

Supplementary Material for “Numerical Simulation of Spin-Dependent Recombination and Hyperfine Structure in Electrically-Detected Magnetic Resonance of 4H-SiC”

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S1. RF EXCITATION AND FIELD MODULATION

The cw-EDMR measurements were performed using a fixed-frequency RF drive at $\nu = 200$ MHz with a nominal output power of 23 dBm.¹ The RF field was applied using a loop coil with an effective diameter of approximately 8 mm, positioned to maximize coupling to the active device region. Under pulsed calibration conditions, the coil corresponds to a $\pi/2$ pulse length of $7.6 \mu\text{s}$ for ^1H at full power.

For the full-sweep spectrum shown in Fig. 2 of the main text, the field modulation amplitude was $B_{\text{mod}} = 1.5$ G peak-to-peak at a frequency of 1 kHz. For the half-field measurements (Fig. 3 of the main text), the modulation amplitude was $B_{\text{mod}} = 2$ G peak-to-peak at a frequency of 1.25 kHz. The EDMR signal was detected using a lock-in amplifier referenced to the modulation frequency, yielding a derivative signal $\partial I / \partial B_0$.

S2. DATA ACQUISITION AND PROCESSING

All spectra were acquired under a constant sample bias voltage of 3.75 V. Individual lock-in traces were averaged over consecutive scans to improve the signal-to-noise ratio. No additional background subtraction, filtering, or other post-processing was applied prior to comparison with simulations. Fig. S1 schematically illustrates the experimental setup.

S3. DERIVATION OF THE SPIN HAMILTONIAN

A. Chosen basis

All Hamiltonian terms are represented as 16×16 matrices. We use the product basis

$$\{|s, m_s\rangle \otimes |m_{\text{Si}}, m_{\text{C}}\rangle\}, \quad (1)$$

with the two-electron subspace written in the coupled basis

$$|s, m_s\rangle \in \{|1, 1\rangle, |1, 0\rangle, |0, 0\rangle, |1, -1\rangle\}, \quad (2)$$

and the two nuclei written in the uncoupled z -projection basis $m_{\text{Si}}, m_{\text{C}} \in \{\pm \frac{1}{2}\}$, ordered as

$$|m_{\text{Si}}, m_{\text{C}}\rangle \in \{|++\rangle, |+-\rangle, |-+\rangle, |--\rangle\}. \quad (3)$$

The full 16-state ordering is taken as the four nuclear states above for each electron state in the order listed.

B. Zeeman Hamiltonian

With $\mathbf{B}_0 = B_0 \hat{\mathbf{z}}$, we take $g_1 \simeq g_2 \simeq g_e$ as in the main text, so that the dominant electron Zeeman term depends only on the total electron spin projection m_s and is diagonal in the chosen basis. We write

$$\mathcal{H}_Z = \mathcal{H}_{Z,e} + \mathcal{H}_{Z,n}, \quad (4)$$

$$\mathcal{H}_{Z,e} = \frac{\mu_B}{\hbar} B_0 g_e (S_{1z} + S_{2z}), \quad (5)$$

$$\mathcal{H}_{Z,n} = -\frac{\mu_N}{\hbar} B_0 (g_{n,\text{Si}} I_{\text{Si},z} + g_{n,\text{C}} I_{\text{C},z}). \quad (6)$$

Equivalently, for the basis ket $|s, m_s\rangle \otimes |m_{\text{Si}}, m_{\text{C}}\rangle$ the Zeeman energy is

$$E_Z = g_e \mu_B B_0 m_s + \mu_N B_0 (g_{n,\text{Si}} m_{\text{Si}} + g_{n,\text{C}} m_{\text{C}}), \quad (7)$$

with $m_s \in \{+1, 0, 0, -1\}$ and $m_{\text{Si}}, m_{\text{C}} \in \{\pm \frac{1}{2}\}$.

Implementation. Since \mathcal{H}_Z is diagonal in the chosen basis, the 16×16 matrix is constructed by assigning Eq. (7) along the diagonal for each ordered basis state. In `sympy`² this is implemented by looping over the ordered quantum numbers and filling only diagonal entries.

C. Hyperfine Hamiltonian

The hyperfine interaction between an electron spin \mathbf{S} and a nuclear spin \mathbf{I} is written in tensor form as³

$$\mathcal{H}_{\text{HF}} = \mathbf{S} \cdot \mathbf{A} \cdot \mathbf{I}. \quad (8)$$

In the principal-axis frame of the interaction we take \mathbf{A} diagonal, $\mathbf{A} = \text{diag}(A_{xx}, A_{yy}, A_{zz})$. For an axially symmetric coupling ($A_{xx} = A_{yy} \equiv A_{\perp}$ and $A_{zz} \equiv A_{\parallel}$),

$$\mathcal{H}_{\text{HF}} = A_{\perp} (S_x I_x + S_y I_y) + A_{\parallel} S_z I_z. \quad (9)$$

Using raising and lowering operators, $S_x = \frac{1}{2}(S_+ + S_-)$, $S_y = \frac{1}{2i}(S_+ - S_-)$ (and similarly for I), the transverse part becomes

$$S_x I_x + S_y I_y = \frac{1}{2} (S_+ I_- + S_- I_+), \quad (10)$$

so that Eq. (9) can be written in the working form

$$\mathcal{H}_{\text{HF}} = A_{\parallel} S_z I_z + \frac{A_{\perp}}{2} (S_+ I_- + S_- I_+). \quad (11)$$

Equation (11) makes the matrix structure transparent in the uncoupled z -projection basis $|m_S, m_I\rangle$ with $m_S, m_I \in \{\pm \frac{1}{2}\}$: the $S_z I_z$ term is diagonal, while the

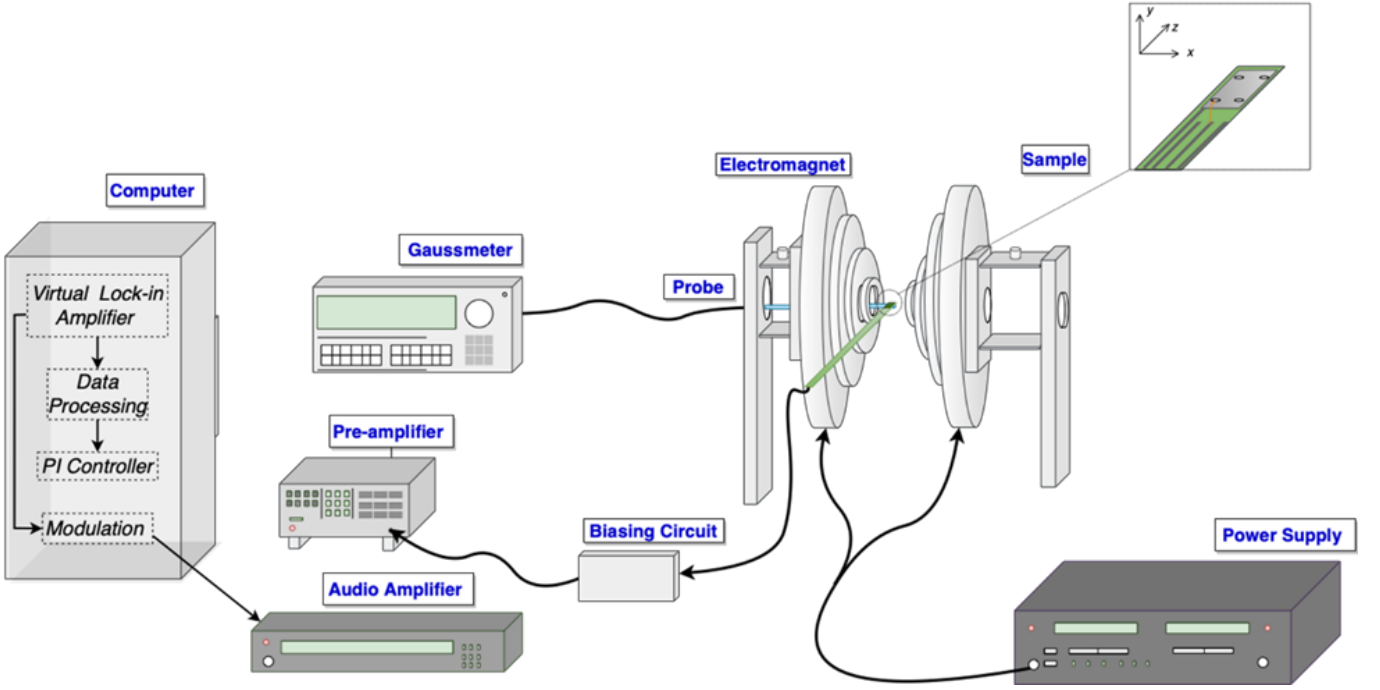


FIG. S1. Schematic of the experimental apparatus used for the cw-EDMR measurements. The figure illustrates the relative placement of the amplifiers, biasing circuit, and the direction of the applied static magnetic field B_0 .

S_+I_- and S_-I_+ terms produce only “flip-flop” couplings $|\uparrow\downarrow\rangle \leftrightarrow |\downarrow\uparrow\rangle$. For spin-1/2, $S_+|\downarrow\rangle = |\uparrow\rangle$, $S_-|\uparrow\rangle = |\downarrow\rangle$ (and likewise for I_{\pm}), so the only off-diagonal matrix elements are

$$\langle\uparrow\downarrow|\mathcal{H}_{\text{HF}}|\downarrow\uparrow\rangle = \langle\downarrow\uparrow|\mathcal{H}_{\text{HF}}|\uparrow\downarrow\rangle = \frac{A_{\perp}}{2}. \quad (12)$$

In the isotropic limit used in the main text $A_{\perp} = A_{\parallel} \equiv A$, Eq. (11) reduces to $\mathcal{H}_{\text{HF}} = A \mathbf{S} \cdot \mathbf{I}$.

1. Two-electron, two-nucleus hyperfine Hamiltonian

With two electrons and two nuclei (Si and C), we sum the electron-nucleus hyperfine couplings over all pairs,

$$\mathcal{H}_{\text{HF}} = \mathbf{S}_1 \cdot \mathbf{A}_{1\text{Si}} \cdot \mathbf{I}_{\text{Si}} + \mathbf{S}_2 \cdot \mathbf{A}_{2\text{Si}} \cdot \mathbf{I}_{\text{Si}} \quad (13)$$

$$+ \mathbf{S}_1 \cdot \mathbf{A}_{1\text{C}} \cdot \mathbf{I}_{\text{C}} + \mathbf{S}_2 \cdot \mathbf{A}_{2\text{C}} \cdot \mathbf{I}_{\text{C}}. \quad (14)$$

Each term is applied as in Eq. (11), acting nontrivially only on the corresponding electron and nuclear subspaces.

Implementation. Although the full simulation basis uses the coupled two-electron states $|s, m_s\rangle$, the flip-flop structure in Eq. (11) is simplest to implement in the uncoupled electron Zeeman basis $\{|\uparrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\downarrow\downarrow\rangle\}$. We therefore construct \mathcal{H}_{HF} in the intermediate product basis

$$\{|m_1, m_2\rangle \otimes |m_{\text{Si}}, m_{\text{C}}\rangle\}, \quad m_1, m_2, m_{\text{Si}}, m_{\text{C}} \in \{\pm\frac{1}{2}\}, \quad (15)$$

by looping over ordered basis kets and adding (i) diagonal contributions from $S_{jz}I_{kz}$ and (ii) the two allowed flip-flop couplings from $S_{j+}I_{k-}$ and $S_{j-}I_{k+}$ for each of the four terms in Eq. (14). In `sympy`² this is implemented by a routine that returns the nonzero targets and coefficients for the action of \mathcal{H}_{HF} on a given ket, and then fills the corresponding matrix entries.

Finally, we transform from the electron Zeeman basis to the coupled basis used in Sec. S3A via the Clebsch-Gordan relations

$$\begin{aligned} |1, 1\rangle &= |\uparrow\uparrow\rangle, \\ |1, 0\rangle &= \frac{|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle}{\sqrt{2}}, \\ |0, 0\rangle &= \frac{|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle}{\sqrt{2}}, \\ |1, -1\rangle &= |\downarrow\downarrow\rangle. \end{aligned} \quad (16)$$

Writing this as a unitary U acting on the four electron states, the full 16×16 basis change is

$$W = U \otimes \mathbb{1}_4, \quad (17)$$

where $\mathbb{1}_4$ acts on the ordered nuclear basis $\{ |++\rangle, |+-\rangle, |-+\rangle, |--\rangle \}$. The hyperfine Hamiltonian in the simulation basis is then

$$\mathcal{H}_{\text{HF}}^{(\text{coupled})} = W \mathcal{H}_{\text{HF}}^{(\text{Zeeman})} W^\dagger. \quad (18)$$

D. Effective dipolar (ZFS) Hamiltonian

The dipole-dipole interaction between two electron magnetic moments separated by \mathbf{r} can be written in the standard form³

$$\mathcal{H}_{\text{dd}} = \frac{\mu_0}{4\pi} \frac{(g_e \mu_B)^2}{\hbar^2 r^3} [\mathbf{S}_1 \cdot \mathbf{S}_2 - 3(\mathbf{S}_1 \cdot \hat{\mathbf{r}})(\mathbf{S}_2 \cdot \hat{\mathbf{r}})], \quad (19)$$

with $\hat{\mathbf{r}} = \mathbf{r}/r$. Introducing the traceless dipolar tensor

$$D_{ij}(\mathbf{r}) = \frac{\mu_0}{4\pi} \frac{(g_e \mu_B)^2}{\hbar^2 r^3} (\delta_{ij} - 3\hat{r}_i \hat{r}_j), \quad \text{Tr } \mathbf{D} = 0, \quad (20)$$

Eq. (19) is equivalently

$$\mathcal{H}_{\text{dd}} = \sum_{i,j} S_{1i} D_{ij} S_{2j}. \quad (21)$$

In the SDR pair model, this anisotropic interaction produces an *effective* splitting within the two-electron triplet manifold of the defect-carrier pair. It is not the intrinsic quartet ZFS of the V_{Si}^- center. Since $\mathbf{S}_1 \cdot \mathbf{S}_2$ is a constant within a fixed total-spin sector, we drop overall energy offsets and write the effective zero-field splitting Hamiltonian acting on the triplet as

$$\mathcal{H}_{\text{ZFS}} = \mathbf{S} \cdot \mathbf{D} \cdot \mathbf{S}, \quad (22)$$

where \mathbf{S} is the total spin-1 operator restricted to the triplet subspace.

1. Axial and transverse parameterization

Diagonalizing \mathbf{D} defines its principal axes, in which $\mathbf{D}_{\text{diag}} = \text{diag}(D_x, D_y, D_z)$ with $D_x + D_y + D_z = 0$. We parameterize the principal values by axial and transverse components

$$D_1 = \frac{3}{2}D_z, \quad D_2 = \frac{1}{2}(D_x - D_y), \quad (23)$$

so that

$$\mathbf{D}_{\text{diag}} = \begin{pmatrix} -\frac{D_1}{3} + D_2 & 0 & 0 \\ 0 & -\frac{D_1}{3} - D_2 & 0 \\ 0 & 0 & \frac{2D_1}{3} \end{pmatrix}. \quad (24)$$

Substituting Eq. (24) into Eq. (22) and using $S_x^2 + S_y^2 + S_z^2 = S(S+1) = 2$ for $S = 1$ gives the standard operator form

$$\mathcal{H}_{\text{ZFS}} = D_1 \left(S_z^2 - \frac{2}{3} \right) + D_2 (S_x^2 - S_y^2). \quad (25)$$

Writing $S_x^2 - S_y^2 = \frac{1}{2}(S_+^2 + S_-^2)$ makes the selection rules explicit: the axial term is diagonal in $|1, m_s\rangle$, while the transverse term couples states with $\Delta m_s = \pm 2$.

2. Matrix elements in the $\{|1, 1\rangle, |1, 0\rangle, |1, -1\rangle\}$ basis

In the triplet basis $\{|1, 1\rangle, |1, 0\rangle, |1, -1\rangle\}$, Eq. (25) has the compact matrix representation

$$\mathcal{H}_{\text{ZFS}}^{(S=1)} = \begin{pmatrix} \frac{D_1}{3} & 0 & D_2 \\ 0 & -\frac{2D_1}{3} & 0 \\ D_2 & 0 & \frac{D_1}{3} \end{pmatrix}. \quad (26)$$

The singlet state $|0, 0\rangle$ is unaffected by \mathcal{H}_{ZFS} in this effective description.

Implementation. In the simulation basis of Sec. S3 A, the electron subspace is ordered as $\{|1, 1\rangle, |1, 0\rangle, |0, 0\rangle, |1, -1\rangle\}$. We therefore embed Eq. (26) into a 4×4 electron matrix by placing the triplet block on the $\{|1, 1\rangle, |1, 0\rangle, |1, -1\rangle\}$ rows/columns and setting the $|0, 0\rangle$ row/column to zero. The full 16×16 operator is then constructed as

$$\mathcal{H}_{\text{ZFS}} = \mathcal{H}_{\text{ZFS}}^{(e)} \otimes \mathbb{1}_4, \quad (27)$$

with $\mathbb{1}_4$ acting on the ordered nuclear basis $\{|++\rangle, |+-\rangle, |-+\rangle, |--\rangle\}$. In `sympy`², this is implemented by directly writing the nonzero entries of $\mathcal{H}_{\text{ZFS}}^{(e)}$ from Eq. (26) and applying a Kronecker product.

E. Exchange Hamiltonian

The exchange interaction between the two electrons is modeled as an isotropic Heisenberg coupling,³

$$\mathcal{H}_{\text{EX}} = J \mathbf{S}_1 \cdot \mathbf{S}_2. \quad (28)$$

Using $\mathbf{S}^2 = (\mathbf{S}_1 + \mathbf{S}_2)^2 = \mathbf{S}_1^2 + \mathbf{S}_2^2 + 2\mathbf{S}_1 \cdot \mathbf{S}_2$ and $\mathbf{S}_1^2 = \mathbf{S}_2^2 = \frac{3}{4}$ for spin-1/2, one finds

$$\mathbf{S}_1 \cdot \mathbf{S}_2 = \frac{1}{2} \left(\mathbf{S}^2 - \frac{3}{2} \right). \quad (29)$$

Therefore, within a sector of definite total spin s ,

$$\mathcal{H}_{\text{EX}} = \frac{J}{2} \left[s(s+1) - \frac{3}{2} \right]. \quad (30)$$

For the triplet ($s = 1$) and singlet ($s = 0$) manifolds this gives

$$E_{\text{EX}}(s = 1) = \frac{J}{4}, \quad E_{\text{EX}}(s = 0) = -\frac{3J}{4}, \quad (31)$$

so exchange produces a singlet-triplet splitting of J .

Implementation. Because \mathcal{H}_{EX} is diagonal in the coupled two-electron basis $\{|1, 1\rangle, |1, 0\rangle, |0, 0\rangle, |1, -1\rangle\}$, the electron-space operator is implemented as

$$\mathcal{H}_{\text{EX}}^{(e)} = \text{diag} \left(\frac{J}{4}, \frac{J}{4}, -\frac{3J}{4}, \frac{J}{4} \right), \quad (32)$$

and the full 16×16 Hamiltonian is constructed as $\mathcal{H}_{\text{EX}} = \mathcal{H}_{\text{EX}}^{(e)} \otimes \mathbb{1}_4$ in the ordered nuclear basis of Sec. S3 A. In `sympy`² this is implemented by filling the four diagonal electron entries and taking the Kronecker product with $\mathbb{1}_4$.

F. The Spin Hamiltonian

Combining the Zeeman, hyperfine, dipolar (effective ZFS), and exchange contributions derived above, the total 16×16 Spin Hamiltonian used in the simulations is

$$\mathcal{H} = \mathcal{H}_Z + \mathcal{H}_{\text{HF}} + \mathcal{H}_{\text{ZFS}} + \mathcal{H}_{\text{EX}}. \quad (33)$$

Each term is constructed in the basis of Sec. S3 A using the procedures described in Secs. S3 B-S3 E, and the re-

sulting matrix is used as the Spin Hamiltonian input to the steady-state SDR simulations in the main text.

¹D. R. Hart, D. M. Deliwala, R. Byrne, and D. J. Spry, in *Spintronics XVIII*, Vol. 13586, edited by J.-E. Wegrowe, J. S. Friedman, M. Razeghi, and H. Jaffrès, International Society for Optics and Photonics (SPIE, 2025) p. 135860F.

²A. Meurer, C. P. Smith, M. Paprocki, O. Čertík, S. B. Kirpichev, M. Rocklin, A. Kumar, S. Ivanov, J. K. Moore, S. Singh, T. Rathnayake, S. Vig, B. E. Granger, R. P. Muller, F. Bonazzi, H. Gupta, S. Vats, F. Johansson, F. Pedregosa, M. J. Curry, A. R. Terrel, v. Roučka, A. Saboo, I. Fernando, S. Kulal, R. Cimrman, and A. Scopatz, *PeerJ Computer Science* **3**, e103 (2017).

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