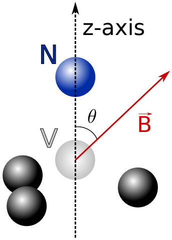
Group meeting : Low-field dipolar interaction between NV centers in diamond

March 24, 2022

Outline

Outline

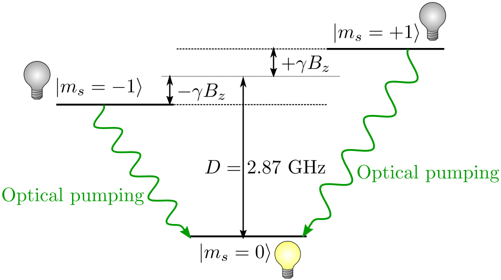
NV$^-$ spin in the fundamental electronic state



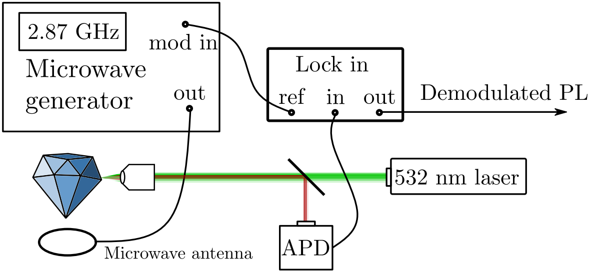
$$\mathcal{H} = D\hat{S}_z^2 + \gamma_e \hat{\mathbf{S}} \cdot \mathbf{B}$$

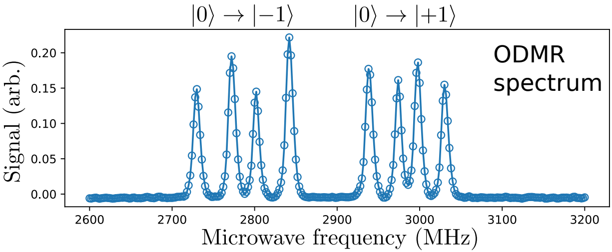
$\mathbf{z}$ of $\hat{S}_z$ defined by the crystalline axis

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



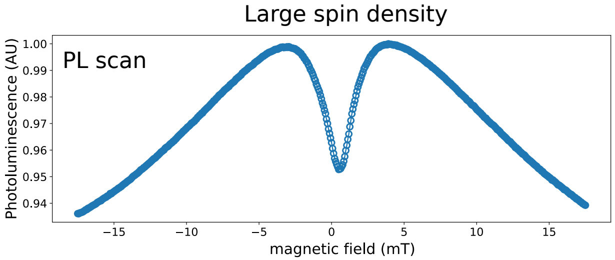
Principle of CW ODMR

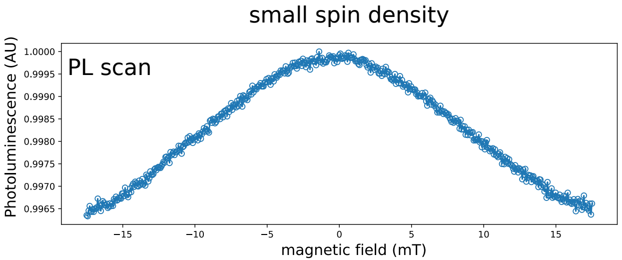




Outline

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

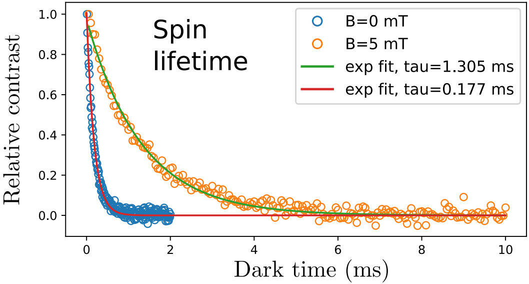


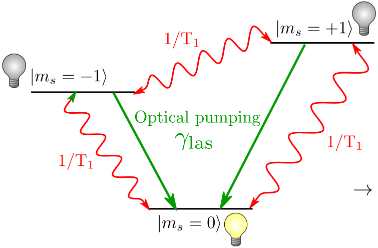


Why does it matter ?

- 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





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

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

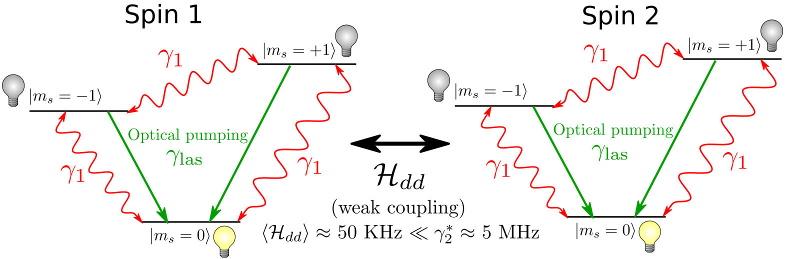
$\rightarrow$ The PL decreases when γ_1 increases

Spin relaxation amplified by dipolar coupling

See animation

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

Homogeneous case

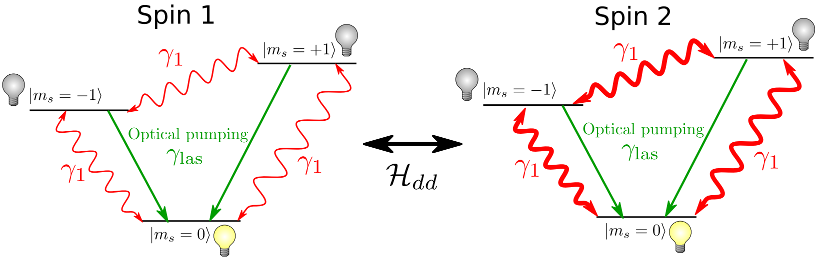


Flip-flop processes ($|-1;0\rangle \to |0;-1\rangle$) preserve the spin population in $|0\rangle$

$\to$ 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 ($\rho_{00}^2 < \rho_{00}^1$)

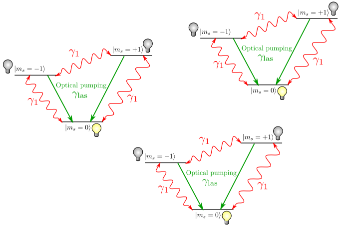
$\rightarrow$ The process $|0;-1\rangle \rightarrow |-1;0\rangle$ is more likely than $|-1;0\rangle \rightarrow |0;-1\rangle$

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

Outline

Theoretical frame : the fluctuator model

"Normal" NVs

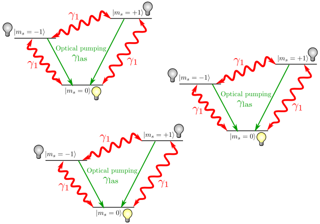


$$\gamma_1 \ll \gamma_{las}$$

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

$$\mathcal{H}_{dd}$$

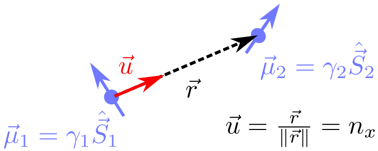
Fluctuator NVs



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

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

The dipole-dipole Hamiltonian



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

$$\mathcal{H}_{dd} = -\frac{\mu_0 \gamma_1 \gamma_2 \hbar^2}{4\pi r^3} [3(\widehat{\mathbf{S}}_\mathbf{1} \cdot \mathbf{u})(\widehat{\mathbf{S}}_\mathbf{2} \cdot \mathbf{u}) - \widehat{\mathbf{S}}_\mathbf{1} \cdot \widehat{\mathbf{S}}_\mathbf{2}]$$

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

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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- 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 :

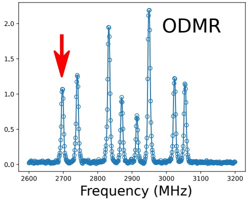
- n_f is the fluctuator density
- γ_f is the fluctuator decay rate

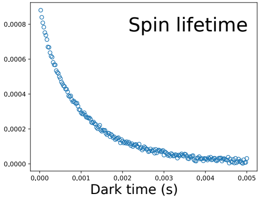
and :

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

Examples

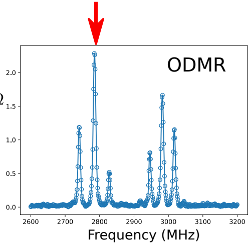
$$\bar{\eta} = \frac{1}{4}\int_{\theta,\phi} |\eta(\theta,\phi)| \mathrm{d}\Omega$$
$$= \frac{1}{4} \times 0.3849$$

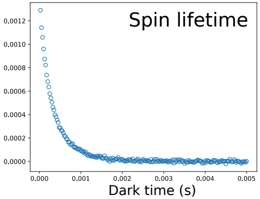




$$\boxed{\mathrm{T} \propto \frac{1}{\bar{\eta}^2}}$$

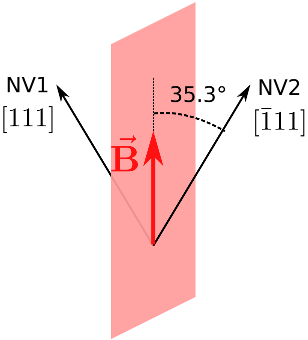
$$\bar{\eta} = \frac{1}{4}\int_{\theta,\phi} |\eta(\theta,\phi)| \mathrm{d}\Omega + \frac{1}{4}\int_{\theta,\phi} |\eta'(\theta,\phi)| \mathrm{d}\Omega$$
$$= \frac{1}{4} * 0.3849 + \frac{1}{4} * 0.8328$$





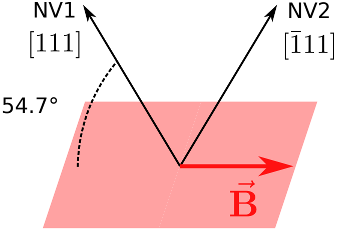
Non-trivial prediction of the model

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



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

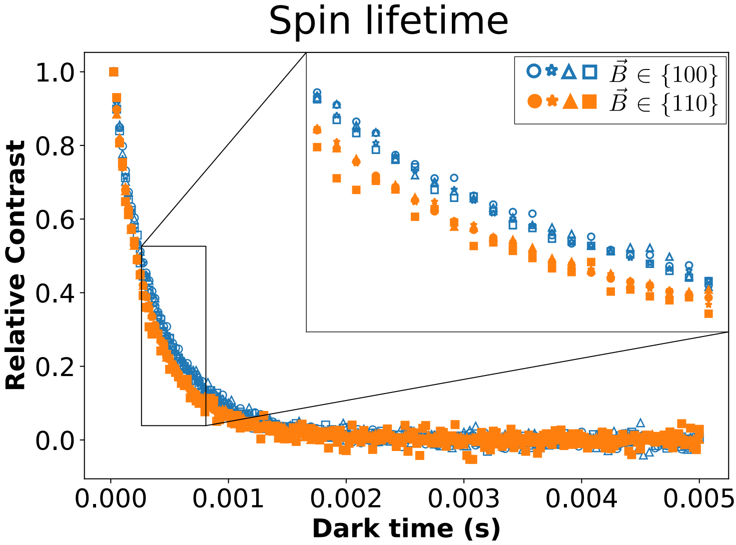
$\vec{B} \in \{011\}$



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

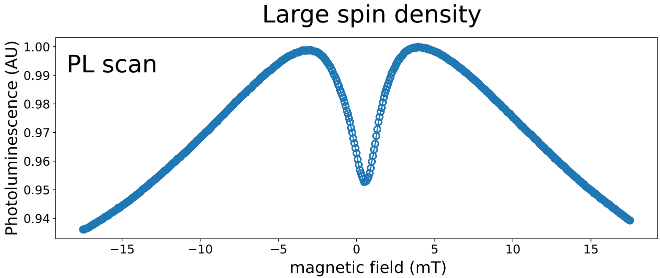
$$\boxed{\begin{array}{c} \bar{\eta}^{100} < \bar{\eta}^{011} \\ \rightarrow \text{We should observe } T_1^{100} > T_1^{011} \end{array}}$$

Non-trivial prediction of the model



Outline

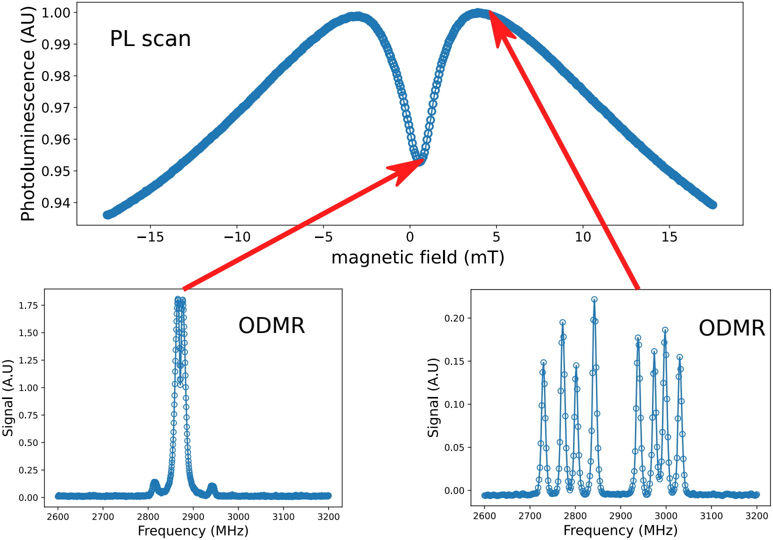
The reasons behind the dip in zero-field



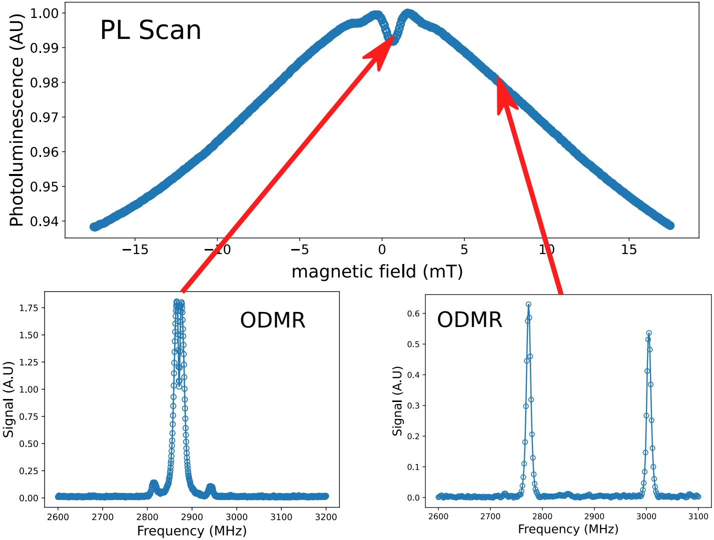
There are 3 parts, all linked to dipolar coupling :

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 $\mathcal{H}_0$

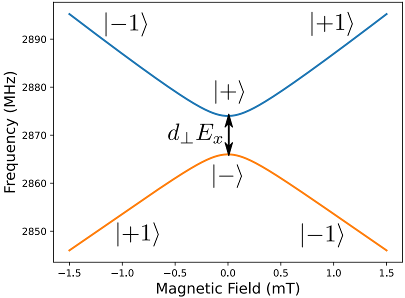
$$\boxed{\mathcal{H}_0 = \begin{pmatrix} D - \gamma_e B_z & \gamma_e B_x & d_\perp E_x \\ \gamma_e B_x & 0 & \gamma_e B_x \\ d_\perp E_x & \gamma_e B_x & D + \gamma_e B_z \end{pmatrix}}$$

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 $(|-\rangle, |0\rangle, |+\rangle) \otimes (|-\rangle, |0\rangle, |+\rangle)$:

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

$\eta_{\mathrm{flip-flop}} = 3n_x^2 - 1$ or $3n_y^2 - 1 \neq \frac{3}{2}(n_x^2 + n_y^2) - 1$

$\rightarrow \bar{\eta}_{\mathrm{flip-flop}}(|\pm 1\rangle) = 0.3849 < \bar{\eta}_{\mathrm{flip-flop}}(|\pm\rangle) = 0.7698$

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

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

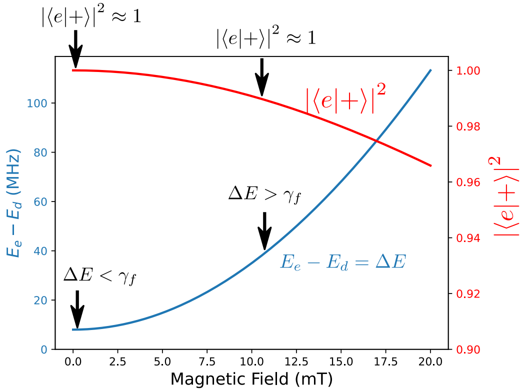
$|e\rangle \approx |+\rangle$

$|d\rangle = |-\rangle$

$|g\rangle \approx |0\rangle$

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

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

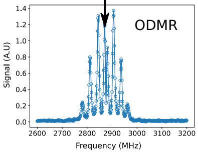


$$\bar{\eta} \propto \frac{\gamma_f^2}{(\Delta E)^2 + \gamma_f^2}$$

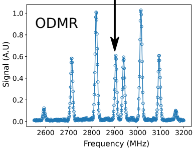
$$\gamma_f \approx 15 \text{ MHz}$$

Experimental results with transverse field

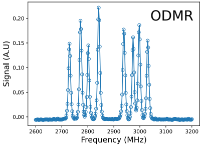
Weak transverse field :
$\rightarrow |\pm\rangle$ basis AND double flips

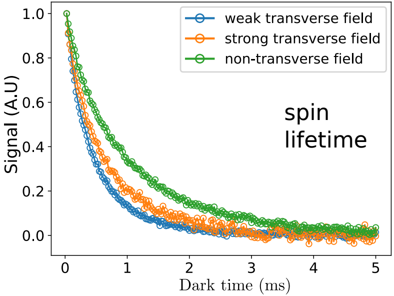


Strong transverse field :
$\rightarrow |\pm\rangle$ basis, no double flips



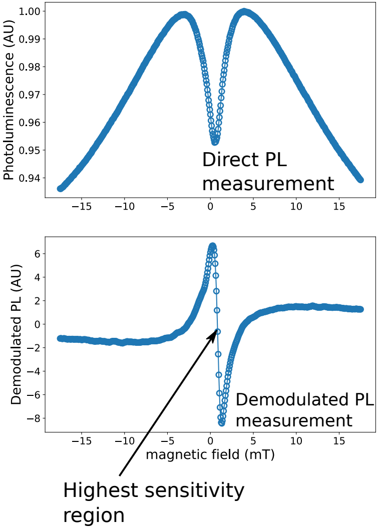
Non-transverse field :
$\rightarrow |\pm1\rangle$ basis, no double flips

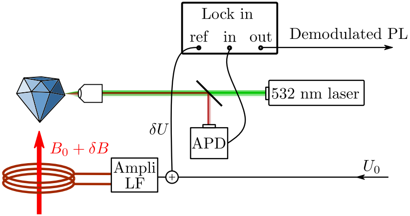




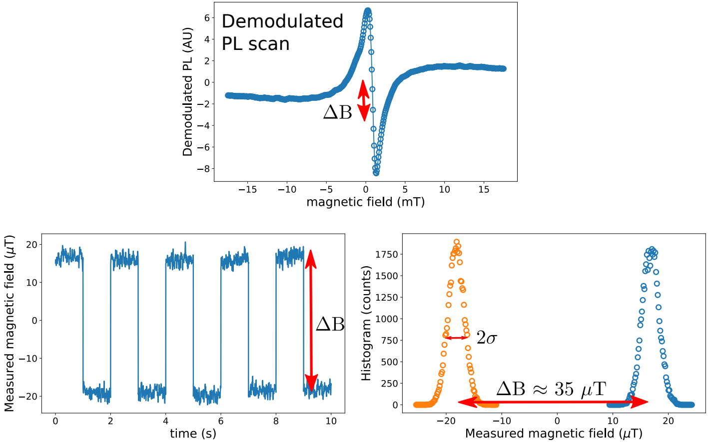
Outline

Low field magnetometry experimental setup





Magnetometry protocol



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

Importance of the various effects on the sensitivity