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Phys. Rev. B 98, 235140 — Published 20 December 2018
DOI: 10.1103/PhysRevB.98.235140
Strongly Anisotropic Spin Relaxation in the Neutral Silicon Vacancy Center in Diamond

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(Dated: October 29, 2018)

Color centers in diamond are a promising platform for quantum technologies, and understanding their interactions with the environment is crucial for these applications. We report a study of spin-lattice relaxation ($T_1$) of the neutral charge state of the silicon vacancy center in diamond. Above 20 K, $T_1$ decreases rapidly with a temperature characteristic of an Orbach process, and is strongly anisotropic with respect to magnetic field orientation. As the angle of the magnetic field is rotated relative to the symmetry axis of the defect, $T_1$ is reduced by five orders of magnitude. The electron spin coherence time ($T_2$) follows the same temperature dependence but is drastically shorter than $T_1$. We propose that these observations result from phonon-mediated transitions to a low lying excited state that are spin conserving when the magnetic field is aligned with the defect axis, and we discuss likely candidates for this excited state.

I. INTRODUCTION

Solid state defects are attractive candidates for quantum technologies because they can have long spin coherence time and can be integrated into nanofabricated devices. However, interactions with phonons can lead to spin decoherence. Color centers in diamond with exceptionally long spin coherence time have been identified, such as the nitrogen-vacancy center ($NV^−$), and these are promising candidates for a wide range of applications including quantum sensing\textsuperscript{1}, quantum information processing\textsuperscript{2,3}, and quantum networks\textsuperscript{4-6}. More recently, the negatively-charged silicon vacancy center in diamond has been demonstrated to have better optical homogeneity, but poor spin coherence at 4 K because of a phonon-mediated orbital relaxation process\textsuperscript{7,8}. We recently demonstrated that the neutral charge state of the silicon vacancy center ($SiV^0$) can be implanted with high conversion efficiency, it exhibits excellent optical coherence with more than 90% of its emission into a nearly transform-limited zero-phonon line which does not exhibit any spectral diffusion, and it displays long spin coherence times at temperatures up to 20 K\textsuperscript{9}. These properties make it an ideal candidate for a single atom quantum memory in a quantum network. However, we also observed that temperatures above 20 K both $T_1$ and $T_2$ decrease exponentially with temperature\textsuperscript{10,12}. Understanding the origin of this process is crucial for extending the operation range of $SiV^0$ to higher temperatures, to enable new applications in quantum information processing and nanoscale sensing.

In this manuscript we investigate the spin-lattice relaxation of $SiV^0$ in detail. The exponential temperature dependence of $T_1$ above 20 K is consistent with an Orbach process\textsuperscript{13-15} with an activation energy ($E_a$) of 16.8 meV, and we observe that the relaxation rate has a sharp dependence on the angle ($θ$) of the magnetic field ($B$) relative to the symmetry axis of the defect (Fig. 1b). As the angle of the magnetic field is rotated away from the crystallographic axis of $SiV^0$ by just 5 degrees, $T_1$ decreases by almost two orders of magnitude. $T_2$ follows the same temperature dependence as $T_1$ but at a rate that is three orders of magnitude faster when the magnetic field is aligned with the defect symmetry axis.

We propose that the strong intrinsic anisotropy in the spin-lattice relaxation of $SiV^0$ and the significantly shorter spin coherence time originate from phonon-mediated transitions to an excited state that are spin-conserving when the magnetic field is aligned with the quantization axis of the center. This is fundamentally similar to previous observations in $SiV^−$ at 4 K, in which a fast orbital relaxation ($T_{1,\text{orbital}} = 38$ ns) is spin-conserving but spin-dephasing, giving rise to a relatively long $T_{1,\text{spin}} = 2$ ms, while $T_2$ is limited by the orbital relaxation rate\textsuperscript{16}. For $SiV^−$, spin relaxation arises from differing spin-orbit coupling in the two low-lying orbital states, and when the external magnetic field is aligned with the $SiV^−$ axis, this spin relaxation is suppressed, leading to $T_{1,\text{spin}} \gg T_{1,\text{orbital}}$. However, in a large off-axis magnetic field, the eigenstates mix, and the spin relaxation rate increases rapidly with angle\textsuperscript{17}.

The identity of the state at 16.8 meV implied by the activation energy is unknown. Likely candidates are a low-lying singlet state or a triplet vibronic mode\textsuperscript{12}. For both candidates, we develop models that explain the observed suppression of spin relaxation when the magnetic field is aligned with the quantization axis, the temperature and orientation dependence of $T_1$, and the ratio between $T_1$ and $T_2$. We show that a model incorporating a singlet excited state closely reproduces our data, and we outline the physical requirements for a triplet excited state that would account for the observations.
FIG. 1. (a, top) Ball and stick model of SiV$^0$. Gray spheres are carbon atoms. The interstitial Si atom (blue sphere) and split vacancy (red spheres) are aligned along the (111) directions in the diamond lattice, and the magnetic field ($\mathbf{B}$) forms angle $\theta$ with the defect axis. (a, bottom) Ground state SiV$^0$ spin levels at $\mathbf{B} = 0$. (b, top) Ground state spin levels at magnetic fields such that the Zeeman energy is large compared to the zero field splitting, $g\mu_B \mathbf{B} \gg D$. (b, bottom) Magnetic field dependence of spin levels with $\mathbf{B} \parallel [111]$. Arrows indicate the magnetic field required for each spin transition to match the $9.7 \text{ GHz}$ resonator mode. (c) Pulsed ESR spectrum of SiV$^0$ (zero field splitting, $D = 940 \text{ MHz}$), and was previously described in reference $\text{A}$). The second diamond (D2) was doped during growth with boron ($\theta = 2.6^\circ$ (D1), $\theta = 0.8^\circ$ (D2) and [111]: $\theta = 106.9^\circ$ (D1), $\theta = 108.7^\circ$ (D2). Lines correspond to the best fit of Eq. 2.

III. RESULTS

A. Temperature dependence of $T_1$ and $T_2$

The ground state spin Hamiltonian is given by $\text{Eq. 2}$.

$$\hat{H} = \hat{S}^\dagger \hat{D} \hat{S} + \mu_B \hat{g} \hat{B},$$

with electron spin $S = 1$, zero field splitting tensor ($\hat{D}_g$) with axial part $D_g = 0.94 \text{ GHz}$ (at $T = 4.8 \text{ K}$), electron $g$ tensor ($\hat{g}$) with parallel and perpendicular components $g_\parallel = 2.0042$ and $g_\perp = 2.0035$ respectively, and the Bohr magneton, $\mu_B$. The $\hat{D}$ and $\hat{g}$ tensors are both aligned along the (111) directions. With the field aligned along [111], there are two inequivalent orientations (Fig. [1]): one orientation aligned with the field, $\theta = 0^\circ$ ([111], outer ESR peaks), and three equivalent orientations aligned off-axis, $\theta = 109.5^\circ$ ([111], [111], [111], inner ESR peaks).

We performed time-resolved measurements of spin relaxation and decoherence for both inequivalent orientations. At low temperatures, $T_1$ and $T_2$ are constant (Fig. [2]). In sample D2, we previously reported that the SiV$^0$ low temperature $T_2$ is dominated by spectral diffusion from the $1.1\%$ abundance of $^{13}\text{C}$ nuclei, $T_2 = 0.954 \pm 0.025 \text{ ms}$. By contrast, the SiV$^0$ density in D1 is large enough that $T_2$ is limited by instantaneous diffusion, $T_2 = 0.48 \pm 0.03 \text{ ms}$ (see Appendices [B], [C]).

At low temperature, $T_1$ is constant, $T_1 = 46 \pm 2 \text{ s}$ (D1)
and $T_1 = 45 \pm 4$ s (D2), similar to previous observations of NV$^-$. 

Above 20 K, both $T_1$ and $T_2$ decrease exponentially with increasing temperature. In this regime the two inequivalent orientations exhibit similar $T_2$ but significantly different $T_1$. $T_1$ and $T_2$ exhibit the same Arrhenius slope (the slope of the log of the rate vs. inverse temperature) for both orientations. The data ($T_{1,[11]}$, $T_{1,[11]}$, $T_{2,[11]}$, $T_{2,[11]}$) were fitted according to the equation:

$$
\frac{1}{T_{1,2}} = \frac{1}{T_{sat}} + A_{T_1,T_2}(\theta) e^{-E_a/kT},
$$

where $T_{sat}$ is the saturated decay time at low temperature, $A_{T_1,T_2}(\theta)$ is the orientation-dependent rate prefactor, $E_a$ is the activation energy, and $kT$ is the thermal energy. $E_a = 16.8 \pm 1.5$ meV for all curves, but $A_{T_1,T_2}(\theta)$ varies significantly (Table I).

Unlike $T_1$, $T_2$ exhibits a weak orientation dependence. However, since $T_2$ displays the same activation energy as $T_1$, the two decay times likely result from the same physical process. This is surprising since $T_2$ is not $T_1$-limited; $T_2$ is 4000 times shorter than $T_1$ when $\theta = 0^\circ$. We can rule out that the decoherence is caused by magnetic noise from nearby centers because we do not observe a density dependence in $T_2$ when comparing samples D1 and D2, and we are unable to extend $T_2$ with further dynamical decoupling (see Appendix C). Moreover, numerical simulations of ensemble dipolar interactions fail to account for the observed temperature dependence of $T_2$ (see Appendix C).

### B. Orientation dependence of $T_1$ and $T_2$

We measured the detailed orientation dependence of $T_1$ and $T_2$ at 30 K (Fig. 3), where the Orbach process dominates spin relaxation. At a magnetic field of $\sim 3400$ G (Fig. 1b), the Zeeman frequency (9.7 GHz) is much larger than the zero field splitting (0.94 GHz). The relative orientation of the field was varied by rotating the crystal about a (110) axis from $\theta = 0^\circ$ ($B \parallel D$) to $\theta = 90^\circ$ ($B \perp D$). The ESR spectrum (Fig. 1d) was measured to determine orientation to within 1°. The relaxation time exhibits dramatic anisotropy, and as the crystal is rotated away from $\theta = 0$°, spin relaxation becomes clearly biexponential (Fig. 3). Near $\theta = 0^\circ$, $T_1$ drops rapidly, and rotating by just 5° increases the relaxation rate by almost two orders of magnitude. Near $\theta = 55^\circ$, the decay is a single exponential with a short timescale that is insensitive to small rotations. Beyond $55^\circ$ the two timescales diverge, differing by over 3 orders of magnitude at $\theta = 90^\circ$ (Fig. 3).

### C. Model for the Orbach process

We propose a model that captures the four salient features of the data: (1) the strong anisotropy of $T_1$, (2) the biexponential nature of $T_1$, (3) the temperature dependence of $T_2$, and (4) the large ratio between $T_1$ and $T_2$. Generically, an Orbach process is a two-phonon process that connects the ground state spin sublevels $m_s = -1,0,+1$ through an excited state ($|\Psi\rangle$) with amplitudes $t_{-1}, t_0,$ and $t_{+1}$, respectively (Fig. 4b). The amplitudes ($t_{-1}, t_0,$ and $t_{+1}$) are overlap parameters between the ground triplet states and the excited state.
The rate $t_{m_{s}} = \langle m_{s} | \Psi \rangle$ (see Appendices C and D). This gives rise to three possible relaxation rates between pairs of ground state spin sublevels $m_s \leftrightarrow m_{s'} = -1 \leftrightarrow 0, 0 \leftrightarrow +1, -1 \leftrightarrow +1:

$$\frac{1}{T_{1,m_s \leftrightarrow m_{s'}}} = C |t_{m_{s}}t_{m_{s'}}|^2 e^{-E_s/kT},$$

where $C$ is a constant.

If the excited state $\Psi$ is a singlet ($S=0$), it is invariant under magnetic field orientation, so the behavior of $T_1$ can be captured by considering the mixing of the ground state (see Appendix C). The mixing of the spin sublevels in the presence of a large off-axis magnetic field leads to:

$$\begin{pmatrix} |t_{-1}|^2 \\ |t_0|^2 \\ |t_{+1}|^2 \end{pmatrix} = \begin{pmatrix} \cos^4 \frac{\theta}{2} & \frac{1}{2} \sin^2 \theta & \sin^4 \frac{\theta}{2} \\ \frac{1}{2} \sin^2 \theta & \cos^2 \theta & \frac{1}{2} \sin^2 \theta \\ \sin^4 \frac{\theta}{2} & \frac{1}{2} \sin^2 \theta & \cos^4 \frac{\theta}{2} \end{pmatrix} \begin{pmatrix} |t_{-1}|^2 \\ |t_0|^2 \\ |t_{+1}|^2 \end{pmatrix},$$

where $|t_{m_s}|^2$ are the overlap parameters at zero magnetic field. Substituting Eq. 4 in Eq. 3 and solving the 3x3 relaxation rate matrix equation for the ground state spin (S=1) provides the $T_1$ relaxation times (see Appendix C). If $|t_{0}| = |t_{-1}| = |t_{+1}|$, then Eqs. 3 and 4 predict that the spin relaxation is isotropic. However, if $|t_{0}| \gg |t_{-1}|, |t_{+1}|$, the spin relaxation is strongly anisotropic with two characteristic times approximated as:

$$\frac{1}{T_{1,a}} = \frac{3}{8} C |t_{0}|^4 \sin^2(2\theta) e^{-E_s/kT},$$

$$\frac{1}{T_{1,b}} = \frac{1}{2} C |t_{0}|^4 \sin^2(\theta) e^{-E_s/kT}.$$

In this limit the model captures the observed angular dependence of the two timescales in $T_1$ (Fig. 3b). By comparing numerical calculations of the orientation dependence of $T_1$ for different ratios of $|t_{0}|/|t_{\pm1}|$ (Fig. 4b), we can place a lower bound on the imbalance between these rates, $|t_{0}|/|t_{\pm1}| > 100$.

We can also predict the effect of this Orbach process on $T_2$. Customarily, the Orbach process is viewed as a spin relaxation process. However, phonon-mediated transitions to the excited state can also lead to decoherence even when the spin projection is preserved, similar to what has been observed for orbital relaxation in SiV$^0$. While the spin relaxation rate relies on a spin flip and therefore the product of the overlap parameters $t_{m_s} \propto |t_{m_s}|^2 |t_{m_{s'}}|^2$ (Fig. 4a, left), the decoherence rate depends on the sum of overlap parameters, $t_{m_s} \propto |t_{m_s}|^2 + |t_{m_{s'}}|^2$ (Fig. 4a, right), if we assume the spin coherence is completely lost after a single excitation to the excited state $\Psi$. The angle-dependent ratio of $T_1$ to $T_2$ will therefore depend on the ratio of overlap parameters. The model predicts (Fig. S5):

$$\frac{T_{1,a}}{T_{2,0\leftrightarrow \pm1}} = \frac{3 |t_{0}|^2 + 2 |t_{\pm1}|^2}{3 |t_{\pm1}|^2}.$$

The orientation dependence of $T_2$ predicted from this model is plotted in Fig. 3a, where we also included the effect of instantaneous diffusion in sample D1, and is plotted in detail in Fig. S7. The anisotropy in $T_2$ is mostly canceled in the sum $|t_{m_s}|^2 + |t_{m_{s'}}|^2$. The model provides the best fit for both the $T_1$ and $T_2$ data when $|t_{0}|/|t_{\pm1}| \approx 125$ (Fig. 3a).

If the excited state $\Psi$ is instead a triplet ($S=1$), the overlap parameters cannot be written compactly, but we analyze this case numerically (see Appendix D). Briefly, phonon-mediated orbital relaxation to a vibronic excited state is generally spin conserving, but differences in the ground and excited state Hamiltonians can lead to mixing during the time spent in the excited state. For SiV$^0$ the ground and excited states can have different zero field splitting tensors ($D_x$ and $D_y$). Since the Zeeman splitting in these measurements is 9.7 GHz, the zero field splittings must differ by a comparable scale in order to reproduce the observed ratio of $T_1$ to $T_2$, and we find that the data can be qualitatively reproduced when $D_x \approx 5 - 7$ GHz (Figs. S6 and S7), compared to the ground state zero field splitting, $D_y = 0.94$ GHz. It is unlikely that the zero field splittings differ by such a large magnitude. Alternatively, the small ratio of $T_1$ to $T_2$ could also arise from incomplete spin dephasing. If
the excited state lifetime is short compared to the spin precession time ($\tau < \pi \hbar / E_{\text{Zeeman}} \sim 50$ ps), then the spin coherence is partially preserved through a single excitation to $\Psi$. A model involving a triplet excited state would therefore require either that $D_x \gg D_y$ or that the excited state lifetime is short enough to partially preserve coherence.

IV. CONCLUSIONS

In summary, we have shown that spin relaxation in SiV$^0$ at high temperature is dominated by an Orbach process that is strongly dependent on the magnetic field orientation, and $T_2$ exhibits the same temperature dependence as $T_1$, but at a significantly faster rate. These observations can be explained by a model for the Orbach process where the overlap parameters from the $m_s = 0$ and $m_s = \pm 1$ spin sublevels to a singlet excited state are drastically different. We note that this is consistent with the preferential optical spin polarization through the intersystem crossing into $m_s = 0$ (Fig. 1). Alternatively, these observations can be qualitatively reproduced by a model with a triplet excited state that either exhibits a much larger zero field splitting than the ground state or a very short excited state lifetime. Although our present results cannot definitively identify the excited state, detailed spectroscopy can help distinguish between these two cases. Absorption spectroscopy could elucidate the vibronic structure and the nature of the singlet state can be explored using time-resolved photon correlation measurements and temperature-dependent intersystem crossing rates. At temperatures well above the activation energy, there should be enough population in the excited state to observe spin resonance transitions associated with a different zero field splitting. We have not observed the existence of additional transitions, but ongoing work includes increasing our sensitivity at higher temperatures to search for such states.

The strong intrinsic anisotropy in SiV$^0$ stands in contrast to prior studies of NV$^-$, in which spin relaxation is mostly insensitive to the magnetic field orientation. To the best of our knowledge, there has not been a detailed study of the orientation and temperature dependence of spin relaxation in NV$^-$ at high magnetic fields, and it would be interesting to perform such measurements in light of our work. Similarly, a more detailed orientation and temperature dependence of $T_{1,\text{spin}}$ in SiV$^-$ would further elucidate analogous spin and orbital relaxation processes, and recent measurements at dilution refrigerator temperatures have started to explore mechanisms for spin relaxation and decoherence.

These observations point to a particularly intriguing possibility for high temperature operation if $\Psi$ is a spin singlet. The small overlap parameters between $m_s = \pm 1$ and $\Psi$ implies that superpositions of $m_s = \pm 1$ could have longer spin coherence time than even the measured single-quantum $T_1$, as they should be limited instead by the double-quantum $T_1$ (see Appendix A). Future experiments include double-quantum spin resonance to interrogate the coherence of such superposition states.

ACKNOWLEDGMENTS

This work was supported by the Princeton Center for Complex Materials, an NSF MRSEC (DMR-1420541) and the NSF EFRI ACQUIRE program (1640959). G. Thiering and A. Gali were supported by the NKP-17-3-III New National Excellence Program of the Ministry of Human Capacities and the EU Commission (DI-ADEMS Contract No. 611143). The authors gratefully acknowledge Jeff Thompson, Shimon Kolkowitz, and Ashok Ajoy for helpful discussions, as well as Sorawis Sangtawesin and Zihuai Zhang for help in proofreading the manuscript.

Appendix A: EXPERIMENTAL DETAILS

Electron spin resonance was performed on a modified Bruker Elexys 580 system with a 1.4 T electromagnet (Fig. 5b). This is done by driving the TE011 mode of a cylindrical dielectric resonator (ER-4118X-MD5) (9.7 GHz) with an external vector microwave source (Agilent E8267D). Temperature dependent measurements were performed in an Oxford CF935 helium-flow cryostat. The microwave excitation channel consists of a vector microwave source (Agilent, E8267D), a variable gain solid state amplifier (AR 2084G11), a protection PIN diode (Hittite switch), and a variable attenuator (ARRA P4844-30). This allows for arbitrary waveform control as well as flexibility in the microwave power delivered to the resonator which is necessary for handling the range of quality factors and excitation bandwidths needed across several samples. The microwave excitation is coupled capacitively to the resonator through a microwave waveguide terminated with an open-loop antenna and the reflected microwaves are returned through a fast Hittite switch (HMC547) redirecting the microwaves to the detection channel. The reflected microwave signal is sent through a cryogenic low noise amplifier (Low Noise Factory, LNF-LNC4,16B) and then through a room temperature low noise amplifier (Bruker). To avoid saturating and damaging the cryogenic amplifier it must be protected from the microwave excitation during the ring down period of the resonator. This protection was supplied by the fast Hittite microwave switch, which was used to couple the reflected microwave excitation out of the resonator back to the excitation pathway. After the ring down period the switch is opened up to the cryogenic amplifier so that any spin signal outside of the ring down period is amplified. The amplified signal is sent to a quadrature detector (Bruker) where it is mixed with a reference signal from the vector source. The in-phase (I) and quadrature (Q) signals coming out of the mixer
are read with a fast digitizing board (Bruker SpecJet). Triggering in this experiment was controlled with a Pulse-Blaster ESR-PRO board.

Spin coherence measurements (T_2) were made using a two-pulse Hahn echo sequence with an initial laser pulse to boost the echo strength by polarizing the spins preferentially into m_s = 0 (Fig. 5a). Spin relaxation measurements (T_1) were made using a three pulse inversion recovery sequence (Fig. 5a). First a laser pulse is used to polarize the SiV^0 spins preferentially into m_s = 0. After a small delay to wait for any laser induced noise to die out, a microwave π pulse is applied to invert the spin polarization even further away from Boltzmann equilibrium. The waiting time in this inverted state (T) is varied and a two-pulse Hahn echo sequence (π/2 − τ − π − τ -echo) is used to read out the polarization at the end of the waiting period. For the strongest signal τ is chosen to be much smaller than T_2.

Appendix B: INSTANTANEOUS DIFFUSION IN SAMPLE D1

At low temperatures, the coherence time in sample D1 is limited by instantaneous diffusion, which arises when a microwave pulse induces spin flips on a dense bath of paramagnetic centers. If we consider a central spin surrounded by its neighbors, then the pulse will induce rotations of the neighbors as well as the central spin. Phase resulting from the pulse-induced change in the dipolar magnetic field from the neighbors is not refocused during a Hahn echo sequence, limiting the coherence time of the central spin to T_{2(ID)}

The effect of instantaneous diffusion can be mitigated by using a smaller rotation angle (θ_0) for the second microwave pulse of a Hahn echo sequence, since the change in the net dipolar magnetic field scales as sin^2(θ_0/2). This results in a proportionally smaller accumulation by the central spin and a decoherence rate 1/T_2 ∝ T_{2(ID)}sin^2(θ_0). Using a smaller rotating angle will enhance T_2, but it will also decrease the bulk echo signal by the same factor. In sample D1, the apparent decoherence rate increases linearly with sin^2(θ_0/2) (Fig. 6). The data were fit according to the following:

\[
\frac{1}{T_2} = \frac{1}{T_{2(SD)}} + \frac{1}{T_{2(ID)}} \sin^2(\theta_0/2),
\]

where T_{2(SD)} is the spectral diffusion decay time. The fit results in T_{2(SD)} = 0.95 ± 0.22 ms, most likely arising from the 1.1% of 13C nuclei, and T_{2(ID)} = 0.319 ± 0.056 ms.

We note that the Hahn echo spin coherence times reported for sample D1 in Figs. 6, 10, and 11 (T_2 = 0.28 ms) is not the same as the spin coherence time reported in Fig. 2 in the main text (T_2 = 0.48 ms). This arises from the nonuniform population distribution of SiV^0 centers in this sample over the four inequivalent crystal orientations ([111], [111], [111], [111]), which has been reported previously as sample C in reference. The data in Figs. 3, 11, and 10 is taken using the [111] orientation (larger SiV^0 concentration), while the data in Fig. 2 is taken using the [111] orientation (smaller SiV^0 concentration).

Appendix C: DECOHERENCE ARISING FROM T_1-INDUCED SPIN FLIPS OF FAST RELAXING NEIGHBORS

An alternative hypothesis for the observed temperature dependence of T_2 (Fig. 2 in the main text) and its relative magnitude with respect to T_1 is that rapid dephasing arises from dipolar interactions with other SiV^0 spins in the bath, such as those misaligned with the external magnetic field. We can immediately rule out spectral diffusion from SiV^0 spin flip-flops and instantaneous diffusion mechanisms arising from dipolar interactions between SiV^0 centers since these mechanisms would be independent of temperature. Instead, we consider the contribution of spectral diffusion arising from the fast T_1 relaxation of nearby SiV^0 centers. This decoherence mechanism is strongest when T_1 of the spin bath is comparable to the T_2 of the central spin under consideration.

In our samples, T_2 ≈ 0.5 ms at low temperatures, which is comparable to T_1 ≈ 1 ms of the three equivalent SiV^0 orientations misaligned with the magnetic field (θ ≈ 100°) at temperatures above 20 K. We numerically model the contribution to the Hahn echo decay from these three equivalent off-axis sites for the range of densities in samples D1 and D2. The electron spin Hamiltonian describing a pair of SiV^0 spins, S_1 and S_2, is given by:

\[
\hat{H} = \hbar \omega_1 \hat{S}_{1z} + \hbar \omega_2 \hat{S}_{2z} + \hbar A (r_{12}) \hat{S}_{1z} \hat{S}_{2z} + \hat{V}_2(t),
\]

with dipolar interaction between the SiV^0 spins

\[
A (r_{12}) = g_1 g_2 \mu_B h^{-1} (1 - 3 \cos^2(\theta_{12})) r_{12}^{-3},
\]

where ω_1 and ω_2 are the transition frequencies of the spins, r_{12} is the distance between the spins, θ_{12} is the angle between r_{12} and B, g_1 and g_2 are the longitudinal components of the g tensors. For our model we consider that S_1 is a slow relaxing spin (θ = 0°) whose coherence time is being measured, and S_2 is a fast relaxing spin (θ ≈ 100°) whose spontaneous T_1 flips induce decoherence of S_1. The term \hat{V}_2(t) accounts for the fast Orbach spin relaxation rate of S_2 spins by inducing random spin flips at a rate W. The contribution to the echo signal decay for S_1 is:

\[
V (2\tau) = \left[ \cosh(R\tau) + \frac{W}{R} \sinh(R\tau) \right]^2 + \frac{A^2 (r_{12})}{4R^2} \sinh(R\tau) \exp(-2W\tau),
\]
FIG. 5. (a) Experimental pulse sequence for bulk electron spin resonance measurements of $T_1$ and $T_2$. (b) Electron spin resonance setup. A vector microwave source is amplified and coupled capacitively to the TE011 mode of a sapphire-dielectric cylindrical resonator. The diamond sample sits in the center of the resonator where the microwave field ($B_{mw}$) is uniform. The spin echo is coupled out of the resonator through a different path set by a fast Hittite microwave switch and amplified by a low noise cryogenic preamplifier. Optical excitation from a green laser (532 nm) is controlled with a mechanical shutter and fiber coupled to the sample.

FIG. 6. Decoherence rates for SiV$^0$ centers in sample D1 measured at 5 K as a function of the rotation angle ($\theta_2$) of the second pulse in a Hahn echo sequence. The linear dependence confirms that $T_2$ is limited by instantaneous diffusion. The black curve is a fit according to Eq. 2.

where $\tau$ is the inter-pulse delay in a Hahn echo sequence, $W = 1/T_{1,111}$ (fast relaxing sites, Fig. 4 blue line), $R^2 = W^2 - A^2 (r_{12})/4$, and $r_{12} = n^{-1/3}$ is the average inter-spin distance. This expression is averaged over all angles $\theta_{12}$ and added to the Hahn echo decay that arises from $^{13}$C spectral diffusion alone ($T_{2(SD)} = 0.95$ ms). The resulting calculated Hahn echo decay times are shown in Fig. 6 for several SiV$^0$ densities in and above the range of the two samples studied here, which have SiV$^0$ concentrations of less than $5 \cdot 10^{16}$ cm$^{-3}$. The density required to account for the data would need to be 100 times higher. Furthermore, at high temperatures, motional narrowing should lead to an increase in $T_2$, which does not qualitatively agree with the observed temperature dependence (Figs. 2 and 7).

Appendix D: DYNAMICAL DECOUPLING USING CPMG

We previously reported dynamical decoupling measurements using the Carr-Purcell-Meiboom-Gill sequence...
FIG. 7. Arrhenius plot of simulations of $T_2$ (red lines) resulting from spectral diffusion arising from fast relaxing SiV$^0$ centers. The blue dashed line is a fit of Eq. 2 to the temperature dependence of $T_1$ (blue squares) of fast relaxing SiV$^0$ sites. This fit is incorporated in Eq. C3 to simulate $T_2$ for a range of defect densities (labels in units of cm$^{-3}$). The simulations indicate that for the range of densities studied, the temperature dependence of $T_2$ is not significant and is inconsistent with the observed temperature dependence of $T_2$ (red dots).

FIG. 8. Orientation dependence of $T_1$ (blue) and $T_2$ (red) measured at 30 K on the $m_s = -1 \leftrightarrow 0$ transition. The lines were simulated for the Orbach model with a singlet excited state using Eqs. G12 and assuming $|g_s^0|/|g_s^0| = 125$: (solid line) $T_{1,a}$, (dashed line) $T_{1,b}$, and (long dash) $T_2$.

on sample D$^{11,36}$. The Hahn echo $T_2$ displays a plateau below 20 K corresponding to $^{13}$C spectral diffusion, but is limited by an Orbach process above 20 K. We observed that $T_{2,CPMG}$ is unchanged above 20 K and follows the temperature dependence of $T_2$. However below 20 K $T_{2,CPMG}$ becomes substantially longer than $T_2$ and follows the extrapolated temperature dependence of the Orbach process. We hypothesize that the CPMG experiment refocuses slow spectral diffusion that arises from the $^{13}$C nuclei, but it does not refocus fast effects from the Orbach process, as expected. All of the points in the CPMG measurement lie along the same curve $T_{2,CPMG} = A \exp (-E_a/kT)$, where $A = 1180 \pm 210$ kHz and $E_a = 16.8 \pm 1.5$ meV in the entire measured temperature range 5 K - 60 K.

Appendix E: ORIENTATION DEPENDENCE OF $T_1$ AND $T_2$ MEASURED ON $m_s = -1 \leftrightarrow 0$

In the main text we presented the orientation dependence of the $T_1$ and $T_2$ times for SiV$^0$ for measurements on the $m_s = 0 \leftrightarrow +1$ transition. We also repeated the same measurements on the $m_s = -1 \leftrightarrow 0$ transition and find that it gives a nearly identical orientation dependence (Fig. 8). Since $T_{1,-1\leftrightarrow0} \approx T_{1,0\leftrightarrow+1}$, we can conclude that $|t_{t+1}^0| \approx |t_{t-1}^0|$. Additionally, because of the 1 GHz zero field splitting of SiV$^0$, the measurements on the $m_s = -1 \leftrightarrow 0$ transition were made at a field that was $\sim 300$ G larger (when aligned with the [111] direction) compared to the measurements on the $m_s = 0 \leftrightarrow +1$ transition in Fig. 3. This implies that the Orbach process has a weak dependence on the magnetic field strength.

Appendix F: RATIO OF $T_1$ TO $T_2$

The singlet model predicts that the observed ratio of $T_1$ to $T_2$ in Figs. 2 and 3 is strongly dependent on the ratio of the overlap parameters at zero field. The analytical form of this dependence is shown in Eqs. 4 and 6 which is plotted in Fig. 9. This figure shows that this ratio is strongly dependent on the orientation of the magnetic field, indicating that the best way to extract the ratio of the zero field overlap parameters is by performing a global fit across all orientations (Fig. 3).

Appendix G: MODEL FOR SPIN RELAXATION: ORBACH PROCESS WITH A SINGLET EXCITED STATE

Here we present a detailed analytical derivation of the spin relaxation of SiV$^0$ for an Orbach process mediated by a spin singlet excited state. The neutral silicon vacancy center has $D_{3d}$ symmetry with a ground spin-triplet state ($3A_{2g}$), and the first excited singlet state is expected to be $^1E_g$. The splitting between these states is unknown. At zero magnetic field the triplet and singlet states can mix through spin-orbit coupling assisted by phonons$^{37,49}$. 

A general rotation, $R$, can be expressed in terms of Euler angles:

$$ R(\alpha, \beta, \gamma) = R_2(\gamma)R_1(\beta)R_3(\alpha), $$

where $\Omega = (\alpha, \beta, \gamma)$ is the set of Euler angles following the "passive" convention. Under this rotation the irreducible tensors in the spin Hamiltonian $T_{J,m}$ transform to $\rho_{J,m}$ as:

$$ \rho_{J,m} = R(\alpha, \beta, \gamma)T_{J,m}R^{-1}(\alpha, \beta, \gamma) = \sum_{m'} D^J_{m',m}(\alpha, \beta, \gamma)T_{J,m'}, $$

where $D^J_{m',m}(\Omega)$ is the Wigner matrix of rank $J$. The elements of this matrix are:

$$ D^J_{m',m}(\alpha, \beta, \gamma) = \exp(-im'\alpha)d^J_{m',m}(\beta)\exp(-im\gamma), $$

with

$$ d^J_{m',m}(\beta) = \int d\Omega \ Y^*_{Jm'}(\theta, \phi) e^{-iJm\theta}Y_{Jm}(\theta, \phi), $$

where $Y_{Jm}(\theta, \phi)$ are the standard spherical harmonic functions and $J_\hbar$ is the component of the total angular momentum along $\hbar \parallel (110)$. Then for $J = S = 1$:

$$ D^{S=1}_{m',m}(\alpha, \beta, \gamma) = \begin{pmatrix}
\frac{1+\cos(\beta)}{2} & -i\frac{1}{\sqrt{2}} \sin(\beta) & -i\alpha \\
\frac{1}{\sqrt{2}} \sin(\beta) & \cos(\beta) & 1 \\
-i\alpha & 1 & \frac{1-\cos(\beta)}{2}
\end{pmatrix} e^{i(\alpha-\gamma)} \tag{G6}
$$

If we specifically define $R$ as the rotation away from (111) about the (110) axis, so that $\alpha = \varphi, \beta = \theta$, and $\gamma = 0^\circ$ define the orientation of the magnetic field, the mixing of the transition amplitudes is given by:

$$ t_{m'} = \sum_m D^{S=1}_{m',m}(\varphi, 0) t^0_m, \tag{G7} $$

From this we obtain the transition rates $|t_{m'}|^2$ by invoking the random phase approximation to neglect the cross terms (averaging over $\varphi$). The physical origin of the random phase approximation can arise from taking an ensemble average over a bath of phonons that randomly induce transitions to the excited state through spin-orbit coupling. The result is:

$$ |t_{m'}|^2 = \sum_m |\langle D^{S=1}_{m',m}(\varphi, 0) \rangle|^2 |t^0_m|^2, \tag{G8} $$

where $|\langle D^{S=1}_{m',m}(\varphi, 0) \rangle|^2 = \begin{pmatrix}
\cos^4(\theta/2) & \frac{1}{2} \sin^2(\theta) & \sin^4(\theta/2) \\
\frac{1}{2} \sin^2(\theta) & \cos^2(\theta) & \frac{1}{2} \sin^2(\theta) \\
\sin^4(\theta/2) & \frac{1}{2} \sin^2(\theta) & \cos^4(\theta/2)
\end{pmatrix} \tag{G9} \begin{pmatrix}
\cos^4(\theta/2) \\
\frac{1}{2} \sin^2(\theta) \\
\sin^4(\theta/2)
\end{pmatrix}$. 

In the main text, Eqs. 3 for the overlap coefficients in the presence of an off-axis magnetic field are obtained by substituting Eq. (G9) into Eq. (G8).

Next, the transition rate matrix (Eq. (G9)) can be used to model the spin relaxation processes for $S = 1$ where the populations $P = (P_{-1}, P_0, P_{+1})$ evolve according to:

$$ \frac{dP(t)}{dt} = \hat{R}P(t), \tag{G10} $$
where the rate matrix $\tilde{R}$ is given by:

$$\tilde{R}_{m,m'} = C(1 - \delta_{m,m'}) \mu^{m-m'} |t_m|^2 |t_{m'}|^2 - C\delta_{m,m'} \left( \sum_{m'' \neq m} |t_m|^2 |t_{m''}|^2 \mu^{m''-m} \right),$$

(Eq. G11)

where $\delta_{m,m'}$ is the Kronecker delta function and $\mu = \exp(hf/kT)$ is the Boltzmann factor at $T = 30$ K and $f = 9.7$ GHz. Assuming that $|t_{0 \pm 1}| = |t_{0 \pm 1}'|$ and $\mu = 1$ ($hf \ll kT$), this results in two distinct rate eigenvalues $\lambda_1$, $\lambda_2$ corresponding to $T_{1,a} = \lambda_1 e^{-E_a/kT}$ and $T_{1,b} = \lambda_2^{-1} e^{-E_a/kT}$:

$$T_{1,a} = \frac{2e^{-E_a/kT}}{3C |t_0|^2 \left( |t_{0+1}|^2 + |t_{0-1}|^2 \right)}$$

$$T_{1,b} = \frac{e^{-E_a/kT}}{C |t_{0-1}|^2 \left( 2 |t_{0+1}|^2 + |t_0|^2 \right)}.$$  

(Eqns. G12)

These were used to simulate the angular dependence of $T_1$ as a function of $|t_0|/|t_{0 \pm 1}|$ in Figs. 3a and 4b. If we assume that $|t_0| \gg |t_{0 \pm 1}|$ then Eqns. G12 reduce to Eqns. 5.

Appendix I: ORIENTATION DEPENDENCE OF $T_2$

Our model for the Orbach process predicts a weak orientation dependence of $T_2$. The orientation dependence fits shown in Fig. 3a utilize the same overlap amplitudes ($t_0^0, t_{0 \pm 1}^0$) to explain both $T_1$ and $T_2$. The actual expression used in fitting the $T_2$ dependence in Fig. 3a is given by:

$$\frac{1}{T_{2,0\pm 1}} = \frac{1}{3} C \left( |t_0^0|^2 + 2 |t_{0 \pm 1}^0|^2 \right) \left( |t_0^0|^2 + |t_{0 \pm 1}^0|^2 \right) + \frac{1}{T_{2(ID)}} + \frac{1}{T_{2(SD)}},$$

(Eq. H1)

which in addition to the Orbach process also includes instantaneous diffusion and $^{13}$C spectral diffusion mechanisms. We used $T_{2(ID)} = 0.319$ ms and $T_{2(SD)} = 0.95$ ms in these simulations.

The orientation dependence of $T_2$ is shown in Fig. 11 with the simulated fits according to the singlet (Fig. 11a).
FIG. 10. (a) Model for the Orbach process of SiV\textsuperscript{0} with a singlet excited state. The transition rates ($|t_{m_s}|^2$) are determined by spin orbit coupling and depend on the overlap between the electronic wavefunctions of the ground triplet state and the excited singlet state. (b) Model for the Orbach process of SiV\textsuperscript{0} with a triplet excited state. The transition rates between the ground state spin sublevels depend on the overlap between the spin eigenstates of the ground state and excited state, and must be summed over all spin sublevels in the excited state. (c) $T_1$ and $T_2$ orientation dependence from Fig. 3a, plotted against calculated fits from the singlet (black) and triplet (red) Orbach models: (solid line) $T_1$, (dashed line) $T_1$, and (long dash) $T_2$. The singlet model fit assumes $|t_0^0/t_{\pm 1}| = 125$. The triplet model fit assumes a coaxial excited state ZFS tensor with $D_e = 5$ GHz.

FIG. 11. Orientation dependence of the SiV\textsuperscript{0} electron spin coherence time, $T_2$ (replotted from Fig. 3a). (a) Predicted orientation dependence using an Orbach model with a singlet excited state (black curve, Eq. 11) with no free parameters. Measurements were made on both the $m_s = 0 \leftrightarrow +1$ transition (red points) and the $m_s = -1 \leftrightarrow 0$ transition (blue points) at each orientation. (b) Predicted orientation dependence of an Orbach model with a triplet excited state. The simulated curves are shown for several values of $D_e$ (labeled in units of GHz). The singlet model has no other free fitting parameters, and we plot Eq. 11 for the singlet model assuming that $|t_0^0/t_{\pm 1}| = 125$ as determined from the fit of the $T_1$ orientation dependence (Fig. 3a). The singlet model predicts the magnitude of $T_2$ with reasonable accuracy.

The triplet model has four free parameters, two angles that set the quantization axis of the excited state, the axial part of the zero field splitting tensor, and the rhombic part of the zero field splitting tensor. We only consider the case where the zero field splitting tensor of
the excited state is axial and aligned with the symmetry axis of the defect since this is the case that best produces the measured $T_1$ orientation dependence (Fig. 10). The dependence for several values of $D_m$ is shown and the best fit occurs with $D_m \approx 5$–7 GHz. Alternatively, if the spin does not fully decohere through a single cycle through the excited state, the magnitude of $T_2$ can be larger than the simulated values.

Appendix J: DOUBLE QUANTUM COHERENCE TIME

The Orbach process affects the double quantum transition ($m_s = -1 \leftrightarrow +1$) through the overlap parameters $t_{+1}, t_{-1}$ and from the anisotropy of $T_1$ (Fig. 3) we know that $|t_{+1}^0|, |t_{-1}^0| \ll |t_0^0|$. It follows that $\frac{1}{t_{+1, -1}} \propto |t_{+1}^0| + |t_{-1}^0| \ll |t_0^0| + |t_0^0| = \frac{1}{t_2^0, \pm 1}$ so that the double quantum coherence time should be longer at zero field or with the magnetic field aligned along the defect axis. Moreover for small angles of the magnetic field the double quantum coherence time can be longer than the single quantum $T_1$ time, even when the single quantum coherence time is much shorter (Fig. 12).

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