All-optical dc nanotesla magnetometry using silicon vacancy fine structure in isotopically purified silicon carbide

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We uncover the fine structure of a silicon vacancy in isotopically purified silicon carbide (4H-28SiC) and reveal not yet considered terms in the spin Hamiltonian, originated from the trigonal pyramidal symmetry of this spin-3/2 color center. These terms give rise to additional spin transitions, which would be otherwise forbidden, and lead to a level anticrossing in an external magnetic field. We observe a sharp variation of the photoluminescence intensity in the vicinity of this level anticrossing, which can be used for a purely all-optical sensing of the magnetic field. We achieve dc magnetic field sensitivity better than 100 nT/√Hz within a volume of 3 × 10⁻⁷ mm³ at room temperature and demonstrate that this contactless method is robust at high temperatures up to at least 500 K. As our approach does not require application of radiofrequency fields, it is scalable to much larger volumes. For an optimized light-trapping waveguide of 3 mm² the projection noise limit is below 100 fT/√Hz.

I. INTRODUCTION

The vacancy-related color centers in CMOS-compatible material silicon carbide (SiC) are perspective for chip-scale quantum technologies based on ensembles as well as on single centers. Similar to the spin S = 1 nitrogen-vacancy (NV) defect in diamond – which has become a standard solid-state system for the application of quantum sensing under ambient conditions – the silicon vacancy (VSi) in SiC possesses selectively addressable spin states through optically detected magnetic resonance (ODMR). Unlike the spin-1 defects, the higher half-integer spin S = 3/2 of VSi provides additional degree of freedom and functionality, but it is usually unutilized. A major obstacle is that the structure of high-spin centers being far more complicated is not known yet. Meanwhile, it is the level fine structure that is the key to understand spin dynamics and relaxation processes, which sets up limits for the performance of potential devices.

Here, we reveal the fine structure of the VSi ground and excited states (GS and ES, respectively) in external magnetic fields. We show that the C₃ᵥ point group of the VSi defect gives rise to additional terms in the spin Hamiltonian, which have not been considered so far. Particularly, the trigonal pyramidal symmetry of the VSi defect enables spin transitions with a change of the spin projection Δm_S = ±2. As compared to the commonly studied spin transitions with Δm_S = ±1, they are induced by counter circularly polarized radiation and their energies shift with the double slope in a magnetic field. Moreover, we observe two GS level anticrossings between the m_S = −3/2 and both m_S = −1/2 (GSLAC-1) and m_S = +1/2 (GSLAC-2) spin sublevels. The GSLAC-2 can fundamentally occur for color centers with the spin S ≥ 3/2 only. We develop a theory of the VSi fine structure, which precisely takes into account the real atomic arrangement of the vacancy and quantitatively describes the experimental findings. The photoluminescence (PL) intensity demonstrates resonance-like behavior in the vicinity of LACs, and the sharpest resonance is detected for GSLAC-2, determined by the parameters related to the trigonal pyramidal symmetry of the VSi center. In the following, we show that this optical phenomenon can be used to measure dc magnetic fields without a need to apply radiofrequency (RF) fields and we demonstrate nanotesla resolution within sub-1000 μm³. The effect is robust up to at least 500 K, suggesting a simple, contactless method to monitor weak magnetic fields in a broad temperature range.

Our approach is easily-scalable, and for a probe volume on the order of 1 mm³ with improved pump/collection ef-
ficiency, we expect magnetic field sensitivity to be about hundred femtotesla per square root of Hertz. While coming close to the sensitivity of other benchmark chip-scale magnetic field sensors [43 45], this technique relies neither on RF fields, as for the NV defects in diamond [24], nor on vapour heating, as for the microfabricated rubidium cells [35]. Furthermore, the proposed method is not restricted to magnetic sensing and can potentially be extended for radiofrequency-free sensing of other physical quantities, particularly temperature and axial stress.

II. EXPERIMENT

In natural SiC, the ODMR spectra of the V_{Si} defects are affected by the hyperfine interaction with the 29Si-isotope nuclear spin \( I = 1/2 \) [33 36]. In order to elude this interaction, we use isotopically purified SiC with above 99.0% of 28Si nuclei with \( I = 0 \). To obtain such a crystal, we first synthesize polycrystalline SiC with the use of silicon and carbon powders, the former being enriched with the 28Si isotope. The polycrystalline substance is then used as a source for the growth of 4H-28SiC crystals by the sublimation method in a tantalum container [37]. The growth is performed in vacuum on 4H-SiC substrates at a temperature of 2000°C. The growth rate is approximately 0.25 mm/hour. Afterwards, we polish out the substrate, obtaining the sample with a thickness of about 500 µm. In order to introduce the silicon vacancies, the sample is irradiated with neutrons in a nuclear reactor with a fluence of \( 1 \times 10^{16} \text{ cm}^{-2} \), resulting in a nominal V_{Si} density of \( 2 \times 10^{14} \text{ cm}^{-3} \) [27].

To optically address the V_{Si} spin states, we use a 785-nm laser diode. The optical excitation followed by the spin-dependent recombination leads to a preferential population of the spin-dependent recombination leads to a preferential population of the spin sublevels \( m_S = \pm 1/2 \) along the crystal symmetry c-axis [1]. The PL from V_{Si} occurs in the near infrared spectral range [38], and it is selected and detected using a Si photodiode and a 900-nm long-pass filter. The PL intensity is spin dependent: in case of the V_{Si} center studied here — so-called V2 center — it is higher when the system is in the \( m_S = \pm 3/2 \) states and lower when the system is in the \( m_S = \pm 1/2 \) states [3 16 36]. The laser beam is focused onto the sample using a 20× optical objective (N.A. = 0.3), optimized for the near infrared light, and the PL is collected through the same objective. The nominal excitation volume is 330 µm². To additionally manipulate the V_{Si} spin states, we apply RF field, provided by a signal generator. RF is then amplified, guided to a 500-µm-thick stripline and terminated with a 50-Ω impedance. A static magnetic field can be applied in an arbitrary direction, using a 3D coil arrangement in combination with a permanent magnet. The field direction and strength are calibrated using a 3D Hall sensor.

In the absence of an external magnetic field, the V_{Si} GS is split in two Kramers degenerate spin sublevels \( m_S = \pm 3/2 \) and \( m_S = \pm 1/2 \) with the zero-field splitting \( 2D = 70 \text{ MHz} \) [16 36]. When an external magnetic field \( B \) is applied parallel to the c-axis, the spin states are further split and the splitting is linear with \( B \) (\( z|c \)), as schematically shown in Fig. 1(a). A resonance RF induces magnetic dipole transitions between the spin-split sublevels (\( \pm 1/2 \rightarrow \pm 3/2 \)), resulting in a change of the PL intensity (\( \Delta PL \)). The room-temperature evolution of the ODMR spectrum (i.e., the RF-dependent ODMR contrast \( \Delta PL/PL \)) with the external magnetic field \( B_z \) is presented in Fig. 1(b).

We first discuss the case of \( B_z = 0 \) [Fig. 1(c)]. At a low RF power of 9 dBm, we detect a single ODMR line at a frequency \( \nu_0 = 70 \text{ MHz} \), which is equal to 2\( D \) in the GS. At much higher RF power (40 dBm), we detect another ODMR line at a frequency of 410 MHz. Below, we establish that it corresponds to the zero-field splitting \( 2D' \) in the ES [Fig. 1(a)]. We would like to mention that similar resonance was observed previously [16] and ascribed to the Frenkel pair [39].

Upon application of a magnetic field \( B_z \), one of the RF driven transitions is (\( -1 \rightarrow -3/2 \)) with \( \Delta m_S = -1 \), and the corresponding ODMR line \( \nu_1 = \left| \nu_0 - g_1\mu_B B_z/h \right| \) shifts linearly with \( B_z \). Another RF-driven transition is (\( +1 \rightarrow +3/2 \)) with \( \Delta m_S = +1 \), and the corresponding ODMR line \( \nu_2 = \nu_0 + g_2\mu_B B_z/h \) shifts linearly towards higher frequencies. These two transitions are indicated by the thick arrows in Fig. 1(a) and the corresponding ODMR lines are clearly seen in Fig. 1(b), being in agreement with the previous results [16 21]. Remarkably, at a magnetic field \( B_{G1} = h\nu_0/g_1\mu_B = 2.5 \text{ mT} \) the frequency of the \( \nu_1 \) ODMR line tends to zero [Fig. 1(b)], which is due to the GSLAC-1 [Fig. 1(a)].

We analyze the relative contrast of the \( \nu_1 \) and \( \nu_2 \) ODMR lines as a function of \( B_z \) and observe two pronounced dips [Fig. 2(a)]. One of them is at \( B_{G1} = 2.5 \text{ mT} \) (i.e., exactly at GSLAC-1) and the other one is at \( B_{G1} = 15 \text{ mT} \). The dashed lines in Fig. 1(b) represent the calculated evolution of the ODMR spectrum associated with the \( 2D' = 410 \text{ MHz} \) resonance assuming the effective g-factor \( g_1 \approx 2.0 \). As expected, the GSLAC-1 occurs at \( B_{G1} \). We hence can reconstruct the ES spin structure, as shown in Fig. 1(a). It agrees with the conclusion drawn from another recent experiment [20]. The observation of the dip at 15 mT in the \( \nu_1 \) rather than in the \( \nu_2 \) ODMR signal unambiguously determines the order of the spin sublevels in the ES, i.e., the \( m_S = \pm 3/2 \) state has higher energy than the \( m_S = \pm 1/2 \) state (\( D' > 0 \)).

The appearance of dips in the ODMR signal of Fig. 2(a) is explained by modification of the optical pumping cycle in the vicinity of LACs either in the GS or ES, which, in turn, results in a change of the PL intensity, as previously reported for some other systems and techniques [40 45]. This suggests that LACs can be detected even without application of RF, simply by monitoring the PL intensity as a function of \( B_z \). A scheme of this experiment is presented in the inset of Fig. 2(b). In order to increase the sensitivity, we modulate the dc magnetic field \( B_z \) by additionally applying a small oscillating field...
The transition between the magnetic field. The level splittings at both GSLAC-1 and GSLAC-2 grow linearly with magnetic field. The ODMR peaks calculated for the $\Delta m = \pm 2$ transitions in the GS and $\Delta m = \pm 1$ transitions in the ES, respectively. Low-RF-power (9 dBm) and high-RF-power (40 dBm) ODMR spectra in zero magnetic field.

In order to understand the origin of two GSLACs, we measure the evolution of the ODMR spectrum as a function of $B_z$ with higher precision, in particular, with compensated transverse components of the geomagnetic field, $B_x = B_y = 0 \mu T$. The zoom-in of the spectral evolution in the vicinity of GSLAC-1 and GSLAC-2 is shown in Fig. 3(a). Beside the $\nu_1$ line, the ODMR spectrum contains an additional ($\nu_3$) line, corresponding to the spin transition between the $m_S = +1/2$ and $m_S = -3/2$ GSs [sketched by a thin arrow in Fig. 1(a)]. The turning points of the $\nu_1$ and $\nu_3$ lines correspond to GSLAC-1 and GSLAC-2, respectively. The corresponding spectral shifts of the turning points $\nu_{AC}^1$ and $\nu_{AC}^3$, which are direct measures of the splitting values, are clearly detectable in Fig. 3(b) for various perpendicular component $B_\perp$ of the magnetic field. The level splittings at both GSLAC-1 and GSLAC-2 grow linearly with $B_\perp$, but with different slopes [Fig. 3(c)]. We emphasize that the amplitude of the $\nu_3$ line would rapidly tend to zero and the GSLAC-2 would disappear in an uniaxial model of the defect for magnetic fields being only slightly misaligned from the $c$-axis and hence should be vanishingly small in the experiment, assuming this model. We present below the exact calculation of the relative spin transition rates. Contrarily, the $\nu_3$ ODMR line is clearly detectable in Fig. 3(a) with the relative strength of the ODMR transitions $\Delta PL_3/\Delta PL_1 = 0.12 \pm 0.02$ and the most pronounced feature in Fig. 2(b) relates to the GSLAC-2.

While the magnetic disorder caused by hyperfine interaction with the residual $^{29}$Si nuclei or inaccurate orientation of the external magnetic field along the $c$-axis can in principle give rise to GSLAC-2 and the $\nu_3$ line, we conclude that they are not responsible for the above effects. Calculations (see Appendix A) yield that, for the average nuclear field seen by the $V_{Si}$ centers of about $(h/2\mu B) \times 1 \text{ MHz}$ or magnetic field misalignment of $1^\circ$, the contrast ratio between the $\nu_3$ and $\nu_1$ ODMR lines is estimated to be $10^{-3}$, which is by two orders of magnitude lower than that observed in our experiments. Moreover, we observe that the amplitude of the $\nu_3$ line is the same in the natural (presented below) and isotopically purified SiC samples, while the abundance of the spin-carrying $^{29}$Si nuclei differs by a factor of five. Considering these results, a whole new approach to the spin structure of the $V_{Si}$ defect is needed and will be thoroughly assembled below.
TABLE I. The $g$-factors in Hamiltonian \( H \) which, together with the zero-field splitting $2D = 70$ MHz and $g_0 \approx g_\perp \approx 2.0$, describe the GS fine structure of the silicon vacancy (V2 center in 4H-SiC) in a magnetic field.

|       | $g_\parallel$ | $g_\perp$ | $g_3 + g_\perp/2$ | $g_3 - g_\perp/2$ |
|-------|---------------|-----------|-------------------|-------------------|
| 0.0±0.1 | 0.0±0.1       | 0.5±0.2   | −0.1±0.4          |

III. SILICON VACANCY FINE STRUCTURE

Our findings can be explained in the framework of the spin Hamiltonian, which precisely takes into account the real microscopic $C_{3v}$ group symmetry of the defect \[17\]. The effective Hamiltonian to the first order in the magnetic field can be presented as a sum of three contributions

\[
\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{1\parallel} + \mathcal{H}_{1\perp}, \tag{1}
\]

where $\mathcal{H}_0$ is the Hamiltonian in zero magnetic field, $\mathcal{H}_{1\parallel} \propto B_\parallel$ and $\mathcal{H}_{1\perp} \propto B_\perp = (B_x, B_y)$ are the magnetic-field-induced terms. The Hamiltonians $\mathcal{H}_0$, $\mathcal{H}_{1\parallel}$, and $\mathcal{H}_{1\perp}$ can be constructed applying the theory of group representations \[18\], see Appendix \[B\] for details. In the $C_{3v}$ group, the magnetic field component $B_\parallel$ and the spin operator $S_\perp$ transform under the irreducible representation $A_2$, the pairs of the in-plane components $(S_x, S_y)$ and $(B_x, B_y)$ transform under the representation $E$, and the Hamiltonian must be invariant (representation $A_1$). Using the multiplication table for the representations, one can construct all possible invariant combinations of the magnetic field components and the first, second and third powers of the spin operator components. The forth and higher powers of the spin-3/2 operator can be reduced to the operators of lower powers. Finally, taking into account that the Hamiltonian must be invariant with respect to the time reversal and, therefore, $\mathcal{H}_0$ is even in $\mathbf{S}$ while $\mathcal{H}_{1\parallel}$ and $\mathcal{H}_{1\perp}$ are odd in $\mathbf{S}$, we obtain

\[
\mathcal{H}_0 = D \left( S_x^2 - \frac{5}{4} \right),
\]

\[
\mathcal{H}_{1\parallel} = \left[ g_\parallel S_z + g_\parallel S_x \left( S_z^2 - \frac{5}{4} \right) + g_\parallel \frac{S_+^2 - S_-^2}{4i} \right] \mu_B B_\parallel,
\]

\[
\mathcal{H}_{1\perp} = g_\perp \mu_B S_\perp \cdot B_\perp + 2 g_{2\perp} \mu_B \left\{ S_\perp \cdot B_\perp, S_z^2 - \frac{3}{4} \right\} + g_{3\perp} \mu_B \left\{ S_\perp^2, S_z \right\} B_\perp - \left\{ S_\perp^2, S_z \right\} B_\parallel. \tag{2}
\]

Here, $S_x, S_y, S_z$ are the spin-3/2 operators, $S_\perp = (S_x, S_y)$, $S_\parallel = S_z \pm i S_y$, $B_\parallel = B_x \pm i B_y$, \( \{A, B\} = (AB + BA)/2 \) is the symmetrized product, $z$ is parallel to $c$-axis, $x$ and $y$ are the perpendicular axes with $y$ lying in a mirror reflection plane, and $\mu_B$ is the Bohr magneton. Six $g$-factors introduced in Eq. \[2\] are linearly-independent in a structure of the $C_{3v}$ point group. They can be determined from experimental data, as we do below, or obtained from ab-initio calculations, which is out of scope of the present manuscript. The difference $g_\parallel - g_\perp$
The solid lines are calculations as explained in the text. (c) The GSLAC splittings as a function of the magnetic field.

The perpendicular components of the geomagnetic field are compensated, measured for non-zero $B_z$. The vertical arrows indicate the positions of the turning points $\nu^{AC}$ and $\nu^{CC}$, which are a direct measure of the level splittings at GSLAC-1 and GSLAC-2, respectively. (c) The GSLAC splittings as a function of $B_z$. The solid lines are calculations as explained in the text.

as well as the non-zero values of $D$, $g_{2\parallel}$ and $g_{2\perp}$ are due to non-equivalence of the $z$ axis and the perpendicular axes. The $g$-factors $g_{3\parallel}$ and $g_{3\perp}$ emerge due to the trigonal pyramidal symmetry of the defect. Hamiltonian (2) can also be presented in the equivalent matrix form (Appendix C).

The parameters of Hamiltonian (2) can be determined from the experimental data. First from the ODMR lines in the parallel $B_z$ [Fig. 3(b)] and perpendicular $B_z$ (Appendix D) magnetic fields, we confirm that to the second digit accuracy $g_{2\parallel}, g_{2\perp} \approx 2.0$, in agreement with earlier studies [38] and $|g_{2\parallel}|, |g_{2\perp}| \ll 1$. Then, using a procedure that is independent of the values of $g_{2\parallel}$ and $g_{2\perp}$, we estimate the ratios $g_{3\parallel}/g_{2\parallel}$ and $g_{3\perp}/g_{2\perp}$ using Eqs. (3) and (5) (Appendix D).

Furthermore, the Hamiltonian (2) describes the opening of spectral gaps due to the perpendicular field component $B_{\perp}$ at GSLAC-1 and GSLAC-2. The corresponding splittings $\Lambda_1$ and $\Lambda_2$ scale linearly with the perpendicular field for $\mu_B B_{\perp} \ll D$. Up to linear terms in $g_{2\parallel}, g_{2\perp}$ and quadratic terms in $g_{3\parallel}, g_{3\perp}$ the splittings in small fields are given by

$$\Lambda_1 \approx \sqrt{3} \left( 1 + \frac{g_{2\perp}}{g_{2\parallel}} - \frac{g_{3\parallel}}{g_{2\parallel}} \frac{g_{3\perp}}{g_{2\perp}} - \frac{g_{3\parallel}^2}{8g_{2\perp}^2} \right) g_{3\perp} \mu_B B_{\perp},$$

$$\Lambda_2 \approx \sqrt{3} \left( \frac{g_{3\parallel}}{g_{2\parallel}} + \frac{g_{3\perp}}{2g_{2\perp}} \right) g_{3\perp} \mu_B B_{\perp}. \quad (3)$$

The GSLAC-2 emerges due to the trigonal asymmetry of the silicon vacancy, and the corresponding energy splitting is expected to be smaller than that in the GSLAC-1, $\Lambda_2 < \Lambda_1$. Exactly such a behavior is observed in the experiment of Fig. 3. We fit the positions of the turning points as $\nu^{AC}_i = [\Lambda_i^2(2) + \Lambda_i^4]^{1/2}$. Here $\Lambda_i/h = 2.5$ MHz accounts for finite ODMR linewidth and inhomogeneity. From the best fit [the solid lines in Fig. 3(c)] we first obtain $g_{3\parallel} + g_{3\perp}/2 = 0.5 \pm 0.2$ using the data for $\Lambda_2$ and then estimate the value $g_{3\perp} - g_{3\parallel}/2 = -0.1 \pm 0.4$ using the data for $\Lambda_1$. All the $g$-factors of the Hamiltonian (2) are summarized in table I. It is instructive to compare the results with a high-symmetry defect of the $T_d$ point group, where one expects the relation $g_{3\parallel}/g_{3\perp} = 2/3$ (Appendix C).

We are now in the position to explain the appearance of the $\nu_3$ and $\nu_4$ ODMR lines in Figs. 3(b), 3(a) and 3(b), even when $B_{\perp} = 0$. It follows from the Hamiltonian (2) that the matrix elements of the allowed magnetic dipole transitions have the form

$$M_{\mp 3/2, \mp 1/2} = \sqrt{3} \left( 1 + \frac{g_{2\perp}}{g_{2\parallel}} \right) g_{3\perp} \mu_B B_{1,\sigma^\pm},$$

$$M_{\mp 3/2, \pm 1/2} = -i \sqrt{3} \left( \frac{g_{3\parallel}}{g_{2\parallel}} + \frac{g_{3\perp}}{2g_{2\perp}} \right) g_{3\perp} \mu_B B_{1,\sigma^\pm}, \quad (4)$$

where $B_1$ is the RF magnetic field and $B_{1,\sigma^\pm} = B_{1,x} \mp iB_{1,y}$. The transitions $(+1/2 \rightarrow -3/2)$ and $(-1/2 \rightarrow -3/2)$
+3/2), responsible for the $\nu_3$ and $\nu_4$ ODMR lines, respectively, occur due to the trigonal pyramidal symmetry of the spin-3/2 defect and are induced by the $\sigma^+$ and $\sigma^-$ circularly polarized RF radiation. There are two microscopic contributions to these transitions: (i) coupling of the $m_S = +3/2$ and $m_S = -3/2$ states by the longitudinal static field $B_z$ (parameter $g_{3l}$)\cite{49,50} followed by the RF driven transitions with $\Delta m = \pm 1$ and (ii) direct coupling of the $m_S = +3/2$ and $m_S = -1/2$ as well as $m_S = -3/2$ and $m_S = +1/2$ states by the transverse RF magnetic field (parameter $g_{3l}$). Far from LACs, the ratio of the $\nu_3$ and $\nu_4$ ODMR line intensities for linearly polarized RF field is given by $|M_{-3/2, 1/2}/M_{-3/2, -1/2}|^2 \approx (g_{3l} + g_{3l})^2/4$. Using $g_{3l} + g_{3l}/2 = 0.5$ from the fit of $\nu_4^{AC}$ in Fig. 1(c), we obtain for the relative intensity 0.06. It is somewhat smaller than the measured value of $\Delta PL_3/\Delta PL_1 \approx 0.1$. Detailed comparison of the experimental and theoretical ODMR contrasts requires the study of the linewidths and resonance shapes in the vicinity of GSLACs, which is beyond the scope of the present paper. Given this uncertainty, we find the agreement between the theory and experiment satisfactory.

IV. ALL-OPTICAL MAGNETOMETRY

Having established the fine structure, we propose to use its unique properties for all-optical magnetometry. The experimental procedure is straightforward and requires no RF field. First, we tune our system in the GSLAC-2, characterized by the narrowest resonance in Fig. 2(b). We then monitor the PL intensity through the lock-in in-phase photovoltage $U_x$, which is simply proportional to the deviation of the measured magnetic field from the bias field $B_{G2}$ (provided this deviation is small) [Fig. 2(a)]. By applying sub-$\mu$T magnetic fields, we calibrate the lock-in signal $U_x/(B_z - B_{G2}) = 39 \mu V/\mu T$ [Fig. 3(b)]. The quadrature component $U_y$ of the lock-in signal, being independent of the magnetic field, is used to measure the noise level. Each data point in Fig. 3(b) corresponds to an integration time of 4 s, and the dc magnetic field sensitivity is obtained to be $\delta B = 87 \text{nT}/\sqrt{\text{Hz}}$. Indeed, magnetic fields below 100 nT can be clearly resolved in Fig. 3(b).

We use an isotopically enriched crystal to exclude possible contributions related to the hyperfine interaction with $^{29}$Si nuclei to spin Hamiltonian of Eq. (2). Figure 5 demonstrates that the proposed all-optical magnetometry can also be performed using SiC with natural isotope abundance. By alignment of the bias magnetic field along the symmetry axis with an accuracy better that one degree (the inset of Fig. 5(a)), it is possible to clearly separate the spin-carrying isotope contributions. The GSLAC-2 is generally narrower and less sensitive to the magnetic field misalignment in comparison to the GSLAC-1, as can be seen in Fig. 5(b). It is indeed expected that $\Lambda_2 < \Lambda_1$, because the peak-to-peak width $\Delta$, as determined in Fig. 5(a), scales with the LAC splitting $\Lambda$ and according to Eq. (3) $\Lambda_2 < \Lambda_1$.

The dynamic range of the proposed magnetometry is relatively small, several tens of $\mu$T. It can be extended by applying a transverse magnetic field at the expense of sensitivity. On the other hand, there are many applications where large dynamic range is not required\cite{51}. We would like to clarify that the proposed magnetometry is highly sensitive to one particular orientation of the magnetic field ($B_z$) and, therefore, designed for applications where weak magnetic variations in a certain direction must be measured with high accuracy. In order to align the magnetometer, it is necessary to conduct several preliminary measurements of magnetic field sweeps around $B_{G2}$ in different orientation bias magnetic fields, until the maximal slope is obtained. An advantage is that the LAC can be observed even for short spin lifetimes which occur, e.g., in excited states. The detection of the ODMR signal in these conditions may be difficult because it would require the application of highly intense RF fields. Contrary, a variation of the PL intensity at ESLAC when the ODMR signal is not detectable has been clearly demonstrated using GaAs/AlGaAs superlattices\cite{12}. Our approach is robust and can be applied in a broad temperature range up to 520 K [Fig. 4(a)]. A crucial factor for field sensitivity is the PL intensity and stability of the pump laser. The latter factor can be compensated using a balanced detection scheme. By increasing the irradiation fluence, the $V_{Si}$ density can be increased by more than two orders of magnitude\cite{27}, and the projected sensitivity in this case is a few nT/$\sqrt{\text{Hz}}$ within...
the same volume of 330 μm³. Alternatively, one can use light-trapping waveguides in bigger samples [34]. For a waveguide of 3 mm × 3 mm × 300 μm with improved collection efficiency by three orders of magnitude [24] and a VSi density of 4 × 10¹⁶ cm⁻³ [27], we estimate the projection noise limit to be below 100 fT/√Hz. In order to realize such an extremely high sensitivity, drift-compensation schemes [31, 34] and magnetic noise screening similar to that usually used for optical magnetometry based on vapour cells [35] are necessary to be applied. The use of completely spin-free samples of high crystalline quality, containing ²⁸Si and ¹²C isotopes only, can lead to further improvement due to the suppression of magnetic fluctuations caused by nuclear spins. In addition, rhombic SiC polytypes (15R) with even lower magnetic fields corresponding to the GSLACs [46] may have advantage compared to hexagonal 4H-SiC.

In conclusion, we reconstruct for the first time the fine structure of the VSi center in 4H-SiC, quantifying its contributions. The presence and behaviour of two GSLACs [46] may have advantage corresponding to the GSLACs [46] may have advantage

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Appendix A: Spin transitions in the uniaxial model

In the uniaxial approximation, the effective Hamiltonian of a spin center has the form

$$H = D \left( S_x^2 - \frac{5}{4} \right) + g_\parallel \mu_B S_z B_z + g_\perp \mu_B \mathbf{S} \cdot \mathbf{B}$$, \hspace{1cm} (A1)

where $g_\parallel$ and $g_\perp$ are the longitudinal and transverse $g$-factors, $B_z$ and $\mathbf{B} = (B_x, B_y)$ are the longitudinal and transverse components of the magnetic field, respectively, and $\mathbf{S}$ is the vector composed of the spin-3/2 operators $S_x$, $S_y$, and $S_z$.

The transverse component of the magnetic field caused by inaccurate orientation of the external magnetic field along the c-axis and/or originated from hyperfine interaction with nuclei couples the +1/2 and −1/2 spin states as well as the −1/2 and −3/2 spin states thereby allowing the $\nu_3$ ODMR line. Straightforward perturbation-theory calculations show that the ratio between the intensities of the $\nu_3$ and $\nu_1$ ODMR lines determined by the corresponding matrix elements of the transitions has the form

$$\frac{|M_{-3/2, +1/2}|^2}{|M_{-3/2, -1/2}|^2} = \left( \frac{2D - g_\parallel \mu_B B_z}{2D - g_\parallel \mu_B B_z \frac{g_\perp B_x}{g_\perp B_z}} \right)^2$$. \hspace{1cm} (A2)

The experimental value of the relative ODMR contrast is about 0.1. To obtain such a contract, e.g., for $g_\parallel \mu_B B_z = D$, one has to assume that $B_z \approx 0.16$ which corresponds to an angle of approximately 10° between the total magnetic field acting upon spin centres and the c-axis or $B_z \approx 0.2$ mT for $B_z \approx 1.25$ mT. Such a transverse magnetic field is an order of magnitude larger than the average nuclear field seen by the VSi centers [46]. The precision of the external magnetic orientation in our experiments is also by an order of magnitude better [the inset of Fig. 5(a)]. For such a precision, the contrast ratio between the $\nu_3$ and $\nu_1$ ODMR lines is estimated from Eq. (A2) to be at most 0.001, which is by two orders of magnitude lower than the experimentally determined ratio.

Appendix B: Effective Hamiltonian for the spin-3/2 center of the $C_{3v}$ point group

We construct the effective spin Hamiltonian using the theory of group representations [35]. In the $C_{3v}$ point group, there are three irreducible representations commonly denoted as $A_1$, $A_2$, and $E$. Accordingly, all physical quantities can be decomposed into the irreducible representations in accordance with their symmetry properties. The magnetic field components $B_\alpha$ ($\alpha = x, y, z$) and all possible linearly independent combinations of the spin operator components $S_\alpha$ are decomposed into the

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irreducible representations as follows

\[ A_1 : S_x^2; \quad S_y(3S_x^2 - S_y^2); \quad (B1) \]
\[ A_2 : B_z; \quad S_x^3; \quad S_y(3S_x^2 - S_y); \]
\[ E : (B_x, B_y); (S_x, S_y); (S_yS_z - S_zS_y); (S_zS_yS_z - S_yS_z); \]
\[ (S_x^2 - S_y^2) - 2(S_xS_y), (2S_xS_yS_z, (S_z^2 - S_y^2)S_z). \]

All other combinations of the spin operator components can be expressed via the above ones taking into account the identity \( S_x^2 + S_y^2 + S_z^2 = 15/4 \) and the fact that the forth and higher powers of the spin-3/2 operator components can be reduced to the operators of lower powers.

The effective Zeeman Hamiltonian is constructed as the sum of all possible products of the magnetic field components and the spin operator combinations which are invariant with respect to (i) symmetry operations of the point group and (ii) time reversal. Each invariant contribution is multiplied by a prefactor which has a physical sense of an effective \( g \)-factor component. The condition (i) implies that the invariant products are constructed from quantities belonging to the same irreducible representation. The condition (ii) implies that the Zeeman Hamiltonian is odd in \( S \). Taking both conditions into account we obtain 6 linearly independent contributions to the Zeeman Hamiltonian which are given in Eq. (2).

**Appendix C: Fine structure of spin-3/2 centers of the \( C_{3v} \) point group**

The spin Hamiltonian given by Eqs. (1)-(2) can be rewritten in the equivalent matrix form

\[
\mathcal{H} = \begin{pmatrix}
D + \frac{3}{2}(1 + \frac{g_\|}{g_\perp})g_\| \mu_B B_z & \frac{\sqrt{3}}{2}(1 + \frac{g_\perp}{g_\perp})g_\perp \mu_B B_z & -i\frac{\sqrt{3}}{2}g_\perp \mu_B B_z & -i\frac{\sqrt{3}}{2}g_\| \mu_B B_z \\
\frac{\sqrt{3}}{2}(1 + \frac{g_\perp}{g_\perp})g_\perp \mu_B B_z & -D + \frac{3}{2}(1 - \frac{g_\perp}{g_\perp})g_\perp \mu_B B_z & (1 - \frac{g_\perp}{g_\perp})g_\perp \mu_B B_z & i\frac{\sqrt{3}}{2}g_\| \mu_B B_z \\
i\frac{\sqrt{3}}{2}g_\perp \mu_B B_z & (1 - \frac{g_\perp}{g_\perp})g_\perp \mu_B B_z & -D - \frac{3}{2}(1 - \frac{g_\perp}{g_\perp})g_\perp \mu_B B_z & \frac{\sqrt{3}}{2}(1 + \frac{g_\perp}{g_\perp})g_\perp \mu_B B_z \\
i\frac{\sqrt{3}}{2}g_\perp \mu_B B_z & i\frac{\sqrt{3}}{2}g_\perp \mu_B B_z & \frac{3}{2}(1 + \frac{g_\perp}{g_\perp})g_\perp \mu_B B_z & D + \frac{3}{2}(1 + \frac{g_\perp}{g_\perp})g_\perp \mu_B B_z
\end{pmatrix},
\]

where \( B_\pm = B_x \pm iB_y = B_\perp e^{\pm i\phi} \), with \( \phi \) denoting the azimuthal angle of \( B \). To derive this matrix, we use the explicit form of the spin-3/2 matrices

\[
S_x = \begin{pmatrix}
0 & \frac{\sqrt{3}}{2} & 0 & 0 \\
\frac{\sqrt{3}}{2} & 0 & 1 & 0 \\
0 & 1 & 0 & \frac{\sqrt{3}}{2} \\
0 & 0 & \frac{\sqrt{3}}{2} & 0
\end{pmatrix}, \quad \text{ (C2)}
\]
\[
S_y = \begin{pmatrix}
0 & -i\frac{\sqrt{3}}{2} & 0 & 0 \\
i\frac{\sqrt{3}}{2} & 0 & -i & 0 \\
0 & i & 0 & -i\frac{\sqrt{3}}{2} \\
0 & 0 & i\frac{\sqrt{3}}{2} & 0
\end{pmatrix}, \quad \text{ (C3)}
\]
\[
S_z = \begin{pmatrix}
\frac{3}{2} & 0 & 0 & 0 \\
0 & \frac{1}{2} & 0 & 0 \\
0 & 0 & -\frac{1}{2} & 0 \\
0 & 0 & 0 & -\frac{3}{2}
\end{pmatrix}, \quad \text{ (C4)}
\]

1. **Zeeman splitting of spin sublevels**

Application of a magnetic field along the c-axis leads to the splitting of the spin sublevels. The energies of the states with the spin projections \( m_s = \pm 1/2 \) and \( m_s = \pm 3/2 \) are given by

\[
E_{\pm 1/2} = -D \pm \frac{1}{2}g_\| \mu_B B_z, \quad \text{ (C5)}
\]
\[
E_{\pm 3/2} = D \pm \frac{3}{2}g_\| \mu_B B_z, \quad \text{ (C6)}
\]

where the effective \( g \)-factors are

\[
g_\| = g_\|, \quad \text{ (C7)}
\]
\[
g_\| = \sqrt{(g_\| + g_\perp)^2 + g_\perp^2}. \quad \text{ (C8)}
\]

The spin sublevel \( m_s = -3/2 \) crosses the spin sublevels \( m_s = -1/2 \) and \( m_s = +1/2 \) at the magnetic fields

\[
B_{G1} = \frac{4D}{(3g_\| + g_\perp)\mu_B} \quad \text{and} \quad \text{ (C9)}
\]
\[
B_{G2} = \frac{4D}{(3g_\| + 2g_\perp)\mu_B}, \quad \text{ (C10)}
\]

respectively.

As described in the main text, application of a small additional perpendicular magnetic field \( B_\perp \) leads to level anticrossings, GSLAC-1 and GSLAC-2 [Fig. 4(a)], at \( B_{G1} \) and \( B_{G2} \), respectively.

In a magnetic field applied perpendicular to the c-axis \( B_\perp = (B_x^2 + B_y^2)^{1/2} \) and \( B_z = 0 \), the energy spectrum is given by
Comparing the Hamiltonians (C1) and (C16) we obtain
\[ E_{3/2} = \pm \frac{1}{2}(g_\perp - g_{2\perp})\mu_B B_\perp + \frac{1}{2}\sqrt{2D \mp (g_\perp - g_{2\perp})\mu_B B_\perp}^2 + 3[(g_\perp + g_{2\perp})^2 + g_{3\perp}^2]\mu_B^2 B_\perp^2, \]
\[ E_{1/2} = \pm \frac{1}{2}(g_\perp - g_{2\perp})\mu_B B_\perp - \frac{1}{2}\sqrt{2D \mp (g_\perp - g_{2\perp})\mu_B B_\perp}^2 + 3[(g_\perp + g_{2\perp})^2 + g_{3\perp}^2]\mu_B^2 B_\perp^2. \] (C11)

Particularly, for small magnetic fields \((\mu_B B_\perp \ll 2D)\), the linear-in-\(B_\perp\) splitting is described by the effective \(g\)-factors
\[ g_{\perp,3/2} = 0, \quad (C12) \]
\[ g_{\perp,1/2} = 2(g_\perp - g_{2\perp}). \] (C13)

2. Relation between \(g\)-factors in the \(T_d\) point group

The spatial arrangement of carbon atoms around the single silicon vacancy is close to the tetragonal structure, which is described by the \(T_d\) point group symmetry. The \(T_d\) group symmetry is higher than the real \(C_{3v}\) group symmetry of the vacancy but properly takes into account the three-fold rotation \(c\)-axis and allows for non-zero values of both \(g_{3\parallel}\) and \(g_{3\perp}\). Thus, one can expect that the relation between \(g_{3\parallel}\) and \(g_{3\perp}\) of the Si vacancy is close to that for the defect of the \(T_d\) point group.

In the \(T_d\) point group, the effective Zeeman Hamiltonian of spin-3/2 defect in the cubic axes \(x' \parallel [100]\), \(y' \parallel [010]\), and \(z' \parallel [001]\) is described by two linearly independent parameters \(g\) and \(q\) and reads \([52]\)
\[ \mathcal{H}_{T_d} = g\mu_B \mathbf{S} \cdot \mathbf{B} + q\mu_B \left( J_{x'}^3 B_{x'} + J_{y'}^3 B_{y'} + J_{z'}^3 B_{z'} - \frac{41}{20} \mathbf{S} \cdot \mathbf{B} \right). \] (C14)

In order to obtain the Hamiltonian in the axes \(x \parallel [1\bar{1}0]\), \(y \parallel [112]\), and \(z \parallel [111]\), relevant to the vacancy orientation, we use the relation between the components of the vector \(\mathbf{B}\) in two coordinate frames,
\[ B_{x'} = \frac{1}{\sqrt{2}}B_x + \frac{1}{\sqrt{6}}B_y + \frac{1}{\sqrt{3}}B_z, \]
\[ B_{y'} = -\frac{1}{\sqrt{2}}B_x + \frac{1}{\sqrt{6}}B_y + \frac{1}{\sqrt{3}}B_z, \]
\[ B_{z'} = -\frac{2}{\sqrt{6}}B_y + \frac{1}{\sqrt{3}}B_z, \] (C15)
and similar equations for the components of the spin operator \(\mathbf{S}\). This yields
\[ \mathcal{H}_{T_d} = \mu_B \begin{pmatrix} \left( \frac{1}{2}g - \frac{1}{5}q \right) B_z & \frac{\sqrt{3}}{2} \left( \frac{1}{2}g + \frac{3}{5}q \right) B_+ & -\sqrt{\frac{3}{8}}iqB_+ & -\frac{1}{\sqrt{2}}iqB_+ \\ \frac{\sqrt{3}}{2} \left( \frac{1}{2}g + \frac{3}{5}q \right) B_- & \left( \frac{3}{2}g + \frac{1}{5}q \right) B_z & \left( \frac{g - \frac{1}{5}q \right) B_- & \sqrt{\frac{3}{8}}iqB_+ \\ \frac{1}{\sqrt{2}}iqB_+ & -\sqrt{\frac{3}{8}}iqB_- & \left( \frac{3}{2}g + \frac{1}{5}q \right) B_z & \left( \frac{1}{2}g - \frac{1}{5}q \right) B_- \\ \frac{1}{\sqrt{2}}iqB_+ & -\sqrt{\frac{3}{8}}iqB_- & \left( \frac{1}{2}g - \frac{1}{5}q \right) B_- & \left( \frac{3}{2}g + \frac{1}{5}q \right) B_z \end{pmatrix}. \] (C16)

Comparing the Hamiltonians \([C1]\) and \([C16]\) we obtain that \(g_{3\parallel}\) and \(g_{3\perp}\) are related to each other by
\[ g_{3\parallel}/g_{3\perp} = 2/3 \] (C17)
for a defect of the \(T_d\) group symmetry.

Appendix D: Extracting the fine structure parameters

To obtain the value of \(g_{2\parallel}\), we use the ODMR spectra, recorded in the magnetic field applied parallel to the \(c\)-axis. The linear shift of the ODMR lines is given by
\[ \nu_{1,2} = \nu_0 \pm \left( \frac{3}{2}g_{\parallel,3/2} - \frac{1}{2}g_{\parallel,1/2} \right) \mu_B B_\parallel/h \]
\[ \approx \nu_0 \pm (g_\parallel + 2g_{2\parallel}) + 3g_{3\parallel}^2/4g_\parallel) \mu_B B_\parallel/h, \] (D1)
\[ \nu_{1,4} = \nu_0 \pm \left( \frac{3}{2}g_{\parallel,3/2} + \frac{1}{2}g_{\parallel,1/2} \right) \mu_B B_\parallel/h \]
\[ \approx \nu_0 \pm (2g_\parallel + 2g_{2\parallel} + 3g_{3\parallel}^2/4g_\parallel) \mu_B B_\parallel/h, \] (D2)
where \(\nu_0 = 2D/h\). The experimentally measured ratio of the magnetic fields corresponding to the GSLAC-2 and GSLAC-1 points, \(B_{G2}/B_{G1} = 0.503 \pm 0.005\) independent of the magnetic field calibration, allows us to extract the
The matrix element for \(-\text{ODMR spectrum in the magnetic field } B\) is zero (for first iteration). We neglect the term \(\propto \nu B\) linear fit to Eq. (D5) with a slope of 0.995 ± 0.01.

value of \(g_{\square}/g\) using the formula

\[
\frac{B_{G2}}{B_{G1}} \approx \frac{1}{2} + \frac{3g_{\|}}{4g} + \frac{3g_{\|}^2}{16g^2}.
\]

For the first iteration we neglect the term \(\propto g_{\|}^2\).

To determine \(g_{\|}\), we exploit the evolution of the ODMR spectrum in the magnetic field \(B_{\|}\) applied perpendicular to the \(c\)-axis, presented in Fig. 6(a). From Eq. (E1) we obtain the positions of the \(\nu_1\) and \(\nu_2\) ODMR lines up to \(B_{\|^2}\)

\[
\nu_{1,2} = \nu_0 \mp (g_{\|} - g_{\perp}) \frac{\mu_B B_{\|^2}}{\hbar} + \frac{3((g_{\|} + g_{\perp})^2 + g_{\parallel}^2)}{2\nu_0} \left(\frac{\mu_B B_{\|^2}}{\hbar}\right)^2.
\]

One can represent the quadratic shift \((\nu_2 + \nu_1)/2\) vs the Zeeman splitting \(\nu_2 - \nu_1 = 2(g_{\|} - g_{\perp})B_{\|^2}/\hbar\), using the theoretical expression

\[
\frac{\nu_2 + \nu_1}{2} - \nu_0 = \frac{3((g_{\|} + g_{\perp})^2 + g_{\parallel}^2)}{8(g_{\|} - g_{\perp})^2} \left(\nu_2 - \nu_1\right) / \nu_0 \\
\approx \left(1 + \frac{4g_{\parallel}}{g_{\perp}} + \frac{g_{\parallel}^2}{g_{\perp}^2}\right) \frac{3(\nu_2 - \nu_1)^2}{8\nu_0}.
\]

From the linear fit of the experimental data in Fig. 6(b), we obtain the value of \(g_{\|}/g_{\perp}\). Again, for the first iteration we neglect the term \(\propto g_{\|}^2\).

Finally, we use the GSLAC-1 and GSLAC-2 splittings, described by Eqs. (3), to determine the values of \(g_{\parallel}\) and \(g_{\perp}\). By iterating the described procedure, we obtain the values of the \(g\)-factors, as presented in table I.

Appendix E: Electron spin resonance

The spin transition rates induced by the RF magnetic field \(B\) are determined by the matrix elements of Hamiltonian (C1). For a static magnetic field \(B \| z\), the matrix elements of the transitions up to linear in \(g_{\parallel}, g_{\|1}, g_{\perp}\), and \(g_{\perp}\) terms are given by Eqs. (4)-(6) in the main text, and the full expressions have the form

\[
M_{-3/2, -1/2} = \sqrt{\frac{3}{8}} \left[ (g_{\|} + g_{\perp}) \sqrt{1 + \frac{g_{\|}}{g_{\|1}} + \frac{g_{\perp}}{g_{\perp1}}} \right] \mu_B (B_{1,x} + iB_{1,y}) ,
\]

\[
M_{3/2, 1/2} = \sqrt{\frac{3}{8}} \left[ (g_{\|} + g_{\perp}) \sqrt{1 + \frac{g_{\|}}{g_{\|1}} + \frac{g_{\perp}}{g_{\perp1}}} \right] \mu_B (B_{1,x} - iB_{1,y}) ,
\]

\[
M_{-3/2, 1/2} = -i \sqrt{\frac{3}{8}} \left[ \frac{g_{\|}(g_{\|} + g_{\perp})}{\sqrt{g_{\|1}^2 + g_{\perp}^2 + g_{\perp1}^2}} + g_{\perp} \sqrt{1 + \frac{g_{\|}}{g_{\perp1}}} \right] \mu_B (B_{1,x} - iB_{1,y}) ,
\]

\[
M_{3/2, -1/2} = -i \sqrt{\frac{3}{8}} \left[ \frac{g_{\parallel}(g_{\|} + g_{\perp})}{\sqrt{g_{\|1}^2 + g_{\perp}^2 + g_{\perp1}^2}} + g_{\perp} \sqrt{1 + \frac{g_{\|}}{g_{\perp1}}} \right] \mu_B (B_{1,x} + iB_{1,y}) .
\]

The matrix element for \((-1/2 \leftrightarrow +1/2\) has the form

\[
M_{1/2, -1/2} = (g_{\|} - g_{\perp}) \mu_B (B_{1,x} - iB_{1,y}) ,
\]

while the spin transition \((-3/2 \leftrightarrow +3/2\) is forbidden for \(B \| z\).

[1] P G Baranov, A P Bundakova, I V Borovykh, S B Or-llinskii, R Zondervan, and J Schmidt, “Spin polarization

induced by optical and microwave resonance radiation in
a Si vacancy in SiC: A promising subject for the spectroscopy of single defects,” Journal of Experimental and Theoretical Physics Letters 86, 202–206 (2007).

[2] J R Weber, W F Koehl, J B Varley, A Janotti, B B Buckley, C G Van de Walle, and D D Awschalom, “Quantum computing with defects,” Proceedings of the National Academy of Sciences 107, 8513–8518 (2010).

[3] Pavel G Baranov, Anna P Bundakova, Alexandra A Soltamova, Sergei B Orlinskii, Igor V Borovyykh, Rob Zondervan, Rogier Verberk, and Jan Schmidt, “Silicon vacancy in SiC as a promising quantum system for single-defect and single-photon spectroscopy,” Physical Review B 83, 125203 (2011).

[4] D Riedel, F Fuchs, H Kraus, S Váth, A Sperlich, V Dyakonov, A Soltamov, P Baranov, V Ilyin, and G V Astakhov, “Resonant Addressing and Manipulation of Silicon Vacancy Qubits in Silicon Carbide,” Physical Review Letters 109, 226402 (2012).

[5] F Fuchs, V A Soltamov, S Váth, P G Baranov, E N Mokhov, G V Astakhov, and V Dyakonov, “Silicon carbide light-emitting diode as a prospective room temperature source for single photons,” Scientific Reports 3, 1637 (2013).

[6] Stefania Castelletto, Brett C Johnson, and Alberto Boretti, “Quantum Effects in Silicon Carbide Hold Promise for Novel Integrated Devices and Sensors,” Advanced Optical Materials 1, 609–625 (2013).

[7] Bólint Somogyi and Adam Gali, “Computational design of in vivobiomarkers,” Journal of Physics: Condensed Matter 26, 143202 (2014).

[8] A Muzha, F Fuchs, N V Tarakina, D Simin, M Trupke, V A Soltamov, E N Mokhov, P G Baranov, V Dyakonov, A Krueger, and G V Astakhov, “Room-temperature near-infrared silicon carbide nanocrystal emitters based on optically aligned spin defects,” Applied Physics Letters 105, 243112 (2014).

[9] Greg Calusine, Alberto Politi, and David D Awschalom, “Silicon carbide photonic crystal cavities with integrated color centers,” Applied Physics Letters 105, 011123 (2014).

[10] William F Koehl, Bob B Buckley, F Joseph Heremans, Greg Calusine, and David D Awschalom, “Room temperature coherent control of defect spin qubits in silicon carbide,” Nature 479, 84–87 (2011).

[11] Victor A Soltamov, Alexandra A Soltamova, Pavel G Baranov, and Ivan I Proskuryakov, “Room Temperature Coherent Spin Alignment of Silicon Vacancies in 4H- and 6H-SiC,” Physical Review Letters 108, 226402 (2012).

[12] Abram L Falk, Bob B Buckley, Greg Calusine, William F Koehl, Viatcheslav V Dobrovitski, Alberto Politi, Christian A Zorman, Philip X L Feng, and David D Awschalom, “Polytype control of spin qubits in silicon carbide,” Nature Communications 3, 187601 (2014).

[13] H Kraus, V A Soltamov, F Fuchs, D Simin, A Sperlich, P G Baranov, G V Astakhov, and V Dyakonov, “Magnetic field and temperature sensing with atomic-scale spin defects in silicon carbide,” Scientific Reports 4, 5303 (2014).

[14] Li-Ping Yang, Christian Burk, Matthias Widmann, Sang-Yun Lee, Jörg Wrachtrup, and Nan Zhao, “Electron spin decoherence in silicon carbide nuclear spin bath,” Physical Review B 90, 241203 (2014).

[15] Olger V Zwier, Danny O’Shea, Alexander R Omr, and Caspar H van der Wal, “All-optical coherent population trapping with defect spin ensembles in silicon carbide,” Scientific Reports 5, 10931 (2015).

[16] Abram L Falk, Paul V Klimov, Viktor Ivády, Krisztián Szász, David J Christle, William F Koehl, Adam Gali, and David D Awschalom, “Optical Polarization of Nuclear Spins in Silicon Carbide,” Physical Review Letters 114, 247603 (2015).

[17] S G Carter, Ö O Soykal, Pratibha Dev, Sophia E Economou, and E R Glaser, “Spin coherence and echo modulation of the silicon vacancy in 4H-SiC at room temperature,” Physical Review B 92, 161202 (2015).

[18] D Simin, F Fuchs, H Kraus, A Sperlich, P G Baranov, G V Astakhov, and V Dyakonov, “High-Precision Angle-Resolved Magnetometry with Uniaxial Quantum Centers in Silicon Carbide,” Physical Review Applied 4, 014009 (2015).

[19] Sang-Yun Lee, Matthias Niethammer, and Jörg Wrachtrup, “Vector magnetometry based on $S=3/2$ electronic spins,” Physical Review B 92, 115201 (2015).

[20] S Castelletto, B C Johnson, V Ivády, N Stavrias, T Umeda, A Gali, and T Ohshima, “A silicon carbide room-temperature single-photon source,” Nature Materials 13, 151–156 (2013).

[21] Stefania Castelletto, Brett C Johnson, Cameron Zachre son, David Beke, István Balogh, Takeshi Ohshima, Igor Aharonovich, and Adam Gali, “Room Temperature Quantum Emission from Cubic Silicon Carbide Nanoparticles,” ACS Nano 8, 7938–7947 (2014).

[22] David J Christle, Abram L Falk, Paolo Andrich, Paul V Klimov, Jawad ul Hassan, Nguyen T Son, Erik Janzné, Takeshi Ohshima, and David D Awschalom, “Isolated electron spins in silicon carbide with millisecond coherence times,” Nature Materials 14, 160–163 (2015).

[23] Matthias Widmann, Sang-Yun Lee, Torsten Rendler, Nguyen Tien Son, Helmut Fedder, Seoyoung Paik, Li-Ping Yang, Nan Zhao, Sen Yang, Ian Booker, Andrej Denisenko, Mohammad Jamali, S Ali Momenzadeh, Ilja Gerhardt, Takeshi Ohshima, Adam Gali, Erik Janzné, and Jörg Wrachtrup, “Coherent control of single spins in silicon carbide at room temperature,” Nature Materials 14, 164–168 (2015).

[24] F Fuchs, B Stender, M Trupke, D Simin, J Pfau, V Dyakonov, and G V Astakhov, “Engineering near-infrared single-photon emitters with optically active spins in ultrapure silicon carbide,” Nature Communications 6, 7578 (2015).

[25] A Lohrmann, N Iwamoto, Z Bodrog, S Castelletto, T Ohshima, T J Karle, A Gali, S Prawer, J C McCal-
lum, and B C Johnson, “Single-photon emitting diode in silicon carbide,” Nature Communications 6, 7783 (2015).

[29] J R Maze, P L Stanwix, J S Hodges, S Hong, J M Taylor, P Cappellaro, L Jiang, M V Gurudev Dutt, E Topan, A S Zibrov, A Yacoby, R L Walsworth, and M D Lukin, “Nanoscale magnetic sensing with an individual electronic spin in diamond,” Nature 455, 644–647 (2008).

[30] Gopalakrishnan Balasubramanian, I Y Chan, Roman Kolesov, Mohammad Al-Hmoud, Julia Tisler, Chang Shin, Changdong Kim, Aleksander Wojcik, Philip R Hemmer, Anke Krueger, Tobias Hanke, Alfred Leitenstorfer, Rudolf Bratschitsch, Fedor Jelezko, and Jörg Wrachtrup, “Nanoscale imaging magnetometry with diamond spins under ambient conditions,” Nature 455, 648–651 (2008).

[31] Thomas Wolf, Philipp Neumann, Kazuo Nakamura, Hitoshi Sumiya, Takeshi Oshima, Junichi Isoya, and Jörg Wrachtrup, “Subpicotesla Diamond Magnetometry,” Physical Review X 5, 041001 (2015).

[32] N Mizuochi, S Yamasaki, H Takizawa, N Morishita, T Oshihina, H Itoh, and J Isoya, “Continuous-wave and pulsed EPR study of the negatively charged silicon vacancy with $S=3/2$ and $C_{3v}$ symmetry in n-type 4H-SiC,” Physical Review B 66, 235202 (2002).

[33] Benjamin P Lanyon, Marco Barbieri, Marcelo P Almeida, Thomas Jennewein, Timothy C Ralph, Kevin J Resch, Geoff J Pryde, Jeremy L O’Brien, Alexei Gilchrist, and Andrew G White, “Simplifying quantum logic using higher-dimensional Hilbert spaces,” Nature Physics 5, 134–140 (2008).

[34] Hannah Clevenson, Matthew E Trusheim, Carson Teale, Tim Schröder, Danielle Braje, and Dirk Englund, “Broadband magnetometry and temperature sensing with a light-trapping diamond waveguide,” Nature Physics 11, 393–397 (2015).

[35] Vishal Shah, Svenja Knappe, Peter D D Schwindt, and John Kitching, “Subpicotesla atomic magnetometry with a microfabricated vapour cell,” Nature Photonics 1, 649–652 (2007).

[36] E Sörman, N Son, W Chen, O Kordina, C Hallin, and E Janzén, “Silicon vacancy related defect in 4H and 6H SiC,” Physical Review B 61, 2613–2620 (2000).

[37] S Yu Karpov, A V Kulik, I A Zhmakin, Yu N Makarov, E N Mokhov, M G Ramm, M S Ramm, A D Roenkov, and Yu A Vodakov, “Analysis of sublimation growth and vacancy-related defect in 4H-SiC,” Nature Communications 6, 7783 (2015).

[38] T C Hain, F Fuchs, V A Soltonam, P G Baranov, G V Astakhov, T Hertel, and V Dyakonov, “Excitation and recombination dynamics of vacancy-related spin centers in silicon carbide,” Journal of Applied Physics 115, 135508 (2014).

[39] H J von Bardeleben, J L Cantin, L Henry, and M F Barthe, “Vacancy defects in p-type 6H-SiC created by low-energy electron irradiation,” Physical Review B 62, 10841–10846 (2000).

[40] Eric van Oort and Max Glasbeek, “Fluorescence detected level-anticrossing and spin coherence of a localized triplet state in diamond,” Chemical Physics 152, 365–373 (1991).

[41] J P D Martin, N B Manson, D C Doetschman, M J Sellars, R Neuhaus, and E Wilson, “Spectral hole burning and Raman heterodyne signals associated with an avoided crossing in the NV centre in diamond,” Journal of Luminescence 86, 355–362 (2000).

[42] P.G. Baranov and N.G. Romanov, “Magnetic resonance in micro- and nanostructures,” Applied Magnetic Resonance 21, 165–193 (2001).

[43] R J Epstein, F M Mendoza, Y K Kato, and D D Awschalom, “Anisotropic interactions of a single spin and dark-spin spectroscopy in diamond,” Nature Physics 1, 94–98 (2005).

[44] L J Rogers, R L McMurtrie, M J Sellars, and N B Manson, “Time-averaging within the excited state of the nitrogen-vacancy centre in diamond,” New Journal of Physics 11, 063007 (2009).

[45] J P Tetienne, L Rondin, P Spinicelli, M Chipaux, T Debuisscher, J F Roch, and V Jacques, “Magnetic-field-dependent photodynamics of single NV defects in diamond: an application to qualitative all-optical magnetic imaging,” New Journal of Physics 14, 103033 (2012).

[46] V. A. Soltonam, B. V. Yavkin, D. O. Tolsmachev, R. A. Babunts, A. G. Badalyan, V. Yu. Davydov, E. N. Mokhov, I. I. Proskuryakov, S. B. Orlinskii, and P. G. Baranov, “Optically addressable silicon vacancy-related spin centers in rhombic silicon carbide with high breakdown characteristics and endor evidence of their structure,” Phys. Rev. Lett. 115, 247602 (2015).

[47] N Mizuochi, S Yamasaki, H Takizawa, N Morishita, T Oshihina, H Itoh, and J Isoya, “EPR studies of the isolated negatively charged silicon vacancies in n-type 4H- and 6H-SiC: Identification of $C_{3v}$ symmetry and silicon sites,” Physical Review B 68, 165206 (2003).

[48] G. L. Bir and G. E. Pikus, “Symmetry and Strain-induced Effects in Semiconductors” (Wiley, New York, 1974).

[49] N. S. Averkiev, V. M. Asnin, Yu. N. Lomasov, G. E. Pikus, A. A. Rogachev, and N. A. Rud, “Radiation polarization of a coupled exciton in Ge(As) in a longitudinal magnetic field,” Sov. Phys. Solid State 23, 1851 (1981).

[50] M V Durnev, M M Glazov, E L Ivchenko, M Jo, T Mano, and B C Johnson, “Single-photon emitting diode in silicon carbide,” Nature Communications 6, 7783 (2015).

[51] Rima Schirhagl, Kevin Chang, Michael Loretz, and Christian L Degen, “Nitrogen-Vacancy Centers in Diamond: Nanoscale Sensors for Physics and Biology,” Annual Review of Physical Chemistry 65, 83–105 (2014).

[52] E. L. Ivenchenko and G. E. Pikus, “Superlattices and Other Heterostructures: Symmetry and Optical Phenomena”, 2nd ed. (Springer-Verlag, Berlin, 1997).