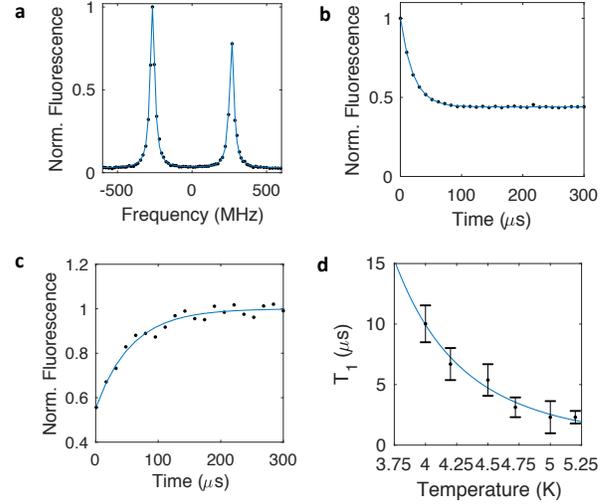


FIG. 4. Spin polarization of SnV centers. (a) PLE spectrum of a SnV center under an applied magnetic field of about 0.2 T along the [001] direction. The two spin-conserving transitions A1 and B2 are shown. (b) Spin polarization of an SnV center. Phonon sideband fluorescence decays as a function of time under resonant pumping of the A1 transition as level 2 is populated. (c) Polarization decay. The fluorescence of the A1 transition recovers as a function of time between the excitation and readout pulses, indicating repopulation of the ground spin level 1. Blue curve: fit to an exponential with $T_1 = 57 \mu\text{s}$. (d) Spin decay time T_1 measured at different temperatures (black dots) for another SnV center. The blue solid curve is a fit in $1/n_{th}(\Delta_g, T)$ [29], with n_{th} the Bose-Einstein distribution of phonons resonant with the ground state orbital splitting $\Delta_g = 850 \text{ GHz}$ and T the measured temperature with an offset of 1 K likely due to imperfect thermal anchoring or local heating of the sample.

increase in orbital splitting via strain engineering would allow to suppress phonon-mediated spin dephasing and decay at 4 K, as demonstrated for SiV [26, 31].

Our spin-resolved spectroscopy shows that the SnV center is a promising spin-photon interface that can operate without the need to cool down to millikelvin temperatures. The combination of transform-limited linewidths in nanostructures and a spin lifetime that is about an order of magnitude greater than the SiV and GeV under the same conditions are essential ingredients for the development of quantum information processing and quantum photonic devices based on the SnV. The addition of more elaborate nanophotonic structures for enhanced light-matter interaction [34] and the use of microwave pulses for spin manipulation [17, 24] will enhance the integrability of the SnV center as an optically interfaced spin qubit in a quantum network node.

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