

# 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.<sup>1</sup> 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 <sup>1</sup>H 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  $16 \times 16$  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

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

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  $16 \times 16$  matrix is constructed by assigning Eq. (7) along the diagonal for each ordered basis state. In `sympy`<sup>2</sup> 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 as<sup>3</sup>

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

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

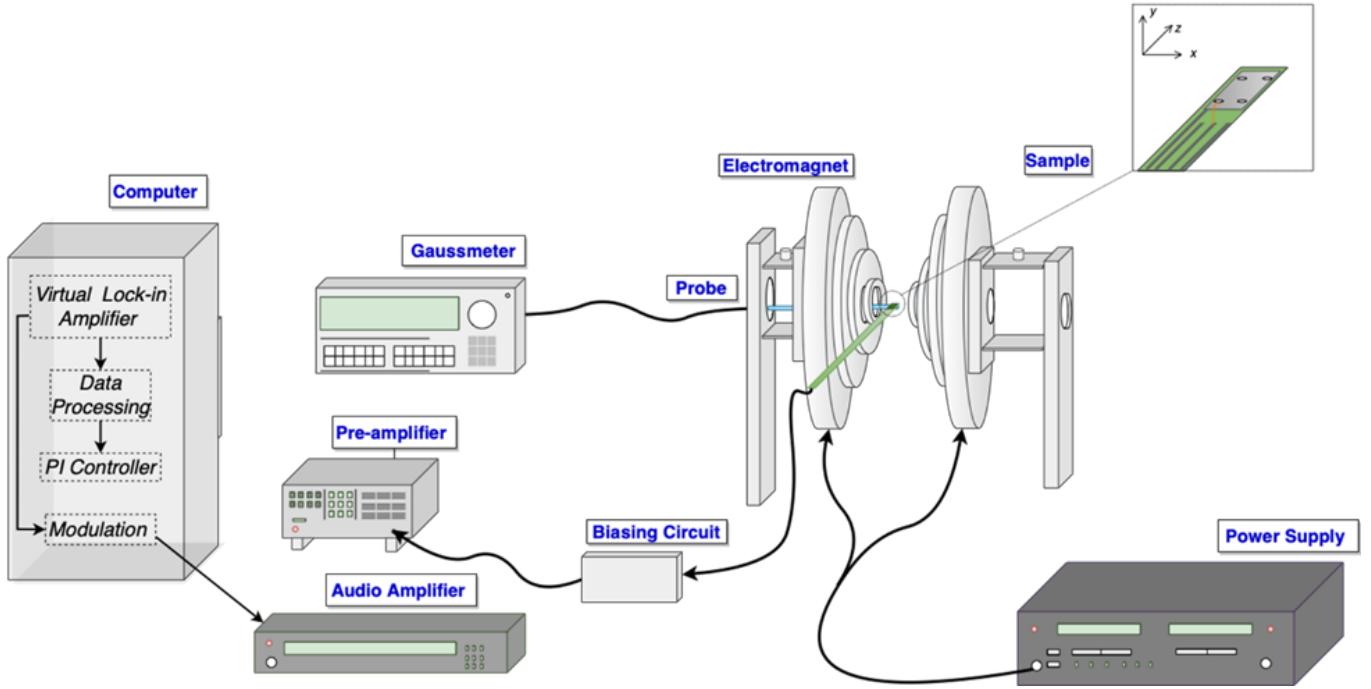


FIG. S1. Schematic of the experimental apparatus used for the cw-EDMR measurements. The figure illustrates the relative placement of the amplifiers, biasing circuit, and the direction of the applied static magnetic field  $B_0$ .

$S_+I_-$  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  $\mathcal{H}_{\text{HF}}$  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`<sup>2</sup> this is implemented by a routine that returns the nonzero targets and coefficients for the action of  $\mathcal{H}_{\text{HF}}$  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. S3 A via the Clebsch-Gordon 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  $16 \times 16$  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 form<sup>3</sup>

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

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  $V_{\text{Si}}^-$  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  $\mathcal{H}_{\text{ZFS}}$  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  $4 \times 4$  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  $16 \times 16$  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`<sup>2</sup>, 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,<sup>3</sup>

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

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  $\mathcal{H}_{\text{EX}}$  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  $16 \times 16$  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`<sup>2</sup> 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  $16 \times 16$  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.

<sup>1</sup>D. 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. Jaffr  s, International Society for Optics and Photonics (SPIE, 2025) p. 135860F.

<sup>2</sup>A. Meurer, C. P. Smith, M. Paprocki, O.  ert k, 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. Rou ka, A. Saboo, I. Fernando, S. Kulal, R. Cimrman, and A. Scopatz, PeerJ Computer Science **3**, e103 (2017).

<sup>3</sup>G. Jeschke, “Physical chemistry iv. part 2: Electron paramagnetic resonance,” Lecture notes, ETH Z  rich (2016).