Hyperfine Structure of Transition Metal Defects in SiC

Benedikt Tissot* and Guido Burkard†

Department of Physics, University of Konstanz, D-78457 Konstanz, Germany

Transition metal (TM) defects in silicon carbide (SiC) are a promising platform in quantum technology, especially because some TM defects emit in the telecom band. We develop a theory for the interaction of an active electron in the D-shell of a TM defect in SiC with the TM nuclear spin and derive the effective hyperfine tensor within the Kramers doublets formed by the spin-orbit coupling. Based on our theory we discuss the possibility to exchange the nuclear and electron states with potential applications for nuclear spin manipulation and long-lived nuclear-spin based quantum memories.

For several applications in quantum technology, such as quantum networks, memories, emitters, and many more [1–4], a quantum system needs to be coherently controlled and isolated from unwanted noise at the same time. Hybrid quantum systems [5–7], consisting of a part that can couple strongly to external fields as well as a part that is better shielded from its environment are promising platforms to fulfill this requirement. In these systems one can benefit from short gate times of one quantum system as well as long coherence times of the other. A much studied system of this type is the nitrogen vacancy center in diamond with its neighboring nuclear spins [8–16] (and Refs. [17, 18] for reviews).

Transition metal (TM) defects in silicon carbide (SiC) constitute a similar family of systems that have the benefit of being based on a well established host material as well as having accessible transitions in the telecommunication bands [19–23]. Recent studies made the first steps towards control of nuclear spins via transition metal defects in SiC [22]. While these results are highly promising, a complete theoretical framework is still needed. In this paper, we derive a model of the hyperfine coupling based on the underlying symmetry properties and relevant orbital configuration of the defect in the crystal, explaining the experimental data and leading to additional insights. In particular we derive a sensible form of the interaction of the defect nuclear spin with the spin and orbital angular momentum of the active electron as well as their combined interaction with external fields.

The prime examples for TM defects in SiC are created by neutral vanadium (V) and positively charged molybdenum (Mo) atoms substituting a Si atom in 6H- or 4H-SiC [19–25]. These defects have one active electron in the atomic D-shell and are invariant under the transformations of the C3v point group imposed by the crystal structure surrounding the defect. While the interaction with the nuclear spins of neighboring C and Si isotopes with non-zero nuclear spins is possible, the presence of such non-zero spin isotopes as a nearest neighbour is fairly improbable, because their natural abundances are about 1% for 12C (spin 1/2) and 5% for 29Si (spin 1/2) [26] and the abundance can be further reduced by using isotopically purified SiC [27, 28]. Here, we therefore concentrate on the interaction with the TM nuclear spin. The nuclear spin for the most common V isotope is I = 7/2 (99%) and I = 5/2 for about 25% of the stable Mo isotopes and I = 0 for the remaining isotopes of Mo [26, 29].

In order to model the hyperfine coupling between the electron and nuclear spins in a TM defect in SiC, we start from the full Hamiltonian

\[
H = H_{el} + H_{hf} + H_{z,nuc} + H_{d,nuc}.
\]

where \(H_{el} = H_{TM} + V_{el} + H_{so} + H_z + V_{el} + H_d\) describes the electronic orbital and spin degrees of freedom without their interaction with the nuclear spin [30], while the remaining terms incorporate the nuclear spin and its interaction with the electron and external fields. The first three terms in \(H_{el}\) describe the zero-field spin-orbit level structure, as shown in Fig. 1. The defect atomic Hamiltonian \(H_{TM}\) localizes the active electron in the d orbital; the crystal potential \(V_{cr}\) reduces the symmetry of the defect to \(C_{3v}\) due to the potentials of the surrounding

FIG. 1. Spin-orbit (a) and hyperfine (b) energy level structure of the active electron bound to the transition metal (TM) defect. The artistic illustration (c) shows the electron with spin-1/2 (yellow arrow) occupying a D-shell (green and violet) which is split by the \(C_{3v}\) symmetric crystal potential, arising from the surrounding crystal atoms (white balls), into two orbital doublets \(E\) and an orbital singlet \(A_1\) (a). The spin-orbit interaction further splits each of the orbital doublets into two Kramers doublets (KD), leading to the final spin-orbit structure given by five KDs. These KDs are then further split into hyperfine levels, due to the interaction with the TM nuclear spin [purple arrow in (c)], shown in (b) for the KD in the red frame.

* benedikt.tissot@uni-konstanz.de
† guido.burkard@uni-konstanz.de
crystal atoms, thereby splitting the $D$-shell levels; the spin-orbit Hamiltonian $H_{\text{so}}$ takes the coupling of spin and orbital angular momentum into account and splits the levels further. In total, this leads to five doubly degenerate levels forming Kramers doublets (KD), pairs of states related by time-reversal symmetry [31]. The time-reversal symmetry protects KDs from coupling via operators that are invariant under time-reversal.

The Zeeman Hamiltonian $H_{z}$ describes the coupling to a static magnetic field which breaks time-reversal symmetry and thus energetically splits the KDs, the electric potential $V_{d}$ denotes the coupling to a static external electric field, and the driving Hamiltonian $H_{d}$ denotes oscillatory external fields. Because $V_{d}$ is invariant under time-inversion, electric fields cannot lift the degeneracy of the KDs, therefore we concentrate on static magnetic fields in the following.

Typically, the $C_{3v}$ symmetric crystal potential is sufficiently large to approximate the spin-orbit coupled electronic system by five pseudo-spin subspaces (the KDs) with distinct zero-field energies $E_{i,\Gamma_{5/6}}$, where we label the orbital configuration with $j = 1, 2$ and $i = 1, 2, 3$ and the irreducible representation (irrep) of $C_{3v}$ pertaining to the KD with $\Gamma_{j}$ [30]. The effective Hamiltonian is already diagonal inside blocks of the same orbital configuration for static fields aligned with the crystal axis, i.e. the threefold rotation axis of $C_{3v}$, which we refer to as the $z$-axis in the following. The reason for this is that a parallel magnetic field only lifts the time-reversal symmetry but keeps the spacial symmetry intact. On the other hand, fields perpendicular to the crystal axis depend on the spin polarization density at the position of the nucleus denoted using the delta distribution $\delta(r)$. The electron is mainly localized in a $D$-shell but the mixing with $s$-orbitals can still lead to a relevant Fermi contact interaction, which is known for TM complexes [32–36].

The nuclear spin can couple to external magnetic fields described by the nuclear Zeeman Hamiltonian

$$H_{\text{z,nuc}} = \mu_N g_N \vec{I} \cdot \vec{B},$$

and the corresponding driving $H_{\text{d,nuc}}$ term for oscillating magnetic fields. These terms are small in comparison to the KD Zeeman part $|\mu_N g_N| \ll g_{\parallel, \Gamma} \mu_B$ [22, 25, 37] and diagonal for $\vec{B}$ parallel to the crystal axis.

While the state describing the active electron shows the transformation properties of a $d$ orbital, due to effects such as the Jahn-Teller effect and covalency [23, 38, 39] there can be an admixture of other orbitals. The Wigner-Eckart theorem [13, 40] enables us to absorb these effects as well as the radial part of the wave-function in reduced matrix elements, in particular to find the minimal set of non-zero matrix elements of (mixed) square components $\varepsilon_{r}$ in Eq. (4), into the orbital basis. Here, we treat reduced matrix elements as parameters that can be obtained experimentally or via ab-inito calculations. Then we (perturbatively) transform to the block diagonal basis of the pure spin-orbit Hamiltonian, where spin and orbital states are entangled. More details are given in the supplemental material [41].

As we did for the mixing due to external fields we neglect off-diagonal blocks between different orbital configurations $i$ due to the crystal field splitting. We find for the effective hyperfine Hamiltonians inside the KDs,
of the KDs (3) and nuclear Zeeman Hamiltonian (5), we further underlined considering measurements by Wolfowicz et al. [22] where the hyperfine coupling strength is at least two orders of magnitude smaller than the spin-orbit splitting in all V defects in 4H- and 6H-SiC for the ground state \( j = 1 \).

We now discuss the projection of Eq. (10) onto the KDs in more detail. Because the \( \Gamma_j \) KDs transform in complete analogy to pure spin states, the coupling has a familiar form in this case. In particular the effective hyperfine coupling in the \([3, \Gamma_4, \sigma]\) KD is the most similar to the simple diagonal dipolar coupling to the nuclear spin, because the crystal potential does not mix the orbital singlet \( (m = 0) \) state with the remaining orbital states and the spin-orbit coupling vanishes in first order in the orbital singlet. Additionally, the form of the symmetry allowed part of the anisotropic hyperfine tensor in this case also agrees with that of the \(^{14}\text{N} \text{NV}^-\) center which has the same symmetry but comprises spins \( S = I = 1 \) [8, 12, 15].

The remaining KDs deviate significantly from this form because their two pseudo-spin states have a mixed spin and orbital wavefunction, due to the interplay of the crystal potential and the spin-orbit coupling. This leads to the (pseudo-)spin-non-conserving coupling, i.e. the non-diagonal coupling \( \sigma^\parallel_{j,\Gamma_5/6}I_\perp \) of the \( \Gamma_5/6 \) KDs as well as the \( \sigma^\perp_{j,\Gamma_4}I_\perp \) coupling of the \( \Gamma_4 \) KDs for \( j = 1, 2 \), see Fig. 2. Furthermore, the magnitude of \( a^\parallel_{j,\Gamma_5/6} \) can deviate significantly from the other two diagonal entries because it can have pure spin contributions.

Group theory further implies that the \( \Gamma_5/6 \) states cannot be coupled by operators transforming according to the \( E \) representation of \( C_{3v} \), e.g. \( I_x, I_y \). This follows from the requirement that in \( C_{3v} \), the spin-orbit operator part of \( H_{\text{hf}} \) has to transform according to the same basis vector of the same irrep as the corresponding nuclear spin operator, because \( H_{\text{hf}} \) as a whole has to transform according to \( A_1 \). On the other hand the counterpart of \( I_z \) (transforming according to \( A_2 \)) can couple these states. Finally, we stress that the pseudo-spin matrices are not angular momentum type operators and, therefore, \( \sigma^\parallel_{j,\Gamma_5/6} \) and \( \sigma^\perp_{j,\Gamma_4} \) can in part transform according to \( A_2 \). For \( \sigma^\parallel_{j,\Gamma_5/6} \) the relevant terms in the hyperfine Hamiltonian are \( xyS_y - (y^2 - x^2)S_z/2 \).

We calculate the second order hyperfine interaction inside the KDs due to the interaction between KDs from the same orbital doublet with a Schrieffer-Wolff transformation [42]. The unperturbed Hamiltonian \( H_0 = \sum_{j,\Gamma_j} E_{j,\Gamma_j} P_{\text{f},\Gamma_j} \) becomes perturbed inside the KDs by \( V_d = H_{\text{hf, bd}} + H_{\text{z, nucl}} + \sum_{j,\Gamma_j} \mu_B g_{j,\Gamma_j} B_{\text{f},\Gamma_j} \) and between KDs with the same orbital origin by \( H_{\text{hf, od}} \), leading to the first order of the Schrieffer-Wolff transformation

\[
S_1 = \sum_{j,\sigma} \frac{1}{\Delta_{j,\sigma}} \left( a^\parallel_{j,\sigma} | j, \Gamma_5/6, \sigma \rangle \langle j, \Gamma_4, \sigma | I_\perp + a^\perp_{j,\sigma} | j, \Gamma_5/6, \sigma \rangle \langle j, \Gamma_4, -\sigma | I_\perp \right) + \text{h.c.},
\]

and the new effective Hamiltonian \( \tilde{H} = H_0 + V_d + [S_1, H_{\text{hf, od}}]/2 \). We obtain the second-order correction

FIG. 2. Non-zero matrix elements of \( H^\text{eff}_{\text{hf}} \) between states of two KDs originating from the same orbital doublet. The matrix elements between states of the same \( \Gamma_4 \) KD are proportional to \( I_\pm \) (red dotted and green dashed arrows) while they are proportional to \( I_z \) (grey dash-dotted arrow) between states of the \( \Gamma_5/6 \) KDs. Inside the KDs the (off-diagonal) hyperfine interaction competes with the Zeeman splitting (brown). The matrix elements between states of the different KDs of the same orbital doublet are proportional to \( I_\pm \) and suppressed by the spin-orbit splitting \( \Delta^\text{so}_j \) (blue).

\[
H^\text{eff}_{\text{hf}} = \frac{1}{2} \left( a_{j,\Gamma_5/6} \sigma^z_{j,\Gamma_5/6} + a^\perp_{j,\Gamma_5/6} \sigma^x_{j,\Gamma_5/6} \right) I_z,
\]

\[
H^\text{eff}_{\text{hf}} = \frac{a_{j,\Gamma_4} \sigma^z_{j,\Gamma_4}}{2} I_z + \frac{a^\perp_{j,\Gamma_4}}{4} \left( \sigma^z_{j,\Gamma_4} I_\perp + \sigma^-_{j,\Gamma_4} I_\perp \right),
\]

\[
H^\text{eff}_{\text{hf}} = \frac{a_{j,\Gamma_5/6} \sigma^z_{j,\Gamma_5/6}}{2} I_z + \frac{a^\perp_{j,\Gamma_5/6}}{4} \left( \sigma^z_{j,\Gamma_5/6} I_\perp + \sigma^-_{j,\Gamma_5/6} I_\perp \right),
\]

for \( j = 1, 2 \) and using the nuclear spin \( \sigma^\parallel_{i,\Gamma_j} = \sigma^\perp_{i,\Gamma_j} = \frac{i}{2} \rho_{i,\Gamma_j} \), as well as nuclear \( I_\pm = I_x \pm iI_y \) ladder operators. The diagonal part of the total effective hyperfine Hamiltonian can thus be written as \( H^\text{hf, bd} = \sum_{j,\Gamma_j} P_{\text{f},\Gamma_j} H^\text{eff}_{\text{hf}} P_{\text{f},\Gamma_j} \). The terms mixing the KDs of the same orbital doublet are

\[
H^\text{hf, od} = \sum_{j,\sigma} a_{j,\sigma} \left( \sigma | j, \Gamma_5/6, \sigma \rangle \langle j, \Gamma_4, \sigma | I_\perp - \sigma | I_\perp \right) + \text{h.c.},
\]

where \( \sigma = \pm 1 \) =↑, ↓. Combined this leads to the effective hyperfine Hamiltonian \( H^\text{eff}_{\text{hf}} = H^\text{hf, bd} + H^\text{hf, od} \). The resulting coupling structure is depicted in Fig. 2.

Combined with the effective spin-orbit Hamiltonians of the KDs (3) and nuclear Zeeman Hamiltonian (5), we obtain our first main result,

\[
H^\text{eff} = H^\text{eff}_{\text{so}} + H^\text{eff}_{\text{hf}} + H_{\text{z, nucl}}.
\]
energy ground states (GS1) correspond to the lowest- 
and the above-mentioned measurement in combina-
tion with our theoretical model suggests that the lowest-
spins. The above-mentioned measurement in combina-
tion with our theoretical model and the energy levels for the 
\(\Gamma_4\) states (b) were calculated using a least squares fit for the eigenvalues of the models for magnetic fields between 2 - 45 mT with 200 data points. While there is a disagreement in the energy levels, the allowed transition rates are compatible with the experimental fit. The magnetic field of 2 mT corresponds to the lowest magnetic field visible in [22]. For the effective hyperfine constants we find \(a_{\text{hf}}^{1/2,\gamma}/h \approx 202.5\) MHz and \(a_{\text{hf}}^{1/3,\gamma}/h \approx 158.2\) MHz, as well as \(a_{\text{hf}}^{1/3,\gamma}/h \approx -174.7 \pm 4.3\) MHz and \(a_{\text{hf}}^{2,\gamma}/h \approx 149.5 \pm 4.2\) MHz, where the fit errors are due to the deviation at small magnetic fields (irrelevant for the transitions). We additionally use \(g_1 r_{1/2}/\gamma = 1.87, g_1 r_{3/2}/\gamma = 2.035, \) and \(\mu_{\text{N}} g_{\text{nuc}}/h = -11.213\) MHz/T from [22].

\[
H_{1,\Gamma_4}^{\text{hf}(2)} = P_{1,\Gamma_4} [S_1, H_{\text{hf,od}}] P_{1,\Gamma_4}/2 \quad \text{with}
\]
\[
H_{1,\Gamma_4}^{\text{hf}(2)} = -H_{1,\Gamma_4}^{\text{hf}(2)} = a_{j}^{\text{od}} [(I^2 - I_z)^2],
\]
for \(j = 1, 2\) and \(H_{1,\Gamma_4}^{\text{hf}(2)} = 0\), with the defect-configuration dependent constant \(a_{j}^{\text{od}} = (a_{j}^2 + a_{j}^2) / \Delta n^2\). Combined with Eqs. (6)–(8) this leads to the second order of the effective hyperfine Hamiltonians for the KDs

\[
H_{1,\Gamma_4} = H_{1,\Gamma_4}^{\text{KD}} + H_{1,\Gamma_4}^{\text{hf}} + H_{2,\text{nuc}} + H_{1,\Gamma_4}^{\text{hf}(2)}.
\]

We stress that the second order contribution of the hyperfine interaction is purely diagonal in the basis where \(z\) points along the crystal axis.

We now compare our results to the recent measurements by Wolfowicz et al. [22], concentrating on the two ground state KDs. In [22], a model for the KDs with a different form of the hyperfine coupling \(\sum k_{x,y,z} a_{\text{hf}}^{k} \sigma_{\gamma}^{k} a_{\text{od}}^{k} I_k/2\) was used for all KDs, additionally allowing a tilt of the quantization axis of the pseudo-spin. The above-mentioned measurement in combination with our theoretical model suggests that the lowest-energy ground states (GS1) correspond to \(|1, \Gamma_4\rangle\) and GS2 to \(|1, \Gamma_5/6\rangle\). The first point we want to highlight is that the measurement confirms that the \(|1, \Gamma_5/6\rangle\) KD states do not couple via \(I_x, I_y\) and that a tilt of the pseudo-spin around the \(y\)-axis corresponding to the coupling of \(I_z\) to \(\sigma_{\gamma}^z\) is found. We highlight that this artificial tilt of the quantization axis needs no further explanation in our theory where the \(\sigma_{\gamma}^z\) coupling emerges naturally from the interplay of the crystal potential and the spin-orbit interaction. This can be seen via the transformation properties of the pseudo-spin operators as well as the form of the wavefunction of the KD states.

The second point is that our model provides a resort to explain the measurements [22] for GS1 without the need for an anisotropy in the hyperfine coupling tensor in the plane perpendicular to the crystal axis. Our model Eq. (7), shows good agreement with the transition frequencies in [22], despite a deviation of two energies in some configurations for small magnetic fields. For larger magnetic fields \((B > 10\) mT\) indeed all energies are in agreement with the model in [22]. Not only does our model provide an explanation without the anisotropy, it furthermore reduces the number of free parameters. For the \(\beta\) configuration of the V defect in 4H-SiC we plot the comparison of the models in Fig. 3.

Additionally, the measurement of \(\beta\) 6H-SiC in [22] includes all relevant electronic energy splittings allowing us to assign \(|3, \Gamma_4\rangle\) to the lowest energy excited state, for which we find that the form of the hyperfine interaction of our theory agrees with their measurement.

Finally we want to use the gained understanding of the effective hyperfine Hamiltonians within the KDs (13) and investigate the consequences. The effective Hamiltonians can be block diagonalised in 2 \(\times\) 2 blocks. In \(\Gamma_5/6\) the KDs are mixed with each other but not with the nuclear spin, such that the resulting states are merely tilted around an axis perpendicular to the crystal axis. On the other hand, the \(\Gamma_4\) KD electronic states are entangled with the nuclear spin, i.e.

\[
|i, \Gamma_4, m_i \rangle = \cos(\phi_{i,\Gamma_4, m_i}) |i, \Gamma_4, \uparrow\rangle |m_i\rangle + \sin(\phi_{i,\Gamma_4, m_i}) |i, \Gamma_4, \downarrow\rangle |m_i\rangle,
\]

and similarly for the corresponding orthogonal states \(|i, \Gamma_4, -m_i \pm 1\rangle\). The analytic diagonalization of the effective hyperfine Hamiltonian (13) including a static external magnetic field can be found in the supplemental material [41]. In combination with the selection rules for the electronic states [30], the mixing leads to the relevant allowed transitions. Here we concentrate on a set of transitions that can be used to transfer the pseudo-spin state of the ground-state KD \(|1, \Gamma_4, \sigma\rangle\) to the nuclear spin and vice versa via a Lambda (\(\Lambda\)) system, i.e. \(\alpha |1, \Gamma_4, +, -I\rangle + \beta |1, \Gamma_4, -, -I\rangle \leftrightarrow \alpha |1, \Gamma_4, -, +I\rangle + \beta |1, \Gamma_4, +, -I\rangle\). When constructing a \(\Lambda\) system using a different KD we can neglect the nuclear driving term \(H_{\text{nuc}}\) because
transitions between the nuclear levels of the same KD pseudo-spin state are highly detuned. Because $|3, \Gamma_4, +, -I\rangle = \cos(\phi_3, \Gamma_4, -I)|1, \Gamma_4, -I\rangle |I + 1\rangle - \sin(\phi_3, \Gamma_4, -I)|1, \Gamma_4, I\rangle |I\rangle$ as well as $|1, \Gamma_4, +, -I\rangle = |1, \Gamma_4, \uparrow\rangle |I\rangle$ via a KD pseudo-spin conserving transition. Therefore, these levels can be used as a $\Lambda$ system driven by a magnetic or electric field perpendicular to the crystal axis, as shown in Fig. 4.

Analogously our effective theory shows that the hyperfine interaction opens the possibility to directly drive the pseudo-spin transition of the KDs for small magnetic fields due to the pseudo spin tilt or the pseudo-spin nuclear entanglement; this was studied using a different framework by Gilardoni et al. [43]. Lastly, when the spin-orbit splitting is sufficiently small, the second order hyperfine interaction can enable optical driving inside the KDs by mixing the KDs of the same orbital doublet.

In summary, we introduced a theory to describe the hyperfine interaction in TM defects in SiC having a single electron in a $D$-shell. The theory yields new insights on previous measurements and reduces the required number of fit parameters of the effective hyperfine coupling tensor. The newly gained insights can be used to construct a $\Lambda$ transition that can be exploited to create a nuclear spin quantum memory.

ACKNOWLEDGMENTS

We thank A. Csóre and A. Gali for useful discussions and acknowledge funding from the European Union’s Horizon 2020 research and innovation programme under grant agreement No 862721 (QuanTELCO).

[1] H. J. Kimble, The quantum internet, Nature 453, 1023 (2008).
[2] I. Aharonovich, D. Englund, and M. Toth, Solid-state single-photon emitters, Nature Photonics 10, 631 (2016).
[3] K. Heshami, D. G. England, P. C. Humphreys, P. J. Bustard, V. M. Acosta, J. Nunn, and B. J. Sussman, Quantum memories: emerging applications and recent advances, J. Mod. Opt. 63, 2065 (2016).
[4] D. Awschalom, K. K. Berggren, H. Bernien, S. Bhave, L. D. Carr, P. Davids, S. E. Economou, D. Englund, A. Faraon, M. Fejer, S. Guha, M. V. Gustaffson, E. Hu, L. Jiang, J. Kim, B. Korzh, P. Kumar, P. G. Kwiat, M. Lončar, M. D. Lukin, D. A. Miller, C. Monroe, S. W. Nam, P. Narang, J. S. Orcutt, M. G. Raymer, A. H. Safavi-Naeini, M. Spirepolu, K. Srinivasan, S. Sun, J. Vucković, E. Waks, R. Walsworth, A. M. Weiner, and Z. Zhang, Development of quantum interconnects (quics) for next-generation information technologies, PRX Quantum 2, 017002 (2021).
[5] A. A. Clerk, K. W. Lehnert, P. Bertet, J. R. Petta, and Y. Nakamura, Hybrid quantum systems with circuit quantum electrodynamics, Nature Phys. 16, 257 (2020).
[6] D. S. Smirnov, T. S. Shamirzaev, D. R. Yakovlev, and M. Bayer, Dynamic polarization of electron spins interacting with nuclei in semiconductor nanostructures, Phys. Rev. Lett. 125, 156801 (2020).
[7] G. Burkard, M. J. Gullans, X. Mi, and J. R. Petta, Superconductor–semiconductor hybrid-circuit quantum electrodynamics, Nature Reviews Physics 2, 129 (2020).
[8] X.-F. He, N. B. Manson, and P. T. H. Fisk, Paramagnetic resonance of photoexcited N-V defects in diamond. I. level anticrossing in the $3^3 A$ ground state, Phys. Rev. B 47, 8809 (1993).
[9] T. Gaebel, M. Domhan, I. Popa, C. Wittmann, P. Neumann, F. Jelezko, J. R. Rabeau, N. Stavrias, A. D. Greentree, S. Prawer, J. Meijer, J. Twamley, P. R. Hemmer, and J. Wrachtrup, Room-temperature coherent coupling of single spins in diamond, Nature Phys. 2, 408 (2006).
[10] L. Childress, M. V. G. Dutt, J. M. Taylor, A. S. Zibrov, F. Jelezko, J. Wrachtrup, P. R. Hemmer, and M. D. Lukin, Coherent dynamics of coupled electron and nuclear spin qubits in diamond, Science 314, 281 (2006).
[11] A. Gali, M. Fyta, and E. Kaxiras, Ab initiosupercell calculations on nitrogen-vacancy center in diamond: Electronic structure and hyperfine tensors, Phys. Rev. B 77, 155206 (2008).
[12] S. Felton, A. M. Edmonds, M. E. Newton, P. M. Martineau, D. Fisher, D. J. Twitchen, and J. M. Baker, Hyperfine interaction in the ground state of the negatively
charged nitrogen vacancy center in diamond, Phys. Rev. B 79, 075203 (2009).

[13] J. R. Maze, A. Gali, E. Togan, Y. Chu, A. Trifonov, E. Kaxiras, and M. D. Lukin, Properties of nitrogen-vacancy centers in diamond: the group theoretic approach, New J. Phys. 13, 025025 (2011).

[14] G. D. Fuchs, G. Burkard, P. V. Klimov, and D. D. Awschalom, A quantum memory intrinsic to single nitrogen–vacancy centres in diamond, Nature Phys. 7, 789 (2011).

[15] L. Busaite, R. Lazda, A. Berzins, M. Auzinsh, R. Ferber, and F. Gahbauer, Dynamic 14N nuclear spin polarization in nitrogen-vacancy centers in diamond, Phys. Rev. B 102, 224101 (2020).

[16] S. S. Hegde, J. Zhang, and D. Suter, Efficient quantum gates for individual nuclear spin qubits by indirect control, Phys. Rev. Lett. 124, 220501 (2020).

[17] M. W. Doherty, N. B. Manson, P. Delaney, F. Jelezko, J. Wrachtrup, and L. C. L. Hollenberg, The nitrogen-vacancy colour centre in diamond, Phys. Rep. 528, 1 (2013).

[18] D. Suter and F. Jelezko, Single-spin magnetic resonance in the nitrogen-vacancy center of diamond, Prog. Nucl. Magn. Reson. Spectrosc. 98-99, 50 (2017).

[19] T. Bosma, G. J. J. Lof, C. M. Gilardoni, O. V. Zwier, F. Hendriks, B. Magnusson, A. Ellison, A. Gällström, I. G. Ivanov, N. T. Son, R. W. A. Havenith, and C. H. van der Wal, Identification and tunable optical coherent control of transition-metal spins in silicon carbide, npj Quantum Inf. 4, 48 (2018).

[20] L. Spindllerber, A. Csóre, G. Thiering, S. Putz, R. Karhu, J. Hassan, N. Son, T. Fromherz, A. Gali, and M. Trupke, Optical properties of vanadium in 4H silicon carbide for quantum technology, Phys. Rev. Appl. 12, 014015 (2019).

[21] C. M. Gilardoni, T. Bosma, D. van Hien, F. Hendriks, B. Magnusson, A. Ellison, I. G. Ivanov, N. T. Son, and C. H. van der Wal, Spin-relaxation times exceeding seconds for color centers with strong spin–orbit coupling in SiC, New J. Phys. 22, 103051 (2020).

[22] G. Wolfowicz, C. P. Anderson, B. Diler, O. G. Poluektov, F. J. Heremans, and D. D. Awschalom, Vanadium spin qubits as telecom quantum emitters in silicon carbide, Sci. Adv. 6, eaaz1192 (2020).

[23] A. Csóre and A. Gali, Ab initio determination of pseudospin for paramagnetic defects in sic, Phys. Rev. B 102, 241201 (2020).

[24] B. Kaufmann, A. Dörnien, and F. S. Ham, Crystal-field model of vanadium in 6H silicon carbide, Phys. Rev. B 55, 13009 (1997).

[25] J. Baur, M. Kunzer, and J. Schneider, Transition metals in SiC polytypes, as studied by magnetic resonance techniques, Phys. Status Solidi (a) 162, 153 (1997).

[26] J. Meija, T. B. Coplen, M. Berghund, W. A. Brand, P. D. Bievre, M. Gröning, N. E. Holden, J. Irrgeher, R. D. Loss, T. Walczyzk, and T. Prohaska, Isotopic compositions of the elements 2013 (IUPAC technical report), Pure Appl. Chem. 88, 293 (2016).

[27] M. Barbouche, R. B. Zaghouni, N. Benammar, V. Aglieri, M. Mosca, R. Macaluso, K. Khirouni, and H. Ezzaouiaia, New process of silicon carbide purification intended for silicon passivation, Superlattices Microstruct. 101, 512 (2017).

[28] V. Mazzocchi, P. Sennikov, A. Bulanov, M. Churbanov, B. Bertrand, L. Hutin, J. Barnes, M. Drozdov, J. Hartmann, and M. Sanquer, 99.992 % 28Si cvd-grown epilayer on 300mm substrates for large scale integration of silicon spin qubits, J. Cryst. Growth 500, 1 (2019).

[29] G. Audi, O. Bersillon, J. Blachot, and A. Wapstra, The NUBASE evaluation of nuclear and decay properties, Nucl. Phys. A 729, 3 (2003).

[30] B. Tissot and G. Burkard, Spin structure and resonant driving of spin-1/2 defects in SiC, Phys. Rev. B 103, 064106 (2021).

[31] M. S. Dresselhaus, G. Dresselhaus, and A. Jorio, Group theory : application to the physics of condensed matter (Springer-Verlag, Berlin, 2010).

[32] M. L. Munzarová, P. Kubáček, and M. Kaupp, Mechanisms of EPR hyperfine coupling in transition metal complexes, J. Am. Chem. Soc. 122, 1900 (2000).

[33] W. A. Coish and J. Baugh, Nuclear spins in nanostructures, Phys. Status Solidi (b) 246, 2203 (2009).

[34] G. Micera and E. Garrubba, Is the spin-orbit coupling important in the prediction of the 31V hyperfine coupling constants of V3+O2− species? ORCA versus gaussian performance and biological applications, J. Comput. Chem. 32, 2822 (2011).

[35] J. Vicha, M. Stráská, M. L. Munzarová, and R. Marek, Mechanism of spin-orbit effects on the ligand NMR chemical shift in transition-metal complexes: Linking NMR to EPR, J. Chem. Theory Comput. 10, 1489 (2014).

[36] S. Gohr, P. Hrobářik, S. Komorovský, K. Ruud, and M. Kaupp, Four-component relativistic density functional theory calculations of EPR g- and hyperfine-coupling tensors using hybrid functionals: Validation on transition-metal complexes with large tensor anisotropies and higher-order spin-orbit effects, J. Phys. Chem. A 119, 12892 (2015).

[37] N. Stone, Table of nuclear magnetic dipole and electric quadrupole moments, At. Data Nucl. Data Tables 95, 70 (2005).

[38] F. S. Ham, Dynamical Jahn-Teller effect in paramagnetic resonance spectra: Orbital reduction factors and partial quenching of spin-orbit interaction, Phys. Rev. 138, A1727 (1965).

[39] F. S. Ham, Effect of linear Jahn-Teller coupling on paramagnetic resonance in a 2E state, Phys. Rev. 166, 307 (1968).

[40] J. F. Cornwell, Group theory in physics : an introduction (Academic Press, San Diego, Calif, 1997).

[41] See supplemental material at [URL will be inserted by publisher] for a more detailed derivation of the hyperfine Hamiltonian in the d orbital constrained to C3v, symmetry of the crystal, the analytic diagonalization of the second order effective Hamiltonians for the KDs, and a table of hyperfine tensors for different V defects corresponding to data from Wolfowicz et al. (2021).

[42] S. Bravyi, D. P. DiVincenzo, and D. Loss, Schrieffer-Wolf transformation for quantum many-body systems, Ann. Phys. 326, 2793 (2011).

[43] C. M. Gilardoni, I. Ion, F. Hendriks, M. Trupke, and C. H. van der Wal, Hyperfine-mediated transitions between electronic spin-1/2 levels of transition metal defects in SiC, in preparation (2021).