

0.1 The dipole-dipole Hamiltonian

The dipole-dipole interaction between two point-like magnetic dipoles can be derived classically by computing the static magnetic field generated by one dipole at the position of the second dipole.

For a magnetic dipole \mathbf{m} , positioned on $\mathbf{0}$, the static magnetic field generated on position $\mathbf{r} = r\mathbf{u}$ is : [1, p. 188]

$$\mathbf{B}(\mathbf{r}) = \frac{\mu_0}{4\pi} \left[\frac{3(\mathbf{m} \cdot \mathbf{u})\mathbf{u} - \mathbf{m}}{r^3} + \frac{8\pi}{3} \mathbf{m} \delta(\mathbf{r}) \right] \quad (1)$$

The interaction energy between two dipole \mathbf{m}_1 and \mathbf{m}_2 with relative position \mathbf{r} is therefore :

$$U = -\mathbf{m}_1 \cdot \mathbf{B}_2(\mathbf{r}) = -\frac{\mu_0}{4\pi} \left[\frac{3(\mathbf{m}_1 \cdot \mathbf{u})(\mathbf{m}_2 \cdot \mathbf{u}) - \mathbf{m}_1 \cdot \mathbf{m}_2}{r^3} + \frac{8\pi}{3} \mathbf{m}_1 \cdot \mathbf{m}_2 \delta(\mathbf{r}) \right] \quad (2)$$

The term in δ , which comes from the necessity to preserve $\nabla \cdot \mathbf{B} = 0$, is at the origin of the Fermi contact energy. This term plays a role for hyperfine coupling when there is an overlap of the electron wave-function with the nucleus, which is the case for s-orbitals in atoms. In the case of NV centers, Fermi contact energy plays an important role to the hyper-fine coupling between the NV^- electron spin and the nuclear spin of the ^{14}N or ^{15}N atom [2] or for nearby ^{13}C atoms [3].

However, in the scope of this manuscript, we will mostly concern ourselves with dipolar interaction between electronic spins separated by several atomic sites, so that the overlap of the electronic wave functions can be mostly neglected. This results in the simplified dipole-dipole coupling energy :

$$U = \frac{\mu_0}{4\pi} \left[\frac{\mathbf{m}_1 \cdot \mathbf{m}_2 - 3(\mathbf{m}_1 \cdot \mathbf{u})(\mathbf{m}_2 \cdot \mathbf{u})}{r^3} \right] \quad (3)$$

For two electronic spins, we can rewrite a quantized version of 3 by replacing the magnetic moments $\mathbf{m}_i = \hbar\gamma_i\hat{\mathbf{S}}_i$ where $\gamma_i = -\frac{g_e\mu_B}{\hbar}$ is the gyromagnetic ratio of the electron. For the NV^- electronic spin, as well as most electronic spin defects in diamond, $g_e \approx 2$ which gives a numerical value for the gyromagnetic ratio $\gamma \approx (2\pi) 28 \text{ GHz/T} \approx (2\pi) 2.8 \text{ MHz/G}$. $\hat{\mathbf{S}}_i = (\hat{S}_x, \hat{S}_y, \hat{S}_z)$ is the spin vector operator for particle i . The quantized version of the dipole-dipole interaction is then:

$$\mathcal{H}_{dd} = \frac{J_0}{r^3} \left[\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2 - 3(\hat{\mathbf{S}}_1 \cdot \mathbf{u})(\hat{\mathbf{S}}_2 \cdot \mathbf{u}) \right] \quad (4)$$

Where $J_0 \equiv \frac{\mu_0\gamma_1\gamma_2\hbar^2}{4\pi}$. For two electronic spins with g -factors close to 2, the numerical value of J_0 is $(2\pi) 52 \text{ MHz} \cdot \text{nm}^3$.

0.2 Flip-Flops, double flips and cross-relaxations

Flip-flop, in the context of dipole-dipole interaction, is the mechanism which exchanges a quantum of spin between two spins. For an initial state of two spins $|m_s^1 = i; m_s^2 = j\rangle$, a flip-flop process will result in a final state $|i+1; j-1\rangle$ or

$|i - 1; j + 1\rangle$. The effective rate of this process depends on the matrix element $\langle i; j | \mathcal{H}_{dd} | i \pm 1; j \mp 1 \rangle$, as well as the resonance condition between the initial and final state. We should note that for two identical spins, the flip-flop process $|i; i \pm 1\rangle \langle i \pm 1, i|$ is always resonant.

Other mechanisms enabled by the dipole-dipole Hamiltonian are the double-flips up and down which couple the states $|i; j\rangle$ and $|i - 1; j - 1\rangle$ or $|i + 1; j + 1\rangle$. For a single spin species in a strong magnetic field, the lift of the various spin levels by the Zeeman effect means that no double-flip process can be resonant. However with weak magnetic field or when two different spin species are present, double flip process can be as relevant as flip-flops, the matrix elements $\langle i; j | \mathcal{H}_{dd} | i \pm 1; j \pm 1 \rangle$ typically being of the same order of magnitude as the flip-flop ones.

When two spin populations of different polarization can be resonantly coupled by either flip-flop or double-flip, cross-relaxation happens, that is a macroscopic transfer of polarization from the most polarized spin group to the least polarized one.

Bibliography

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