Cross-relaxation in dense ensembles of NV centers and application to magnetometry

Clément Pellet-Mary

PhD Defense

Outline

Outline

Quantum sensing and metrology

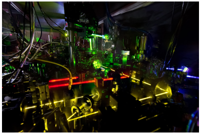
Quantum metrology:

$\boxed{\text{Using quantum}^{*} \text{ properties to create more sensitive measurement protocols.}}$

* quantum $\equiv$ discrete energy levels

Example :

Time measurement



Atomic clock

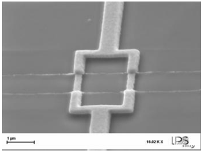
Gravitational wave detection

LIGO

Medical imaging

MRI

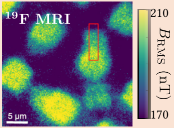
Magnetometry



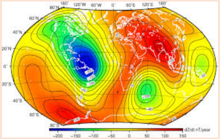
SQUIDs

Magnetometry applications

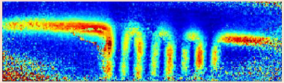
Chemical analysis



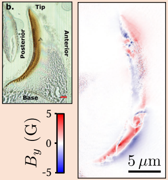
Geomagnetism



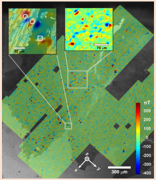
Circuit imaging



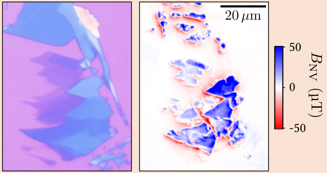
Biomagnetism



Paleomagnetism

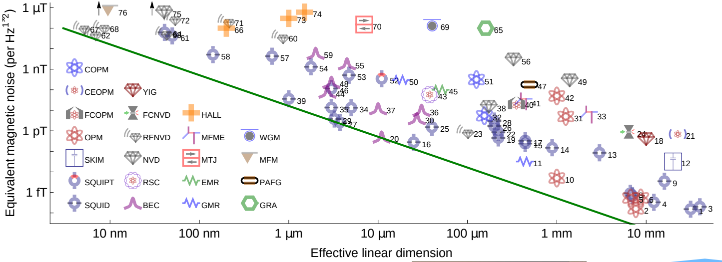


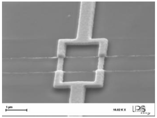
Solid state magnetism



Quantum magnetometers

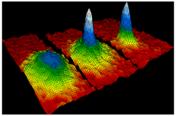
Mitchell, M. W., & Alvarez, S. P. *Reviews of Modern Physics*, 92(2), 021001.





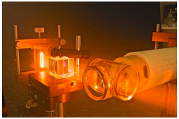
SQUID

✗ Cryogenic temperature



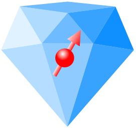
BEC

✗ Ultra high vacuum



SERF / OPM

✗ Large working distance



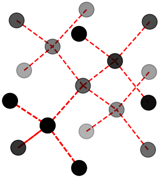
NV center

✗ Hard to scale

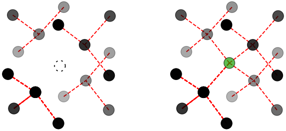
Outline

Colored centers in diamond

Diamond crystal lattice

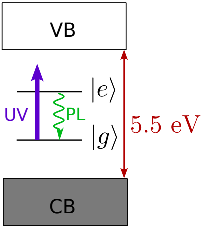


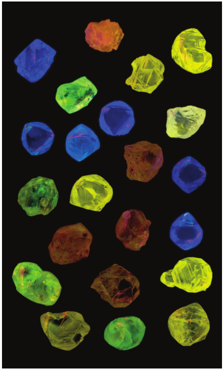
Point-like defects



Vacancy Substitution

Colored center fluorescence

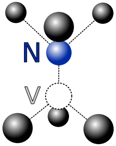




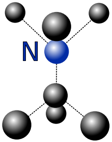
Natural diamonds fluorescence under UV light

Synthetic diamond and NV centers

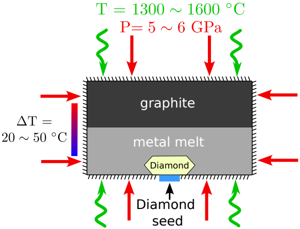
NV center

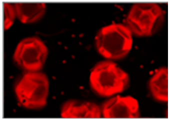


N_s (P1 center)



Highe Pressure High Temperature (HPHT)



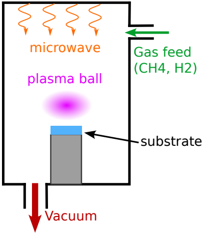


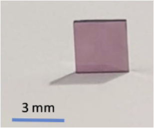
Adamas 15/150 μm

$[N_s] \approx 100$ ppm

$[NV] \approx 3$ ppm

Chemical Vapour Deposition (CVD)



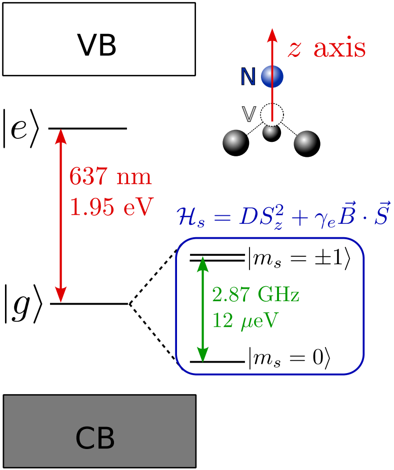


IRCP-LSPM

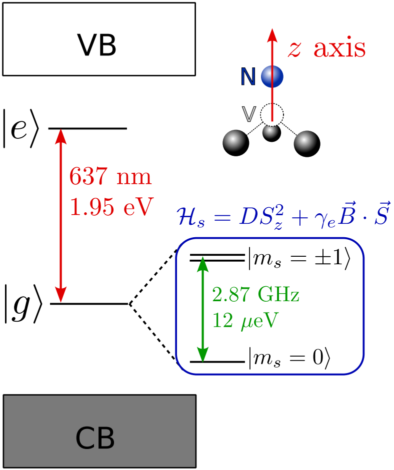
$[N_s] \approx 25$ ppm

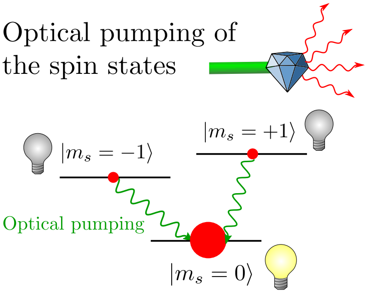
$[NV] \approx 4.5$ ppm

The NV center energy levels



The NV center energy levels

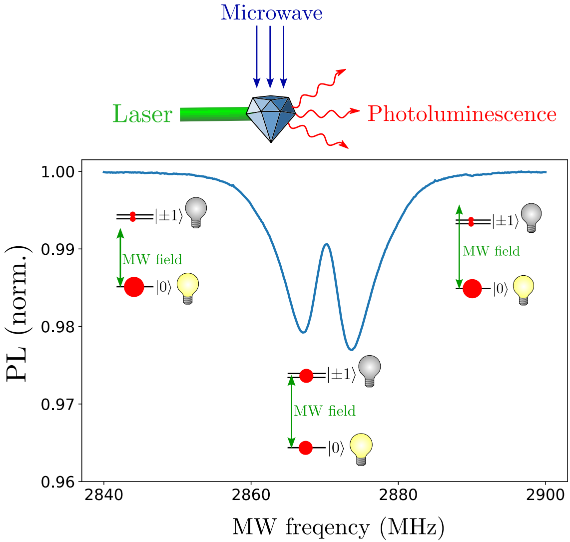




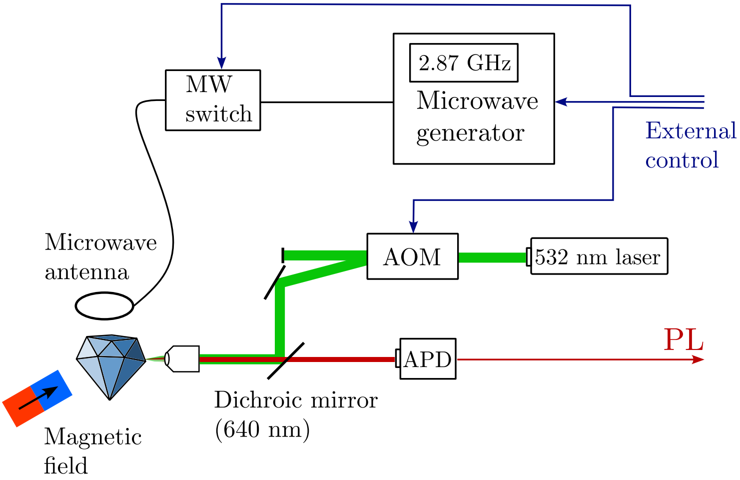
- Population accumulation in the $|0\rangle$ state
  ↳ Initialization of the spin state

- $|0\rangle$ sate brighter than $|\pm 1\rangle$ states
  ↳ Optical readout of the spin state

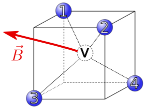
Optically detected magnetic resonance (ODMR)



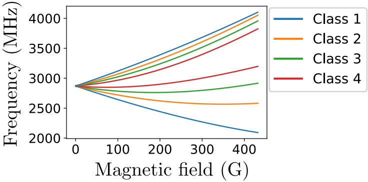
Experimental setup

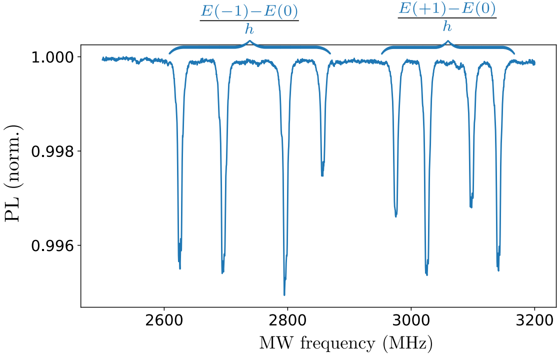


ODMR with NV ensemble



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



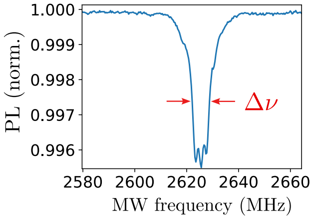


Position of the 8 lines:
$\rightarrow$ 3D reconstruction of $\vec{B}$

Sensitivity limit for NV ensemble magnetometry

Ideal (DC) sensitivity for
N independent NV centers:

$$\boxed{\eta[\mathrm{T}/\sqrt{\mathrm{Hz}}] \approx \frac{\hbar\sqrt{\Delta\nu}}{\mathrm{g}\mu_\mathrm{B}\mathrm{C}\sqrt{\mathrm{N}}}}$$



- $\hbar$: Planck constant
- μ_B : Bohr magneton
- g : NV electron Landé factor

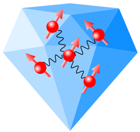
} Constants

- C : Spin readout contrast

} Experimental parameters

- N : Number of NV centers
- $\Delta\nu = \frac{1}{T_2^*}$: Spectral linewidth

} Sample parameters

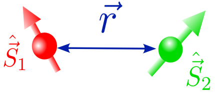


Increased NV density:
$\rightarrow$ Interactions

Dipole-dipole interaction between two electronic spins

Magnetic dipole-dipole Hamiltonian:

$$\boxed{\mathcal{H}_{\mathrm{dd}} = \frac{J_0}{r^3}\left[3(\hat{\vec{S}}_1 \cdot \vec{u})(\hat{\vec{S}}_2 \cdot \vec{u}) - \hat{\vec{S}}_1 \cdot \hat{\vec{S}}_2\right]}$$



$J_0/\hbar \approx 51.8\ \mathrm{MHz} \cdot \mathrm{nm}^3$

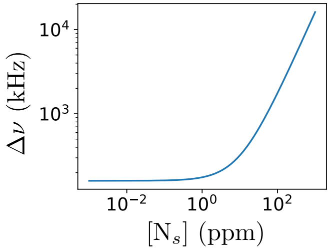
$$\begin{array}{c c}
 & \begin{array}{ccccc} |-1;-1\rangle & |-1;0\rangle & |-1;+1\rangle & |0;-1\rangle & \cdots \end{array} \\
\begin{array}{c} \langle -1;-1| \\ \langle -1;0| \\ \langle -1;+1| \\ \langle 0;-1| \\ \vdots \end{array} &
\begin{pmatrix} J_{zz} & 0 & 0 & 0 & \\ 0 & 0 & 0 & J_{xx}^* & \cdots \\ 0 & 0 & -J_{zz} & 0 & \\ 0 & J_{xx} & 0 & 0 & \\ & \vdots & & & \ddots \end{pmatrix}
\end{array}$$

: Energy shift
→ Spectral broadening

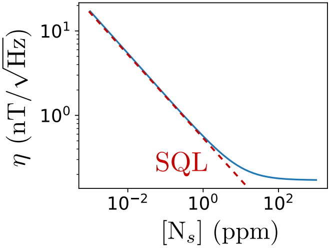
: Spin exchange
→ Flip-flop, relaxation

Interaction limit to NV ensemble magnetometry

Spectral broadening



Projected sensitivity ($C = 10^{-3}$, $V = 1\ \mu\mathrm{m}^3$, $\varepsilon(\mathrm{N}_s \to \mathrm{NV}) \approx 10\%$)

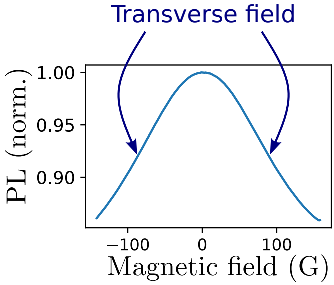


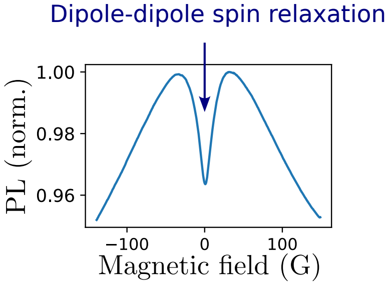
Going beyond the "Interaction limit" ($[\mathrm{N}_s] > 10$ ppm):

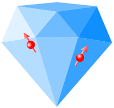
- Decoupling interaction (Hamiltonian engineering)
- Exploiting interactions

Outline

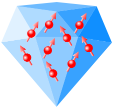
Depolarization of dense NV ensemble at low magnetic field







Low NV density
[NV] ≤ 100 ppb

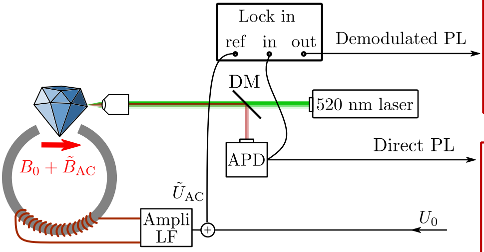


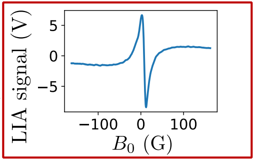
High NV density
[NV] ≥ 1 ppm

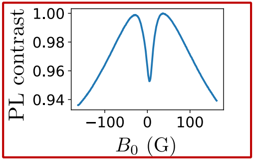
- Better understand the dipole-dipole interaction in dense NV ensembles
- Exploit the PL feature for magnetometry

Pesentation of low field depolarization magnetometry (LFDM)

Experimental setup

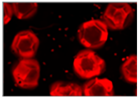






Samples

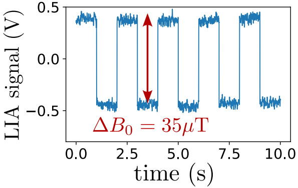
Adamas 1/15/150 µm
fluorescent microdiamond
[N] = 100~200 PPM
[NV] ~ 3 PPM

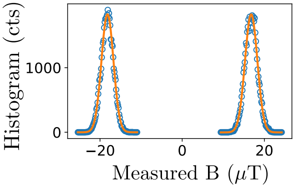


Experimental parameters

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

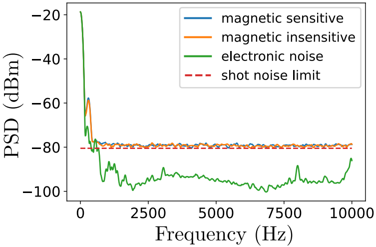
Sensitivity of LFDM





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

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



Shot noise limited for $f_{\mathrm{mod}} > 500$ Hz

Comparison with the state of the art

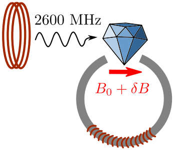
Sensitivity comparison

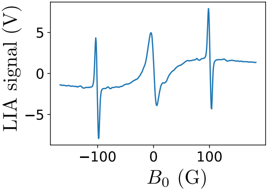
	GSLAC [1]	ODMR [2]	LFDM
η (nT/$\sqrt{\text{Hz}}$)	0.3*	0.015	116
V (μm^3)	??	$5.2 \cdot 10^6$	$3.3 \cdot 10^3$
η_v (nTμm$^{3/2}$Hz$^{-1/2}$)	??	34	6700

[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.

	ODMR	GSLAC	LFDM
Microwave free	✗	✓	✓
Low magnetic field (<10 G)	✓	✗	✓
Robust to T° and B-field inhomogeneities	✗	✗	✓
Orientation free (polycrystalline, powder)	✗	✗	✓

Comparison with CW ODMR



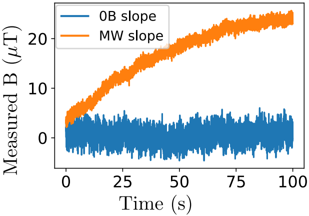


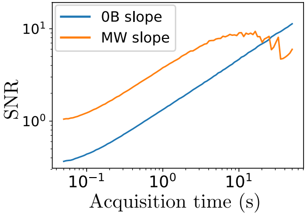
Adding a fixed microwave tone

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

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

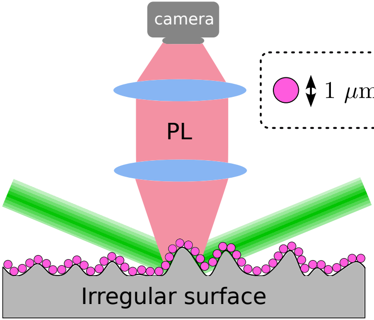
Temporal stability

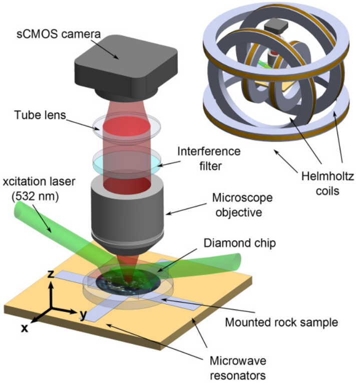




Application: wide-field magnetometry on irregular surfaces

(Commercially available 1 μm diamonds)





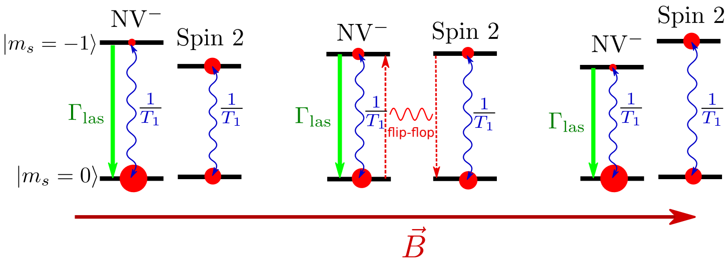
Glenn, D. R. [...] Walsworth, R. L. (2017)
Geochemistry, Geophysics, Geosystems, 18(8), 3254-3267.

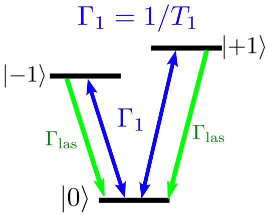
Area normalized sensitivity:
$\eta_S \approx 6\ \mu\mathrm{T} \cdot \mu\mathrm{m}/\sqrt{\mathrm{Hz}}$

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

Outline

Principle of cross-relaxation with NV centers

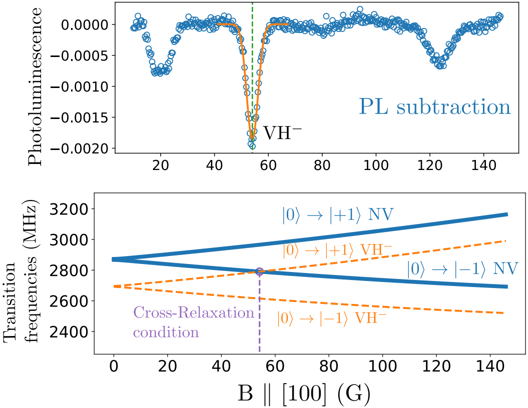


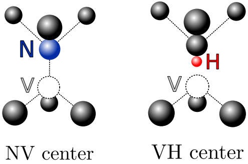


Rate equation: $\rho_{00} = \frac{\Gamma_1+\Gamma_{las}}{3\Gamma_1+\Gamma_{las}}$

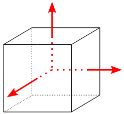
$$\Gamma_1 \nearrow \Rightarrow PL \searrow$$

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





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

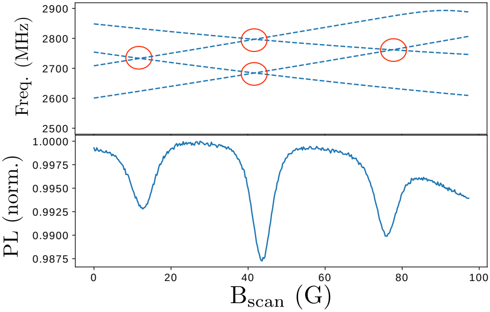


Optical detection of paramagnetic defects in diamond grown by chemical vapor deposition

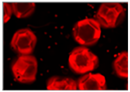
C. Pellet-Mary, P. Huillery, M. Perdriat, A. Tallaire, and G. Hétet
Phys. Rev. B **103**, L100411 – Published 24 March 2021

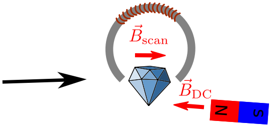
Cross-relaxation between NV centers and NV centers

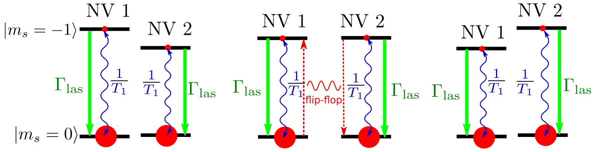
$|0\rangle \rightarrow |-1\rangle$ frequency transition
for all four classes of NV



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

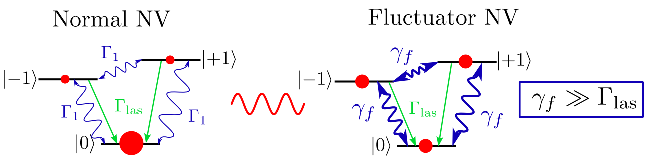






No cross-relaxation for equally polarized spins

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

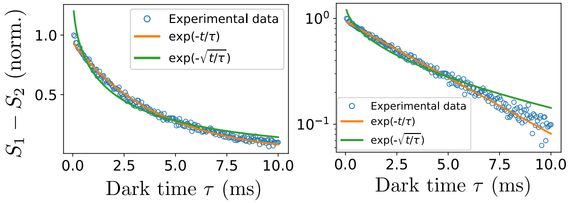
Possible microscopic explanation:

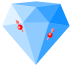
- charge tunneling
- modulation of J-coupling

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

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

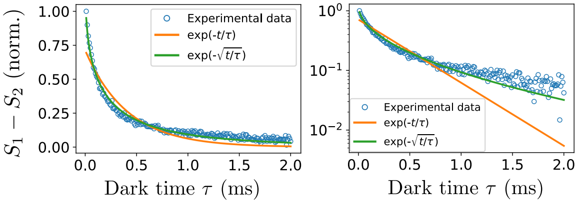
Stretched exponential decay profile

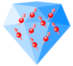




Low NV density

- Exponential profile
- $T_1 \sim 5$ ms





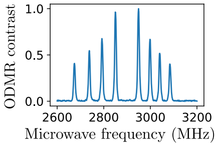
High NV density

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

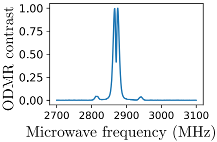
Zero field depolarization sources (theory)

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

Single class $B \neq 0$

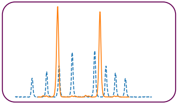


B=0

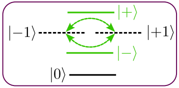


$\boxed{\Gamma_1 = 102\,\Gamma_0^{th}}$

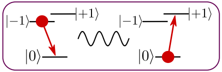
- 4-classes degeneracy
  $\Gamma_1 = \Gamma_0^{th} \to 43\,\Gamma_0^{th}$



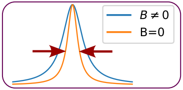
- Eigenbasis change
  $\Gamma_1 = 43\,\Gamma_0^{th} \to 51\,\Gamma_0^{th}$



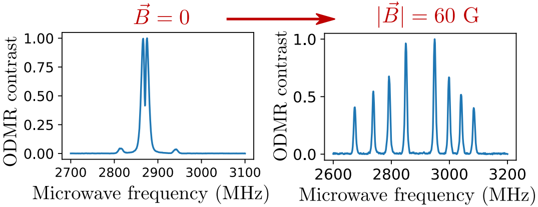
- Double flips
  $\Gamma_1 = 51\,\Gamma_0^{th} \to 94\,\Gamma_0^{th}$



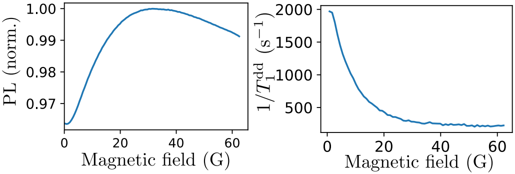
- T_2^* increase
  $\Gamma_1 = 94\,\Gamma_0^{th} \to 102\,\Gamma_0^{th}$



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



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

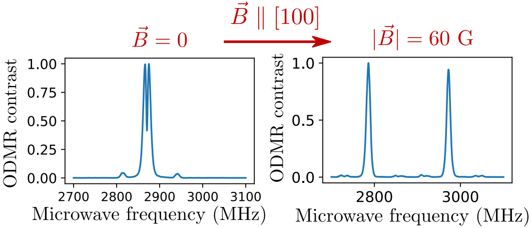


$\Gamma_1(B=0) \approx 10\ \Gamma_1(B \neq 0)$

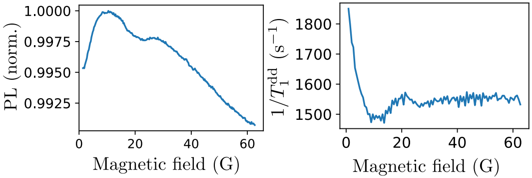
$\sim$ 4% PL contrast

HWHM $\sim$ 9 G

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



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



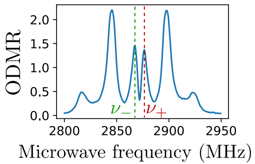
$\Gamma_1(B=0) \approx 1.2\ \Gamma_1(B \neq 0)$

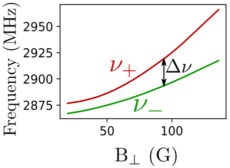
$\sim 0.5\%$ PL contrast

HWHM ~ 2 G

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

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

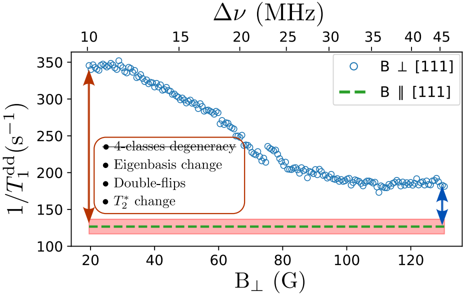




Same eigenbasis :

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

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



cancelling out double flips with transverse field

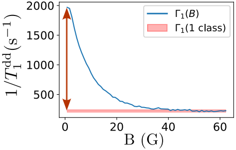
- ~~4-classes degeneracy~~
- Eigenbasis change
- ~~Double-flips~~
- T_2^* change

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

Summary of the experimental observations

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

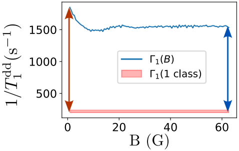
Random $\vec{B}$ direction



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

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

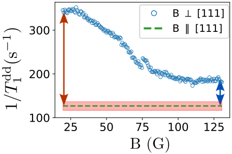
$\vec{B} \parallel [100]$



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

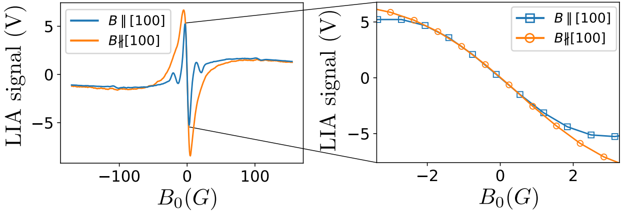
- ~~4-classes degeneracy~~
- Eigenbasis change
- Double-flips
- T_2^* change

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

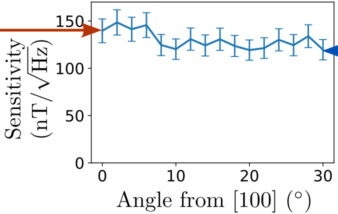


- ~~4-classes degeneracy~~
- Eigenbasis change
- ~~Double-flips~~
- T_2^* change

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

Conclusion

Acknowledgments