Group meeting : Cross-relaxation with NV centers ensemble in diamond

March 23, 2022

Outline

1. (Quick) Reminder on NV center
2. Dipolar interaction and spin lifetime
3. The fluctuator model
4. The reasons behind the dip in zero-field
5. Low-field Magnetometry

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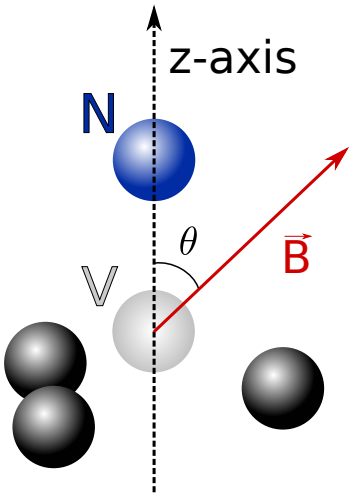
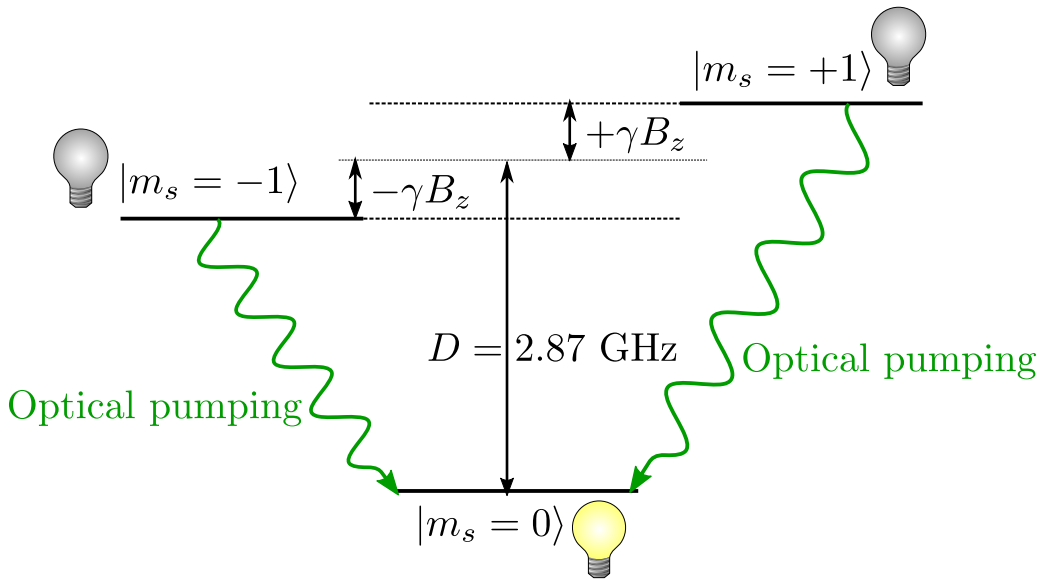
NV^- spin in the fundamental electronic state



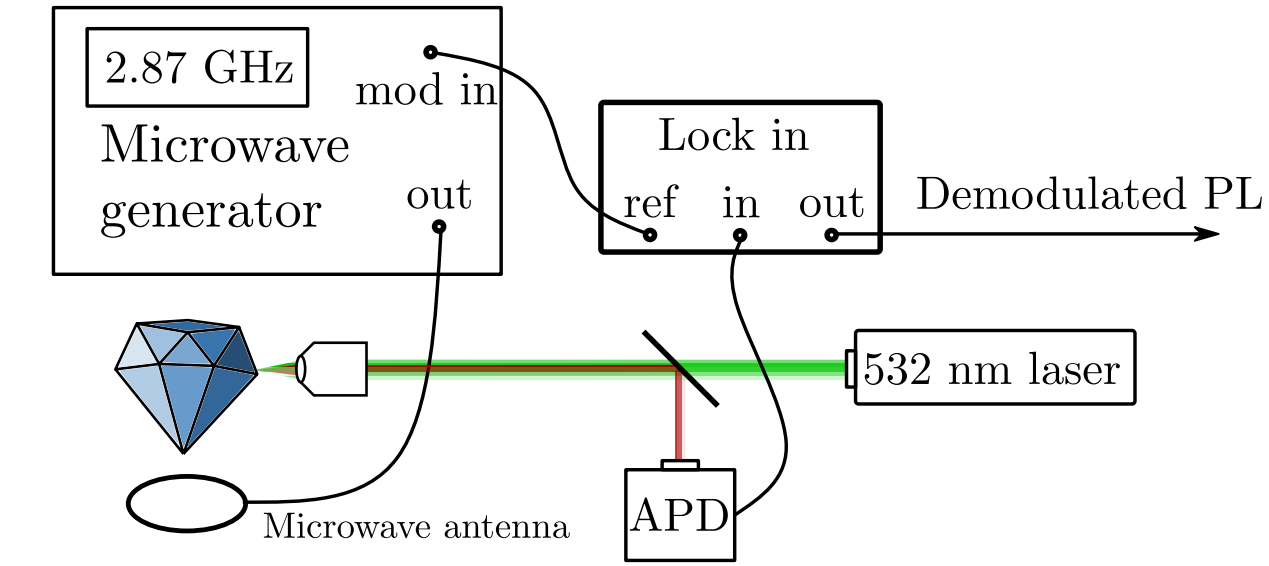
$$\mathcal{H} = D\hat{S}_z^2 + \gamma_e \hat{\mathbf{S}} \cdot \mathbf{B}$$

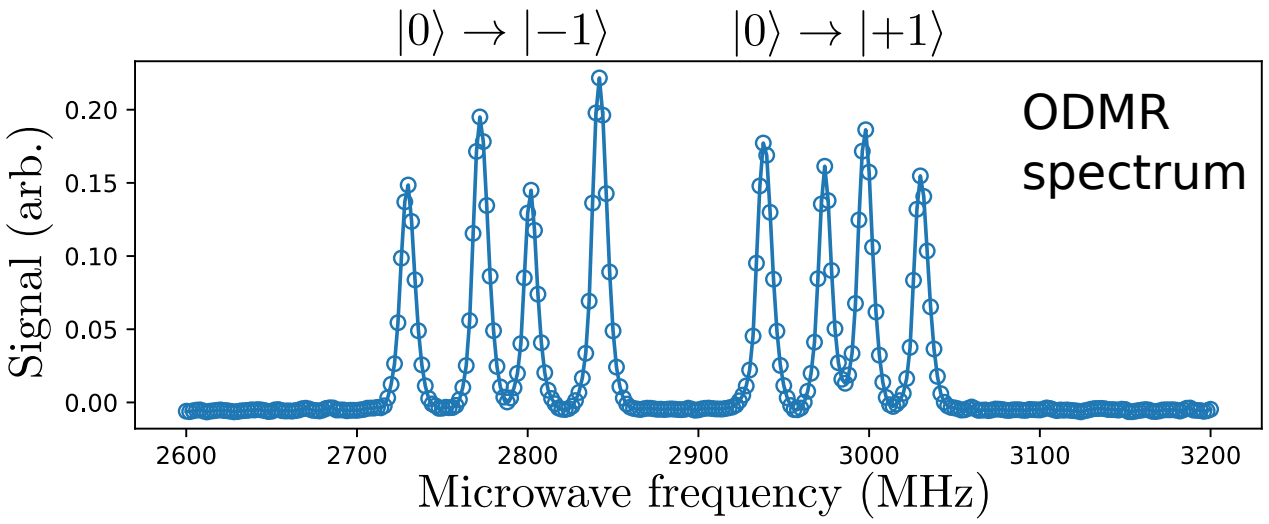
$\mathbf{z}$ of $\hat{S}_z$ defined by the crystalline axis

- $|0\rangle$ state brighter than $|\pm 1\rangle$ state by ~ 30 %
- polarization in $|0\rangle$ state of ~ 80 % (equivalent to ~ 65 μK)



Principle of CW ODMR





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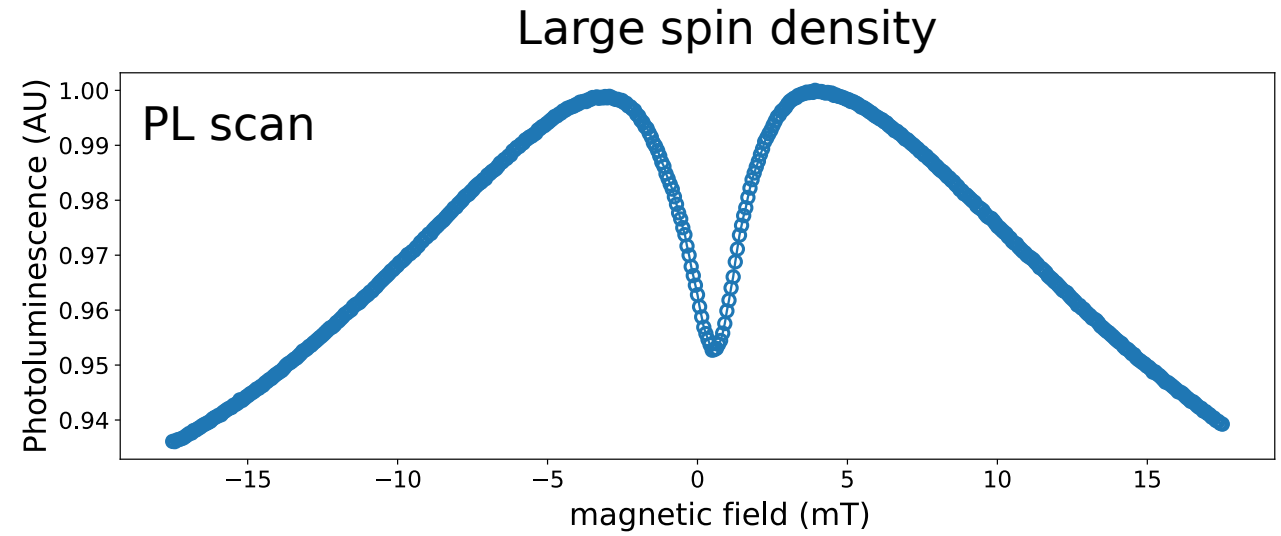
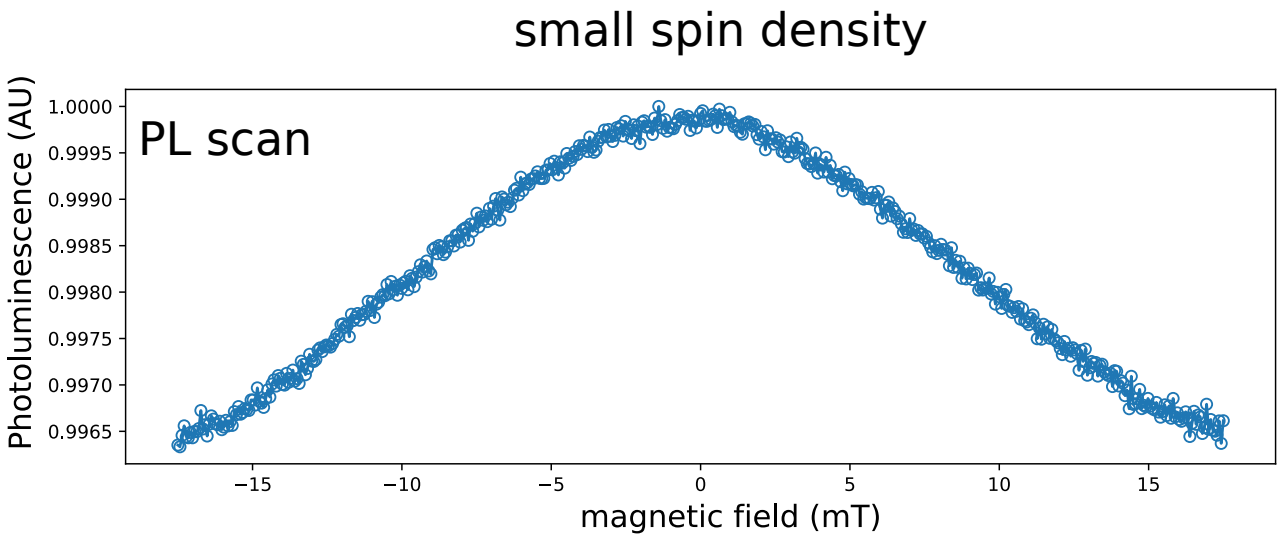
Today's goal : understand the PL dip in zero-field

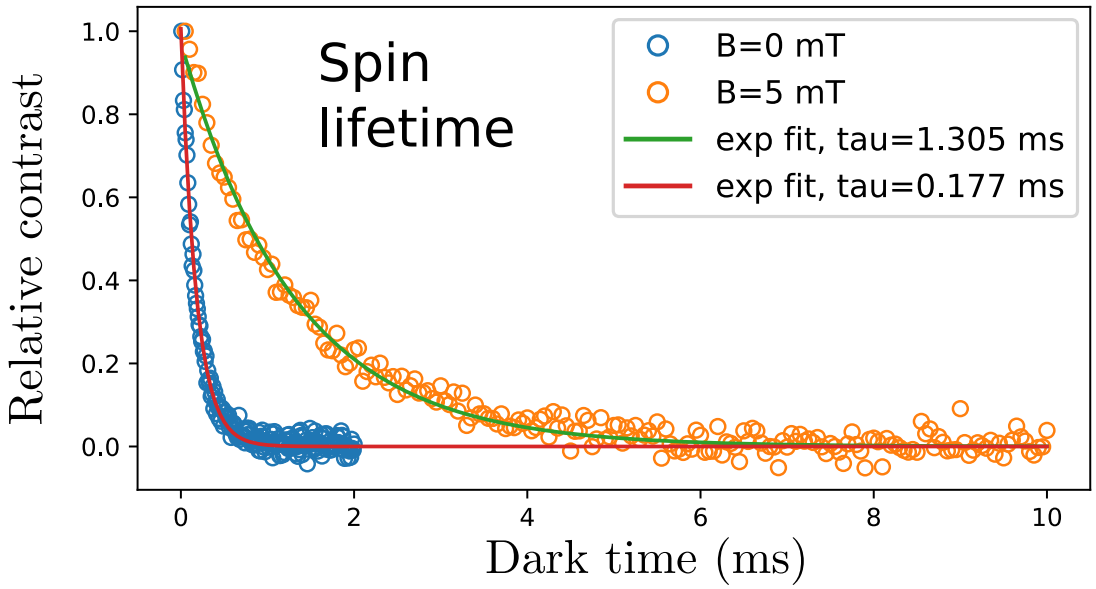


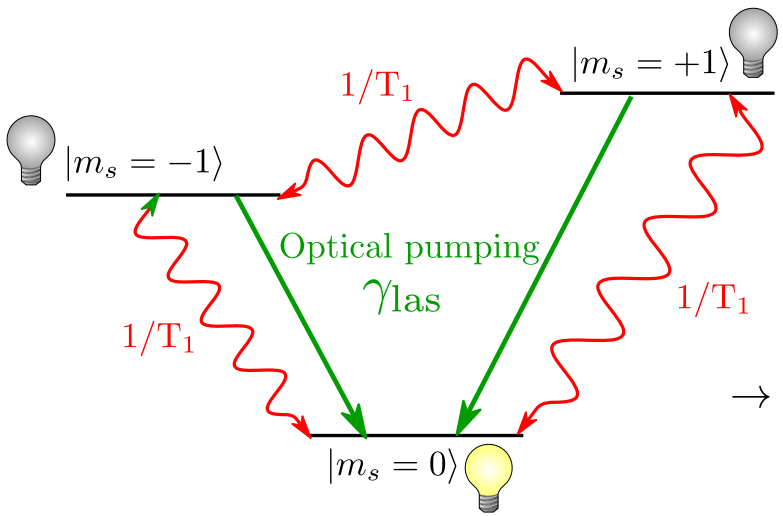


Why does it matter ?

- The concentration of NV^- centers (while maintaining good spectral properties) is increasing.
- Magnetometry in low field is already hard with standard techniques.
- The dip itself can be used to perform low-field, microwave-less magnetometry.

Link between the PL and the spin T1





$$\mathrm{PL} \propto \rho_{00}$$

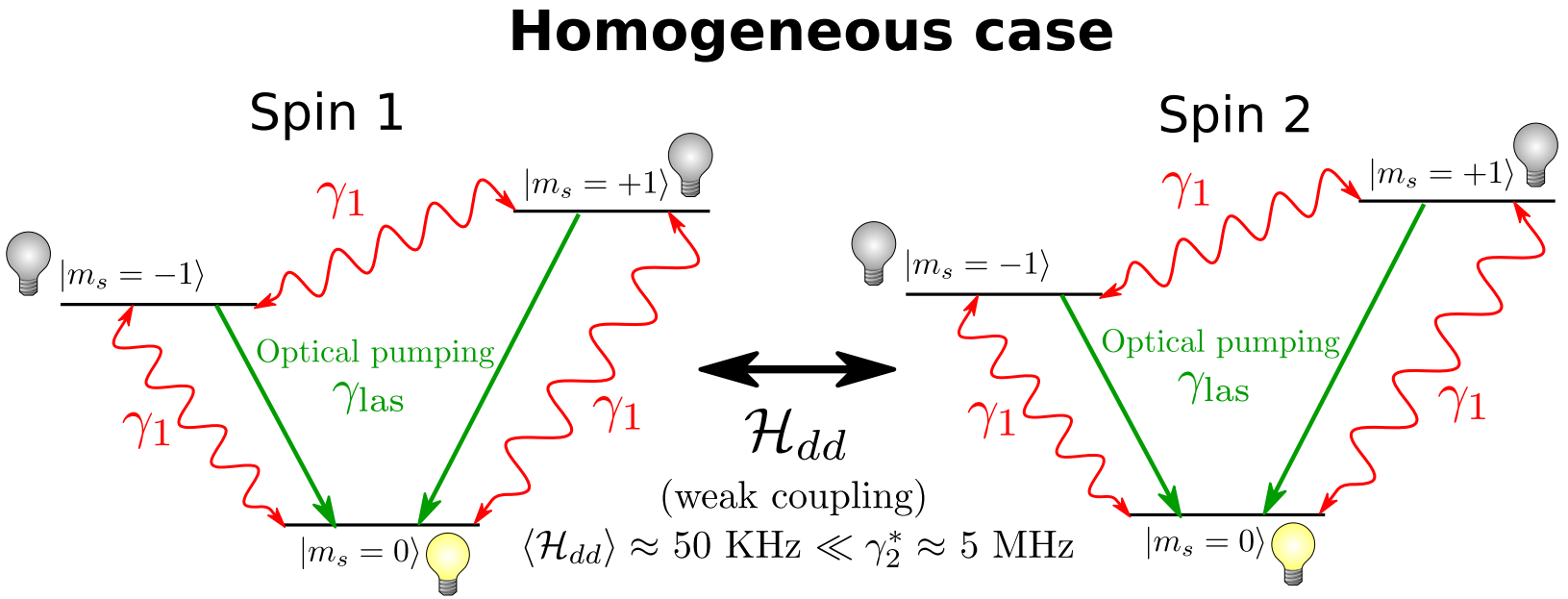
$$\rho_{00} = \frac{\gamma_{las}}{\gamma_{las} + 3\gamma_1} \qquad (\gamma_1 = 1/T_1)$$

$\rightarrow$ The PL decreases when γ_1 increases

Spin relaxation amplified by dipolar coupling

See animation

Dipolar interaction should not modify the total spin polarization...

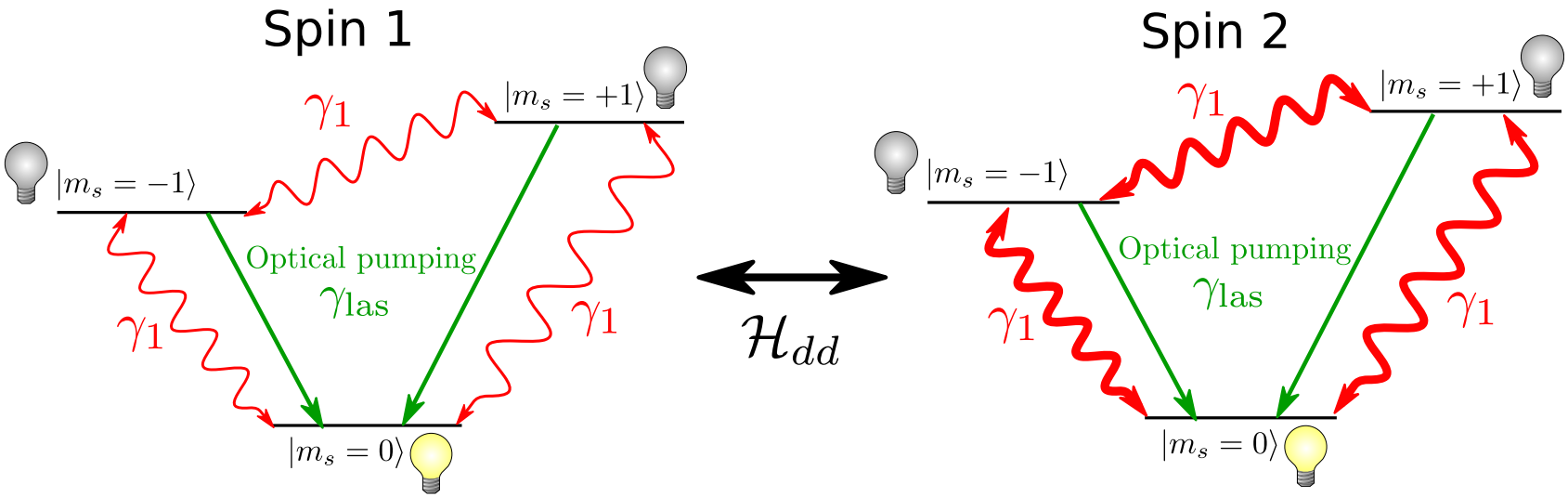


Flip-flop processes ($|-1;0\rangle \rightarrow |0;-1\rangle$) preserve the spin population in $|0\rangle$

$\rightarrow$ Dipolar coupling does not modify the ensemble spin lifetime

...except with inhomogeneities

Inhomogeneous case : $\gamma_1 < \gamma_1'$



Spin 2 is less polarized than spin 1 ($\rho_{00}^2 < \rho_{00}^1$)

$\rightarrow$ The process $|0;-1\rangle \rightarrow |-1;0\rangle$ is more likely than $|-1;0\rangle \rightarrow |0;-1\rangle$

$\rightarrow$ The combined action of the dipolar coupling and the inhomogeneous relaxation rates can reduce the ensemble spin lifetime

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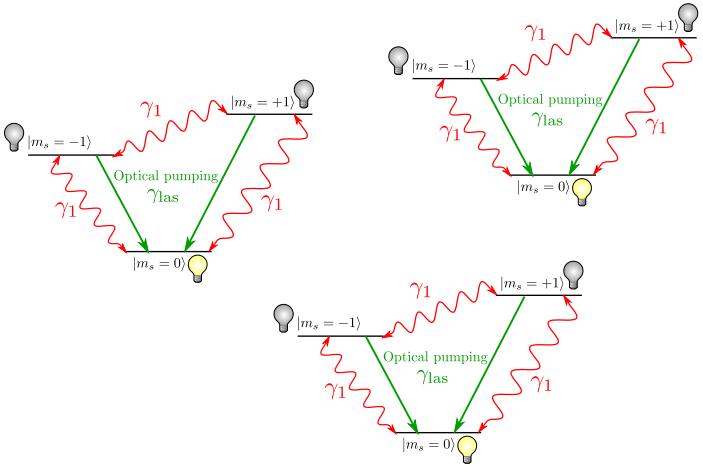
Theoretical frame : the fluctuator model

"Normal" NVs

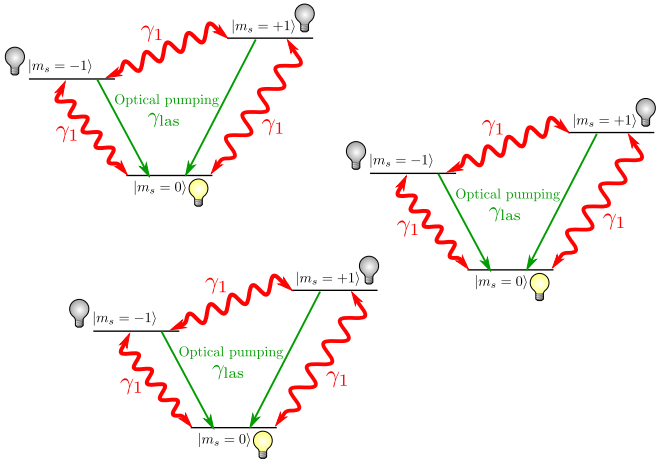


$$\gamma_1 \ll \gamma_{las}$$

$$\hat{\rho} \approx \begin{pmatrix} 0.1 & 0 & 0 \\ 0 & 0.8 & 0 \\ 0 & 0 & 0.1 \end{pmatrix}$$

$\mathcal{H}_{dd}$

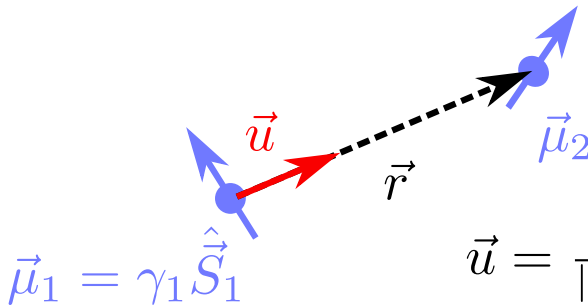
Fluctuator NVs



$$\gamma_1 \gg \gamma_{las}$$

$$\hat{\rho} \approx \begin{pmatrix} 0.33 & 0 & 0 \\ 0 & 0.33 & 0 \\ 0 & 0 & 0.33 \end{pmatrix}$$

The dipole-dipole Hamiltonian



$$\vec{u} = \frac{\vec{r}}{\|\vec{r}\|} = n_x \vec{e}_x + n_y \vec{e}_y + n_z \vec{e}_z$$

$$\mathcal{H}_{dd} = -\frac{\mu_0 \gamma_1 \gamma_2 \hbar^2}{4\pi r^3} [3(\widehat{\mathbf{S}}_1 \cdot \mathbf{u})(\widehat{\mathbf{S}}_2 \cdot \mathbf{u}) - \widehat{\mathbf{S}}_1 \cdot \widehat{\mathbf{S}}_2]$$

Every matrix element $\langle i; \alpha | \mathcal{H}_{dd} | j; \beta \rangle$ can be written as $\xi(\|\vec{r}\|)\eta(\vec{u})$

Example : $\langle 0; -1 | \mathcal{H}_{dd} | -1; 0 \rangle$ is the matrix element for a flip-flop process

$\mathcal{H}_{dd}$ for two aligned spin

In the magnetic basis $(|-1\rangle, |0\rangle, |+1\rangle) \otimes (|-1\rangle, |0\rangle, |+1\rangle)$:

$$\mathcal{H}_{dd} \propto (\frac{3}{2}(n_x^2 + n_y^2) - 1)\,[|0;+1\rangle\langle+1;0| + |-1;0\rangle\langle 0;-1| + h.c.] \tag{1}$$

$$+ \frac{3}{2}(n_x^2 - n_y^2 + i2n_xn_y)\,[|0;+1\rangle\langle-1;0| + |+1;0\rangle\langle 0;-1|] \tag{2}$$

$$+ \frac{3}{2}(n_x^2 - n_y^2 - i2n_xn_y)\,[|0;-1\rangle\langle+1;0| + |-1;0\rangle\langle 0;+1|] \tag{3}$$

$$+ (3n_z^2 - 1)\hat{S}_z^1\hat{S}_z^2 \tag{4}$$

$$+ \mathcal{H}_{\text{other}} \tag{5}$$

Example : $\eta_{\text{flip-flop}} = \frac{3}{2}(n_x^2 + n_y^2) - 1)$

The fluctuator model : calculation steps

- Decay rate induced by a single fluctuator on a single NV :

$$\Gamma_{ij} = \sum_{\alpha,\beta} |\langle i,\alpha|\mathcal{H}_{dd}|j,\beta\rangle|^2 \frac{\gamma_f}{(\omega_{ij}-\omega_{\beta\alpha})^2+\gamma_f^2}$$

The fluctuator model : calculation steps

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$$\Gamma_{ij} = \sum_{\alpha,\beta} |\langle i,\alpha|\mathcal{H}_{dd}|j,\beta\rangle|^2 \frac{\gamma_f}{(\omega_{ij}-\omega_{\beta\alpha})^2+\gamma_f^2}$$

- Each NV will see a total decay rate

$$\gamma = \sum_{\text{fluct}} \Gamma_{ij}(\vec{r})$$

The fluctuator model : calculation steps

- We can compute the distribution of these individual decay rates γ :

$$\rho(\gamma) = \int \{d_{\text{fluct}}\}\, \delta\left(\gamma - \sum_{\text{fluct}} \Gamma_{ij}(\vec{r})\right)$$
$$= \frac{e^{-1/(4\gamma T)}}{\sqrt{4\pi\gamma^3 T}}$$

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- The measured lifetime will be averaged over all the NVs :

$$PL(t) = \int e^{-\gamma t}\rho(\gamma)d\gamma = e^{-\sqrt{t/T}}$$

The fluctuator model : calculation steps

Where :

$$T \equiv \left(\frac{4\pi n_f J_0 \bar{\eta}}{3} \right)^2 \frac{\pi}{\gamma_f}$$

With :

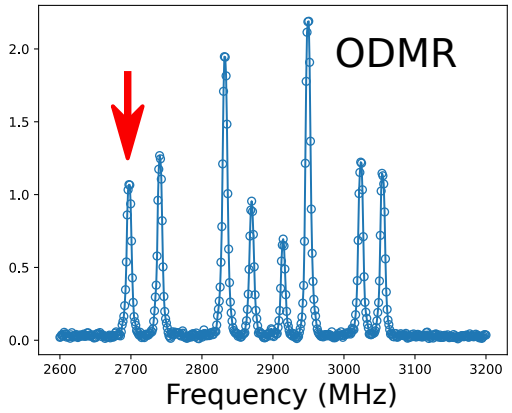
- n_f is the fluctuator density
- γ_f is the fluctuator decay rate

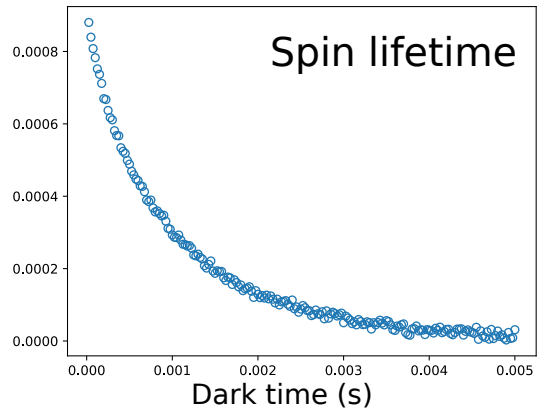
and :

$$\bar{\eta} \equiv \frac{1}{4} \sum_{\text{classes}} \frac{\gamma_f^2}{(\omega_{\text{NV}} - \omega_{\text{fluct}})^2 + \gamma_f^2} \int_{\theta,\phi} |\eta(\theta, \phi)| \mathrm{d}\Omega$$

Examples

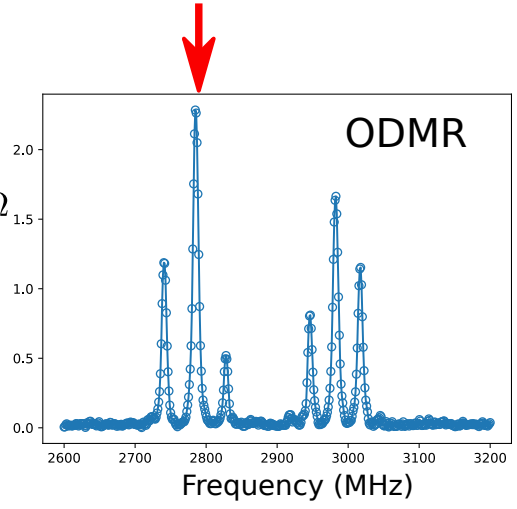
$$\bar{\eta} = \frac{1}{4}\int_{\theta,\phi} |\eta(\theta,\phi)| \mathrm{d}\Omega$$
$$= \frac{1}{4} \times 0.3849$$

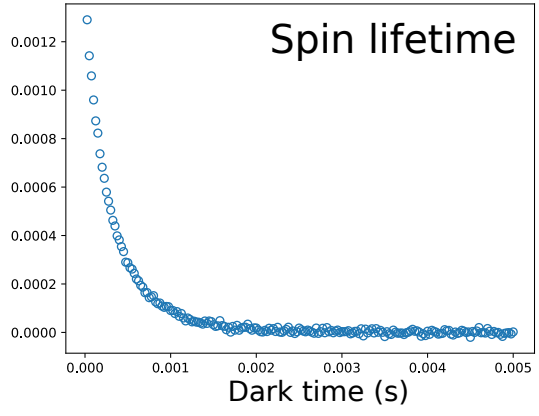




$$\mathrm{T} \propto \frac{1}{\bar{\eta}^2}$$

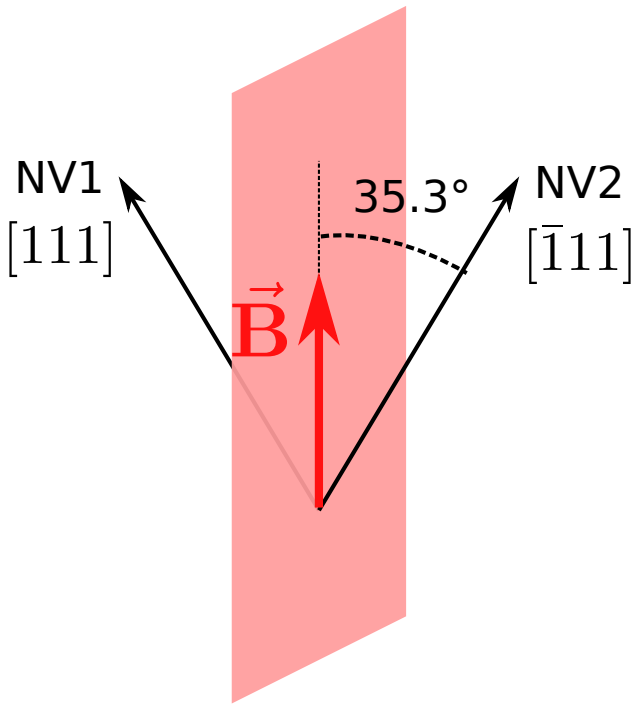
$$\bar{\eta} = \frac{1}{4}\int_{\theta,\phi} |\eta(\theta,\phi)| \mathrm{d}\Omega + \frac{1}{4}\int_{\theta,\phi} |\eta'(\theta,\phi)| \mathrm{d}\Omega$$
$$= \frac{1}{4} * 0.3849 + \frac{1}{4} * 0.8328$$





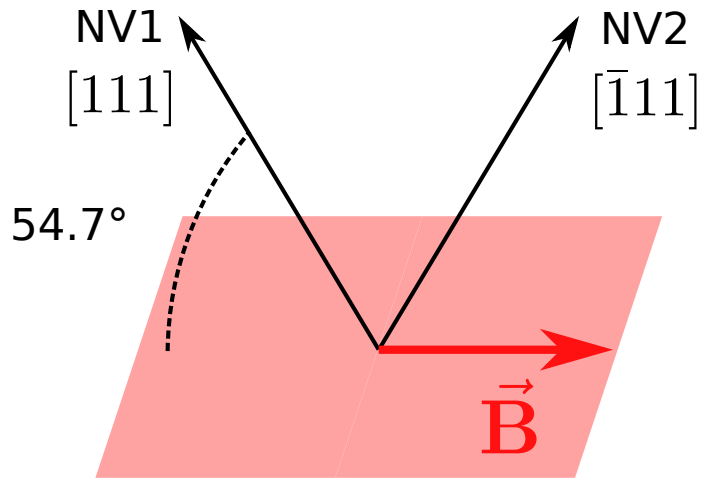
Non-trivial prediction of the model

$\vec{B} \in \{100\}$



$$\bar{\eta} = \frac{1}{4} \times 0.3849 + \frac{1}{4} \times 0.6507$$

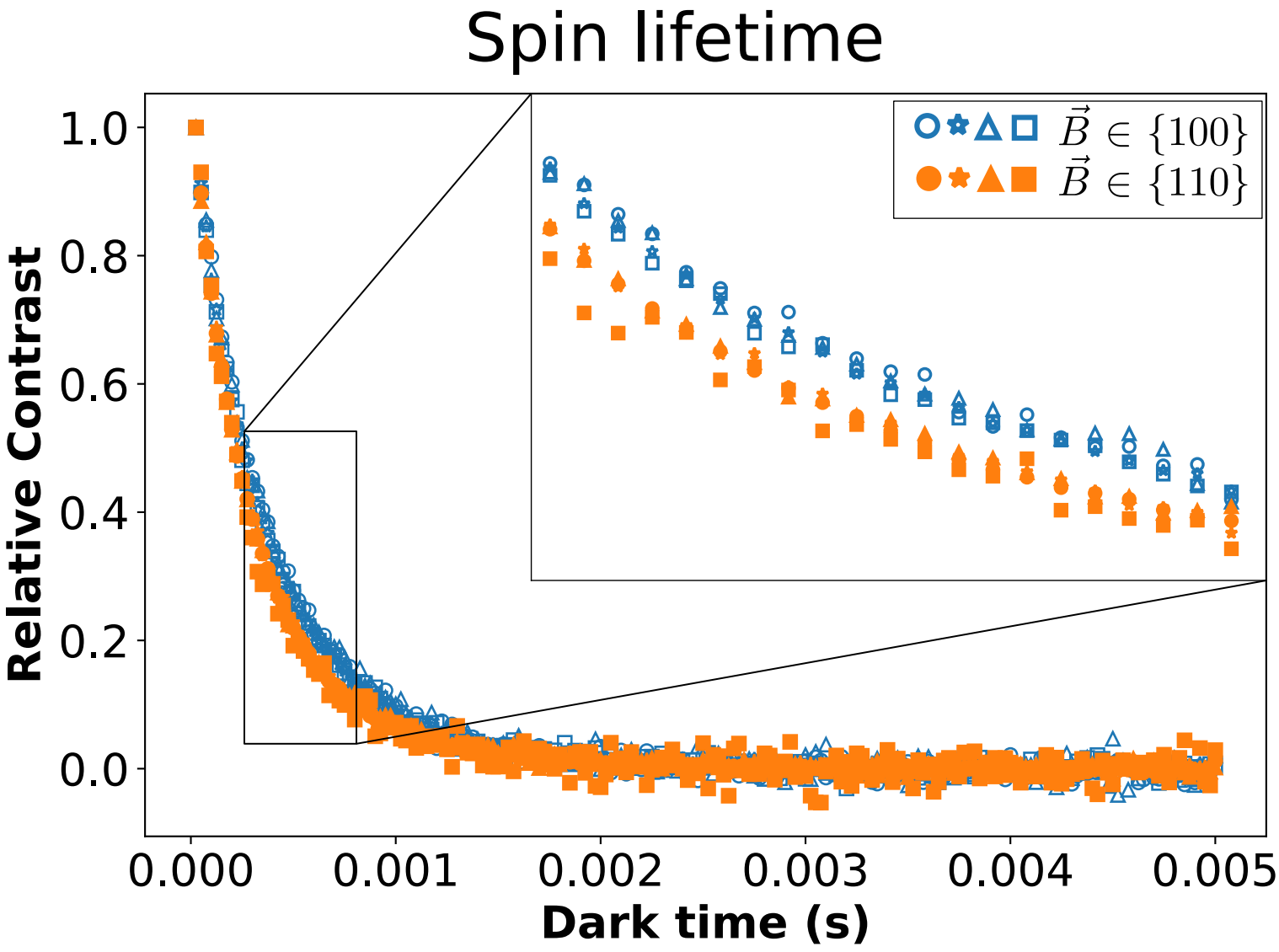
$\vec{B} \in \{011\}$



$$\bar{\eta} = \frac{1}{4} \times 0.3849 + \frac{1}{4} \times 0.8328$$

$$\bar{\eta}^{100} < \bar{\eta}^{011}$$
$$\rightarrow \text{We should observe } T_1^{100} > T_1^{011}$$

Non-trivial prediction of the model



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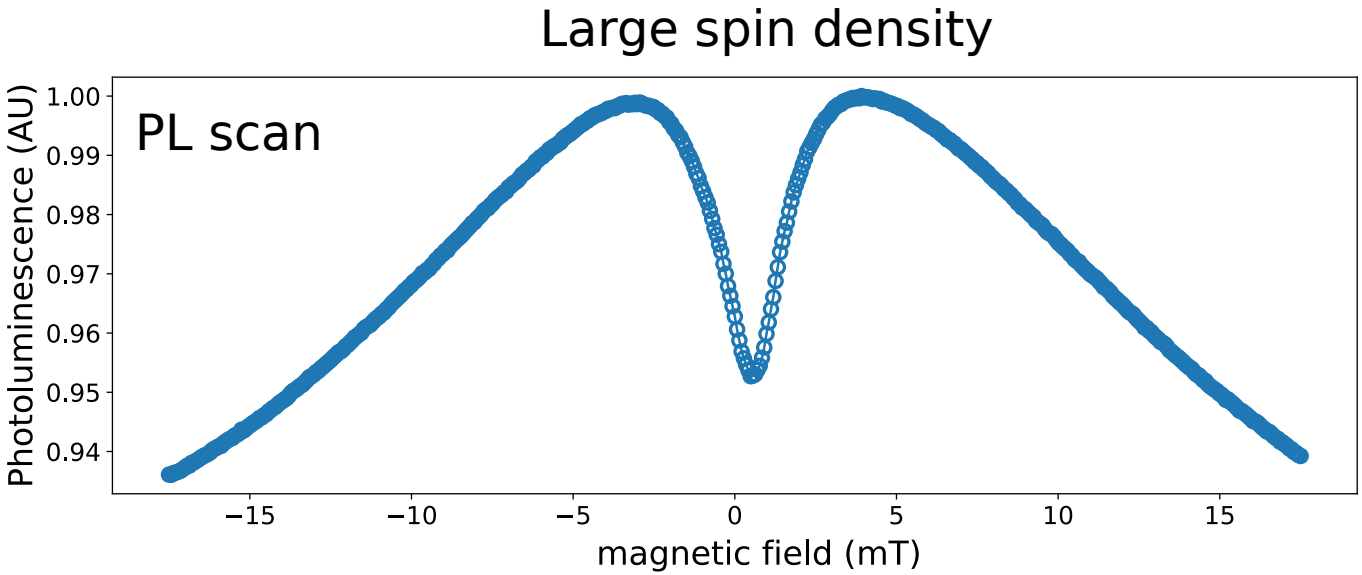
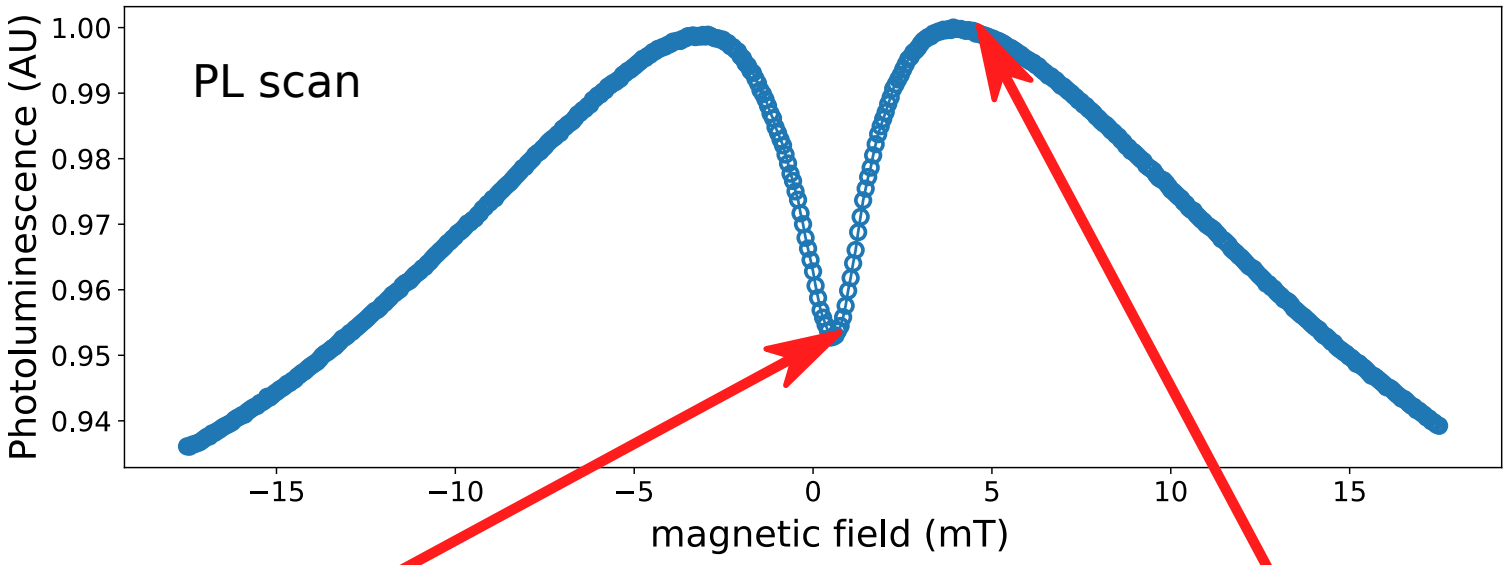
The reasons behind the dip in zero-field

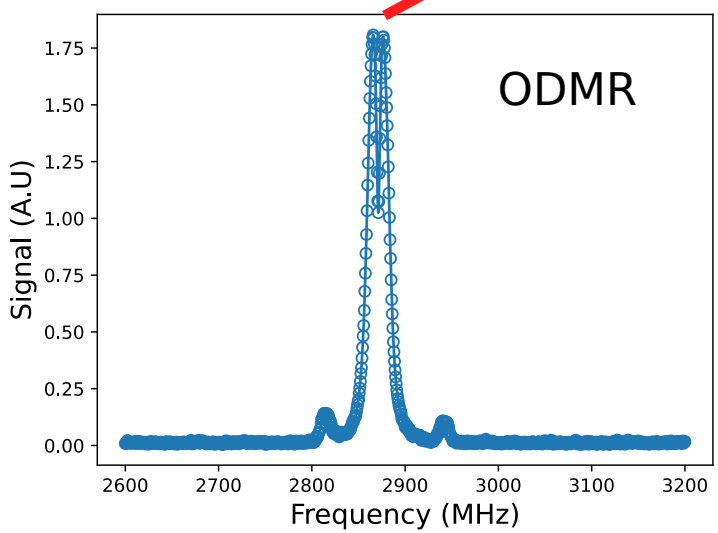


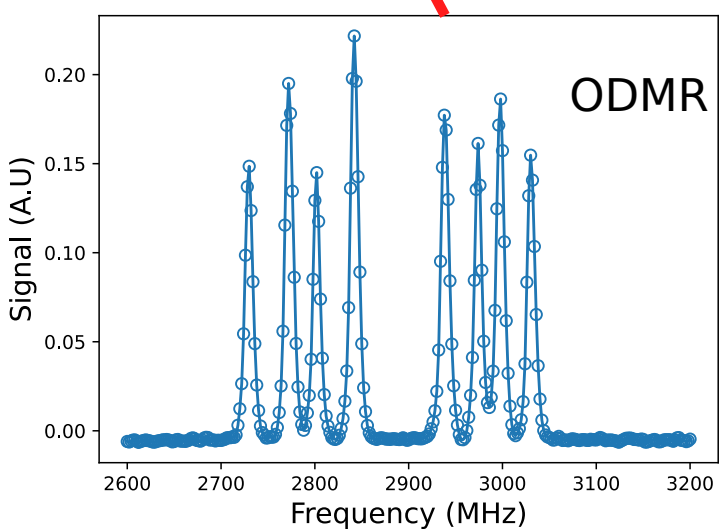
There are 3 parts, all linked to dipolar coupling :

1. The lift of the degeneracies between the four classes
2. The change of the eigenstates in the avoided crossing region
3. The double-flip terms in the dipolar Hamiltonian

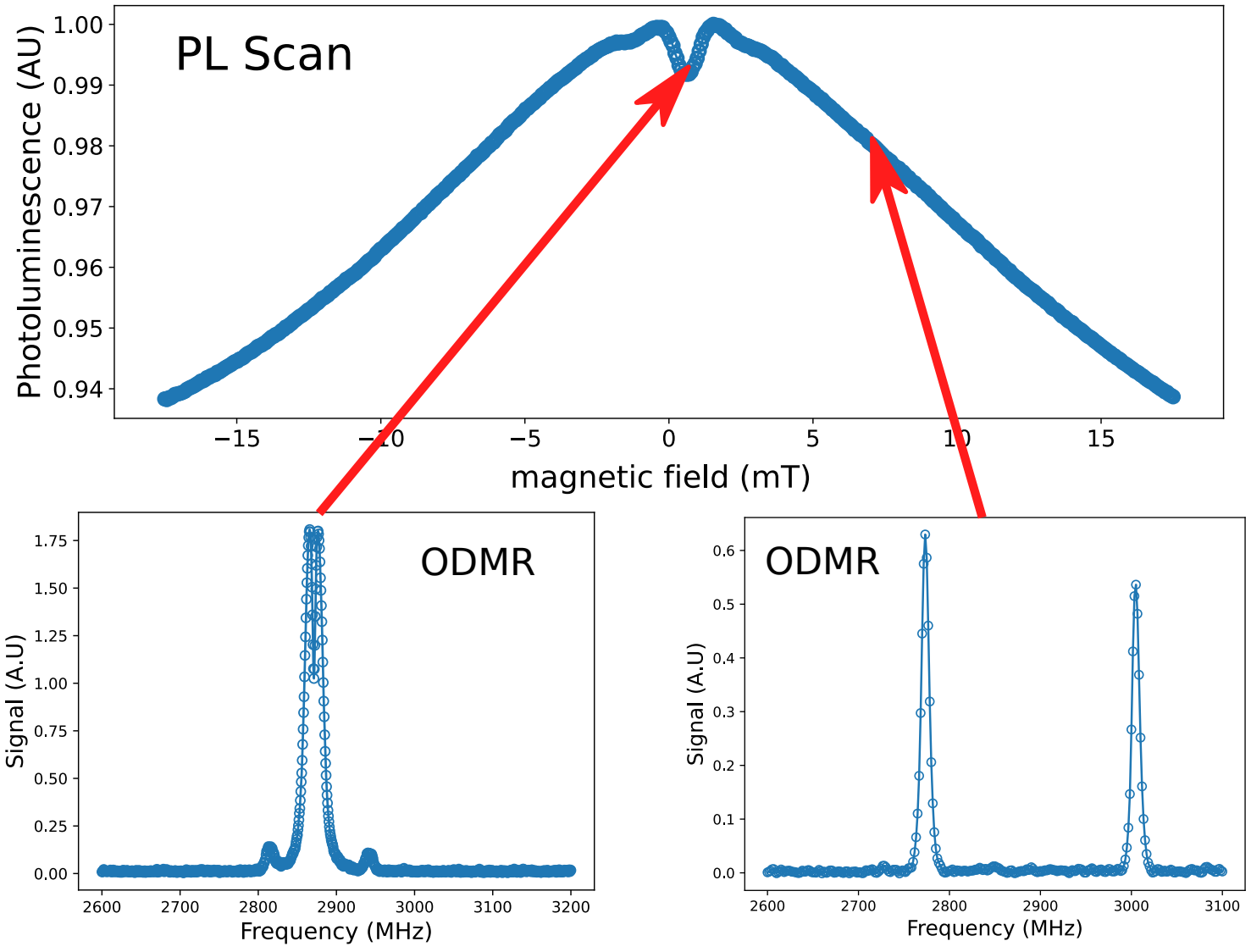
1. The lift of the degeneracies







But it is not the only reason



2. The change in eigenstates of $\mathcal{H}_0$

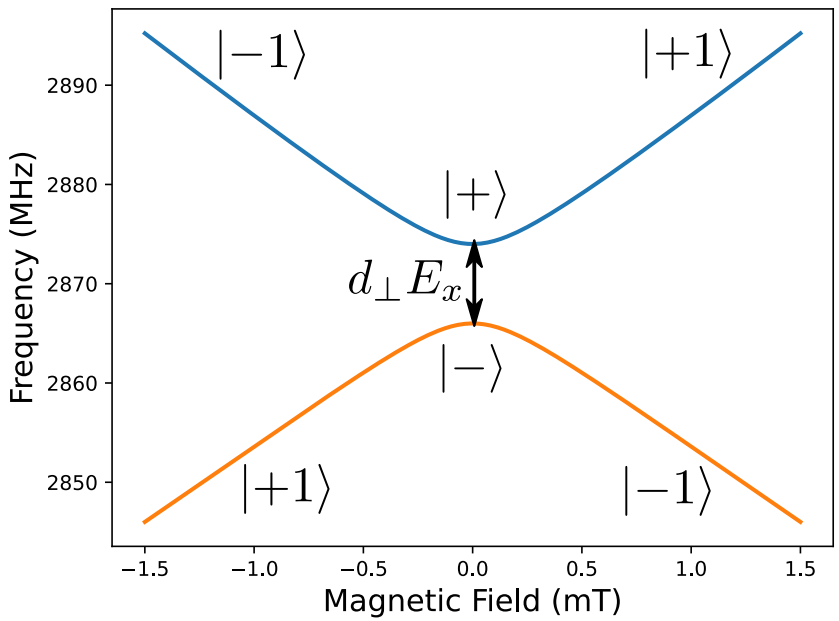
$$\mathcal{H}_0 = \begin{pmatrix} D - \gamma_e B_z & \gamma_e B_x & d_\perp E_x \\ \gamma_e B_x & 0 & \gamma_e B_x \\ d_\perp E_x & \gamma_e B_x & D + \gamma_e B_z \end{pmatrix}$$

D=2870 MHz

$\gamma_e B_x = 0 - 500$ MHz

$\gamma_e B_z = 0 - 500$ MHz

$d_\perp E_x \approx 5$ MHz



$$|+\rangle = \frac{|+1\rangle + |-1\rangle}{\sqrt{2}}$$

$$|-\rangle = \frac{|+1\rangle - |-1\rangle}{\sqrt{2}}$$

Dipolar Hamiltonian in the coupled basis

In the coupled basis $(|-\rangle, |0\rangle, |+\rangle) \otimes (|-\rangle, |0\rangle, |+\rangle)$:

$$\mathcal{H}_{dd} = (3n_x^2 - 1)\left[|0;+\rangle\langle+;0| + h.c.\right] \quad (1)$$
$$+ (3n_y^2 - 1)\left[|0;-\rangle\langle-;0| + h.c.\right] \quad (2)$$
$$+ i3n_x n_y\left[|0;+\rangle\langle-;0| + |+;0\rangle\langle0;-| + h.c.\right] \quad (3)$$
$$+ (3n_z^2 - 1)\left[|+;-\rangle\langle-;+| + h.c.\right] \quad (4)$$
$$+ \mathcal{H}_{\text{other}} \quad (5)$$

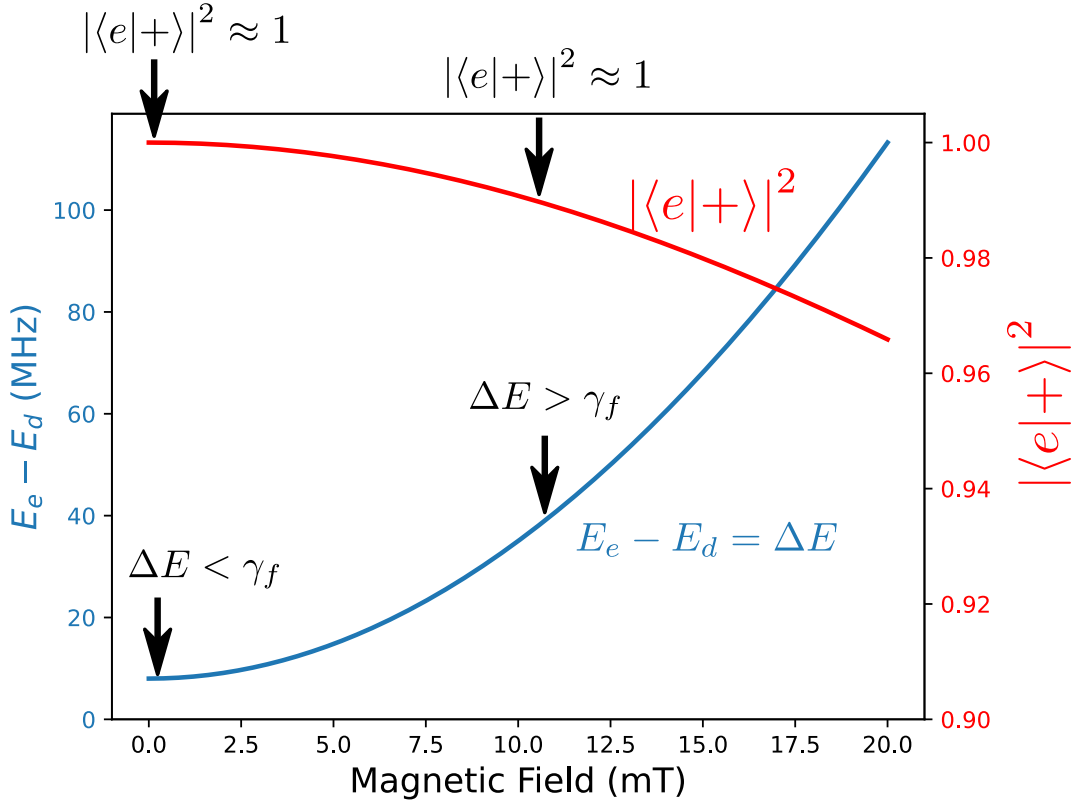
$\eta_{\text{flip-flop}} = 3n_x^2 - 1$ or $3n_y^2 - 1 \neq \frac{3}{2}(n_x^2 + n_y^2) - 1$
$\rightarrow \bar{\eta}_{\text{flip-flop}}(|\pm 1\rangle) = 0.3849 < \bar{\eta}_{\text{flip-flop}}(|\pm\rangle) = 0.7698$

$\mathcal{H}_0$ under pure transverse field

Eigen states of $\mathcal{H}_0$ when $\vec{B} \parallel \vec{e}_x$:

$$\text{——} |e\rangle \approx |+\rangle$$
$$\text{——} |d\rangle = |-\rangle$$
$$\text{——} |g\rangle \approx |0\rangle$$

$$|+\rangle = \frac{|+1\rangle + |-1\rangle}{\sqrt{2}}$$
$$|-\rangle = \frac{|+1\rangle - |-1\rangle}{\sqrt{2}}$$

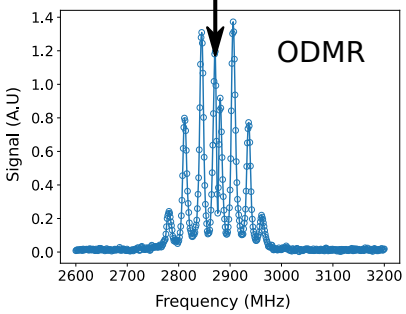


$$\bar{\eta} \propto \frac{\gamma_f^2}{(\Delta E)^2 + \gamma_f^2}$$

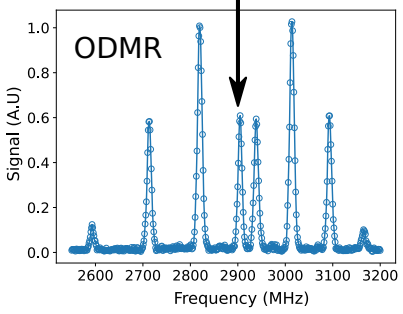
$$\gamma_f \approx 15 \text{ MHz}$$

Experimental results with transverse field

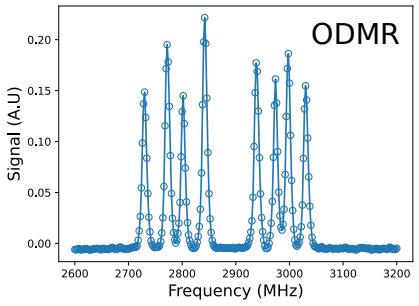
Weak transverse field :
$\rightarrow |\pm\rangle$ basis AND double flips

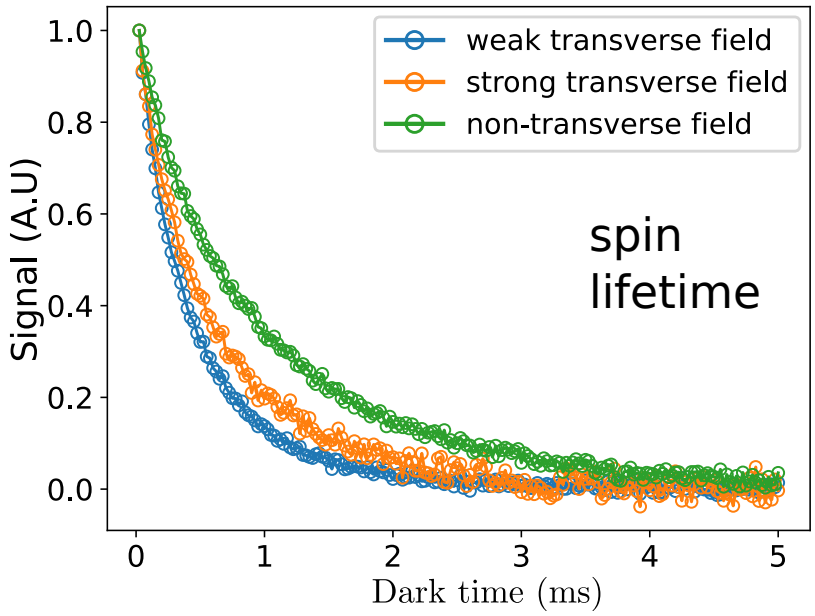


Strong transverse field :
$\rightarrow |\pm\rangle$ basis, no double flips



Non-transverse field :
$\rightarrow |\pm 1\rangle$ basis, no double flips





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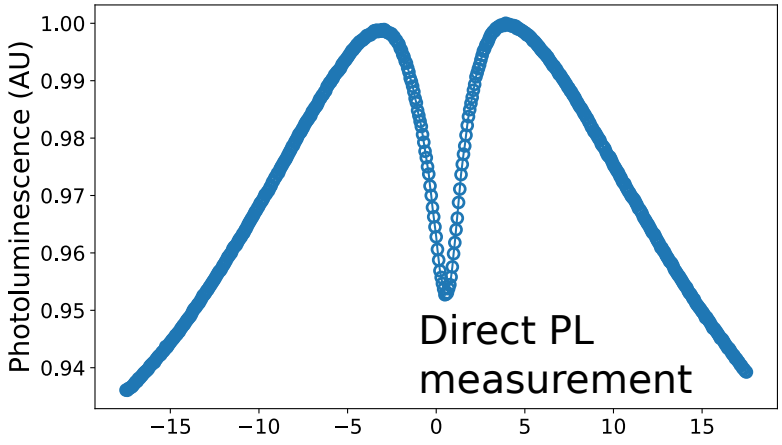
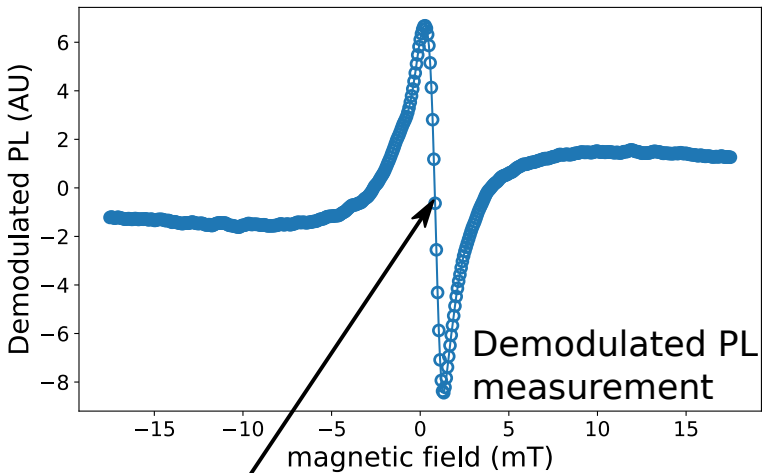
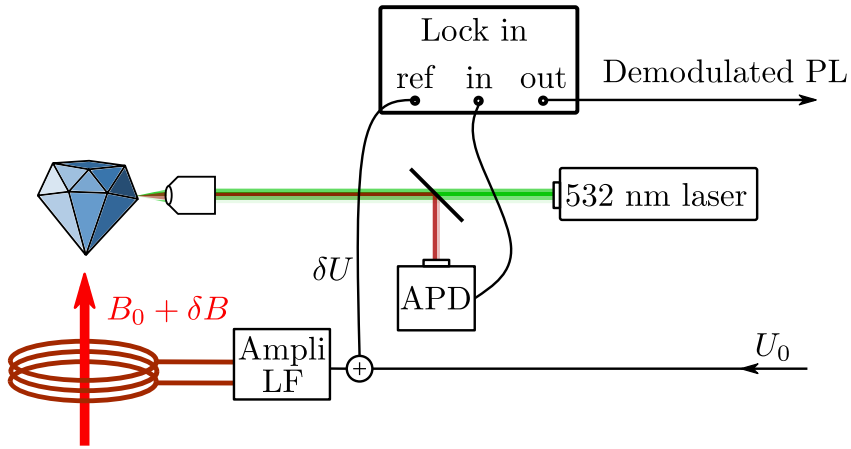
Low field magnetometry experimental setup

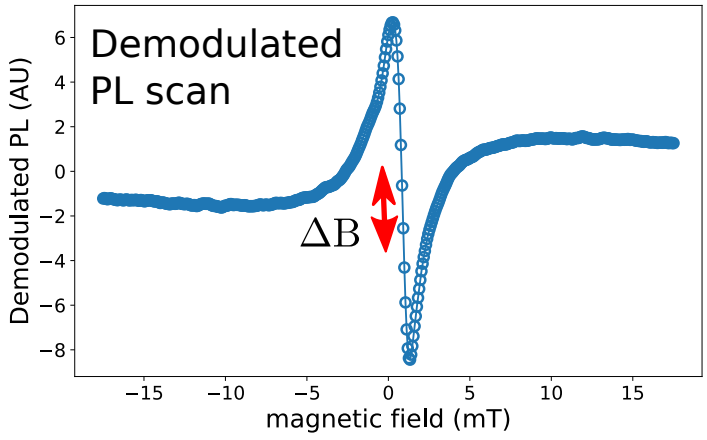


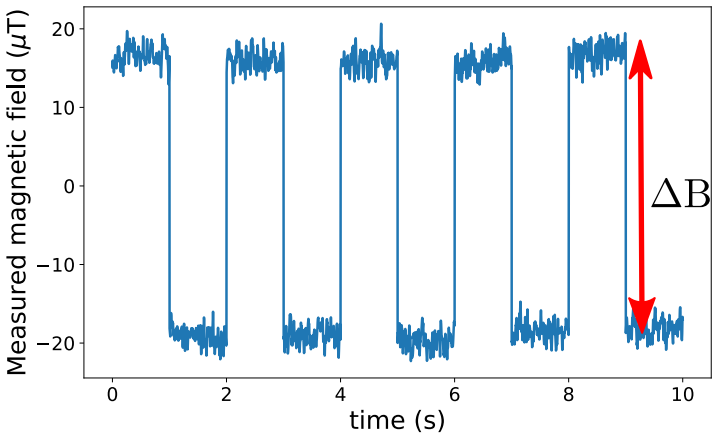


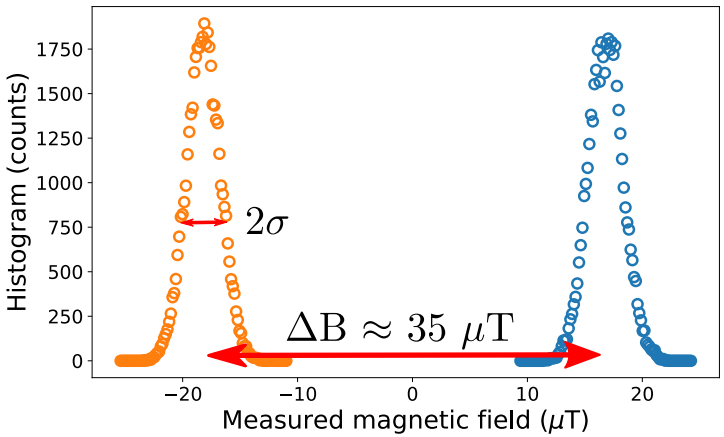
Highest sensitivity region

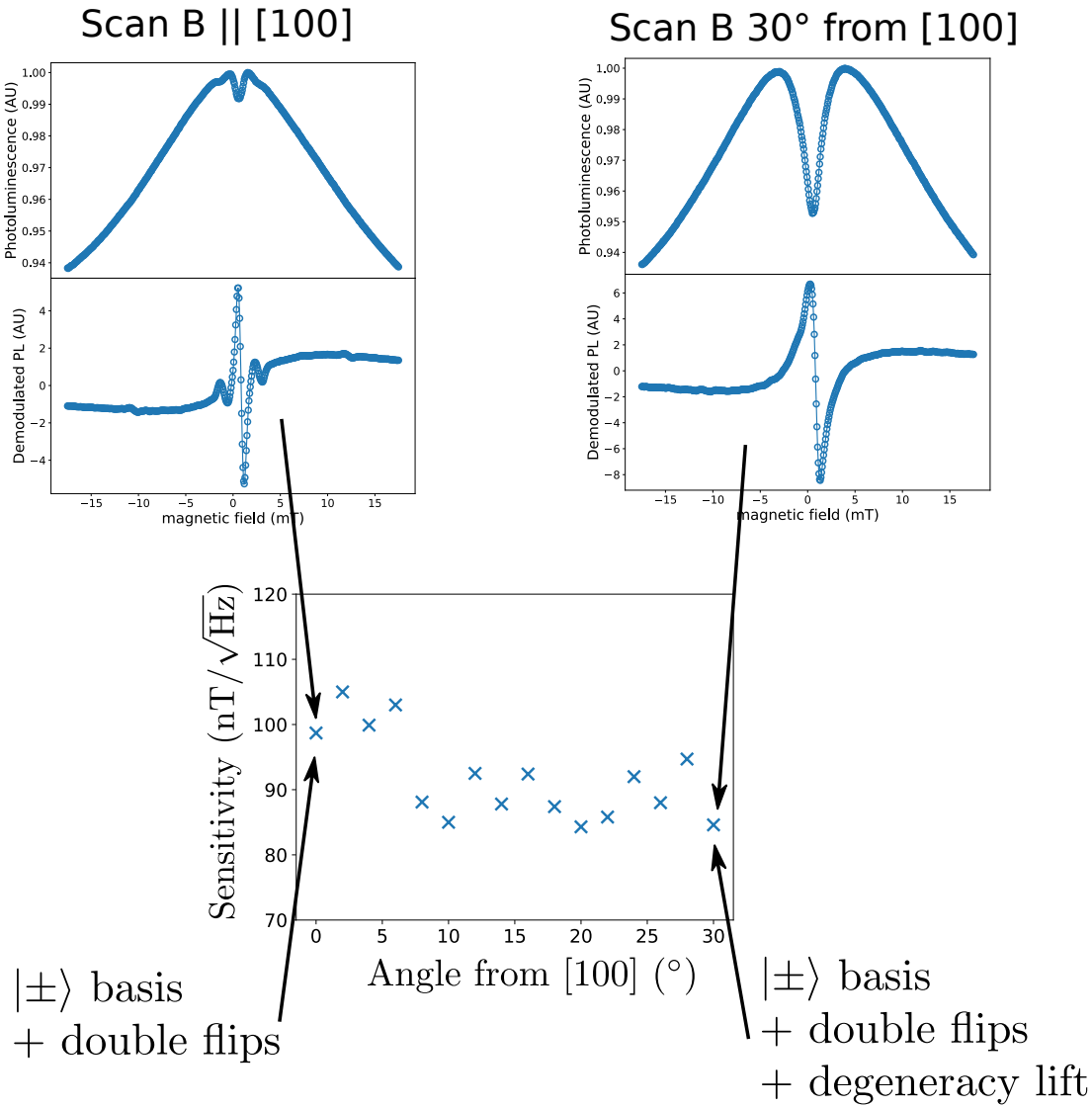


Magnetometry protocol







$$\text{Sensitivity} = \sigma\sqrt{\tau_{\text{meas}}} \approx 100\ \text{nT}/\sqrt{\text{Hz}}$$

Effects of the various effects on the sensitivity