Accelerated EDMR Simulation for On-Chip Sensing

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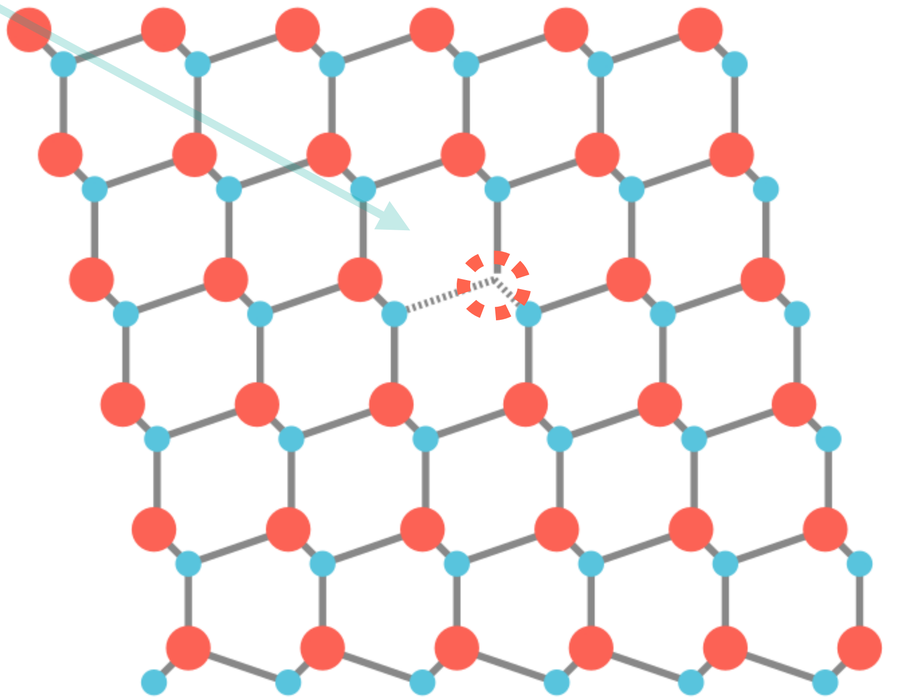
The Silicon Vacancy V_{Si}^{-}

The Silicon Vacancy is a spin-3/2 defect from the absence of a Silicon Atom in the 4H-SiC lattice.

It is theorized to be the primarily active defect responsible for recombination processes (next slide).



4H-SiC *h* lattice

Silicon

Carbon

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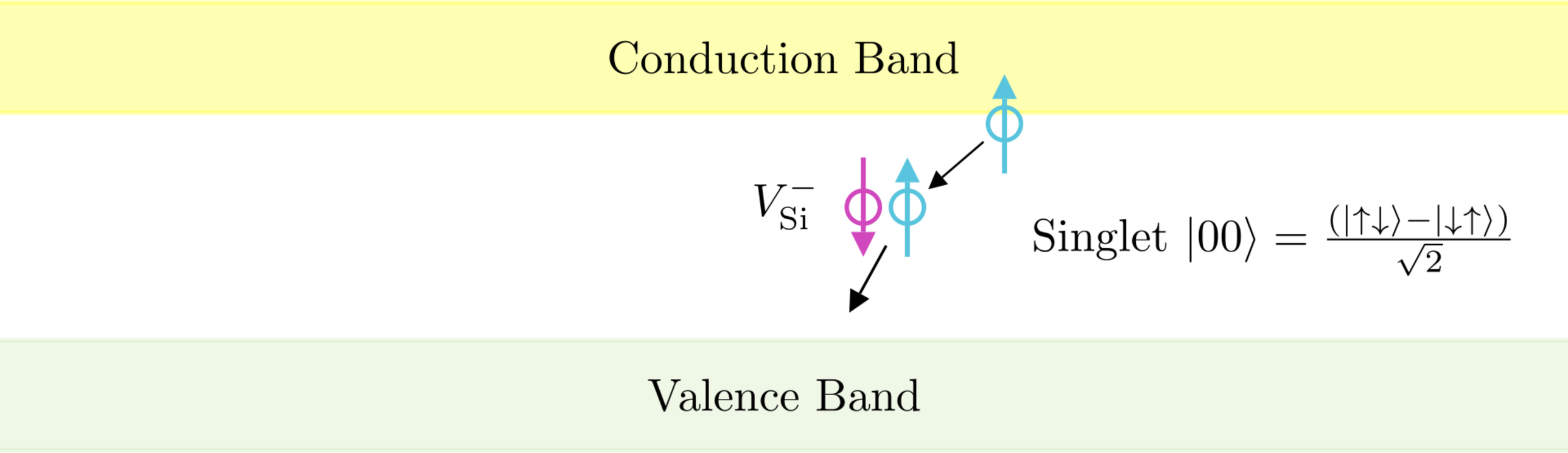
Spin Dependent Recombination (SDR)

SDR involves a carrier electron in the conduction band and a hole at an energy defect.

If carrier $\leftrightarrow$ hole $= |00\rangle$ singlet, the defect can catch the carrier electron and recombine.

Magnetic fields modulate spins $\rightarrow$ change recombination rates $\rightarrow$ change current — EDMR Magnetometry



Spin Selection Rules

Spin is a conserved quantity. After recombination, $S_{\text{tot}} = 0$.

Hence, the initial electron-hole pair must also have total $S = 0$.

Formally, recombination is governed by a matrix element of the operator $\mathscr{H}_{\text{rec}}$ that commutes with total spin $\hat{S}^2$:

$$[\mathscr{H}_{\text{rec}}, \hat{S}^2] = 0$$

Wigner-Eckart —

$$\langle S', m'_S \,|\, \mathscr{H}_{\text{rec}} \,|\, S, m_S \rangle \quad \propto \quad \delta_{S'S}\, \delta_{m'_S m_S}$$

The final state has $S_{\text{tot}} = 0$, hence the allowed initial state has $S = 0$.

The Spin Hamiltonian

The Spin Hamiltonian primarily governs the spin physics of 4H-SiC.

It is the combination of Zeeman, Hyperfine, Zero-field splitting, and Exchange Interactions

$$\mathscr{H} = \hat{H}_Z + \hat{H}_{HF} + \hat{H}_{ZFS} + \hat{H}_{EX}$$

$$\hat{H}_Z = \mu_B \vec{S} \cdot g \cdot \overrightarrow{B}_0 \qquad \hat{H}_{HF} = \hat{S} \cdot A \cdot \hat{I} \qquad \hat{H}_{ZFS} = \hat{S} \cdot D \cdot \hat{S} \qquad \hat{H}_{EX} = -J\hat{S}_a \cdot \hat{S}_b$$

In 4H-SiC SDR, the relevant spin system has 2 spin-1/2 electrons (carrier + defect hole) and two spin-1/2 nuclei (Silicon and Carbon).

Hence, as each particle has two possible states — $|m_s = \pm\frac{1}{2}\rangle \in \{|\uparrow\rangle, |\downarrow\rangle\}$, there are

$$2^4 = 16; \qquad \dim\mathscr{H} = 16$$

basis states spanning the 4H-SiC Hilbert space (with V_{Si}^{-} only).

The Spin Hamiltonian

We define the electron-coupled + nuclear-zeeman basis: $\mathscr{B} = \{\,|s,m\rangle \otimes |m_{I_1}, m_{I_2}\rangle\}$.

The electrons are coupled together, whereas the nuclei I_1, I_2 remain in their Zeeman $|\uparrow,\downarrow\rangle$ basis.

$\|1,1\rangle$ manifold	$\|1,0\rangle$ manifold	$\|0,0\rangle$ manifold	$\|1,-1\rangle$ manifold
$\|1,1\rangle \otimes \left\|+\frac{1}{2}\right\rangle_1 \left\|+\frac{1}{2}\right\rangle_2$	$\|1,0\rangle \otimes \left\|+\frac{1}{2}\right\rangle_1 \left\|+\frac{1}{2}\right\rangle_2$	$\|0,0\rangle \otimes \left\|+\frac{1}{2}\right\rangle_1 \left\|+\frac{1}{2}\right\rangle_2$	$\|1,-1\rangle \otimes \left\|+\frac{1}{2}\right\rangle_1 \left\|+\frac{1}{2}\right\rangle_2$
$\|1,1\rangle \otimes \left\|+\frac{1}{2}\right\rangle_1 \left\|-\frac{1}{2}\right\rangle_2$	$\|1,0\rangle \otimes \left\|+\frac{1}{2}\right\rangle_1 \left\|-\frac{1}{2}\right\rangle_2$	$\|0,0\rangle \otimes \left\|+\frac{1}{2}\right\rangle_1 \left\|-\frac{1}{2}\right\rangle_2$	$\|1,-1\rangle \otimes \left\|+\frac{1}{2}\right\rangle_1 \left\|-\frac{1}{2}\right\rangle_2$
$\|1,1\rangle \otimes \left\|-\frac{1}{2}\right\rangle_1 \left\|+\frac{1}{2}\right\rangle_2$	$\|1,0\rangle \otimes \left\|-\frac{1}{2}\right\rangle_1 \left\|+\frac{1}{2}\right\rangle_2$	$\|0,0\rangle \otimes \left\|-\frac{1}{2}\right\rangle_1 \left\|+\frac{1}{2}\right\rangle_2$	$\|1,-1\rangle \otimes \left\|-\frac{1}{2}\right\rangle_1 \left\|+\frac{1}{2}\right\rangle_2$
$\|1,1\rangle \otimes \left\|-\frac{1}{2}\right\rangle_1 \left\|-\frac{1}{2}\right\rangle_2$	$\|1,0\rangle \otimes \left\|-\frac{1}{2}\right\rangle_1 \left\|-\frac{1}{2}\right\rangle_2$	$\|0,0\rangle \otimes \left\|-\frac{1}{2}\right\rangle_1 \left\|-\frac{1}{2}\right\rangle_2$	$\|1,-1\rangle \otimes \left\|-\frac{1}{2}\right\rangle_1 \left\|-\frac{1}{2}\right\rangle_2$

Able to recombine

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Calculating Hamiltonians

$$\hat{H}_Z \,|\, s, m\rangle = m g \mu_B B \,|\, s, m\rangle \,.$$

$$\hat{H}_{\mathrm{ZFS}} \,|\, s, m\rangle = D\, m^2 \,|\, s, m\rangle \;-\; \frac{D}{3}\, s(s+1) \,|\, s, m\rangle$$

$$+\frac{E}{2}\Big[s(s+1) - m(m+1)\Big] \,|\, s, m+2\rangle$$

$$+\frac{E}{2}\Big[s(s+1) - m(m-1)\Big] \,|\, s, m-2\rangle \,.$$

Forbidden $\Delta m = 2$ transitions

$$\hat{H}_{EX} \,|\, s, m\rangle = -\,J \left[\frac{s(s+1) - \frac{3}{2}}{2}\right] |\, s, m\rangle \,.$$

Hamiltonian Action on Electronic-States

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Calculating Hamiltonians

We calculate the Hyperfine 16×16 Hamiltonian on the full-Zeeman basis and covert to the coupled-zeeman basis via Clebsch-Gordan.

For two electrons with *one* nearby nucleus:

$$\hat{H}_{HF} \left| m_a, m_b, m_I \right\rangle = \sum_{k \in \{a,b\}} \frac{\hbar^2}{4} \left(A_{kx} - A_{ky}\right) \hat{S}_{k+} \hat{I}_{-} \, \delta_{m_k, m_I} \left| -m_k, -, -m_I \right\rangle + \sum_{k \in \{a,b\}} \frac{\hbar^2}{4} \left(A_{kx} - A_{ky}\right) \hat{S}_{k-} \hat{I}_{+} \, \delta_{m_k, -m_I} \left| -m_k, -, -m_I \right\rangle + \sum_{k \in \{a,b\}} A_{kz} \hbar^2 m_k m_I \left| m_k, -, m_I \right\rangle .$$

For two electrons with *two* nearby nuclei:

$$\hat{H}_{HF} \left| m_a, m_b, m_{I_1}, m_{I_2} \right\rangle = \hbar^2 \sum_{p=1}^{2} \left(A_{apz} m_a m_{I_p} + A_{bpz} m_b m_{I_p} \right) \left| m_a, m_b, m_{I_1}, m_{I_2} \right\rangle + \frac{\hbar^2}{4} \left[\left(A_{a1x} - A_{a1y}\right) \delta_{m_a, m_{I_1}} + \left(A_{a1x} + A_{a1y}\right) \delta_{m_a, -m_{I_1}} \right] \left| -m_a, m_b, -m_{I_1}, m_{I_2} \right\rangle + \frac{\hbar^2}{4} \left[\left(A_{a2x} - A_{a2y}\right) \delta_{m_a, m_{I_2}} + \left(A_{a2x} + A_{a2y}\right) \delta_{m_a, -m_{I_2}} \right] \left| -m_a, m_b, m_{I_1}, -m_{I_2} \right\rangle + \frac{\hbar^2}{4} \left[\left(A_{b1x} - A_{b1y}\right) \delta_{m_b, m_{I_1}} + \left(A_{b1x} + A_{b1y}\right) \delta_{m_b, -m_{I_1}} \right] \left| m_a, -m_b, -m_{I_1}, m_{I_2} \right\rangle + \frac{\hbar^2}{4} \left[\left(A_{b2x} - A_{b2y}\right) \delta_{m_b, m_{I_2}} + \left(A_{b2x} + A_{b2y}\right) \delta_{m_b, -m_{I_2}} \right] \left| m_a, -m_b, m_{I_1}, -m_{I_2} \right\rangle .$$

$$\hat{H}_{HF_{\text{coupled}}} = W \left(\hat{H}_{HF_{\text{zeeman}}} \right) W^{\dagger}, \qquad W = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\ 0 & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \otimes \mathbb{I}_{4 \times 4}$$

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5 columns from 16×16 $[\mathscr{H}]_{\mathscr{B}}$

$$
\begin{bmatrix}
0 \\ 0 \\ 0 \\ \frac{\sqrt{2}\hbar^2(A_{a1}+A_{b1})}{4} \\ 0 \\ \frac{B_0 g_{n1}\mu_N}{2} - \frac{B_0 g_{n2}\mu_N}{2} - \frac{2D_1}{3} - \frac{J}{4} \\ 0 \\ 0 \\ 0 \\ \frac{\hbar^2(A_{a1}-A_{a2}-A_{b1}+A_{b2})}{4} \\ 0 \\ 0 \\ \frac{\sqrt{2}\hbar^2(A_{a2}+A_{b2})}{4} \\ 0 \\ 0 \\ 0
\end{bmatrix}
\quad
\begin{bmatrix}
0 \\ 0 \\ 0 \\ \frac{\sqrt{2}\hbar^2(A_{a2}+A_{b2})}{4} \\ 0 \\ \frac{B_0 g_{n1}\mu_N}{2} - \frac{B_0 g_{n2}\mu_N}{2} + \frac{3J}{4} \\ 0 \\ 0 \\ 0 \\ \frac{\hbar^2(-A_{a1}+A_{a2}+A_{b1}-A_{b2})}{4} \\ 0 \\ \frac{\sqrt{2}\hbar^2(A_{a1}+A_{b1})}{4} \\ 0 \\ 0 \\ 0
\end{bmatrix}
\quad
\begin{bmatrix}
0 \\ 0 \\ 0 \\ 0 \\ D_2 \\ 0 \\ 0 \\ -B_0 g_e \mu_B + \frac{B_0 g_{n1}\mu_N}{2} - \frac{B_0 g_{n2}\mu_N}{2} + \frac{D_1}{3} - \frac{J}{4} \\ 0 \\ 0 \\ 0 \\ \frac{\hbar^2(-A_{a1}-A_{a2}+A_{b1}+A_{b2})}{4} \\ 0 \\ \frac{\sqrt{2}\hbar^2(A_{a1}+A_{b1})}{4} \\ \frac{\sqrt{2}\hbar^2(A_{a2}+A_{b2})}{4} \\ 0
\end{bmatrix}
$$

$$
\begin{bmatrix}
0 \\ \frac{\sqrt{2}\hbar^2(-A_{a2}+A_{b2})}{4} \\ \frac{\sqrt{2}\hbar^2(-A_{a1}+A_{b1})}{4} \\ 0 \\ \frac{\hbar^2(A_{a1}+A_{a2}-A_{b1}-A_{b2})}{4} \\ 0 \\ 0 \\ 0 \\ B_0 g_e \mu_B - \frac{B_0 g_{n1}\mu_N}{2} + \frac{B_0 g_{n2}\mu_N}{2} + \frac{D_1}{3} - \frac{J}{4} \\ 0 \\ 0 \\ D_2 \\ 0 \\ 0 \\ 0 \\ 0
\end{bmatrix}
\quad
\begin{bmatrix}
0 \\ 0 \\ 0 \\ \frac{\sqrt{2}\hbar^2(-A_{a1}+A_{b1})}{4} \\ 0 \\ \frac{\hbar^2(A_{a1}-A_{a2}-A_{b1}+A_{b2})}{4} \\ 0 \\ 0 \\ 0 \\ -\frac{B_0 g_{n1}\mu_N}{2} + \frac{B_0 g_{n2}\mu_N}{2} - \frac{2D_1}{3} - \frac{J}{4} \\ 0 \\ 0 \\ \frac{\sqrt{2}\hbar^2(A_{a2}-A_{b2})}{4} \\ 0 \\ 0 \\ 0
\end{bmatrix}
$$

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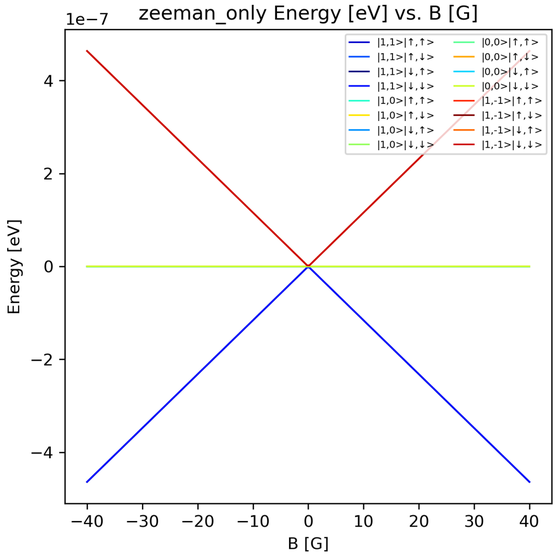
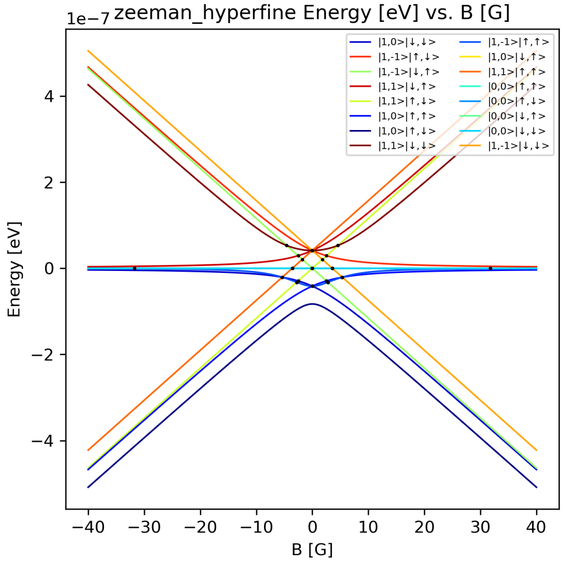
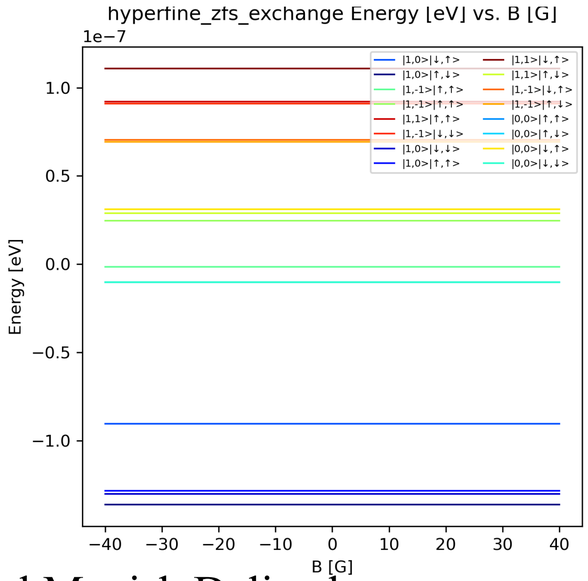
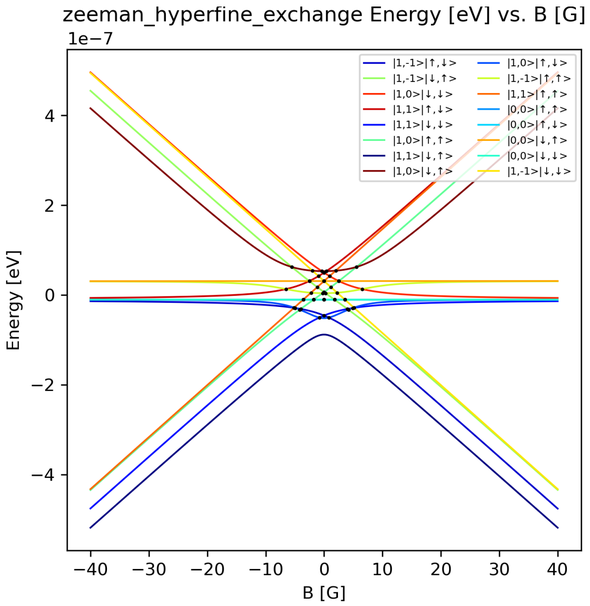
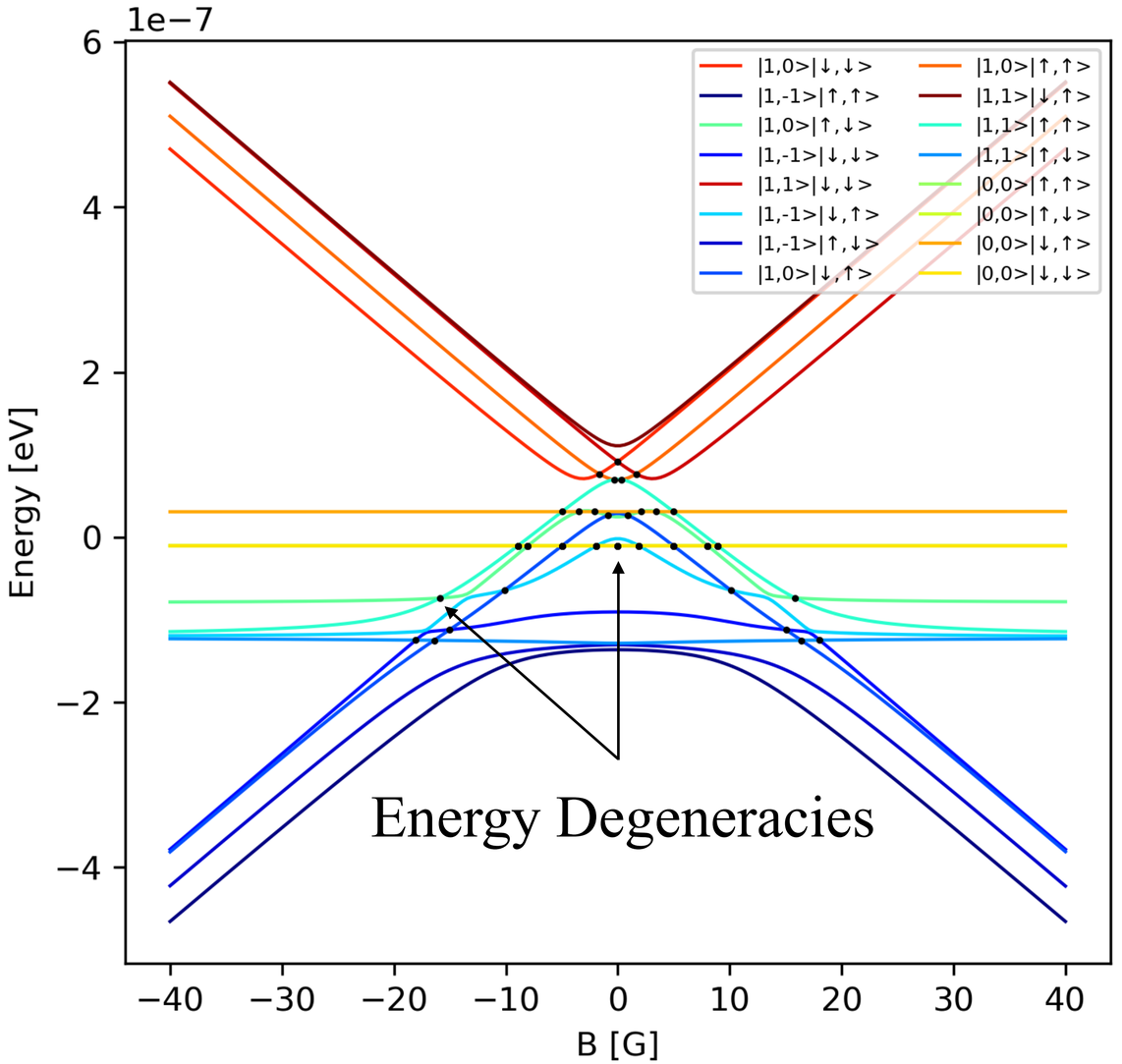
Eigenspectra Simulations











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The Stochastic Liouville Equation (SLE)

The Schrödinger Equation

Dissipation Term at rate k

Generation Term at rate p

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar}[\mathscr{H}, \rho] - \frac{k}{2}\{\Lambda, \rho\} + p\Gamma$$

Describes the evolution of the spin-ensemble *with* environmental interactions.

The *first* term reproduces the unitary Schrödinger Equation in density-matrix formalism — the "Quantum Liouville Equation (QLE)."

The *second* term introduces *dissipation* at rate k: the projection operator Λ removes spin-pairs from its subspace.

The *third* term acts as a *source* — injecting spin-pairs with random orientations at rate p.

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The Stochastic Liouville Equation (SLE)

$$\frac{\partial \rho}{\partial t} = 0 = -\frac{i}{\hbar}[\mathscr{H}, \rho] - \frac{1}{2}(k_S + k_D)\{\Lambda_S, \rho\} - \frac{1}{2}k_D\{\Lambda_T, \rho\} + \frac{1}{16}p\Gamma\,.$$

From Hansen and Pedersen [1].

- The second term, via the singlet projector Λ_S, removes singlet states at the effective rate $(k_S + k_D)$
- The third term, via the triplet projector Λ_T, dissociates triplets at rate k_D
- The final term injects carriers at rate p, with $\Gamma = \mathbb{I}_{16\times16}$. The factor 1/16 normalizes this over the 16-dimensional Hilbert space

We solve the steady-state SLE since carrier generation and annihalation are at constant rates. This will return, the density matrix:

$$\rho(\vec{\theta}), \qquad \vec{\theta} = \langle B, g, \mu_B, \mu_N, A, J, D, \cdots \rangle$$

which encapsulates the full spin-physics — as a function of the Hamiltonian parameters.

$$0 = -\frac{i}{\hbar}[\mathscr{H}, \rho] - \frac{1}{2}(k_S + k_D)\{\Lambda_S, \rho\} - \frac{1}{2}k_D\{\Lambda_T, \rho\} + \frac{1}{16}p\Gamma\,.$$

Rearrange to Sylvester-Lyanupov form:

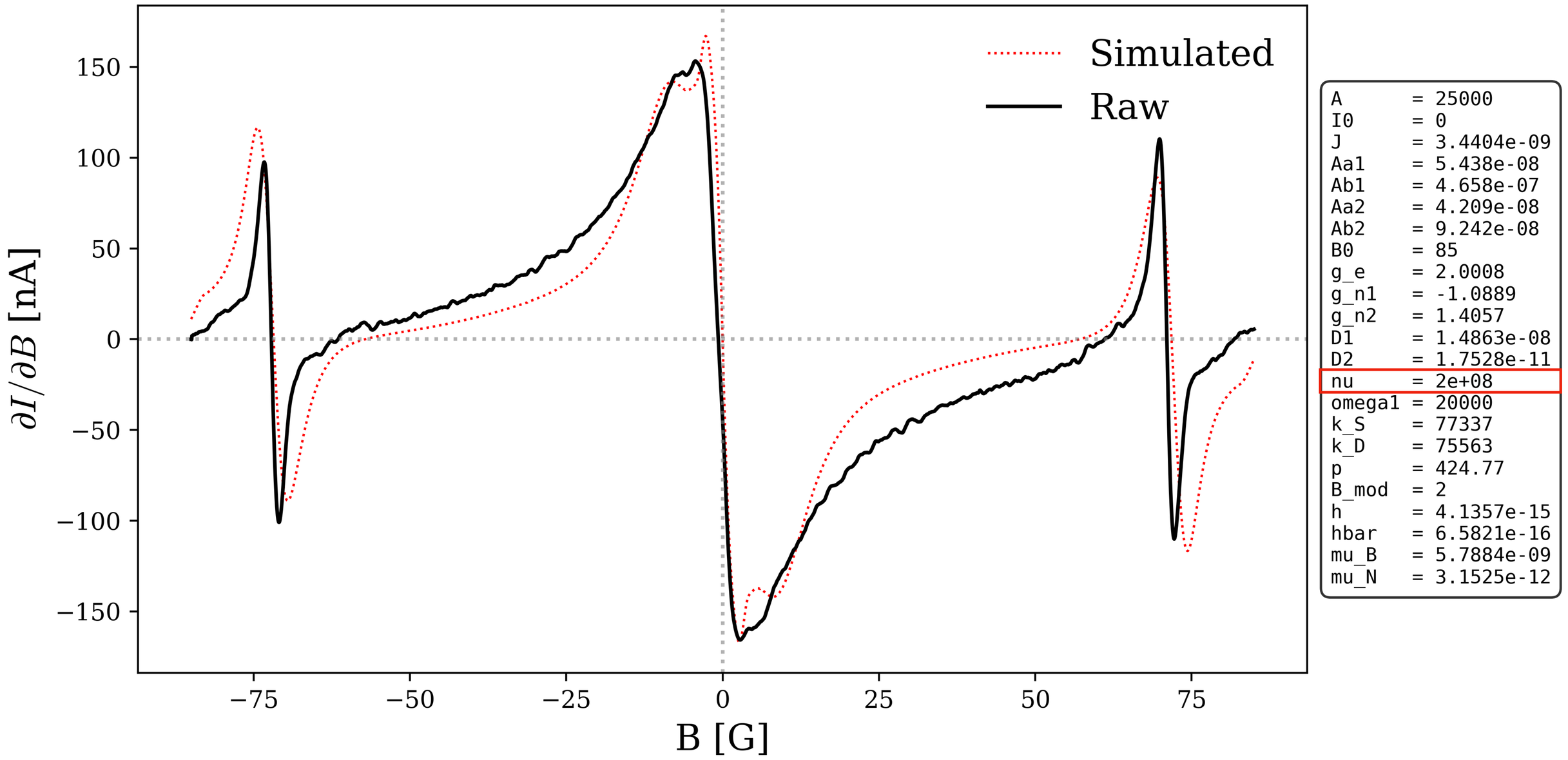
$$A\rho + \rho A^\dagger + g = 0$$

$$A = -\frac{i}{\hbar}\mathscr{H} - \frac{1}{2}(k_S + k_D)\Lambda_S - \frac{1}{2}k_D\Lambda_T, \qquad g = -\frac{1}{16}\Gamma\,.$$

The solution is given by $\rho = \int_0^\infty e^{-iAt} g^{iA^\dagger t}\, dt$, but difficult numerically.

We vectorize into a 256×256 linear system using the identity, with Fortran column ordering

$$\mathrm{vec}(X\rho Y) = \left(Y^T \otimes X\right)\mathrm{vec}(\rho) \qquad \Rightarrow \qquad \left(I \otimes A + A^\dagger \otimes I\right)\mathrm{vec}(\rho) = -\,\mathrm{vec}(g)$$



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Building the NZFMR Model

Now that we have the density matrix $\rho(\vec{\theta}) \simeq \rho(B)$, we can compute the *singlet population* in the silicon-vacancy — carrier electron system as a function of B.

$$\text{Singlet Population (B)} = \text{Tr}(\Lambda_S \rho(B))$$

Since only singlet states recombine and thereby modulate the observed EDMR current, the singlet population is *directly proportional* to the EDMR signal. Therefore, our EDMR model takes the simple form

$$I(B) \simeq A\text{Tr}(\Lambda_S \rho(B)) + I_0$$

However, this is only with Hamiltonian Parameters (NZFMR) — not experimental (RF Freq.) that produce the resonances. We now add the Rotating Wave Approximation.

Rotating Wave Approximation

In Magnetic Resonance, we have

- Large static field $B_0\hat{k}$ giving Larmor precession at RF frequency ν — Zeeman Resonances
- Transverse oscillting field $B_1\cos(2\pi\nu t)$ that drives spin transitions, yielding the extra *drive* hamiltonian:

$$\mathscr{H}_{\text{drive}}(t) = h\omega_1\cos(2\pi\nu t)\hat{S}_x, \qquad \omega_1 = \gamma_e B_1, \ \ [\text{Hz}]$$

This adds time-dependence!, making it too complicated to solve the SLE *for every B*...
Additionally, in the lab-frame, states whip around at $\nu \sim$ GHz — difficult numerically.

We expand the drive hamiltonian in terms of raising/lowering operators in the rotating frame, and drop the fast oscillating $e^{\pm i4\pi\nu t} \sim 0$ terms:

$$\mathscr{H}'_{\text{drive}}(t) = \frac{h\omega_1}{2}\hat{S}_x + \frac{h\omega_1}{2}\left[\hat{S}_+ e^{+i4\pi\nu t} + \hat{S}_- e^{-i4\pi\nu t}\right] \qquad \Rightarrow \qquad \mathscr{H}^{RWA}_{\text{drive}} = \frac{h\omega_1}{2}\hat{S}_x\,.$$

$$\boxed{\mathscr{H}_{\text{effective}} = \mathscr{H} + \mathscr{H}^{RWA}_{\text{drive}}}$$

Final EDMR Model

Lock-in amplication is implemented via numerically solving $X(B_i) = \frac{2}{T}\int_0^T I(B_i + \beta \sin \omega_m t)\sin \omega_m t \, dt$
(the first-harmonic, in phase, idealized lock-in output)

Given the effective Hamiltonian $\mathscr{H}_{\text{effective}}$

For every *B* value:

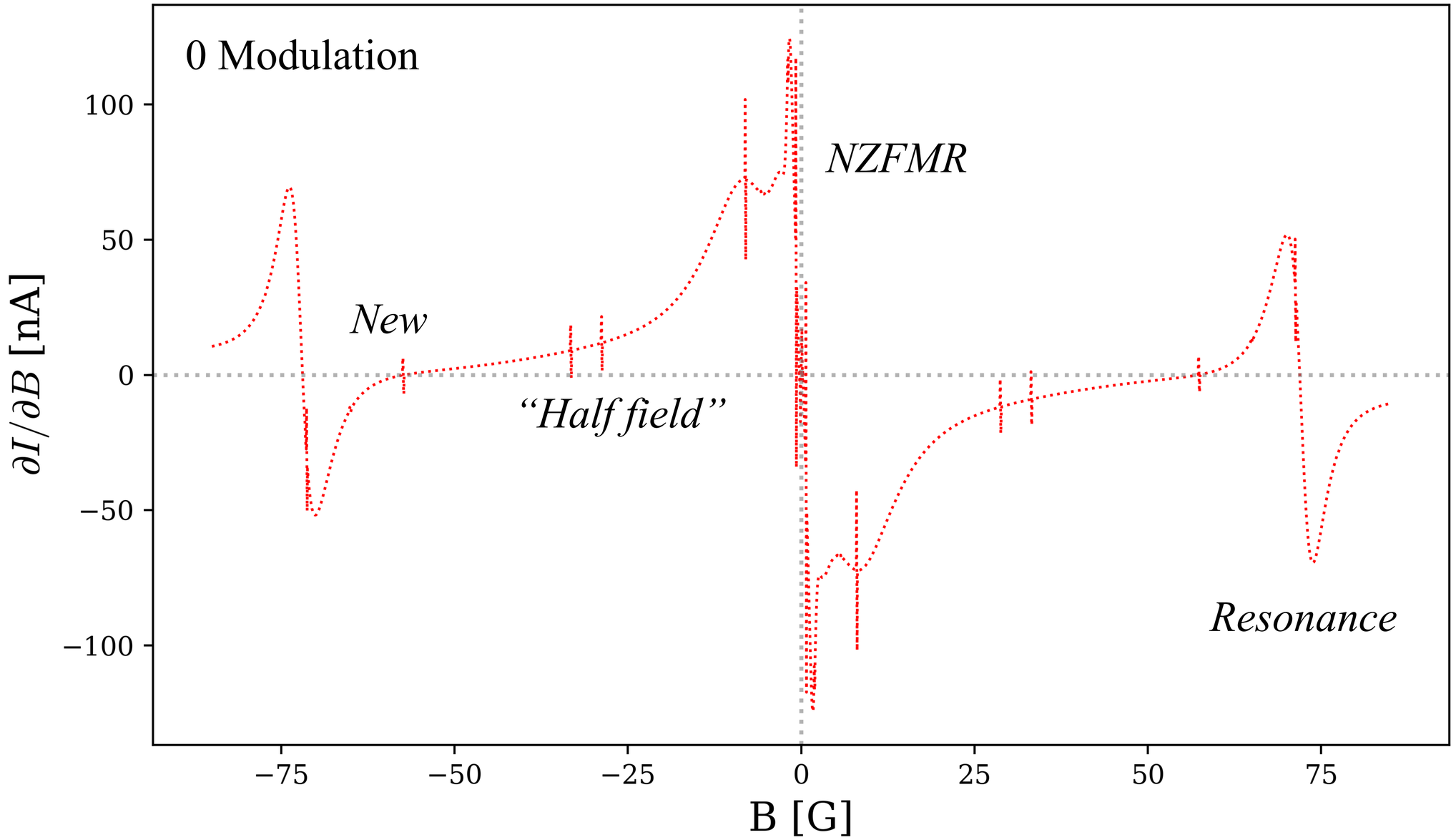
1. Solve the effective SLE for $\rho(\vec{\theta}) \simeq \rho(B)$
2. Calculate Singlet Population(*B)*
3. Perform Lock-in amplification
4. Determine proportionality constants A, I_0 from fitting.

Final EDMR Model

Quoting Jakob Nielsen [2]:

- 0.1 seconds is about the limit for having the user feel that the system is reacting instantaneously.
- 1.0 seconds is about the limit for the user's flow of thought to stay uninterrupted
- 10 seconds is about the limit for keeping the user's attention

A 200-point sweep (-100G to 100G) takes **~0.2s** to render on a *single* M3 silicon core.



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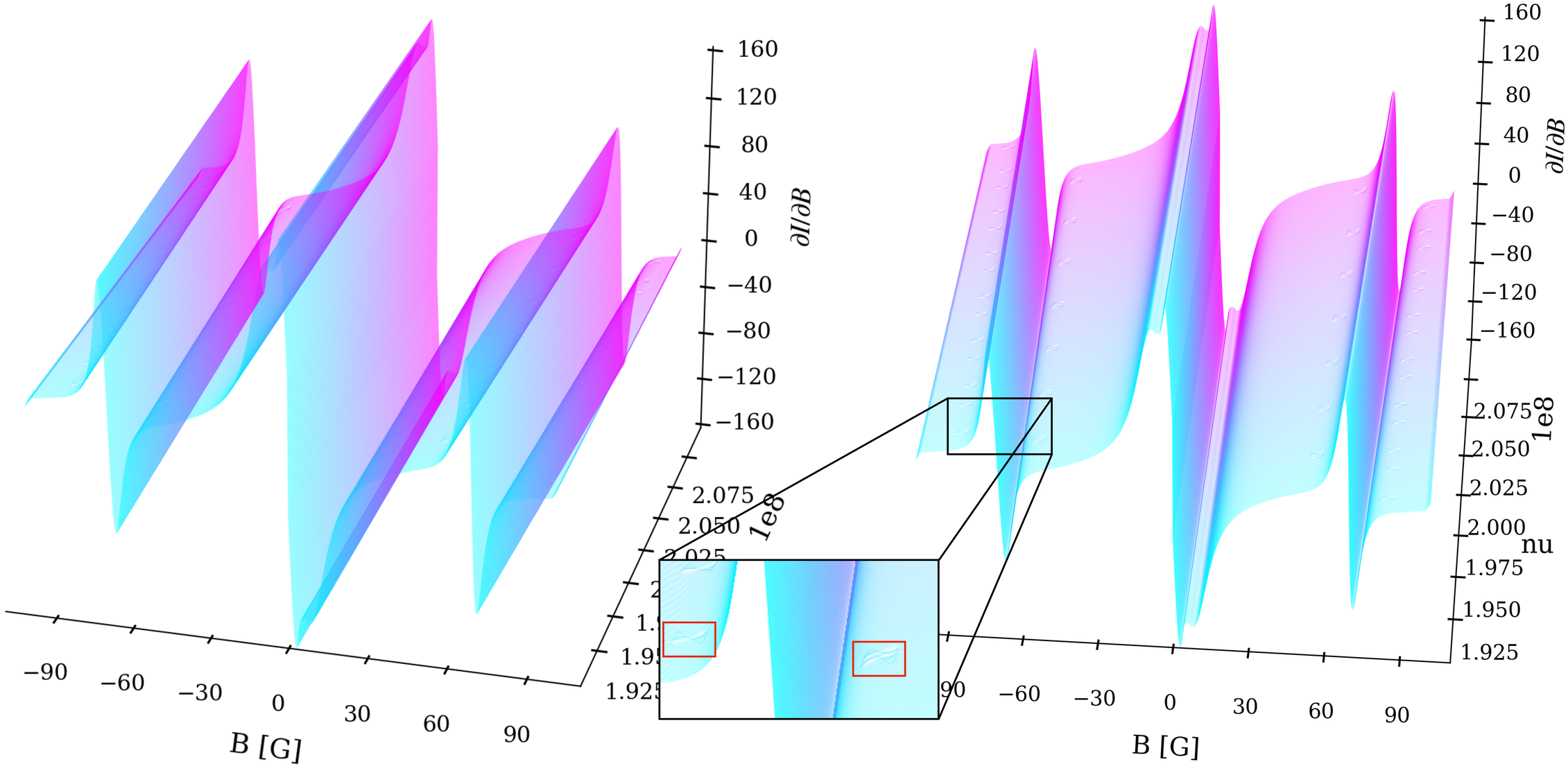

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References

[1] T. Hansen and N. L. Pedersen, “Spin-dependent recombination studied by steady state stochastic Liouville theory,” Chem. Phys. Lett. 363, 636–642 (2002).

[2] Jakob Nielsen. Usability engineering. Morgan Kaufmann, 1994.

[3] D. J. Lépine, “Spin-dependent recombination on silicon surfaces,” Phys. Rev. B 6, 436–441 (1972).

[4] Ö. O. Soykal, P. Dev, and S. E. Economou, “Silicon vacancy center in 4H-SiC: Electronic structure and spin–photon interfaces,” Phys. Rev. B 93, 081207 (2016).

[5] C. J. Cochrane and P. M. Lenahan, “Zero-field detection of spin-dependent recombination with direct observation of electron–nuclear hyperfine interactions in the absence of an oscillating electromagnetic field,” J. Appl. Phys. 112(12), 123714 (2012).