Zeeman and Hyperfine

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The spin Hamiltonian $\mathscr{H}$ governs the spin physics of recombination. It is what we need to implement before simulating EDMR.

$\mathscr{H}$ is a sum of the Zeeman effect, hyperfine interactions, zero-field splitting, and the exchange interaction.

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

Normally $\mathscr{H}$ also includes a nuclear quadrupole interaction term. In 4H-SiC, there are no nuclei with nuclear spin $I > \frac{1}{2}$, so we set that term to zero.

The goal of this post is to derive the first two Hamiltonian terms. The next two are reviewed in the next post.

I use a fancy $\mathscr{H}$ to represent the full spin Hamiltonian. Sub-Hamiltonians are normal H's but with hats (for example, $\hat{H}_Z$).

The Zeeman Effect

$\hat{H}_Z$ determines how spin-state energies split in the presence of an external magnetic field $\vec{B}$. It is the dominant term in the spin Hamiltonian that drives recombination.

A spinning charged particle is a magnetic dipole. Its magnetic dipole moment $\vec{\mu}$ is proportional to its spin angular momentum $\vec{S}$:

$$\vec{\mu} = \gamma \vec{S}.$$

The proportionality constant γ is the gyromagnetic ratio. From the Dirac equation it can be shown

$$\vec{\mu} = \gamma \vec{S} = -g\left(\frac{q}{2m_e}\right)\vec{S} = -\left(\frac{g\mu_B}{\hbar}\right)\vec{S},$$

where $g \approx 2.0023$ is the Landé g-factor for the free electron and $\mu_B \approx 9.2 \cdot 10^{-24}$ J/T is the Bohr magneton.

When a magnetic dipole is placed in a magnetic field $\vec{B}$, it experiences a torque $\vec{\mu} \times \vec{B}$ which tends to align it with the field like a compass. The energy associated with this torque is

$$H = -\vec{\mu} \cdot \vec{B} = -\gamma \vec{B} \cdot \vec{S}.$$

Therefore, the Zeeman Hamiltonian is

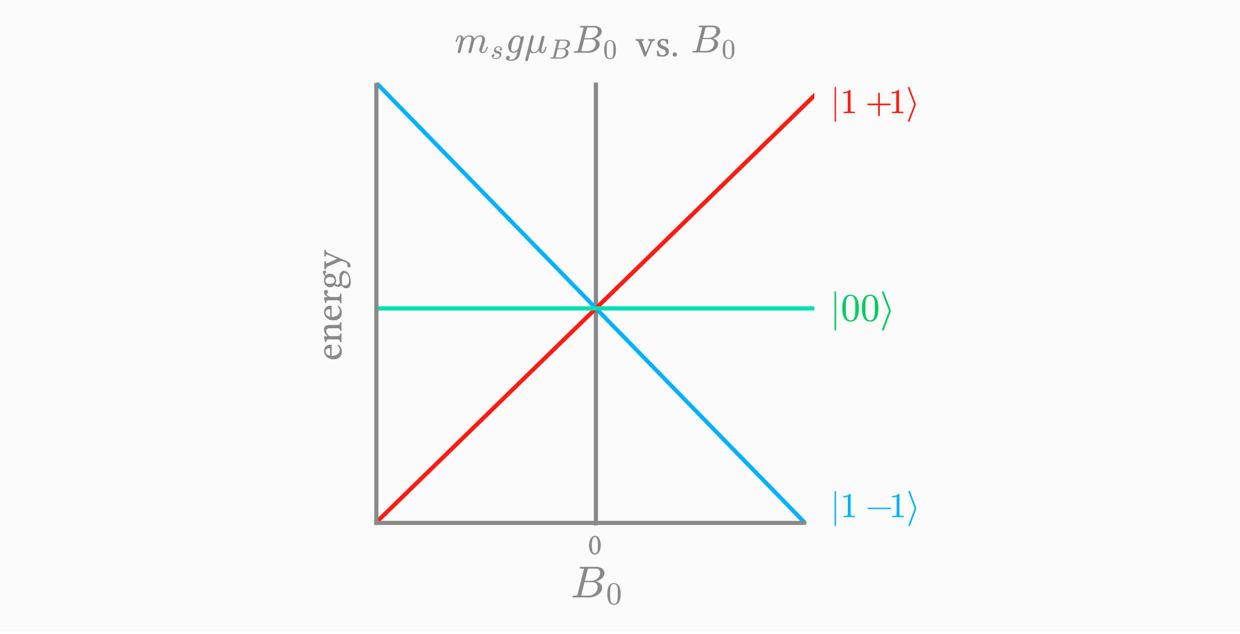
$$\hat{H}_Z = -\vec{\mu} \cdot \vec{B}_0 = \left(\frac{g\mu_B}{\hbar}\right)\vec{S} \cdot \vec{B}_0 = \left(\frac{g\mu_B}{\hbar}\right)B_0 S_z$$

where $S_z = \left(\frac{\hbar}{2}\right)\hat{\sigma}_z$ and σ_z is the Pauli-z spin matrix. Applying $\hat{H}_Z$ on a spin state $|s, m_s\rangle$ gives

$$\hat{H}_Z\ |s, m_s\rangle = \left(\frac{g\mu_B}{\hbar}\right)B_0 S_z\ |s, m_s\rangle$$

$$= \left(\frac{g\mu_B}{\hbar}\right)B_0(m_s\hbar)\ |s, m_s\rangle = m_s g\mu_B B_0\ |s, m_s\rangle.$$

From the equation above, we can calculate $\hat{H}_Z$'s effect on any spin state $|s, m_s\rangle$.



Ordinarily we use an anisotropic g-tensor. Magnetic fields in different directions act differently on $|s, m_s\rangle$. In that case,

$$\hat{H}_Z = \mu_B \vec{S} \cdot g \cdot \vec{B}_0.$$

This can be diagonalized along the principal axis:

$$g_{\mathrm{diag}} = \begin{pmatrix} g_x & 0 & 0 \\ 0 & g_y & 0 \\ 0 & 0 & g_z \end{pmatrix}.$$

For our simulation, we start with $g \approx 2.0023$. Our Zeeman Hamiltonian is

$$\hat{H}_Z\ |s, m_s\rangle = m_s g\mu_B B_0\ |s, m_s\rangle.$$

Hyperfine Interaction

The hyperfine interaction comes from the magnetic field generated by the nucleus $\vec{B}(\vec{r})$ acting on the magnetic moment of an orbiting electron. We will calculate $\vec{B}(\vec{r})$ and use the dipole energy to construct $\hat{H}_{\mathrm{HF}}$.

Maxwell's equations say

$$\nabla \cdot \vec{B} = 0, \quad \nabla \times \vec{B} = \mu_0 \vec{J}.$$

Vector Potential

The first equation tells us there are no magnetic monopoles; the magnetic field $\vec{B}$ has zero divergence. From vector calculus this means there exists a vector field $\vec{A}$ such that

$$\vec{B} = \nabla \times \vec{A}.$$

$\vec{A}$ is not unique, but to make the math simple we can impose the Coulomb gauge condition $\nabla \cdot \vec{A} = 0$. Substituting yields

$$\nabla^2 \vec{A} = -\mu_0 \vec{J}.$$

Current Density

$\vec{J}$ is the current density. If we model a localized nuclear magnetic moment $\vec{\mu}_n$ at the origin, it generates an effective current density

$$\vec{J}_{n(\vec{r}')} = \nabla' \times \left[\vec{\mu}_n \delta^3(\vec{r}')\right].$$

The general solution for $\vec{A}$ is

$$\vec{A}(\vec{r}) = \left(\frac{\mu_0}{4\pi}\right)\int d^3r' \left(\frac{\vec{J}_{n(\vec{r}')}}{|\vec{r} - \vec{r}'|}\right)$$

$$= \left(\frac{\mu_0}{4\pi}\right)\vec{\mu}_n \times \nabla\left(\frac{1}{r}\right) = \left(\frac{\mu_0}{4\pi}\right)\frac{\vec{\mu}_n \times \vec{r}}{r^3}.$$

Then, using the identity

$$\nabla \times (\vec{a} \times \vec{r} f(r)) = \vec{a}\nabla \cdot (\vec{r} f) - (\vec{a} \cdot \nabla)(\vec{r} f),$$

with $\vec{a} = \vec{\mu}_n$ and $f(r) = \frac{1}{r^3}$, we get the canonical expression for a magnetic field from a dipole in classical electrodynamics:

$$\vec{B}(\vec{r}) = \left(\frac{\mu_0}{4\pi r^3}\right)\left[3(\vec{\mu}_n \cdot \hat{r})\hat{r} - \vec{\mu}_n\right] + \left(\frac{2\mu_0}{3}\right)\vec{\mu}_n \delta^3(\vec{r}).$$

Hamiltonian Density

A magnetic moment $\vec{\mu}_e$ at $\vec{r}$ has energy

$$U(\vec{r}) = -\vec{\mu}_e \cdot \vec{B}(\vec{r}).$$

Substituting $\vec{B}(\vec{r})$ with the expression above, we can separate the energy into two terms:

$$U_{\mathrm{dipolar}}(\vec{r}) = -\vec{\mu}_e \cdot \left[\left(\frac{\mu_0}{4\pi r^3}\right)(3(\vec{\mu}_n \cdot \hat{r})\hat{r} - \vec{\mu}_n)\right],$$

$$U_{\mathrm{contact}}(\vec{r}) = -\vec{\mu}_e \cdot \left[\left(\frac{2\mu_0}{3}\right)\vec{\mu}_n \delta^3(\vec{r})\right].$$

Promoting magnetic moments to their quantum operators,

$$\vec{\mu}_e = -g_e \mu_B \left(\frac{\hat{S}}{\hbar}\right), \quad \vec{\mu}_n = +g_n \mu_N \left(\frac{\hat{I}}{\hbar}\right),$$

where the Bohr magneton $\mu_B = \frac{e\hbar}{2m_e}$ and the nuclear magneton $\mu_N = \frac{e\hbar}{2m_p}$.

Now substitute into the energy expression to get the Hamiltonian of the electron in the magnetic field generated by the nucleus:

$$H(\vec{r}) = \left(\frac{\mu_0}{4\pi}\right)\left(\frac{g_e \mu_B g_n \mu_N}{\hbar^2}\right)\left(\frac{\vec{S} \cdot \vec{I} - 3\left(\vec{S} \cdot \hat{r}\right)\left(\vec{I} \cdot \hat{r}\right)}{r^3} + \left(\frac{8\pi}{3}\right)\delta^3(\vec{r})\left(\vec{S} \cdot \vec{I}\right)\right).$$

The prefactor is a constant. Call it C. If an electron occupies a spatial wavefunction $\psi(\vec{r})$, the full hyperfine Hamiltonian is

$$\hat{H}_{\mathrm{HF}} = \int d^3r, |\psi(\vec{r})|^2 H(\vec{r}).$$

Now write the Hamiltonian in index form:

$$H(\vec{r}) = C\left[\left(\frac{1}{r^3}\right)(\delta_{kl} - 3\hat{r}_k \hat{r}_l)S_k I_l + \left(\frac{8\pi}{3}\right)\delta^3(\vec{r})S_k I_k\right],$$

where we sum over repeated indices. The final expression to solve is

$$\hat{H}_{\mathrm{HF}} = C\left[\underbrace{S_k I_l \int d^3r |\psi(\vec{r})|^2 \frac{\delta_{kl} - 3\hat{r}_k \hat{r}_l}{r^3}}_{\text{dipolar term}} + \underbrace{\left(\frac{8\pi}{3}\right)S_k I_k \int d^3r |\psi(\vec{r})|^2 \delta^3(\vec{r})}_{\text{contact (isotropic) term}}\right].$$

The contact term reduces to

$$\left(\frac{8\pi}{3}\right)S_k I_k \int d^3r |\psi(\vec{r})|^2 \delta^3(\vec{r}) = \left(\frac{8\pi}{3}\right)|\psi(0)|^2 S_k I_k$$
$$= A_{\mathrm{iso}} \delta_{kl} S_k I_l,$$

where $A_{\mathrm{iso}} = C\left(\frac{8\pi}{3}\right)|\psi(0)|^2$.

To solve the dipolar term, define

$$A_{kl}^{\mathrm{dip}} = C\int d^3r, |\psi(\vec{r})|^2 \frac{\delta_{kl} - 3\hat{r}_k \hat{r}_l}{r^3}.$$

Then the dipolar term reduces to $S_k I_l A_{kl}^{\mathrm{dip}}$. Define the A_{ij} tensor as

$$A_{ij} = A_{\mathrm{iso}} \delta_{ij} + A_{ij}^{\mathrm{dip}}.$$

Finally,

$$\hat{H}_{\mathrm{HF}} = \hat{S} \cdot A \cdot \hat{I} = \hat{S}_i A_{ij} \hat{I}_j.$$

For multiple nuclei, like in silicon carbide, we sum over every j inequivalent nucleus:

$$\hat{H}_{\mathrm{HF}} = \sum_j \hat{\vec{S}} \cdot A_j \cdot \hat{\vec{I}}_j.$$

For a two-nucleus (silicon + carbon) and two-electron (defect + carrier) system, the Hilbert space is $2^4 = 16$ dimensional. The hyperfine Hamiltonian is a 16×16 matrix.

Making a B_0 versus energy plot for hyperfine, like with Zeeman above, is difficult. In a future post, we'll generate it. I will be writing eigen-energy simulations to show what the hyperfine interaction does to the 16 basis states defining our spin system.