All-Optical Initialization, Readout, and Coherent Preparation of Single Silicon-Vacancy Spins in Diamond

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The silicon-vacancy (SiV−) color center in diamond has attracted attention because of its unique optical properties. It exhibits spectral stability and indistinguishability that facilitate efficient generation of photons capable of demonstrating quantum interference. Here we show optical initialization and readout of electronic spin in a single SiV− center with a spin relaxation time of $T_1 = 2.4 \pm 0.2$ ms. Coherent population trapping (CPT) is used to demonstrate coherent preparation of dark superposition states with a spin coherence time of $T_2^\phi = 35 \pm 3$ ns. This is fundamentally limited by orbital relaxation, and an understanding of this process opens the way to extend coherence by engineering interactions with phonons. Hyperfine structure is observed in CPT measurements with the $^{29}$Si isotope which allows access to nuclear spin. These results establish the SiV− center as a solid-state spin-photon interface.

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Coherent quantum systems which efficiently couple long-lived quantum memories to optical photons are a key resource for realizing quantum networks [1]. Color centers in diamond [2] are attractive candidates owing to unique properties of diamond, which include optical transparency and a high lattice quality that allows spin to function as long-lived quantum memory [3]. The negative silicon-vacancy (SiV−) defect in diamond [4–6] has exceptional optical properties that facilitate efficient generation of indistinguishable photons from multiple distinct emitters [7]. Here we show optical initialization and readout of electronic spin in a single SiV− center with a spin relaxation time of $T_1 = 2.4 \pm 0.2$ ms. Two-photon resonance [8] is used to demonstrate coherent preparation of dark superposition states with a spin coherence time of $T_2^\phi = 35 \pm 3$ ns. This is shown to be limited by orbital relaxation that may be suppressed by engineering interactions with phonons. We present the first evidence of hyperfine interaction with a $^{29}$Si nuclear spin in SiV− which can potentially be used as a memory qubit [9].

Quantum information processing efforts in diamond have mainly focused on the nitrogen-vacancy (NV−) center because of its excellent spin properties at ambient conditions [10]. All-optical access to NV− spin is possible [11–14]; however, its large phonon sideband and spectral diffusion reduce coherent photon generation rates and limit the development of NV− quantum networks [15–17]. The main optical advantage provided by the SiV− center is that 70% of its fluorescence is concentrated in a sharp zero-phonon line (ZPL), making it ideal for single photon source applications [18,19]. It is spectroscopically stable at 737 nm, exhibits line widths limited by the excited state lifetime [20], and can be coupled to optical cavities [21,22]. Physically, the SiV− center consists of a single silicon atom replacing two carbon atoms in the diamond lattice, forming D4h symmetry as illustrated in Fig. 1(a) [4–6,23]. This geometry makes the SiV− center insensitive to small electric fields [7] and therefore adds low inhomogeneous broadening to the set of attractive optical properties. The electronic structure of the SiV− center consists of doubly degenerate $E$ ground and excited state orbitals which both have electronic spin-$1/2$ [4–6]. Optical signatures of the electron spin have been identified [24]; however, control of SiV− spin remains an outstanding challenge.

The technique used here to manipulate and measure SiV− spin involves resonant excitation with narrow-band lasers. A diamond plate with $\{111\}$ surfaces (see Ref. [25] for details of the synthesis) was cooled to around 5 K in a continuous flow cryostat, and imaged using a home-built confocal fluorescence microscope. Resonant excitation spectra of the four fine-structure transitions A–D [Fig. 1(b)] were measured for grown-in SiV− centers by scanning a high-resolution (100 kHz) resonant laser and recording sideband fluorescence.

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through a 750–810 nm band-pass filter. Mounting the diamond on a fixed magnet as shown in Fig. 1(c) lifted the spin degeneracy by Zeeman splitting, and excitation spectra were recorded for a SiV\(^{-}\) center oriented normal to the \{111\} sample surface. (d) Resonant excitation spectroscopy using a single laser reveals the two spin-conserving transitions D2 and D3 (green). Applying a second pump laser on the D2 (orange) or D3 (blue) transition makes the spin-flipping transitions D1 and D4 appear in the excitation spectrum. These optical transitions between the Zeeman-split levels are illustrated beneath the spectrum (bold indicates spin conserving).

Although optical transitions are spin conserving, any off-axis magnetic field results in a different quantization axis between the ground and the excited states because of the difference in spin-orbit couplings [5]. In our experiments, a residual off-axis field makes the spin-flipping transitions (D1 and D4) weakly allowed. These transitions therefore pump the spin into a dark state after a few optical cycles and are not visible in single laser scans. Figure 2(a) shows time-resolved fluorescence measurements where we utilized the D1 transition to optically pump the spin into a dark state and the cycling D2 transition to efficiently read out the resulting spin state. Laser pulses were produced using acousto-optical modulators, giving a rise or fall time of about 60 ns but a constant intensity over the duration of the pulse. The fluorescence produced by the D2 transition had a prominent leading edge when the interpulse delay was small, which is evidence of optical pumping causing spin initialization.

The decay of the leading-edge height \(h\) with increasing pulse separation \(\tau\) establishes the spin relaxation time in the aligned field at \(T_{\text{spin}}^1 = 2.4 \pm 0.2\) ms, as shown in Fig. 2(b).

For a SiV\(^{-}\) center misaligned about 20 degrees from the magnetic field, the spin relaxation time was measured to be \(T_{\text{spin}}^1 = 3.4\) ms, and a 70-degree misalignment reduced this to \(T_{\text{spin}}^1 \sim 60\) ns [25].}

FIG. 1 (color online). Optical access to spin levels of SiV\(^{-}\) in diamond. (a) A SiV\(^{-}\) center consists of a silicon atom between two vacant lattice sites and is aligned along a \(\langle 111\rangle\) crystal bond direction. (b) At zero field the ZPL consists of four transitions, which occur between spin-orbit branches of the doublet ground and excited states as illustrated below the spectrum. Transitions B and C appear weak since the SiV\(^{-}\) is viewed here along its symmetry axis. (c) Zeeman splitting of the spin-1/2 electronic states (\(\uparrow\) and \(\downarrow\) for simplicity) was achieved by mounting the diamond on a neodymium magnet. This produced a field around 4.5 kG closely aligned with the SiV\(^{-}\) centers oriented normal to the \{111\} sample surface. (d) Resonant excitation spectroscopy using a single laser reveals the two spin-conserving transitions D2 and D3 (green). Applying a second pump laser on the D2 (orange) or D3 (blue) transition makes the spin-flipping transitions D1 and D4 appear in the excitation spectrum. These optical transitions between the Zeeman-split levels are illustrated beneath the spectrum (bold indicates spin conserving).
FIG. 2 (color online). Optical initialization and readout of SiV\textsuperscript{−} spin. (a) A laser pulse resonant to transition D1 flips the spin without producing measurable fluorescence. The initialized spin state can be read out using a laser pulse on the cycling transition D2. The spin polarization signal appears as a leading-edge peak of the fluorescence that decays in about 500 ms. (b) The reduction of $P$ with increasing $\tau$ gives the spin relaxation time at $T_{1/2}^{\text{spin}} = 2.4 \pm 0.2$ ms. Interpreting the asymptotic limit $\alpha$ to correspond to thermal spin population suggests a spin initialization fidelity of $h_{\alpha=0}/2\alpha = 78\%$. (c) Similar pulsed measurements give the orbital relaxation time at 4.5 K as $T_{1/2}^{\text{orbital}} = 38 \pm 1$ ns. The orbital relaxation rate $1/T_{1/2}^{\text{orbital}}$ increased linearly with temperature.

The SiV\textsuperscript{−} ground states also have an orbital degeneracy which can introduce additional dynamics beyond spin $T_{1/2}^{\text{spin}}$. Orbital relaxation was isolated from any spin pumping/relaxation by measuring at zero magnetic field. Since the time scale was found to be fast compared to the acousto-optical modulation bandwidth, laser pulses of 80 ns (1 ns rise/fall time) were produced using an electro-optical modulator and applied resonant to transition D. Photon counts were recorded with timing resolution of 200 ps, and a leading-edge peak similar to that in Fig. 2(a) was observed. In this case the decay of the initial intensity corresponds to population being pumped to the other ground-state branch (decaying via transition C). Varying the dark interval between subsequent pulses revealed that the leading-edge peak recovered in an orbital relaxation time of only $T_{1/2}^{\text{orbital}} = 38 \pm 1$ ns at 4.5 K. This is much shorter than $T_{1/2}^{\text{spin}}$, which indicates that the orbital relaxation must be highly spin conserving, as expected for electron-phonon interactions. Measurements of $T_{1/2}^{\text{orbital}}$ were repeated for temperatures up to 22 K, and the rate was found to vary linearly with temperature [Fig. 2(c)]. This is evidence of a single-phonon mechanism for this fast relaxation process [30–32], which has significant implications for the spin coherence time discussed below.

With this understanding of the transitions and the population dynamics, it is possible to identify $\Lambda$ schemes consisting of two allowed transitions from different ground states to a common excited state. Such $\Lambda$ schemes provide the ability to optically prepare a dark superposition of the ground states [8]. The dark state causes a dip in the excitation spectrum with a width limited by the phase lifetime of the superposition. This phenomenon is known as coherent population trapping (CPT) and is used here to probe ground-state spin coherence.

To observe CPT, a pump laser was tuned to transition D2 and a probe laser was scanned across transition D1. The excitation peak for transition D1 was observed to contain a sharp two-photon resonance dip as shown in Fig. 3(a). The minimum width of this dip was probed by generating both excitation frequencies from a single laser, using a high bandwidth electro-optic amplitude modulator operated in the linear response regime. The modulation frequency was scanned across the two-photon detuning using microwave sources, and the CPT linewidths in Figs. 3(b) and 3(c) are therefore insensitive to laser frequency noise. Laser power limitations with this equipment prevented access to the dark transitions for the well-aligned field, and so measurements were made on a second SiV\textsuperscript{−} site oriented at about 70 degrees to the field, which accounts for the lower D1–D2 detuning. The CPT dip width was measured for various laser powers [Fig. 3(b)] and was found to be about 4.5 \pm 0.3 MHz in the absence of power broadening [Fig. 3(c)]. This is far below the transform-limited optical linewidth of 94 MHz [20], which confirms CPT and corresponds to a spin coherence time of $T_{2} = 35 \pm 3$ ns. This coherence time is similar to $T_{1/2}^{\text{orbital}}$, indicating that the rapid switching between orbital branches leads to dephasing of the electronic spin even when its polarization is maintained. Although this dramatically limits the usefulness of electronic spin in SiV\textsuperscript{−}, our identification of
the orbital switching mechanism makes it possible to consider techniques to overcome this limit.

One approach to create a long-lived memory is provided by SiV\(^{-}\) centers containing the \(^{29}\)Si isotope, which has nuclear spin \(I = \frac{1}{2}\). Such SiV\(^{-}\) centers can be identified by an isotopic shift of the ZPL\(^{23}\). We observed a double CPT dip for \(^{29}\)Si SiV\(^{-}\) sites, as shown in Fig. 3(c). This doublet arises because of the hyperfine interaction with the \(^{29}\)Si nuclear spin \(I = \frac{1}{2}\) which gives rise to a two-photon resonance condition at two different detunings, as shown in Fig. 3(d). This observation of \(^{29}\)Si hyperfine splitting in SiV\(^{-}\) raises the possibility of optical access to nuclear spin, which should have much longer coherence times than electron spins\(^{33–35}\).

It may also be possible to improve the coherence time of electronic spin in SiV\(^{-}\). Since \(T_2^*\) is limited by orbital relaxation that arises from single-phonon processes, most of these ideas revolve around limiting the availability of phonons at the relevant frequency (around 47 GHz). For temperatures below 47 GHz (\(\ll 2\) K), the reduced phonon occupation results in a suppression of the orbital relaxation from the lower branch of the ground state. Using the lower branch states as qubits would then enable long-lived spin coherences. Even at 5 K, nanodiamonds with sizes smaller than half the transition phonon wavelength (around 125 nm) would inhibit the orbital relaxation rate\(^{36}\), as the diamond-vacuum interface results in a 3D phonon cavity with a discrete spectrum\(^{37}\). Similarly, recent advances in diamond nanofabrication\(^{38,39}\) suggest that periodic nanostructures can be fabricated on diamond to realize a complete phononic band gap at the phonon transition frequency\(^{40–42}\). We expect both approaches to reduce the orbital relaxation rate and therefore increase the spin coherence time.

We have demonstrated that resonant excitation can be used to read out and coherently prepare electronic spin in individual SiV\(^{-}\) centers. These results open the door for exploration of long-lived quantum memories based on SiV\(^{-}\) centers and the development of spin-photon interfaces benefiting from unique optical properties.

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Note added.—Recently, we became aware of complementary work reporting CPT with SiV− centers [43].

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See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.263602, which includes Ref. [26], for details on sample synthesis, transitions C1−C4, population dynamics, spin relaxation in misaligned field, and CPT on an orbital A scheme.

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