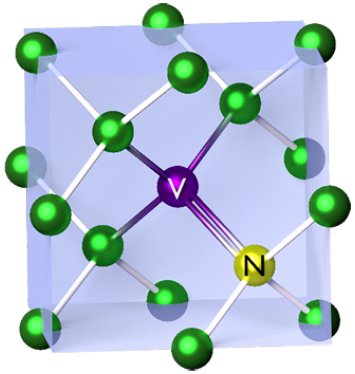
Dipolar interactions in dense ensembles of Nitrogen-Vacancy centers

Clément Pellet-Mary, Maxime Perdriat, Gabriel Hétet

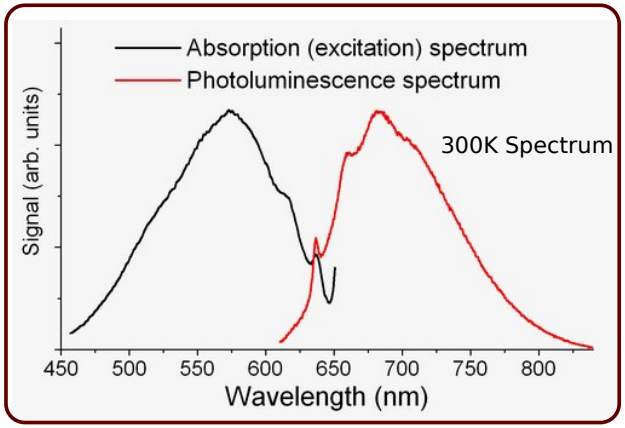
Nano-optics group

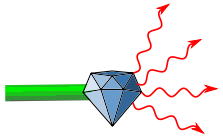
Preamble : the NV center

Crystalline structure



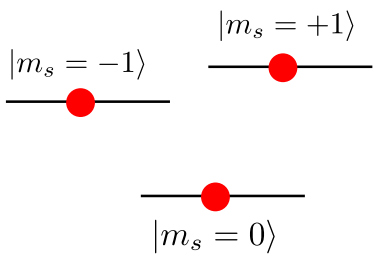
Optical properties





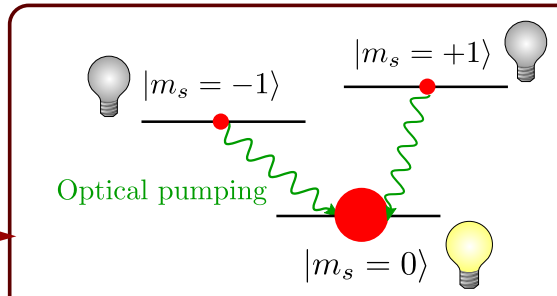
Spin properties

$S = 1$ in the electronic ground state



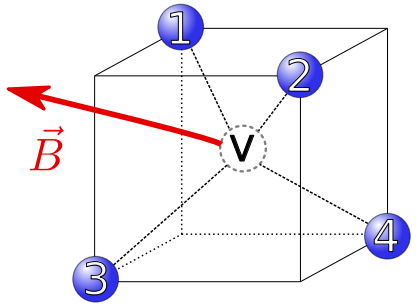
Unpolarized at 300 K :
$\rho_{00} \approx 1/3 \approx \rho_{+1+1} \approx \rho_{-1-1}$

Interplay between spin and light

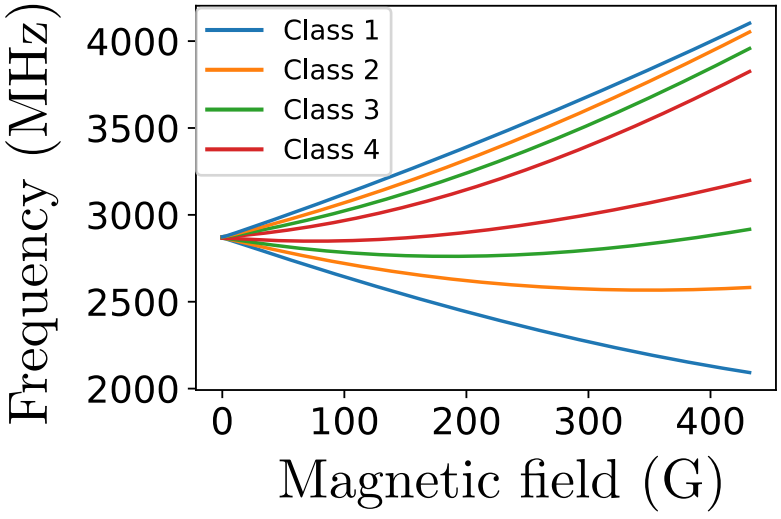


- Optical pumping in the $|m_s = 0\rangle$ state
- $|m_s = 0\rangle$ sate brighter than $|m_s = \pm 1\rangle$

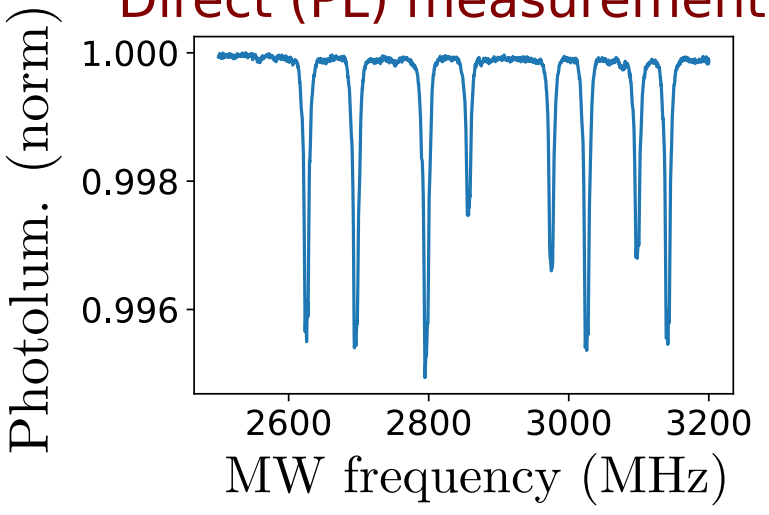
Preamble : the 4 classes of NV centers



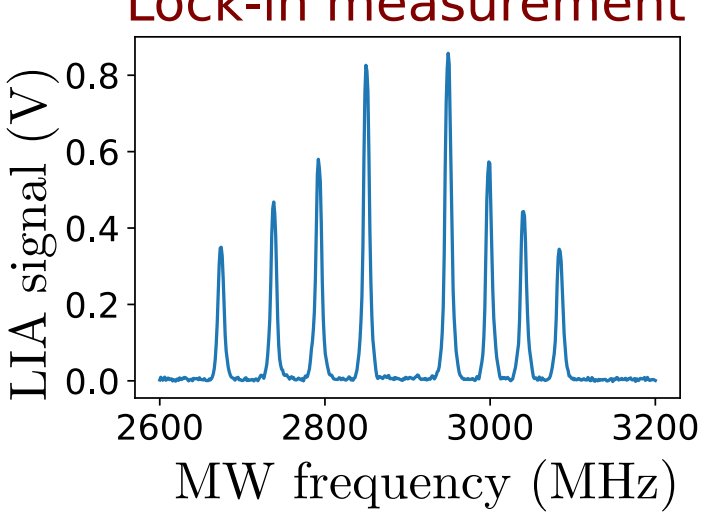
4 different projections of $\vec{B}$ over the 4 possible NV axes
$\rightarrow$ 4 classes of resonances



Direct (PL) measurement

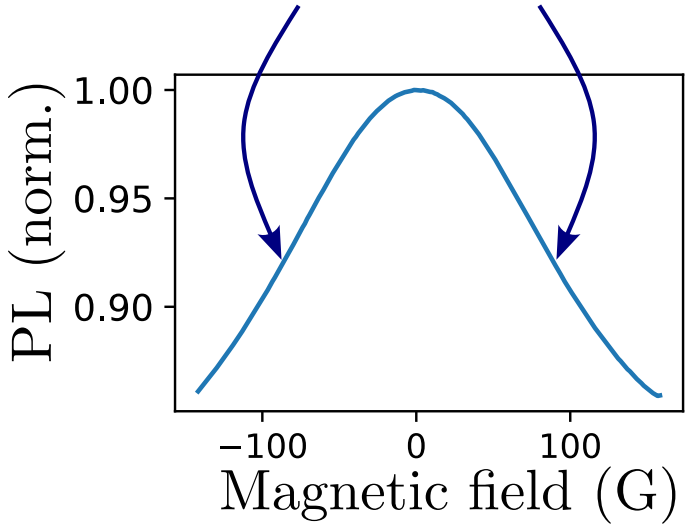


Lock-in measurement

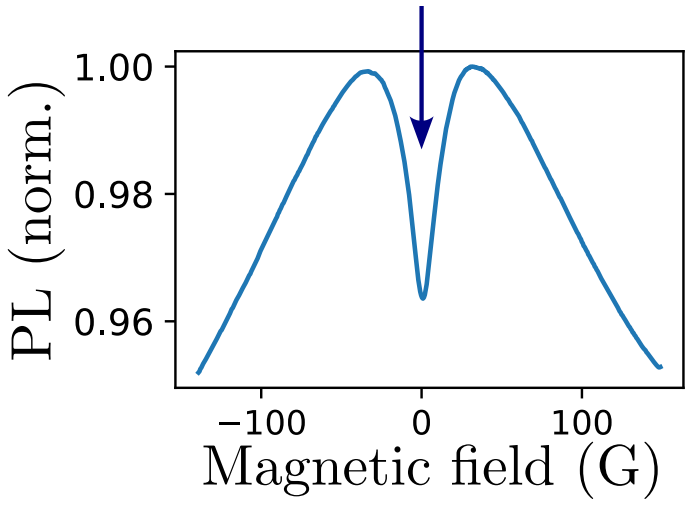


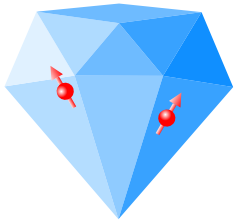
Subject of this presentation

Transverse field

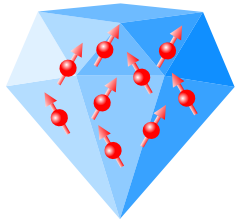


Dipole-dipole spin relaxation





Low NV density
[NV] ≤ 100 ppb

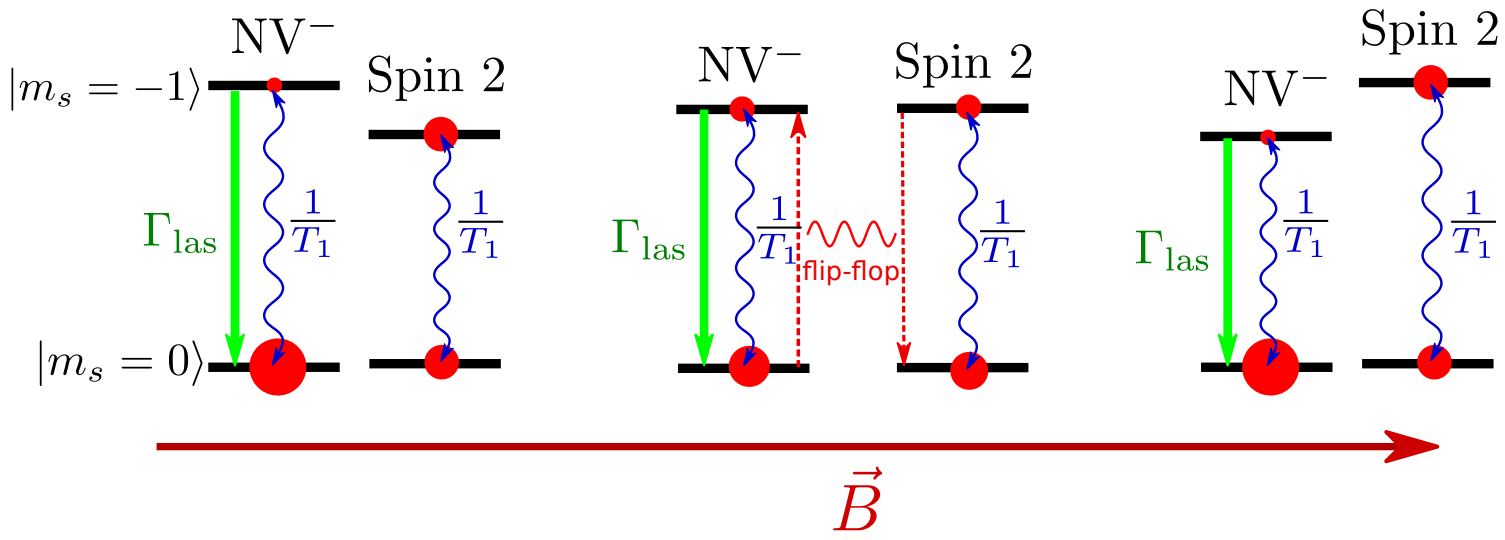


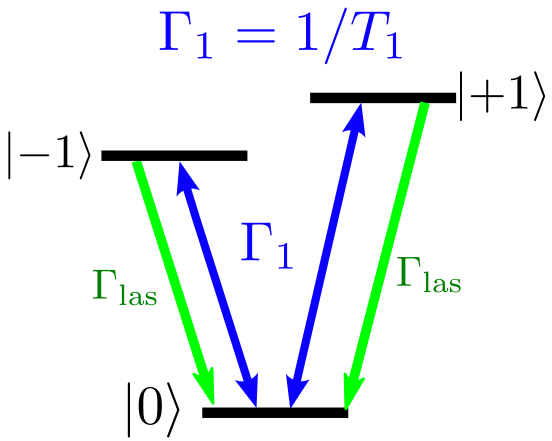
High NV density
[NV] ≥ 1 ppm

- Better understand the dipole-dipole interaction in dense NV ensembles
- Exploit the PL feature for magnetometry

Outline

Principle of cross-relaxation with NV centers

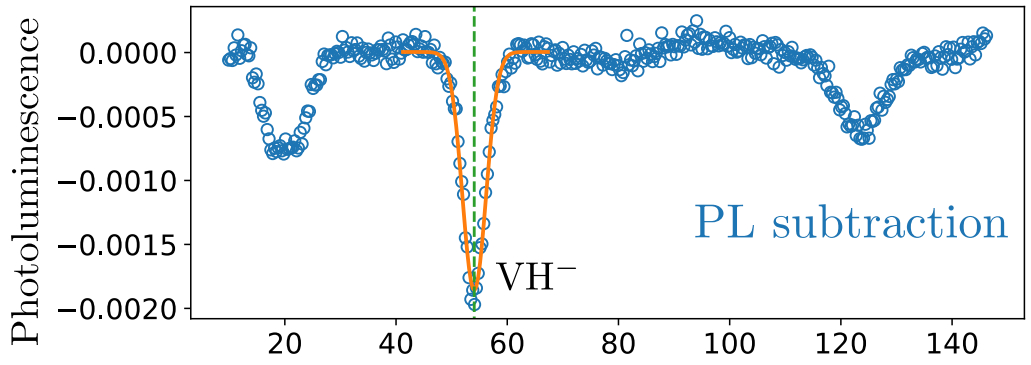


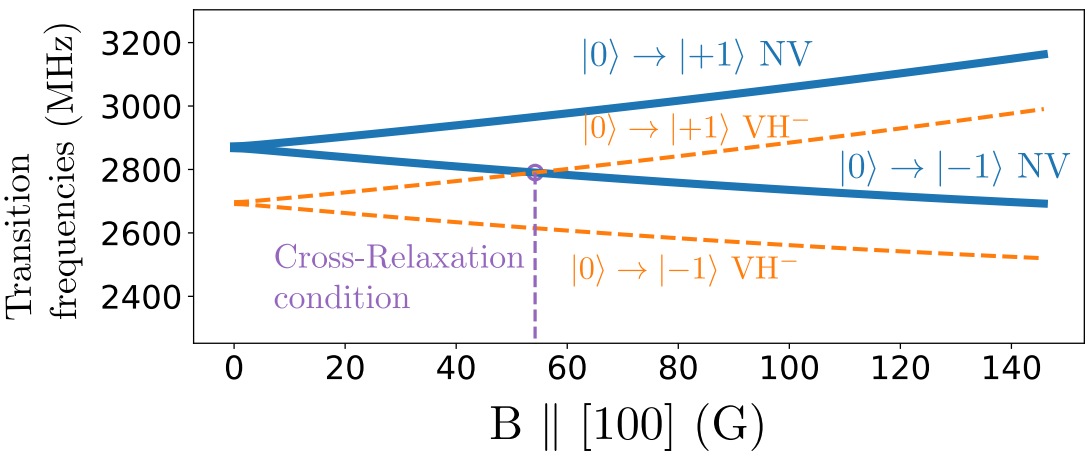


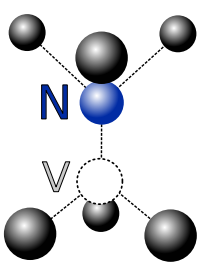
Rate equation: $\rho_{00} = \frac{\Gamma_1 + \Gamma_{\text{las}}}{3\Gamma_1 + \Gamma_{\text{las}}}$

$$\Gamma_1 \nearrow \Rightarrow PL \searrow$$

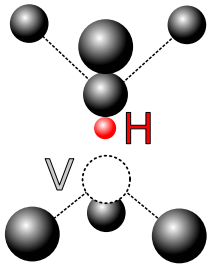
Example: Cross-relaxation between NV centers and VH^-





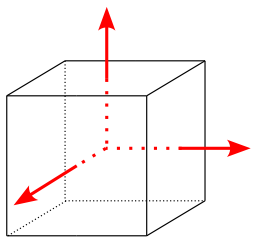


NV center



VH center

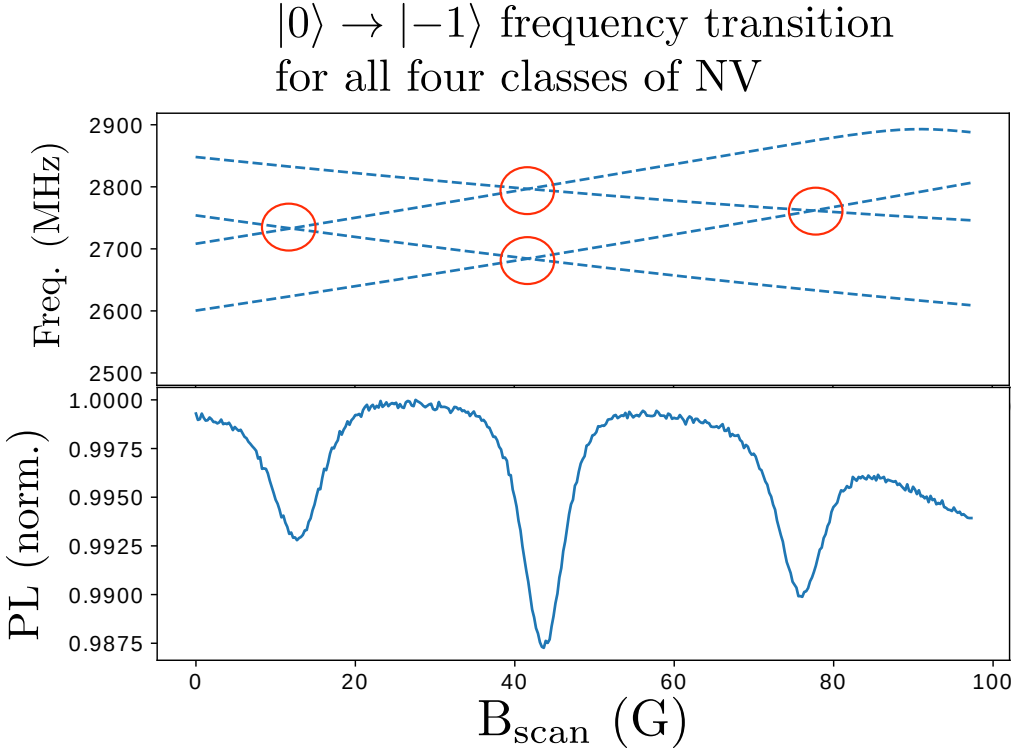
$\vec{B} \parallel \langle 100 \rangle$



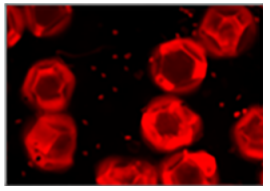
Optical detection of paramagnetic defects in diamond grown by chemical vapor deposition

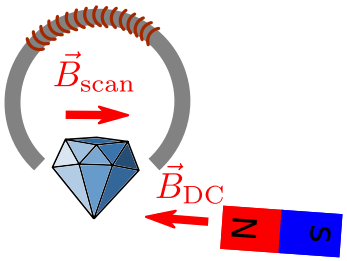
C. Pellet-Mary, P. Huillery, M. Perdriat, A. Tallaire, and G. Hétet
Phys. Rev. B **103**, L100411 – Published 24 March 2021

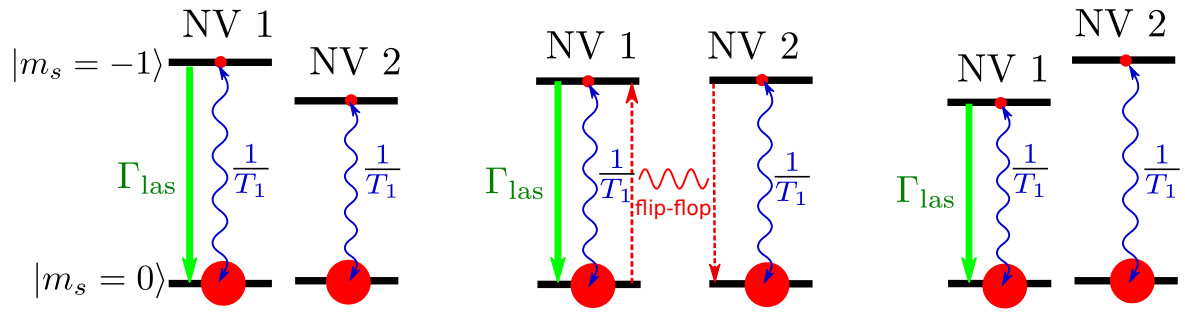
Cross-relaxation between NV centers and NV centers



samples : Adamas 15/150 µm
fluorescent microdiamond
[N] = 100~200 PPM
[NV] ~ 3 PPM

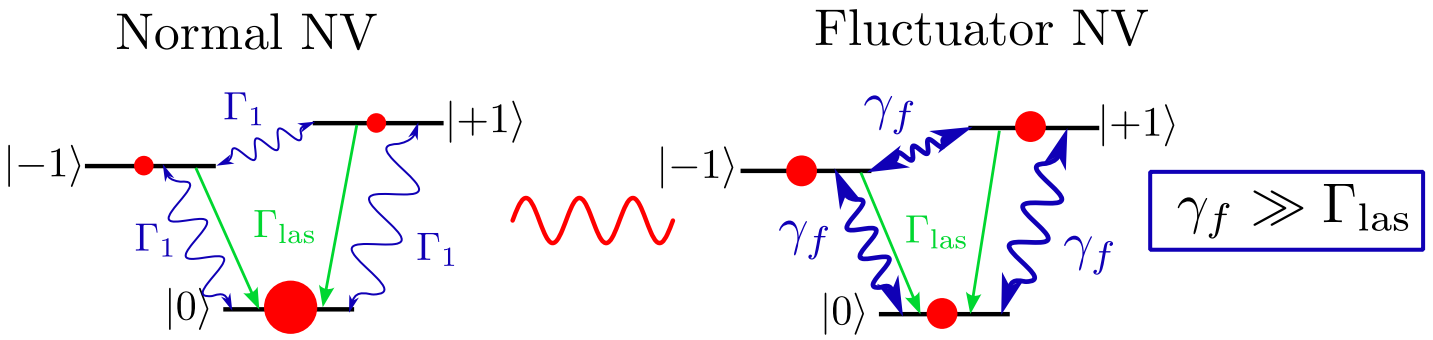






No cross-relaxation for equally polarized spins

Presentation of the fluctuator model



Fluctuators are NV centers with a fast intrinsic depolarization mechanism

→ Localized noise sources with the spectral response of an NV center

Precedents in:
- P-doped Si
- solid-state NMR
- FRET

Possible microscopic explanation:
- charge tunneling
- modulation of J-coupling

Choi, Joonhee, et al. Physical review letters 118.9 (2017): 093601.

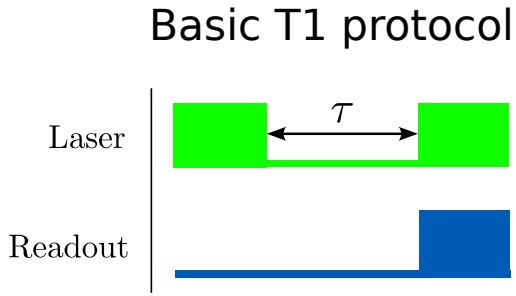
Predictions of the fluctuator model

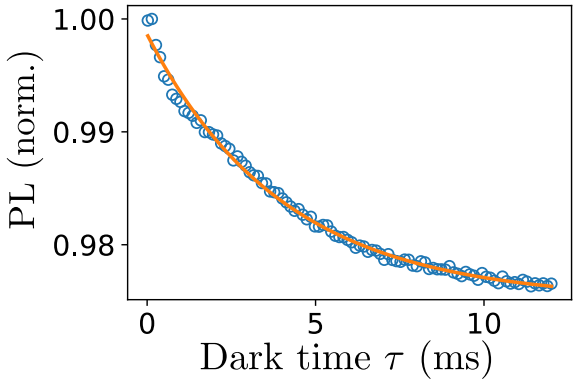
- Γ_1 increases when classes overlap spectrally (increase in the resonant fluctuator density).
- The dipole induced depolarization has a stretched exponential profile:

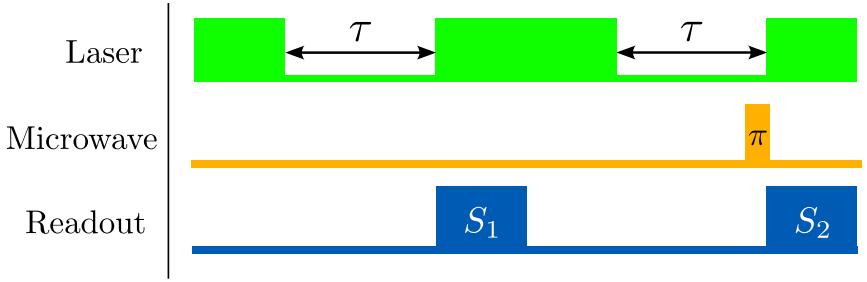
$$\rho_{00}(t) \propto \exp\left(-\sqrt{\frac{t}{T_1}}\right)$$

- The Fluctuators spectral response is broadened by their decay rate γ_f (lifetime limit).

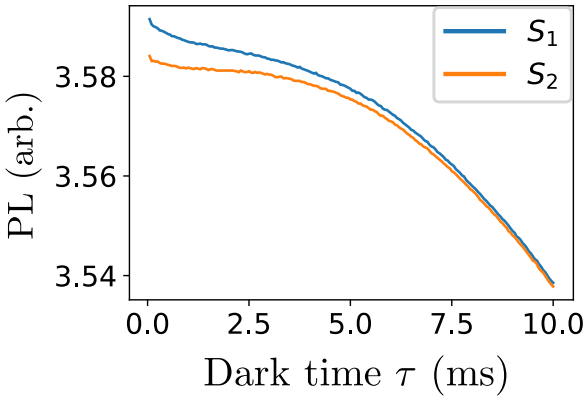
T_1 measurement protocol

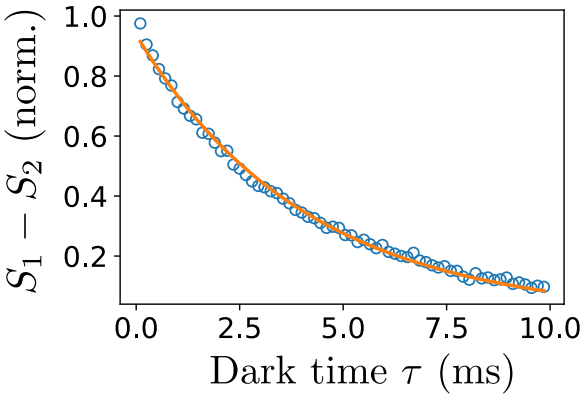




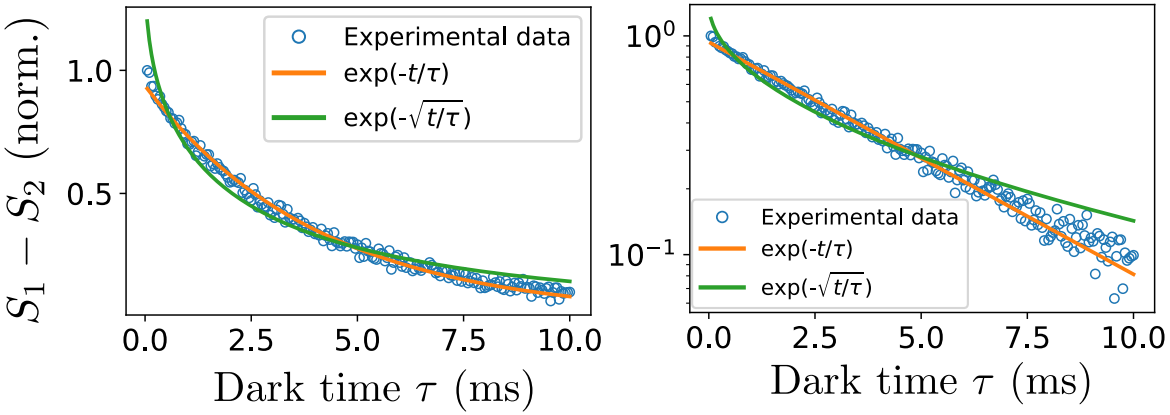


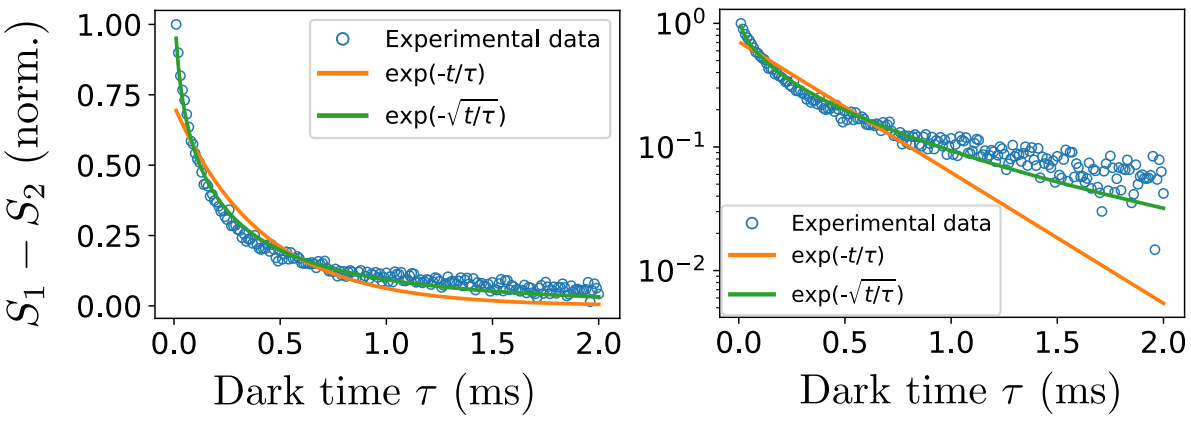
common mode rejection method

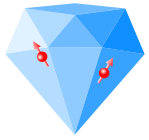




Stretched exponential decay profile

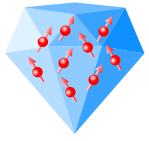






Low NV density

- Exponential profile
- $T_1 \sim 5$ ms

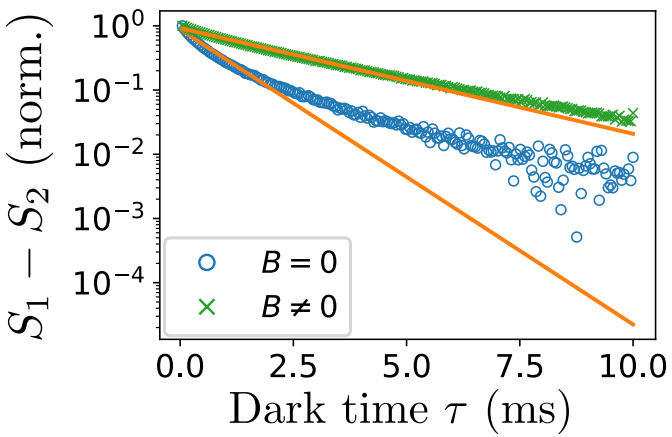


High NV density

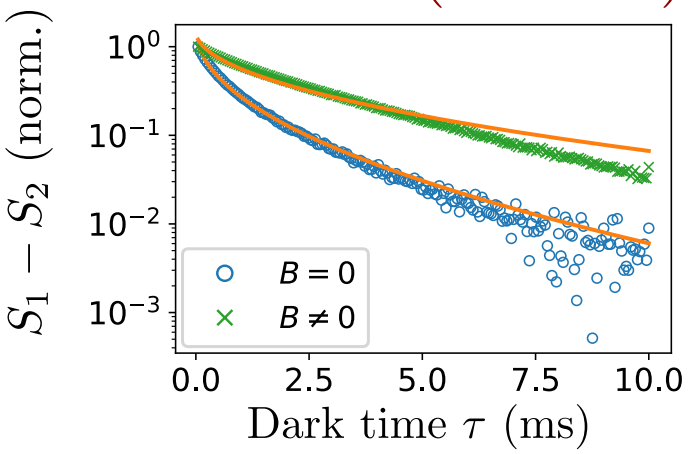
- Stretched exp. profile
- $T_1 \sim 0.5$ ms

Competition between stretched and exponential decay

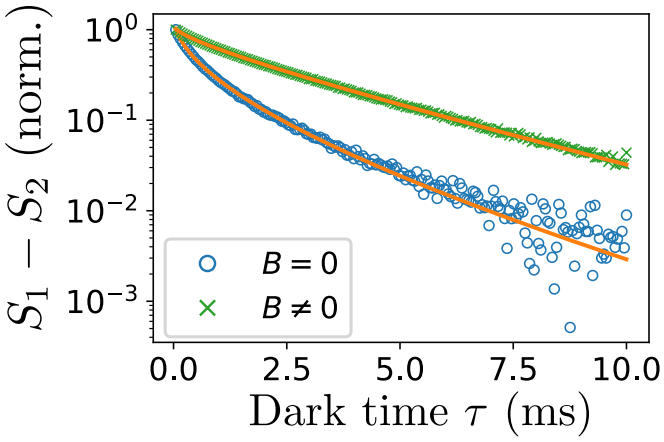
$$f(\tau) = \exp\left(-\tau/T_1^{\mathrm{ph}}\right)$$



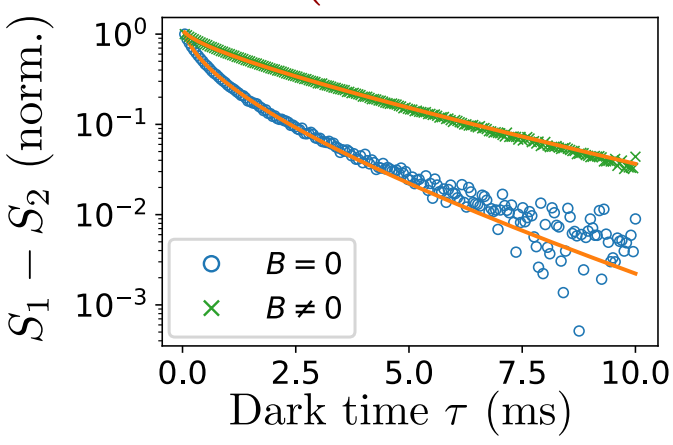
$$f(\tau) = \exp\left(-\sqrt{\tau/T_1^{\mathrm{dd}}}\right)$$



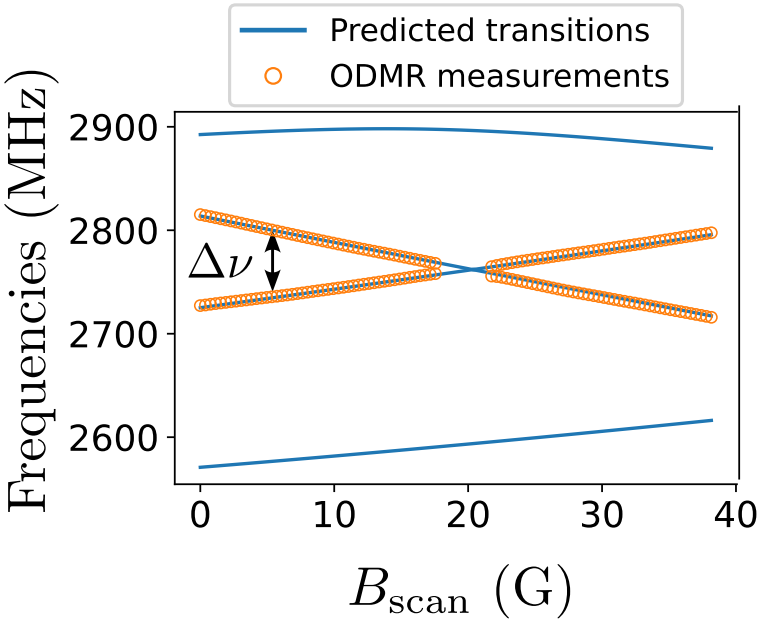
$$f(\tau) = \exp\left(-\tau/T_1^{\mathrm{ph}} - \sqrt{\tau/T_1^{\mathrm{dd}}}\right)$$

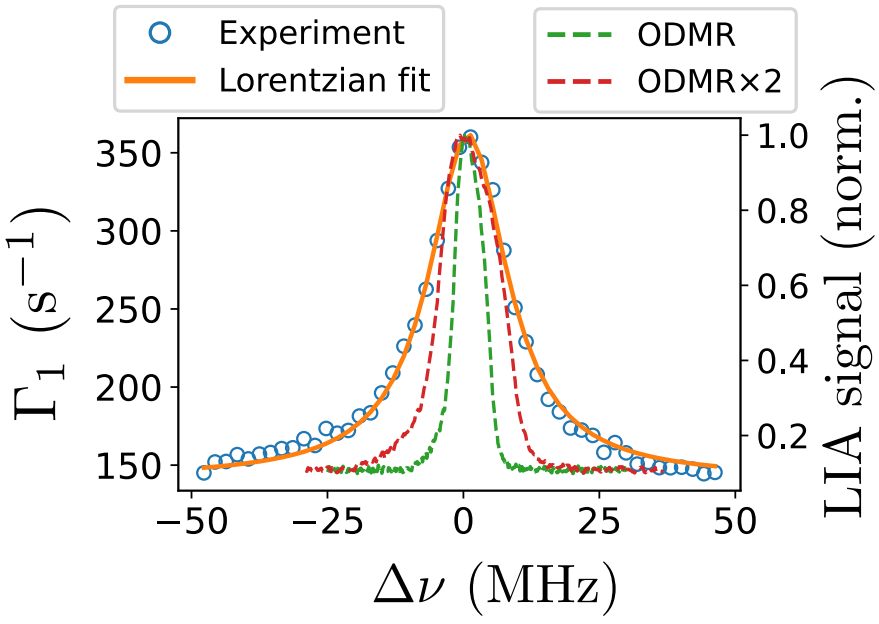


$$f(\tau) = \exp\left(-\tau/5 - \sqrt{\tau/T_1^{\mathrm{dd}}}\right)$$



Spectral response of the fluctuators



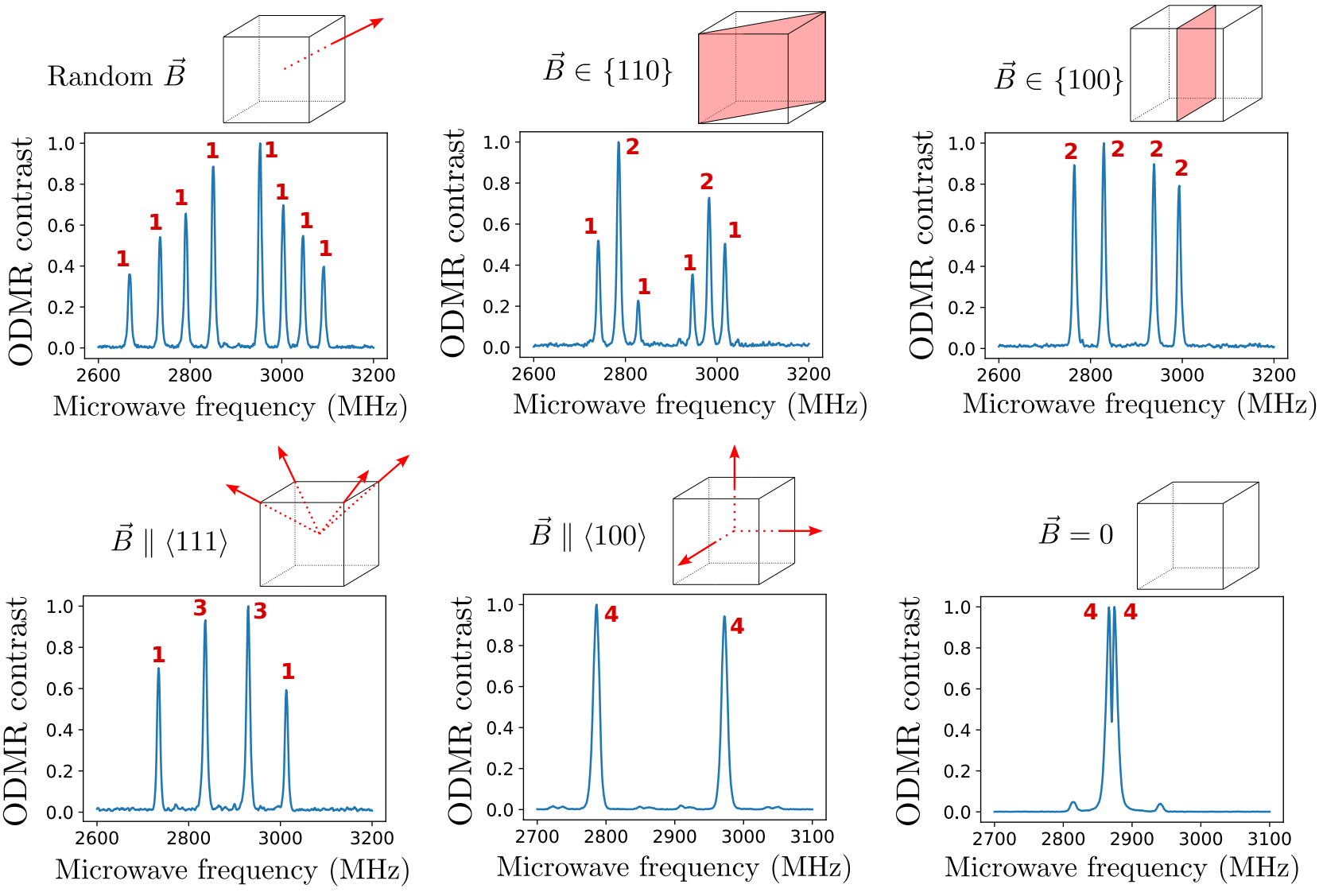


- Γ_1 curve broader than ODMR overlap
- Lorentzian shape

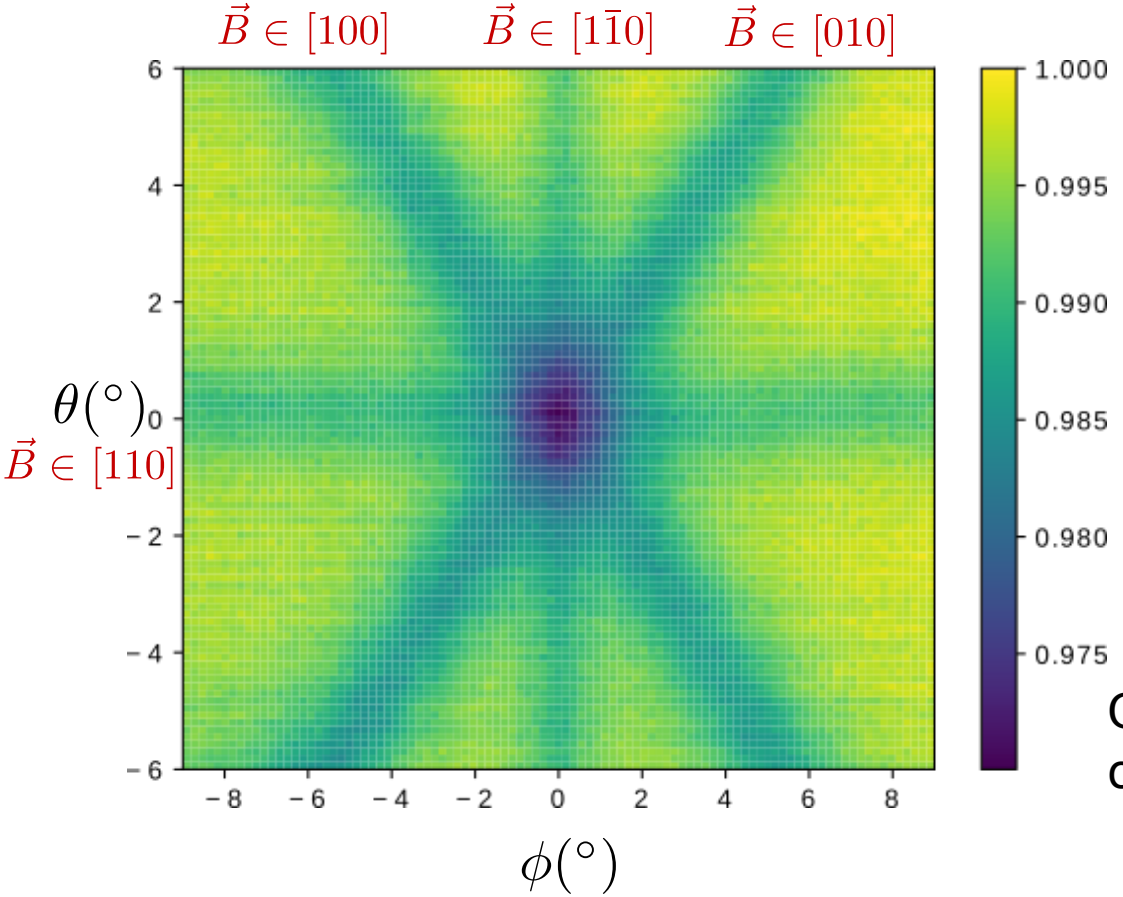
↓

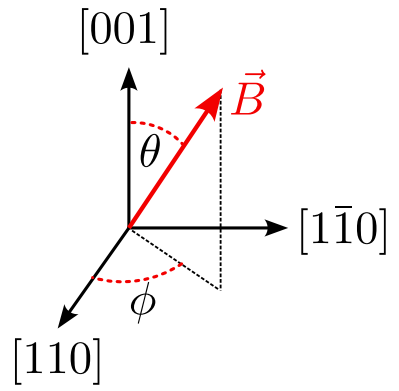
The fluctuator's spectral response (T_2^*) in broadened by γ_f

Geometry conditions for class resonances



PL mapping of the crystalline planes



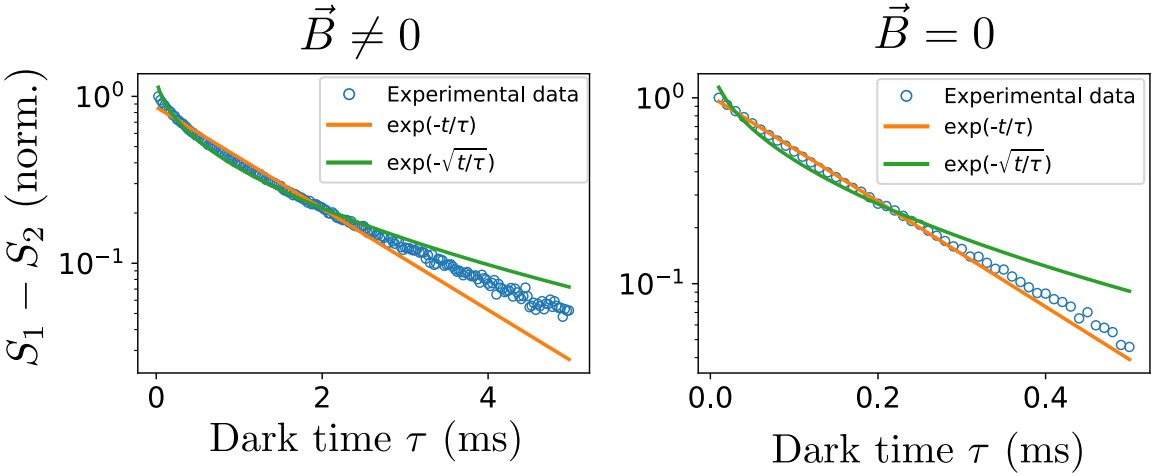


Crystalline planes visible only with PL !

Limitations of the fluctuator model

$\Gamma_1^{\mathrm{dd}}(\mathbf{B})$	Theory	Experimental
random $\mathbf{B}$ (1 class)	Γ_0^{th}	$1.53 \pm 0.04\ \mathrm{ms}^{-1} \equiv \Gamma_0^{\mathrm{exp}}$
$\mathbf{B} \in \{110\}$ (2 classes)	$10.0\ \Gamma_0^{\mathrm{th}}$	$5.2 \pm 0.1\ \Gamma_0^{\mathrm{exp}}$
$\mathbf{B} \in \{100\}$ (2 classes)	$7.24\ \Gamma_0^{\mathrm{th}}$	$4.2 \pm 0.1\ \Gamma_0^{\mathrm{exp}}$
$\mathbf{B} \parallel \langle 111 \rangle$ (3 classes)	$28.4\ \Gamma_0^{\mathrm{th}}$	$11.6 \pm 0.4\ \Gamma_0^{\mathrm{exp}}$
$\mathbf{B} \parallel \langle 100 \rangle$ (4 classes)	$42.8\ \Gamma_0^{\mathrm{th}}$	$14.1 \pm 0.5\ \Gamma_0^{\mathrm{exp}}$
$\mathbf{B} = 0$ (4 classes)	$104\ \Gamma_0^{\mathrm{th}}$	$19.9 \pm 0.8\ \Gamma_0^{\mathrm{exp}}$

Overestimation of the relaxation rate



Exponential lifetime (still dipole-dipole limited)

Improvement of the model:

- Saturation of the fluctuators (non-Markovian)
- NV-NV spin diffusion