

Zeeman and Hyperfine

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

\mathcal{H} is a sum of the [Zeeman effect](#), [hyperfine interactions](#), [zero-field splitting](#), and the [exchange interaction](#).

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

Normally \mathcal{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 \mathcal{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

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

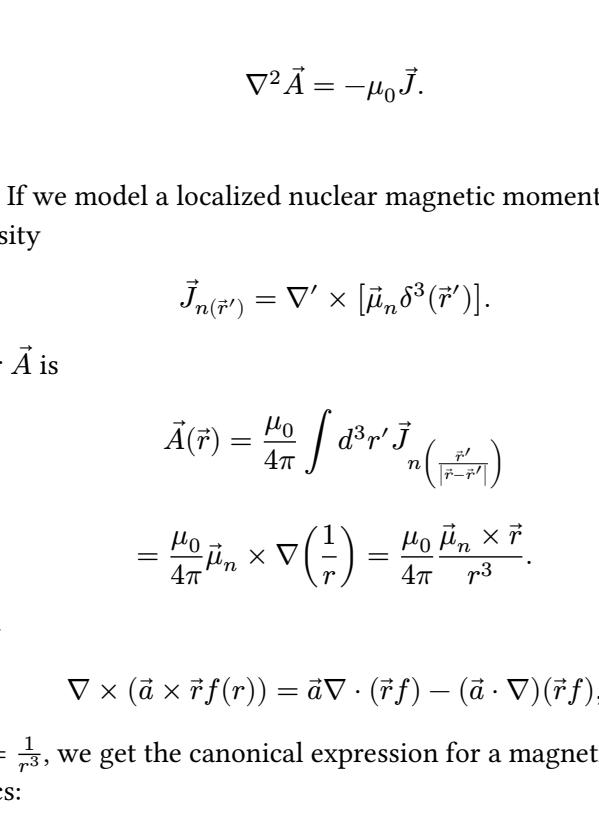
Therefore, the Zeeman Hamiltonian is

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

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

$$\begin{aligned} \hat{H}_Z |s, m_s\rangle &= \frac{g\mu_B}{\hbar} B_0 S_z |s, m_s\rangle \\ &= \frac{g\mu_B}{\hbar} B_0 (m_s \hbar) |s, m_s\rangle = m_s g\mu_B B_0 |s, m_s\rangle. \end{aligned}$$

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,

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

This can be diagonalized along the principal axis:

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

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}_{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

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

The general solution for \vec{A} is

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

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

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:

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

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:

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

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

Promoting magnetic moments to their quantum operators,

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

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:

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

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

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

Now write the Hamiltonian in index form:

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

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

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

The contact term reduces to

$$\begin{aligned} \frac{8\pi}{3} S_k I_k \int d^3 r |\psi(\vec{r})|^2 \delta^3(\vec{r}) &= \frac{8\pi}{3} |\psi(0)|^2 S_k I_k \\ &= A_{\text{iso}} \delta_{kl} S_k I_l, \end{aligned}$$

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

To solve the dipolar term, define

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

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

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

Finally,

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

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

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

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.