

## 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](#).

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

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  $\gamma$  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

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

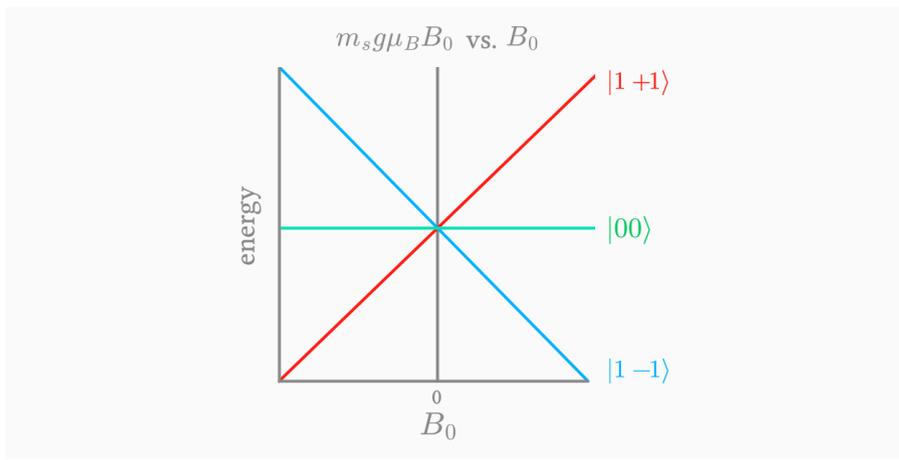
Therefore, the Zeeman Hamiltonian is

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

where  $S_z = \left( \frac{\hbar}{2} \right) \hat{\sigma}_z$  and  $\sigma_z$  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,

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

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  $\hat{H}_{\text{HF}}$ .

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

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

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

$$\begin{aligned} \vec{A}(\vec{r}) &= \frac{\mu_0}{4\pi} \int d^3 r' \vec{J}_n \left( \frac{\vec{r}'}{|\vec{r}-\vec{r}'|} \right) \\ &= \frac{\mu_0}{4\pi} \vec{\mu}_n \times \nabla \left( \frac{1}{r} \right) = \frac{\mu_0}{4\pi} \frac{\vec{\mu}_n \times \vec{r}}{r^3}. \end{aligned}$$

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:

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

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

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

Now write the Hamiltonian in index form:

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

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

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

The contact term reduces to

$$\frac{8\pi}{3} S_k I_k \int d^3 r |\psi(\vec{r})|^2 \delta^3(\vec{r}) = \frac{8\pi}{3} |\psi(0)|^2 S_k I_k$$

$$= A_{\text{iso}} \delta_{kl} S_k I_l,$$

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,

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

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

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

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  $16 \times 16$  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.