Chapter 1

Detection of dark spins via cross-relaxations

In this chapter, we will see how...

1.1 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]

$$\mathbf{B}(\mathbf{r}) = \frac{\mu_0}{4\pi}\left[\frac{3(\mathbf{m}\cdot\mathbf{u})\mathbf{u} - \mathbf{m}}{r^3} + \frac{8\pi}{3}\mathbf{m}\,\delta(\mathbf{r})\right] \tag{1.1}$$

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

$$U = -\mathbf{m}_1 \cdot \mathbf{B}_2(\mathbf{r}) = -\frac{\mu_0}{4\pi}\left[\frac{3(\mathbf{m}_1\cdot\mathbf{u})(\mathbf{m}_2\cdot\mathbf{u}) - \mathbf{m}_1\cdot\mathbf{m}_2}{r^3} + \frac{8\pi}{3}\mathbf{m}_1\cdot\mathbf{m}_2\,\delta(\mathbf{r})\right] \tag{1.2}$$

The term in δ, which comes from the necessity to preserve $\boldsymbol{\nabla}\cdot\mathbf{B} = 0$, 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 :

$$U = \frac{\mu_0}{4\pi}\left[\frac{\mathbf{m}_1\cdot\mathbf{m}_2 - 3(\mathbf{m}_1\cdot\mathbf{u})(\mathbf{m}_2\cdot\mathbf{u})}{r^3}\right] \tag{1.3}$$

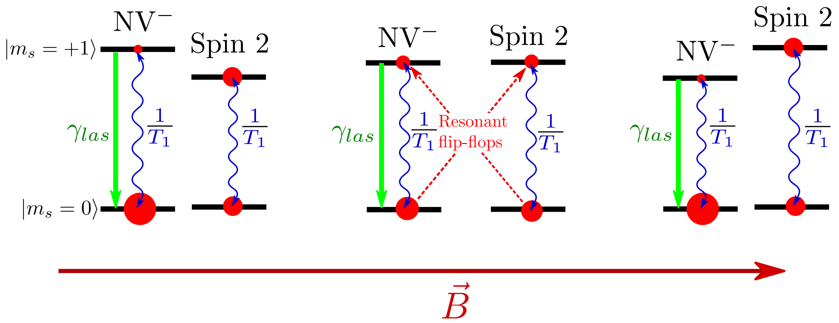

Figure 1.1: Illustration of cross-relaxations between an NV center and an unpolarized spin. Red dots represent the population in each states, and the arrows the various population transfer mechanisms

For two electronic spins, we can rewrite a quantized version of 1.3 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 $\gamma \approx (2\pi)\,28$ GHz/T $\approx (2\pi)\,2.8$ 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:

$$\mathcal{H}_{dd} = \frac{J_0}{r^3}\left[\hat{\mathbf{S}}_1\cdot\hat{\mathbf{S}}_2 - 3(\hat{\mathbf{S}}_1\cdot\mathbf{u})(\hat{\mathbf{S}}_2\cdot\mathbf{u})\right] \tag{1.4}$$

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 $(2\pi)\,52$ MHz $\cdot$ nm^3.

1.2 Flip-Flops, double flips and cross-relaxation

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 $\left|m_s^1 = i; m_s^2 = j\right\rangle$, 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.

Cross-relaxation (CR) is the transfer of polarization from one spin to another (or more generally from one family of spins to another family). This process con occur either through flip-flops, or through double flips, as long as the resonance condition between the two spins is met.

Fig. 1.1 illustrates CR between a polarized NV center and an unpolarized second spin. When the two considered spin transitions become resonant, in this case by tuning a magnetic field, polarization from the NV center will be transferred to the second spin, meaning in this case that the NV center will end up less polarized and the second spin more polarized.

1.3 Detection of dark spins with NV centers via cross-relaxations

Bibliography

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[3] Benjamin Smeltzer, Lilian Childress, and Adam Gali. “13C hyperfine interactions in the nitrogen-vacancy centre in diamond”. In: *New Journal of Physics* 13.2 (2011), p. 025021.