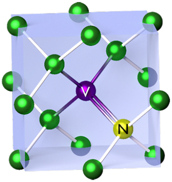
Low field depolarization of dense NV centers ensemble: application to magnetometry

Clément Pellet-Mary, Maxime Perdriat, Paul Huillery, Gabriel Hétet

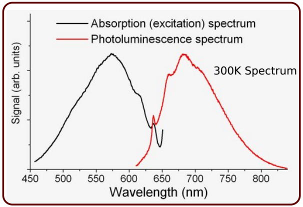
Nano-optics group

Preamble : the NV center

Crystalline structure

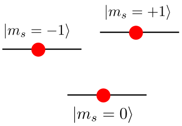


Optical properties

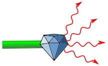


Spin properties

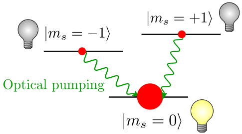
$S = 1$ in the electronic ground state



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

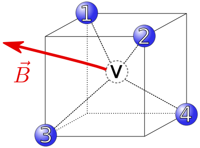


Interplay between spin and light

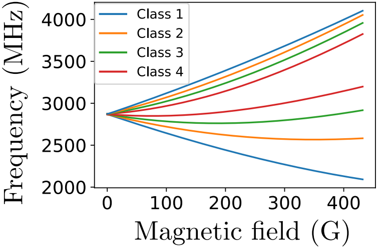


- Optical pumping in the $|m_s = 0\rangle$ state
- $|m_s = 0\rangle$ sate brighter than $|m_s = \pm 1\rangle$

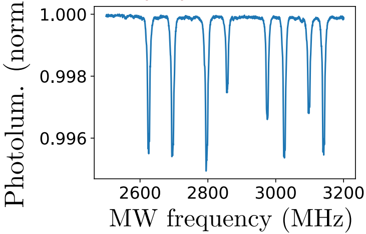
Preamble : the 4 classes of NV centers



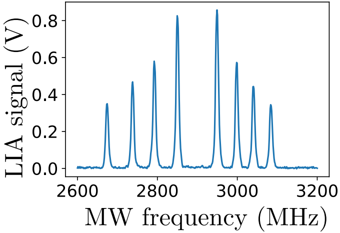
4 different projections of $\vec{B}$
over the 4 possible NV axes
$\rightarrow$ 4 classes of resonances



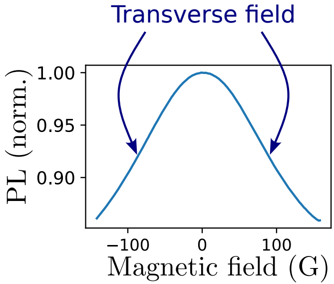
Direct (PL) measurement

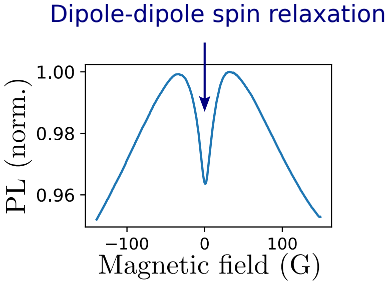


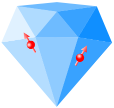
Lock-in measurement



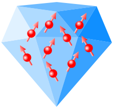
Subject of this presentation







Low NV density
[NV] ≤ 100 ppb



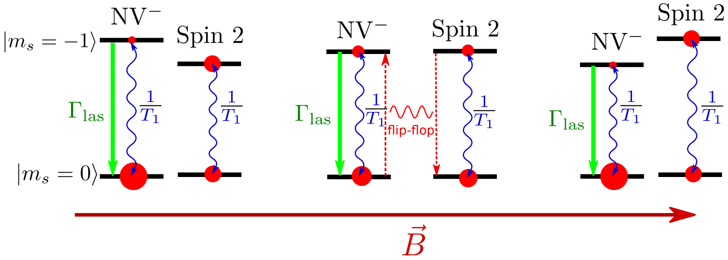
High NV density
[NV] ≥ 1 ppm

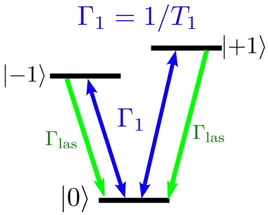
- Better understand the dipole-dipole interaction in dense NV ensembles
- Exploit the PL feature for magnetometry

Outline

Outline

Principle of cross-relaxation with NV centers

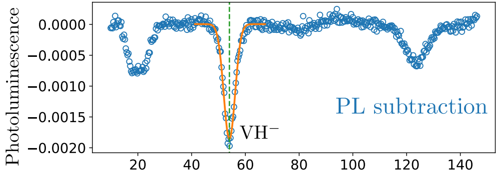


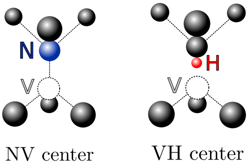


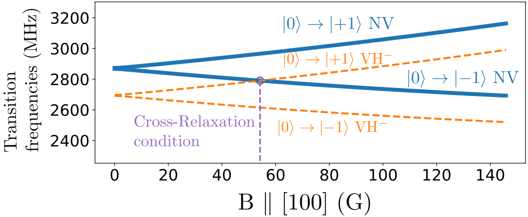
Rate equation: $\rho_{00} = \frac{\Gamma_1 + \Gamma_{\text{las}}}{3\Gamma_1 + \Gamma_{\text{las}}}$

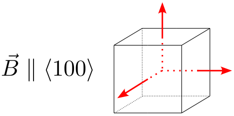
$$\Gamma_1 \nearrow \Rightarrow PL \searrow$$

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







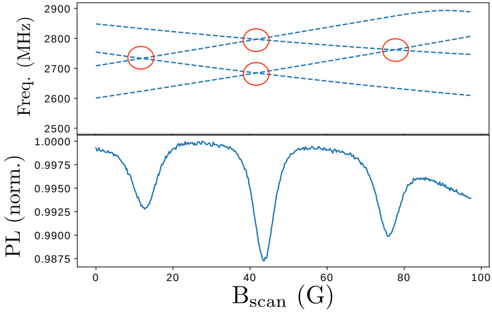


Optical detection of paramagnetic defects in diamond grown by chemical vapor deposition

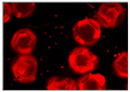
C. Pellet-Mary, P. Huillery, M. Perdriat, A. Tallaire, and G. Hétet
Phys. Rev. B **103**, L100411 – Published 24 March 2021

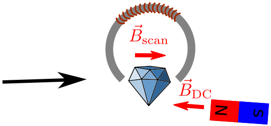
Cross-relaxation between NV centers and NV centers

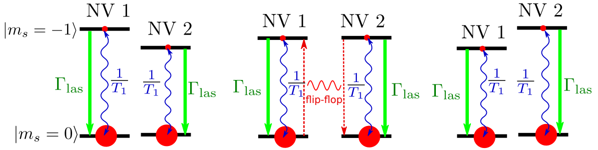
$|0\rangle \rightarrow |-1\rangle$ frequency transition
for all four classes of NV



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



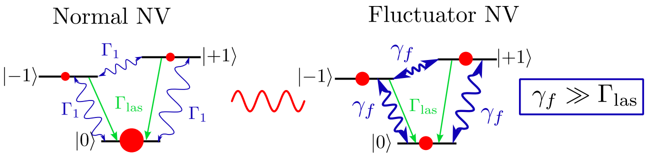




No cross-relaxation for equally polarized spins

Outline

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

Up to 1/3 of all NV centers could be fluctuators

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

Predictions of the fluctuator model

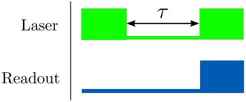
- Γ_1 increases when classes overlap spectrally (increase in the resonant fluctuator density).
- The dipole induced depolarization has a stretched exponential profile:

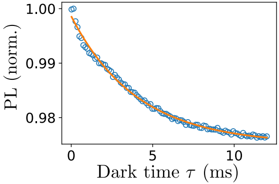
$$\rho_{00}(t) \propto \exp\left(-\sqrt{\frac{t}{T_1}}\right)$$

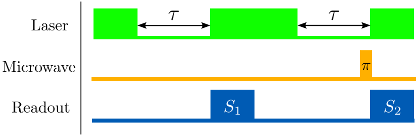
- The Fluctuators spectral response is broadened by their decay rate γ_f (lifetime limit).

T_1 measurement protocol

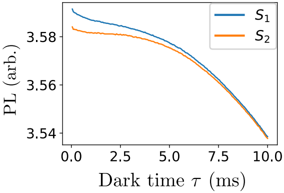
Basic T1 protocol

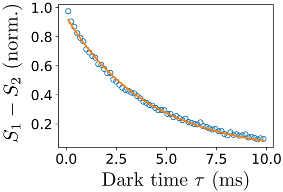




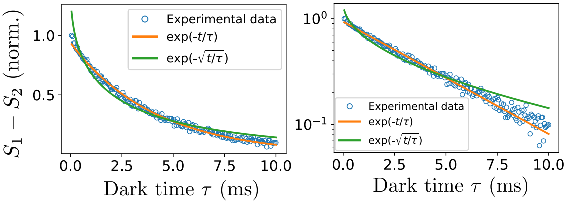


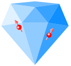
common mode rejection method





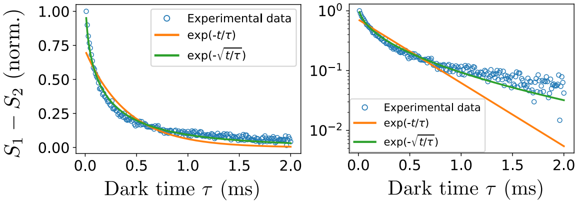
Stretched exponential decay profile

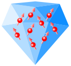




Low NV density

- Exponential profile
- $T_1 \sim 5$ ms

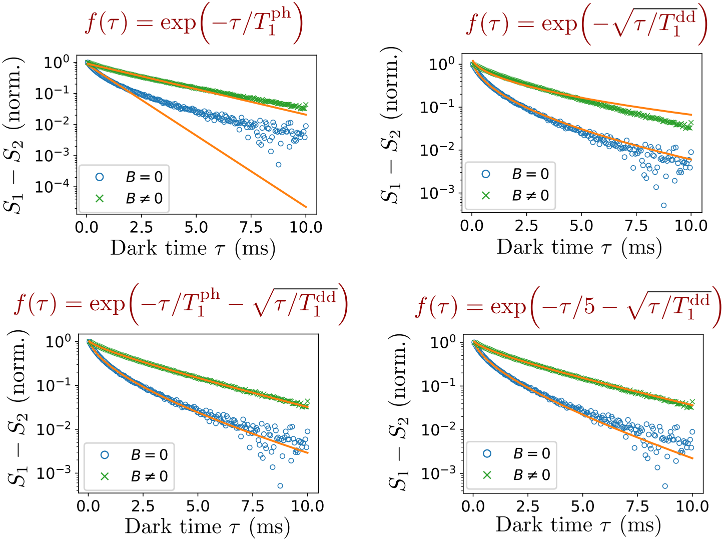




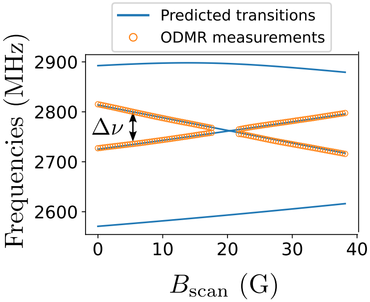
High NV density

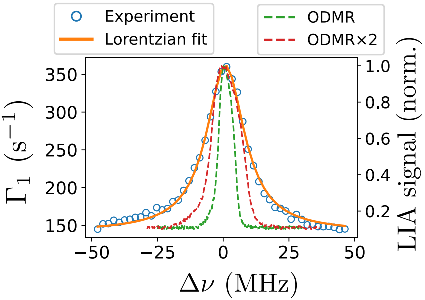
- Stretched exp. profile
- $T_1 \sim 0.5$ ms

Competition between stretched and exponential decay



Spectral response of the fluctuators

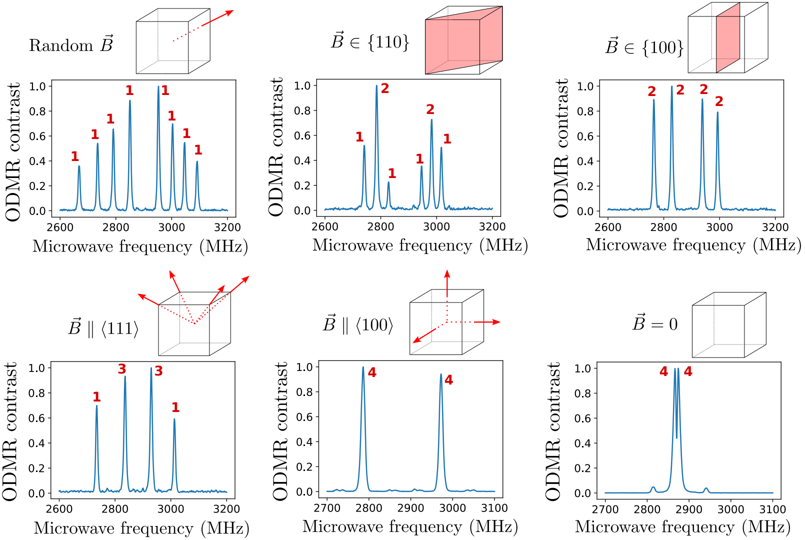




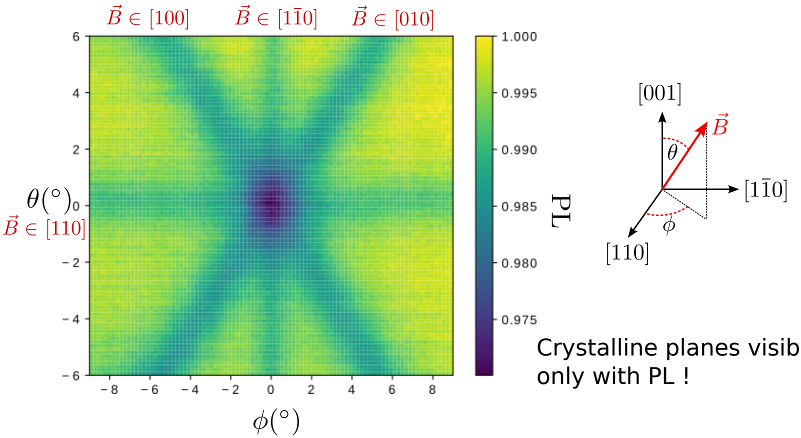
- Γ_1 curve broader than ODMR overlap
- Lorentzian shape

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

Geometry conditions for class resonances



PL mapping of the crystalline planes

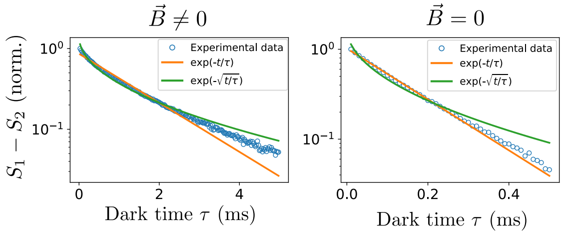


Crystalline planes visible only with PL !

Limitations of the fluctuator model

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

Overestimation of the relaxation rate



Exponential lifetime
(still dipole-dipole limited)

Improvement of the model:

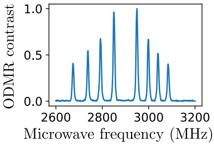
- Saturation of the fluctuators (non-Markovian)
- NV-NV spin diffusion

Outline

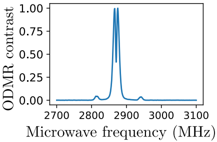
Zero field depolarization sources (theory)

$\boxed{\Gamma_1 = \Gamma_0^{\mathrm{th}}}$

Single class $B \neq 0$

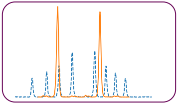


B=0

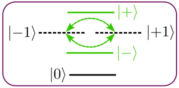


$\boxed{\Gamma_1 = 102\,\Gamma_0^{\mathrm{th}}}$

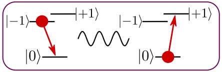
- 4-classes degeneracy
  $\Gamma_1 = \Gamma_0^{\mathrm{th}} \to 43\,\Gamma_0^{\mathrm{th}}$



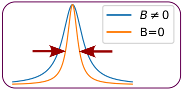
- Eigenbasis change
  $\Gamma_1 = 43\,\Gamma_0^{\mathrm{th}} \to 51\,\Gamma_0^{\mathrm{th}}$



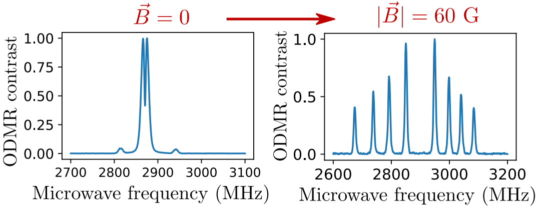
- Double flips
  $\Gamma_1 = 51\,\Gamma_0^{\mathrm{th}} \to 94\,\Gamma_0^{\mathrm{th}}$



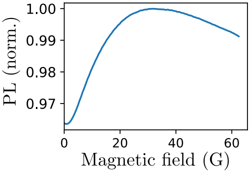
- T_2^* increase
  $\Gamma_1 = 94\,\Gamma_0^{\mathrm{th}} \to 102\,\Gamma_0^{\mathrm{th}}$

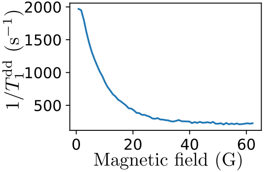


Experiment: $\vec{B}$ in arbitrary direction



- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change



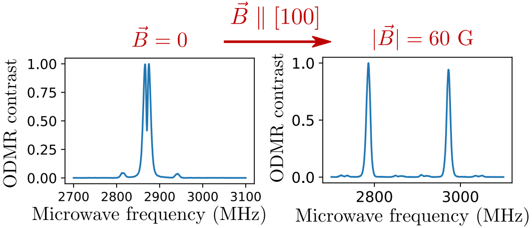


$\Gamma_1(B = 0) \approx 10\ \Gamma_1(B \neq 0)$

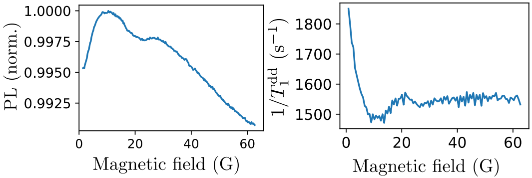
$\sim 4\%$ PL contrast

HWHM ~ 9 G

Experiment: $\vec{B} \parallel [100]$



- 4-classes degeneracy
- ~~Eigenbasis change~~
- ~~Double-flips~~
- ~~T_2^* change~~



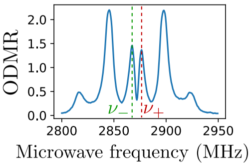
$\Gamma_1(B=0) \approx 1.2\ \Gamma_1(B \neq 0)$

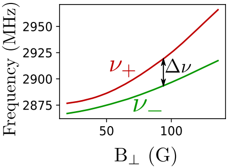
$\sim 0.5\%$ PL contrast

HWHM ~ 2 G

Classes degeneracy is the dominant cause of depolarization at low magnetic field

Experiment: $\vec{B} \perp [111]$

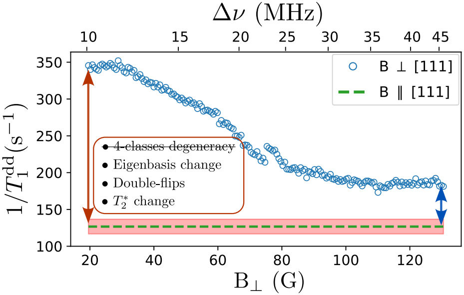




Same eigenbasis :

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

for $\vec{B} \perp [111]$ than for $\vec{B} = 0$



cancelling out double flips with transverse field

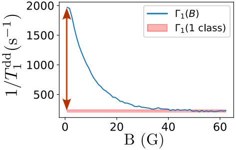
- ~~4-classes degeneracy~~
- Eigenbasis change
- ~~Double-flips~~
- T_2^* change

Double flips are the second dominant cause of depolarization at low magnetic field

Summary of the experimental observations

Random $\vec{B}$ direction

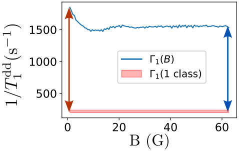
- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change



- ~~4-classes degeneracy~~
- ~~Eigenbasis change~~
- ~~Double-flips~~
- ~~T_2^* change~~

$\vec{B} \parallel [100]$

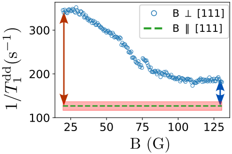
- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change



- 4-classes degeneracy
- ~~Eigenbasis change~~
- ~~Double-flips~~
- ~~T_2^* change~~

$\vec{B} \perp [111]$

- ~~4-classes degeneracy~~
- Eigenbasis change
- Double-flips
- T_2^* change

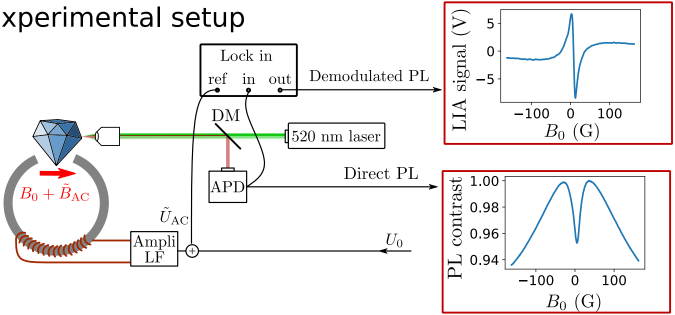


- ~~4-classes degeneracy~~
- Eigenbasis change
- ~~Double-flips~~
- T_2^* change

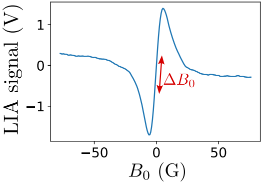
Outline

Principle of LFDM

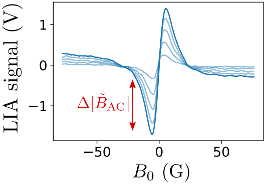
Experimental setup



DC sensing



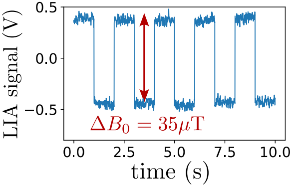
AC sensing

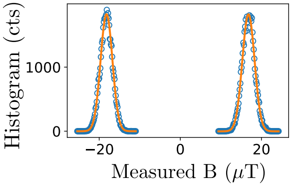


Sample: 15 μm Adamas

- $f_{\mathrm{mod}} \sim 1$ kHz
- $|B_{\mathrm{mod}}| \sim 10$ G
- $I_{\mathrm{las}} \sim 1$ mW
- PL ~ 1 μW

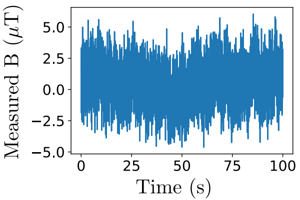
Sensitivity of LFDM

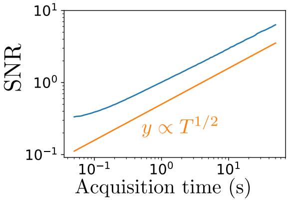




Low pass filter $\tau = 3$ ms $\sqrt{\langle \delta B^2 \rangle} \approx 1.2\ \mu\mathrm{T}$

$\rightarrow$ sensitivity $\eta = \sqrt{\frac{\langle \delta B^2 \rangle}{2\tau}} \approx 116\ \mathrm{nT}/\sqrt{\mathrm{Hz}}$





Temporal stability up to 100+ seconds without feedback

Comparison with the state of the art

Comparison with the best DC NV ensemble magnetometers

	GSLAC [1]	Microwave DC [2]	LFDM
η (nT/$\sqrt{\text{Hz}}$)	0.3^*	0.015	116
V (μm^3)	??	$5.2 \cdot 10^6$	$3.3 \cdot 10^3$
η_v (nTμm$^{3/2}$Hz$^{-1/2}$)	??	34	6700

[1] Zheng, H.[...] Budker, D. (2020). Physical Review Applied, 13(4), 044023.
[2] Barry, J. F. [...] Walsworth, R. L (2016). PNAS, 113(49), 14133-14138.

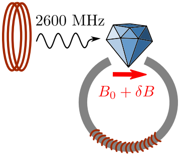
LFDM advantages

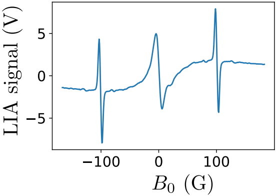
- Microwave free
- Works at low magnetic field
- Insensitive to inhomogeneous/slow noises (strain gradient, T° fluct., ...)
- Magnetic orientation insensitive: diamond powder, polycristalline

Room for improvement

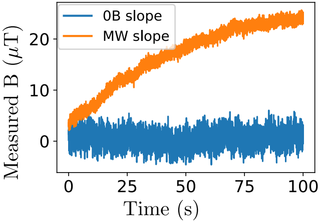
- Material optimization
- Better light collection (NA=0.65, non gaussian laser beam)
- My knowledge of metrology

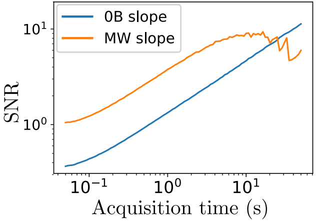
Comparison with CW ODMR





Adding a fixed microwave tone

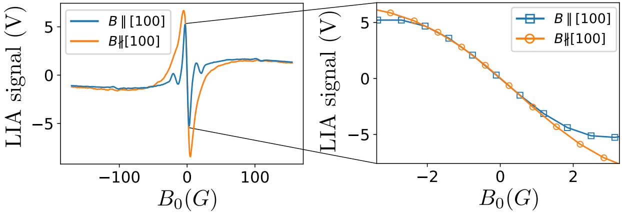




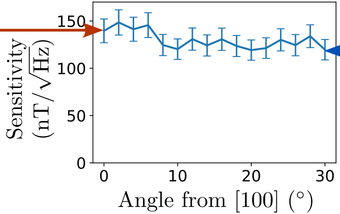
$$\text{MW slope sensitivity: } \eta \approx 40 \text{ nT}/\sqrt{\text{Hz}}$$

$$\text{B=0 sensitivity: } \eta \approx 120 \text{ nT}/\sqrt{\text{Hz}}$$

Angular sensitivity of LFDM



- ~~4-classes degeneracy~~
- Eigenbasis change
- Double-flips
- T_2^* change

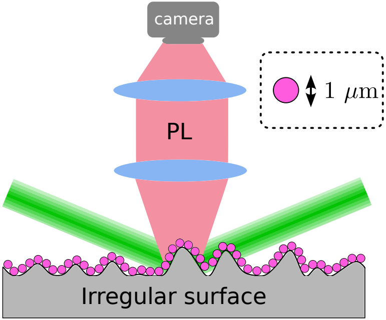


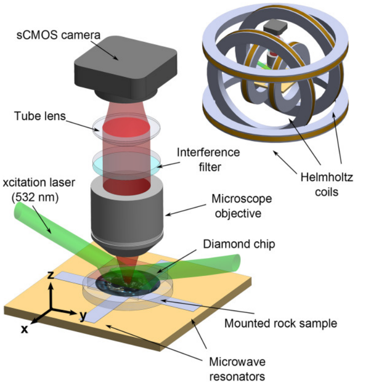
- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change

The 4-classes degeneracy is not the limiting factor of the sensitivity

Application: wide-field magnetometry on irregular surfaces

(Commercially available 1 μm diamonds)



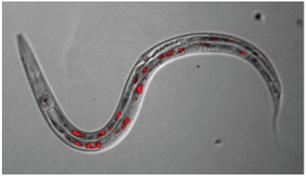


Glenn, D. R. [...] Walsworth, R. L. (2017) Geochemistry, Geophysics, Geosystems, 18(8), 3254-3267.

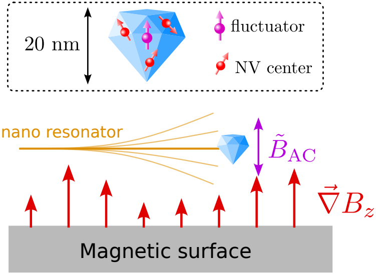
Area normalized sensitivity:
$\eta_S \approx 6\ \mu\mathrm{T} \cdot \mu\mathrm{m}/\sqrt{\mathrm{Hz}}$

Area normalized sensitivity:
$\eta_S \approx 20\ \mu\mathrm{T} \cdot \mu\mathrm{m}/\sqrt{\mathrm{Hz}}$

Other applications



- Backgrounf-free fluorescence imaging
- Real Time in vivo magnetometry



- Microwave-free nano-sensing (AC or DC)

Take home messages

- The spin depolarization in dense NV ensemble is dominated by dipole-dipole interaction with fast decaying centers (fluctuators).
- The leading cause of depolarization in zero field is the spectral overlap of the NV classes, followed by the double flips.
- Low field depolarization magnetometry achieves a sensitivity $\sim 6\ \mu\mathrm{T} \cdot \mu\mathrm{m}^{3/2}/\mathrm{Hz}^{1/2}$ with commercial samples.
- LFDM can be used with powder or polycristalline samples.