

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

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Université
de Paris



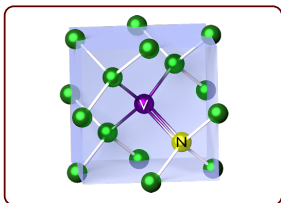
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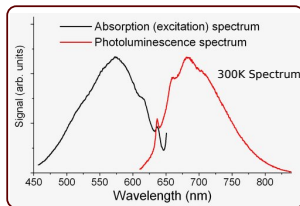
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Preamble : the NV center

Crystalline structure

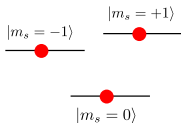


Optical properties



Spin properties

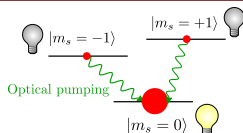
$S = 1$ in the electronic ground state



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

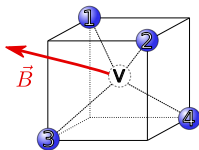


Interplay between spin and light

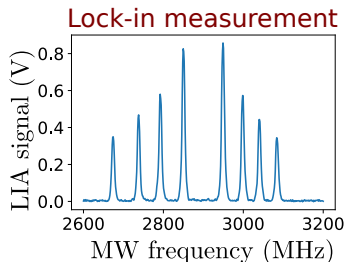
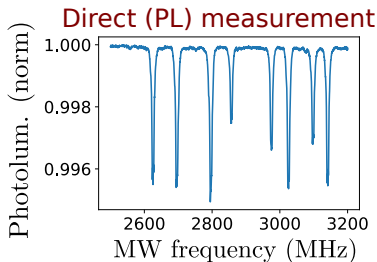
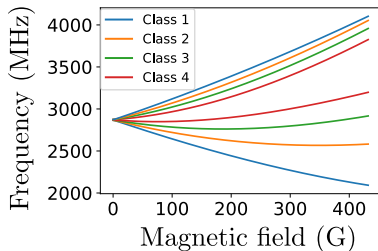


- Optical pumping in the $|m_s = 0\rangle$ state
- $|m_s = 0\rangle$ state brighter than $|m_s = \pm 1\rangle$

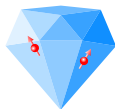
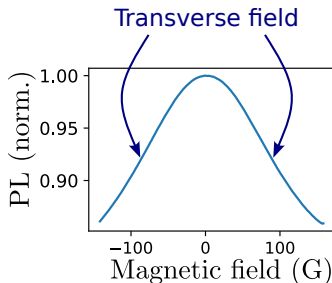
Preamble : the 4 classes of NV centers



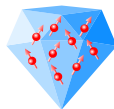
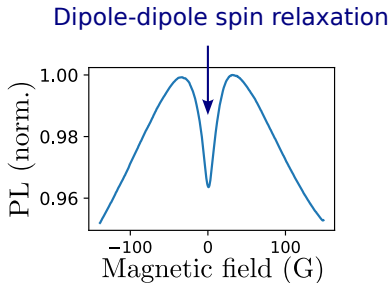
4 different projections of \vec{B}
over the 4 possible NV axes
→ 4 classes of resonances



Subject of this presentation



Low NV density
[NV] \leq 100 ppb



High NV density
[NV] \geq 1 ppm

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

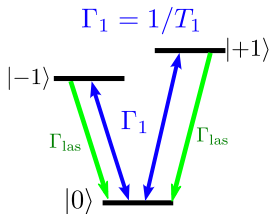
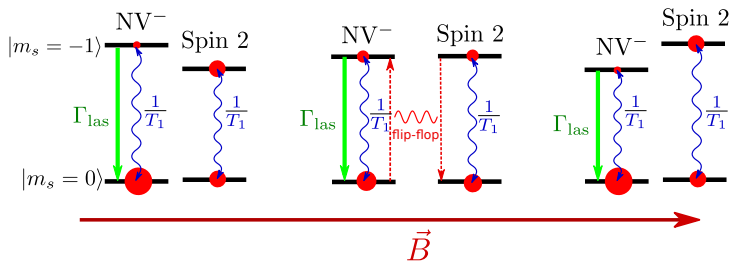
Outline

- 1 Cross-relaxation with NV centers
- 2 The NV-fluctuator model and experimental verification
- 3 Dipole-dipole interaction under low magnetic field
- 4 Low field depolarization magnetometry (LFDM)

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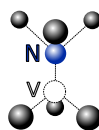
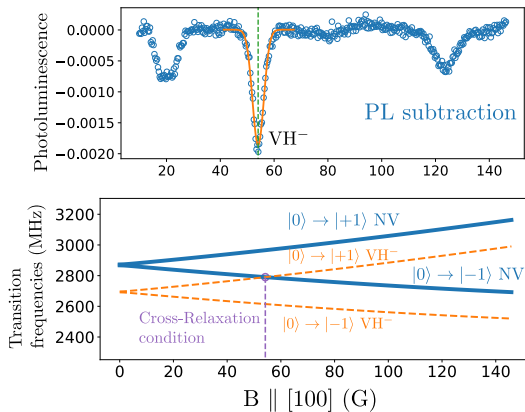
Principle of cross-relaxation with NV centers



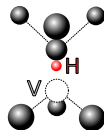
$$\text{Rate equation: } \rho_{00} = \frac{\Gamma_1 + \Gamma_{\text{las}}}{3\Gamma_1 + \Gamma_{\text{las}}}$$

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

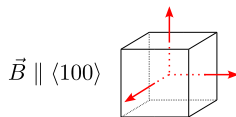
Example: Cross-relaxation between NV centers and VH^-



NV center



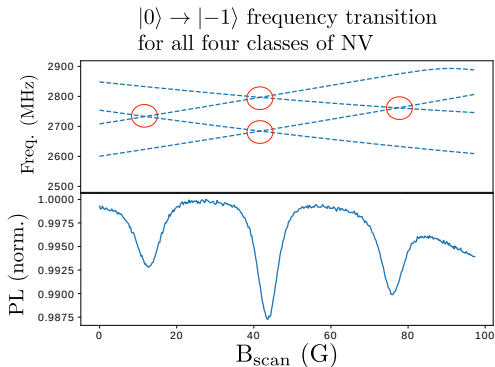
VH center



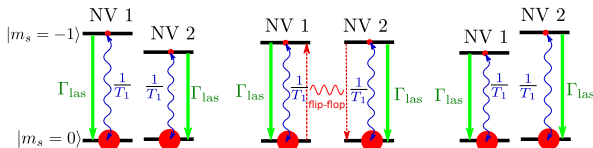
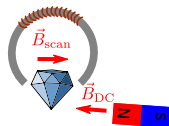
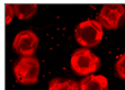
Optical detection of paramagnetic defects in diamond grown by chemical vapor deposition

C. Pellet-Mary, P. Huillery, M. Perdriat, A. Tallaïre, 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] \sim 3 PPM

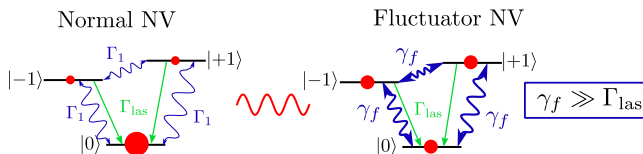


No cross-relaxation
for equally polarized
spins

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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

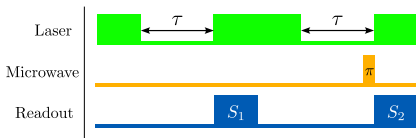
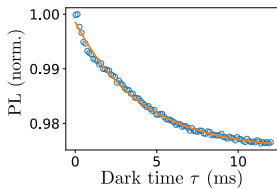
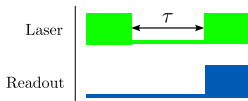
- Γ_1 increases when classes overlap spectrally (increase in the resonant fluctuator density).
- The dipole induced depolarization has a stretched exponential profile:

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

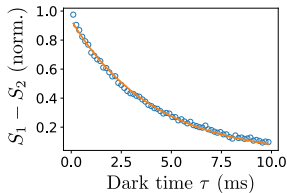
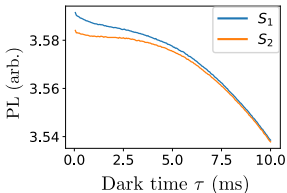
- The Fluctuators spectral response is broadened by their decay rate γ_f (lifetime limit).

T_1 measurement protocol

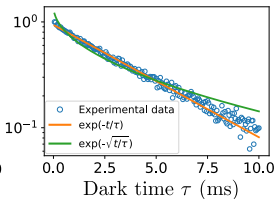
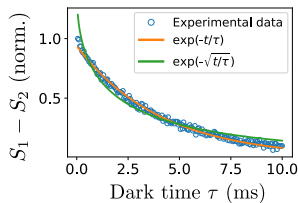
Basic T_1 protocol



common mode rejection method

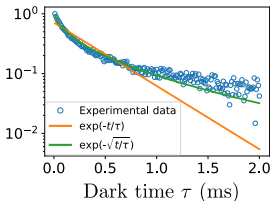
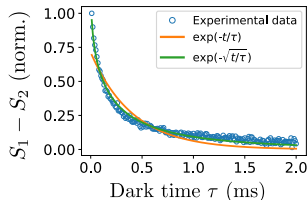


Stretched exponential decay profile



Low NV density

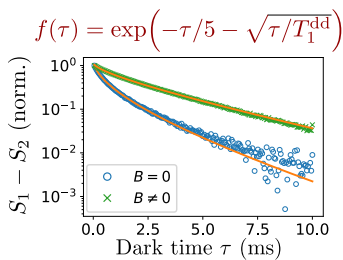
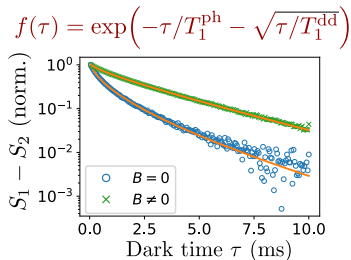
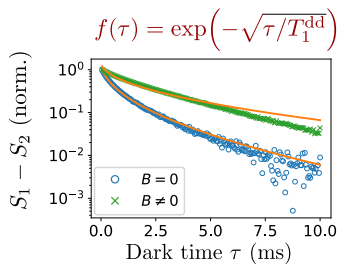
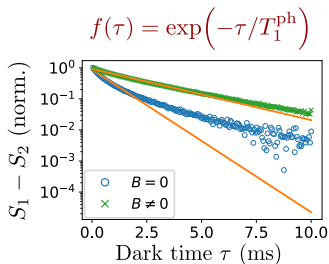
- Exponential profile
- $T_1 \sim 5$ ms



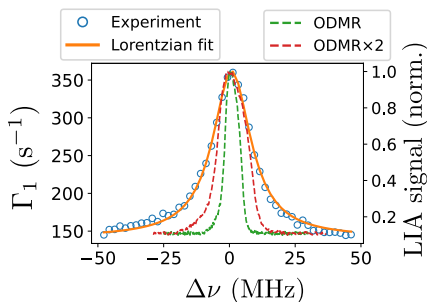
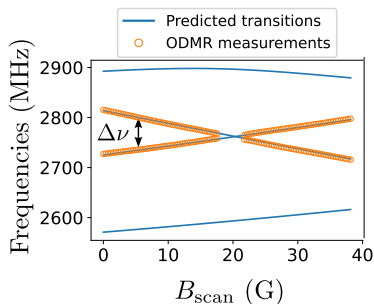
High NV density

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

Competition between stretched and exponential decay



Spectral response of the fluctuators

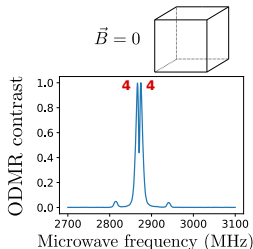
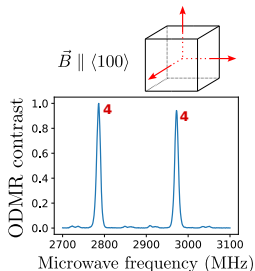
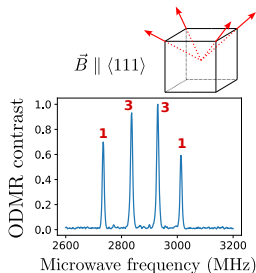
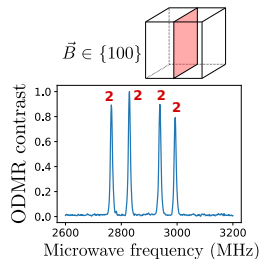
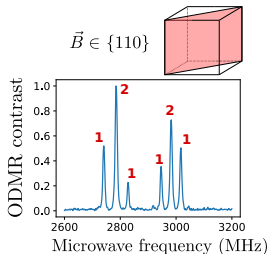
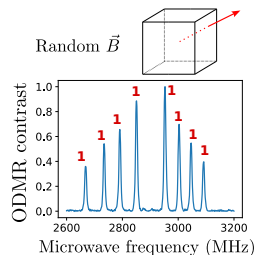


- Γ_1 curve broader than ODMR overlap
- Lorentzian shape

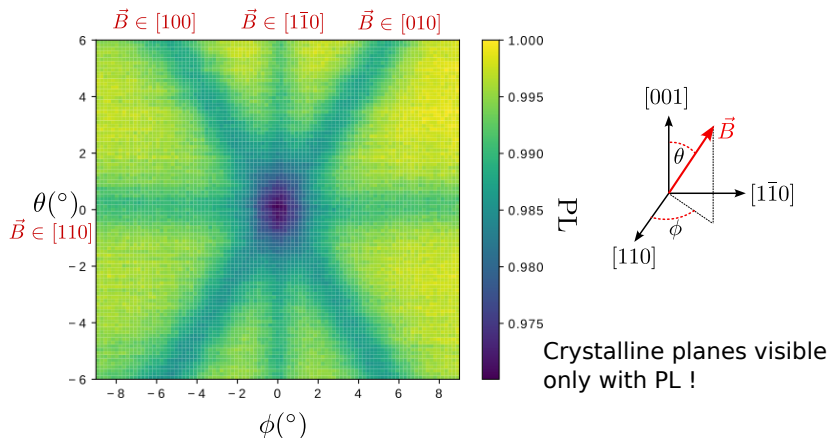


The fluctuator's spectral response (T_2^*) is broadened by γ_f

Geometry conditions for class resonances



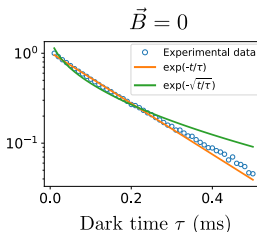
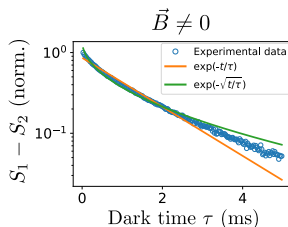
PL mapping of the crystalline planes



Limitations of the fluctuator model

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

Overestimation of
the relaxation rate



Exponential lifetime
(still dipole-dipole limited)

Improvement of the model:

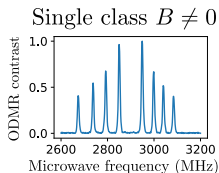
- Saturation of the fluctuators (non-Markovian)
- NV-NV spin diffusion

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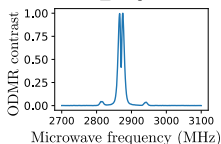
Zero field depolarization sources (theory)

$$\Gamma_1 = \Gamma_0^{\text{th}}$$



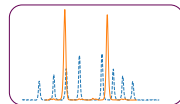
$B=0$

$$\Gamma_1 = 102 \Gamma_0^{\text{th}}$$



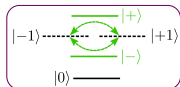
- 4-classes degeneracy

$$\Gamma_1 = \Gamma_0^{\text{th}} \rightarrow 43 \Gamma_0^{\text{th}}$$



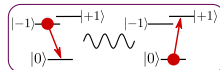
- Eigenbasis change

$$\Gamma_1 = 43 \Gamma_0^{\text{th}} \rightarrow 51 \Gamma_0^{\text{th}}$$



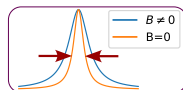
- Double flips

$$\Gamma_1 = 51 \Gamma_0^{\text{th}} \rightarrow 94 \Gamma_0^{\text{th}}$$

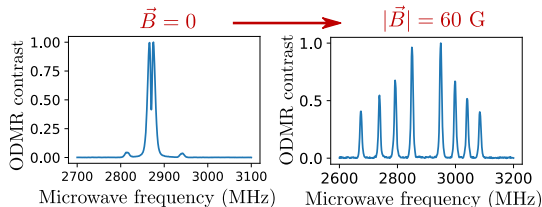


- T_2^* increase

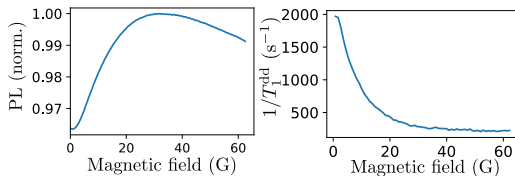
$$\Gamma_1 = 94 \Gamma_0^{\text{th}} \rightarrow 102 \Gamma_0^{\text{th}}$$



Experiment: \vec{B} in arbitrary direction

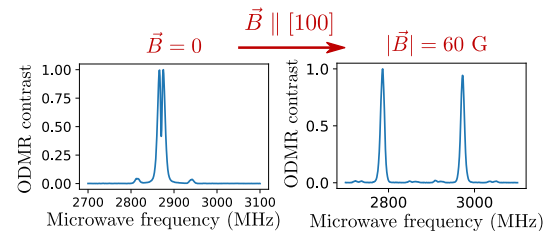


- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change

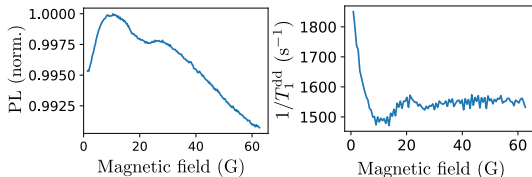


$\Gamma_1(B=0) \approx 10 \Gamma_1(B \neq 0)$
 $\sim 4\%$ PL contrast
HWHM $\sim 9 \text{ G}$

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



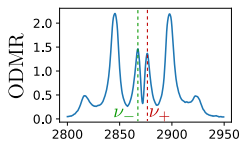
- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change



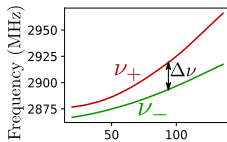
$\Gamma_1(B=0) \approx 1.2 \Gamma_1(B \neq 0)$
 $\sim 0.5\%$ PL contrast
HWHM $\sim 2 \text{ G}$

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

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



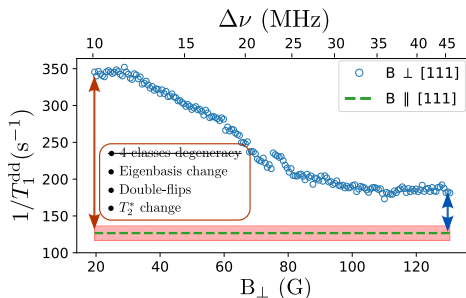
Microwave frequency (MHz)



B_{\perp} (G)

Same eigenbasis :

$$|\pm\rangle = \frac{|+1\rangle \pm |-1\rangle}{\sqrt{2}}$$
 for $\vec{B} \perp [111]$ than for $\vec{B} = 0$



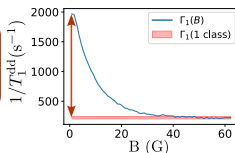
cancelling out double flips
with transverse field

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

Summary of the experimental observations

Random \vec{B} direction

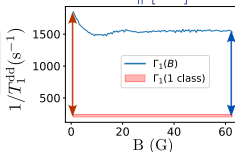
- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change



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

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

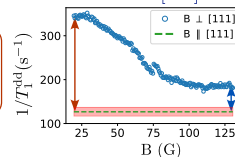
- 4-classes degeneracy
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- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change

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

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



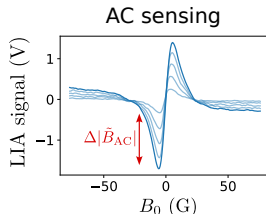
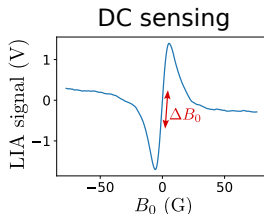
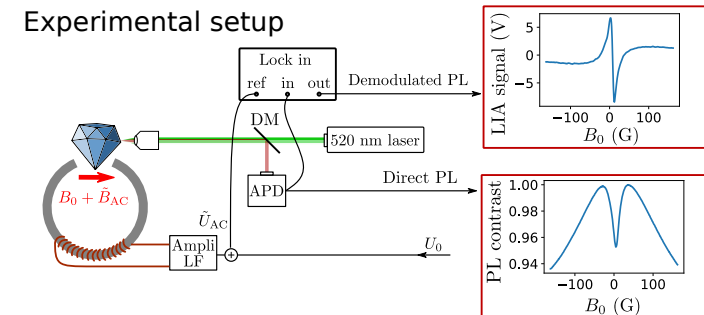
- 4-classes degeneracy
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Principle of LFDM

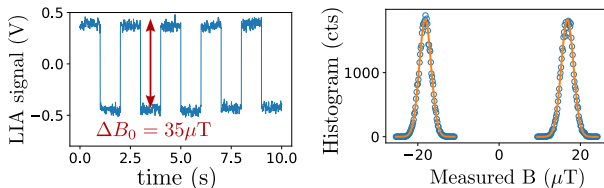
Experimental setup



Sample: 15 μm Adamas

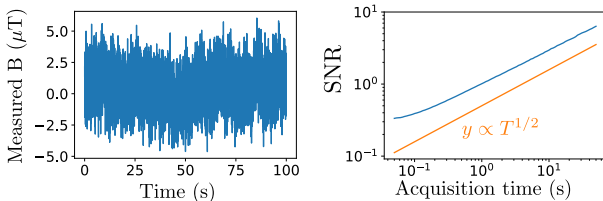
- $f_{\text{mod}} \sim 1 \text{ kHz}$
- $|B_{\text{mod}}| \sim 10 \text{ G}$
- $I_{\text{las}} \sim 1 \text{ mW}$
- $\text{PL} \sim 1 \mu\text{W}$

Sensitivity of LFDM



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

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



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 (nT $\mu\text{m}^{3/2}\text{Hz}^{-1/2}$)	??	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.

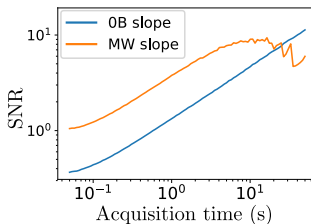
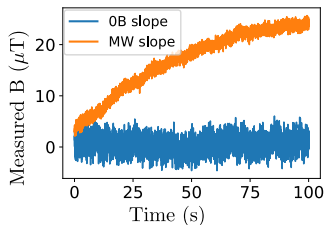
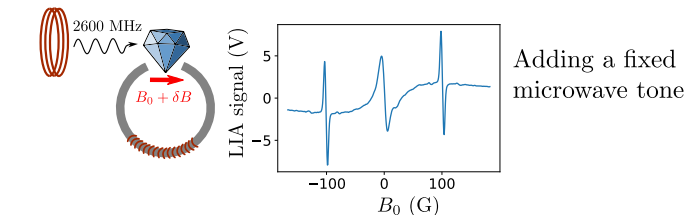
LFDM advantages

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

Room for improvement

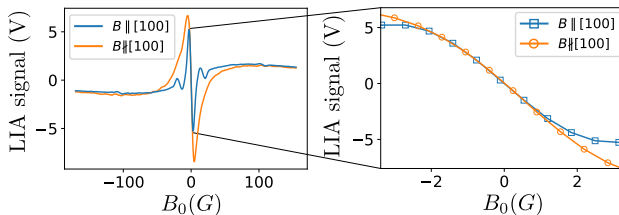
- Material optimization
- Better light collection (NA=0.65, non gaussian laser beam)
- My knowledge of metrology

Comparison with CW ODMR

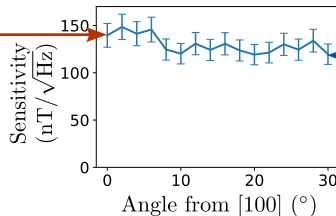


MW slope sensitivity: $\eta \approx 40 \text{ nT}/\sqrt{\text{Hz}}$
B=0 sensitivity: $\eta \approx 120 \text{ nT}/\sqrt{\text{Hz}}$

Angular sensitivity of LFDM



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

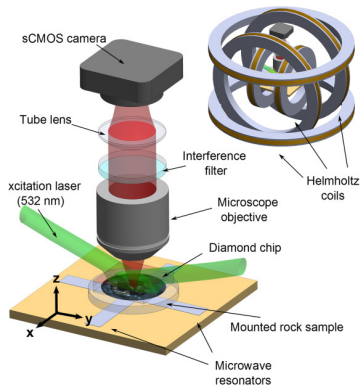
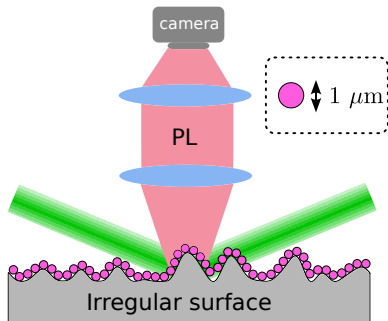


- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change

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

Application: wide-field magnetometry on irregular surfaces

(Commercially available $1\ \mu\text{m}$ diamonds)



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

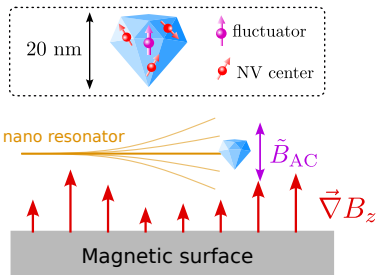
Area normalized sensitivity:
 $\eta_S \approx 6\ \mu\text{T} \cdot \mu\text{m} / \sqrt{\text{Hz}}$

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

Other applications



- Background-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 $\sim 6 \mu\text{T} \cdot \mu\text{m}^{3/2}/\text{Hz}^{1/2}$ with commercial samples.
- LFDM can be used with powder or polycrystalline samples.