

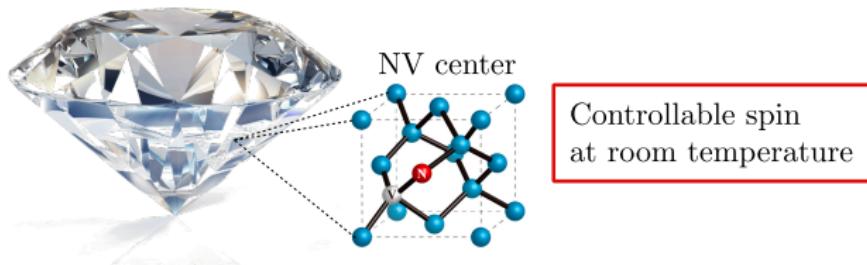
Cross-relaxation in dense ensembles of NV centers and application to magnetometry

Clément Pellet-Mary

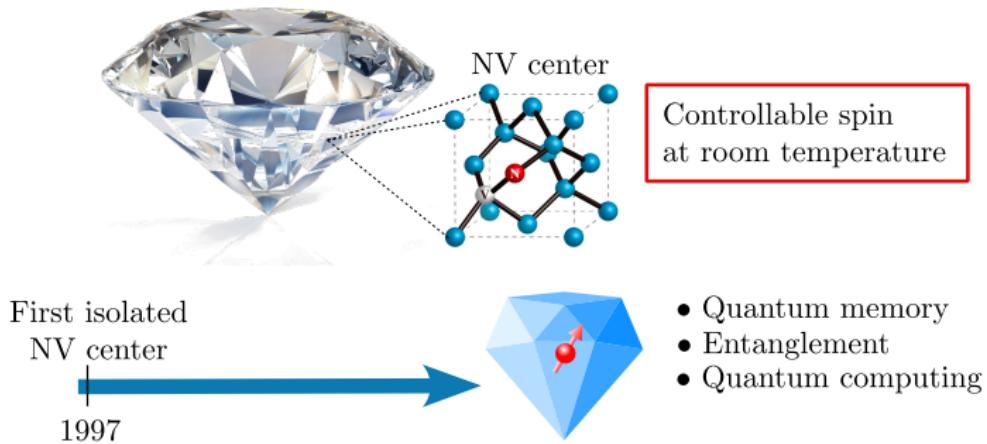
PhD Defense



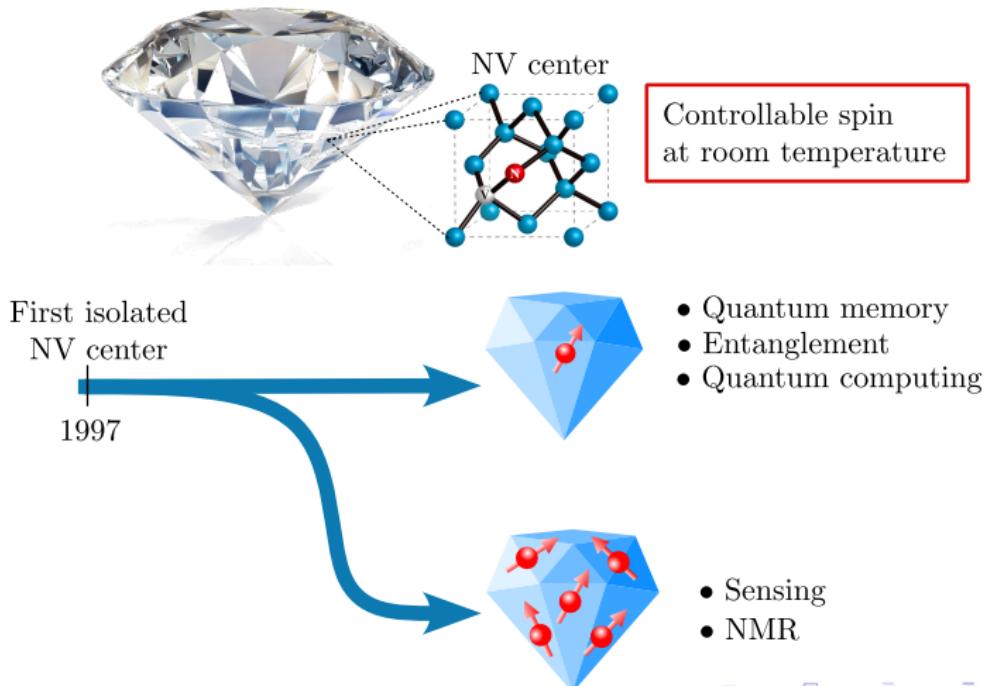
Context of my PhD



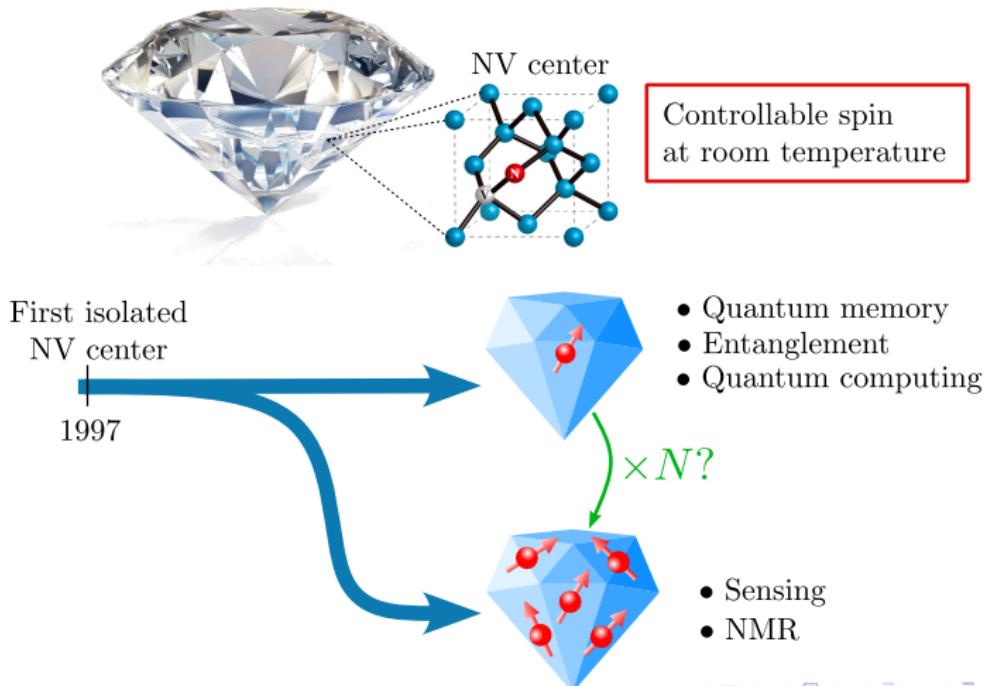
Context of my PhD



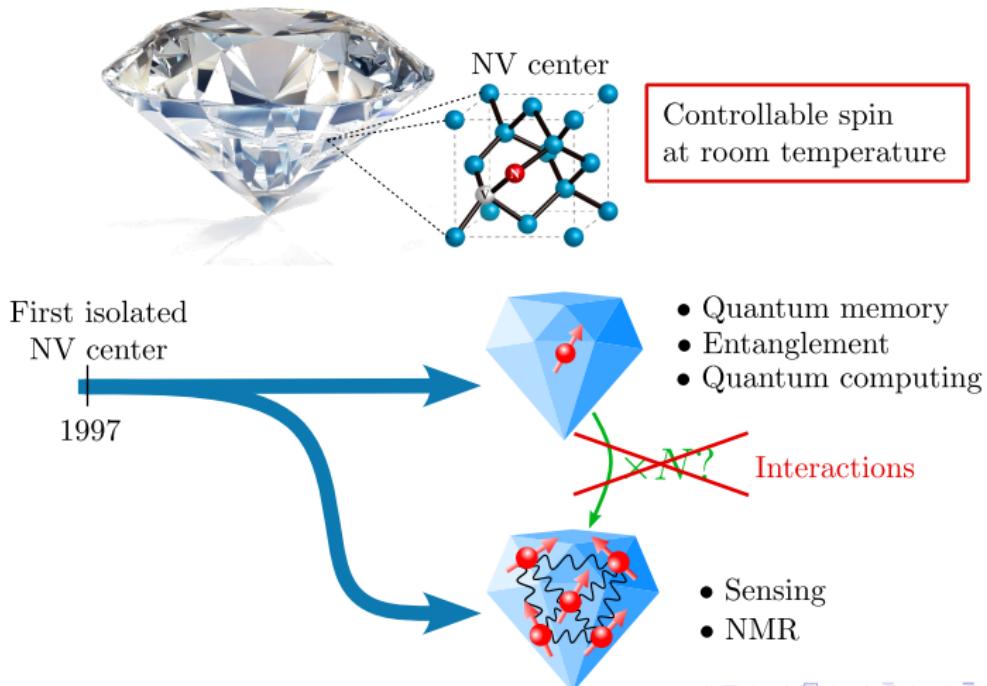
Context of my PhD



Context of my PhD



Context of my PhD



Sensing with quantum mechanics

NV centers and diamonds in practice

Low field depolarization magnetometry (LFDM)

Depolarization mechanisms in dense NV ensemble

Bonus slides

Outline

Sensing with quantum mechanics

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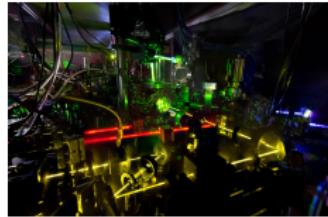
Quantum sensing and metrology

Quantum metrology:

Using quantum* properties to create
more sensitive measurement protocols.

* quantum \equiv discrete energy levels

Time measurement



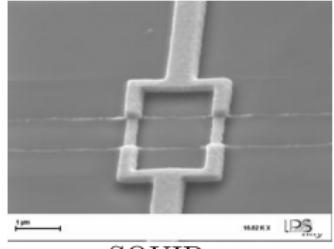
Atomic clock

Medical imaging



MRI

Magnetometry

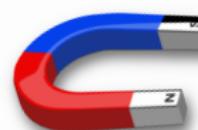
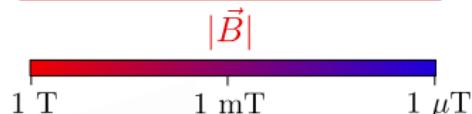


SQUIDs

Key properties of magnetometers

Sensitivity [T/ $\sqrt{\text{Hz}}$]:

Minimum magnetic field value detectable with a signal-to-noise ratio of 1 within 1 second.



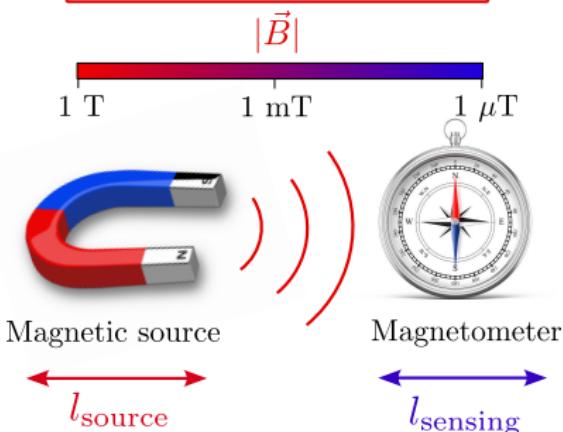
Magnetic source

Magnetometer

Key properties of magnetometers

Sensitivity [T/ $\sqrt{\text{Hz}}$]:

Minimum magnetic field value detectable with a signal-to-noise ratio of 1 within 1 second.



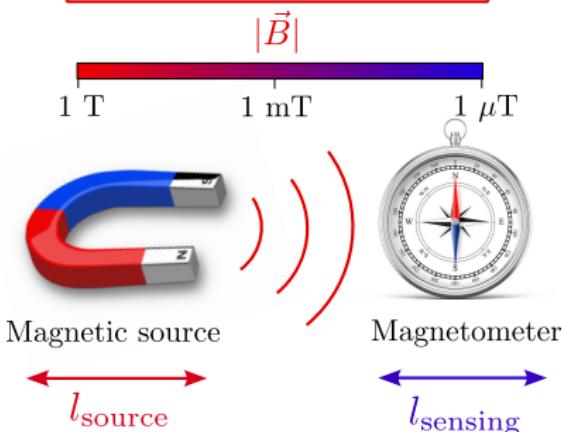
Optimum spatial resolution (imaging):

$$l_{\text{sensing}} \leq l_{\text{source}}$$

Key properties of magnetometers

Sensitivity [T/ $\sqrt{\text{Hz}}$]:

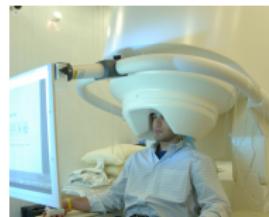
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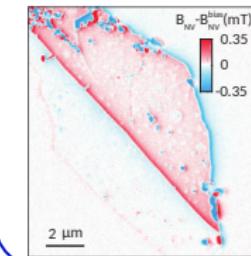
Magnetoencephalography



$$|\vec{B}| \sim 10 \text{ fT}$$

$$l \sim 1 \text{ cm}$$

2D materials magnetism

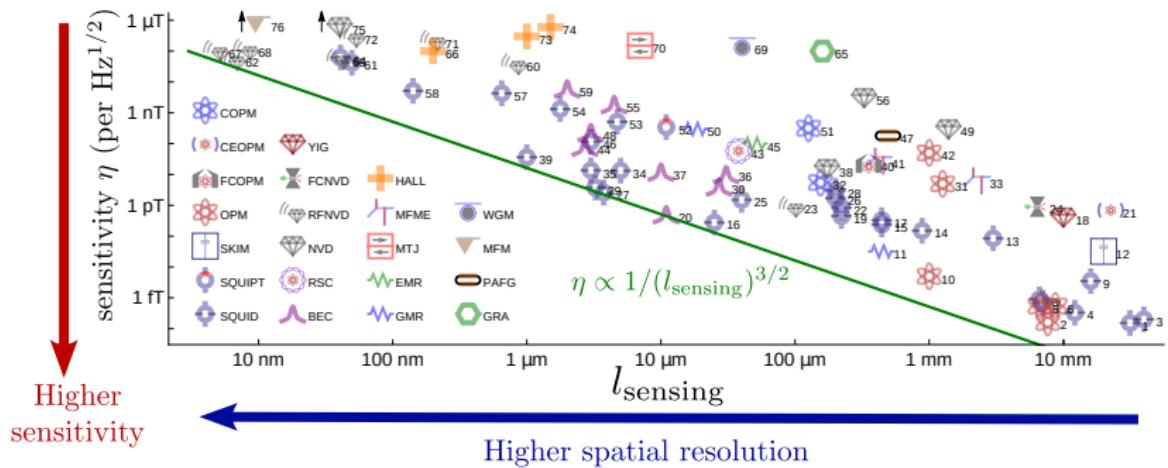


$$|\vec{B}| \sim 100 \mu\text{T}$$

$$l \sim 50 \text{ nm}$$

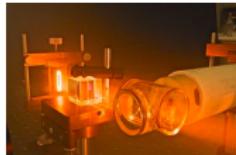
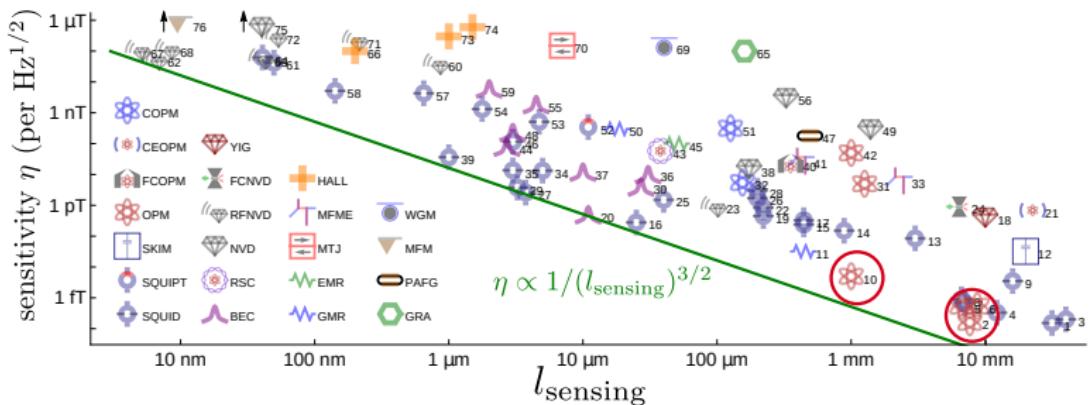
Thiel, L. et al (2019). Science, 364(6444), 973-976.

Sate of the art magnetometers



Mitchell, M. W., & Alvarez, S. P. (2020). Reviews of Modern Physics, 92(2), 021001

Sate of the art magnetometers

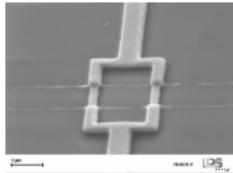
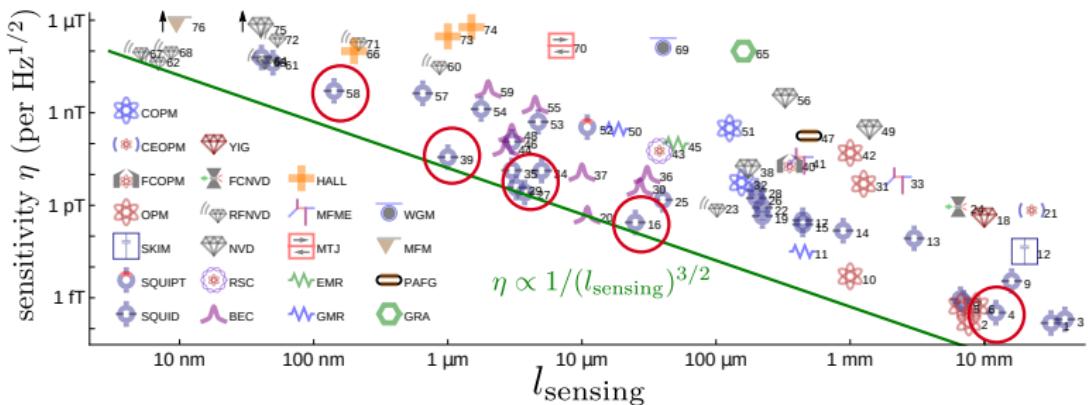


Optically pumped magnetometers (OPM)

- ✓ Very high sensitivity
- ✗ Limited in size

Mitchell, M. W., & Alvarez, S. P. (2020). Reviews of Modern Physics, 92(2), 021001

State of the art magnetometers



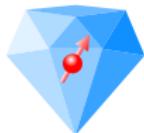
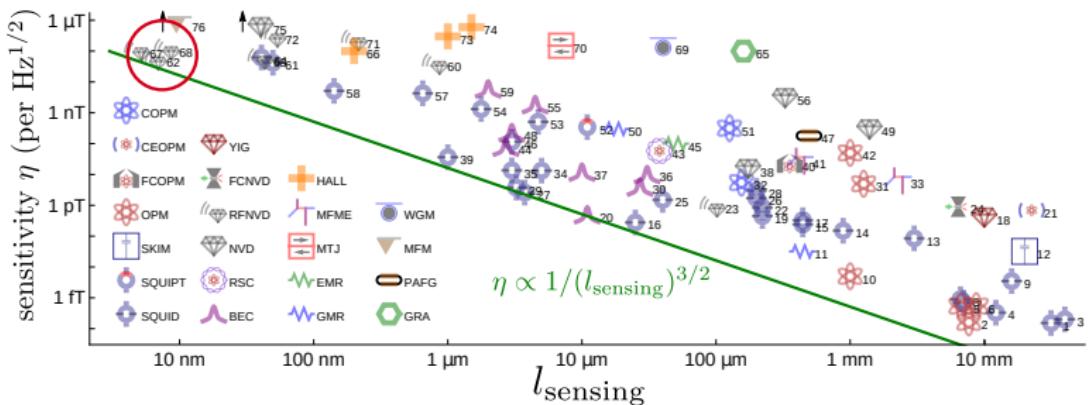
Superconducting quantum interference device (SQUID)

✓ High versatility, mature technology

✗ Requires cryogenic temperatures

Mitchell, M. W., & Alvarez, S. P. (2020). Reviews of Modern Physics, 92(2), 021001

State of the art magnetometers

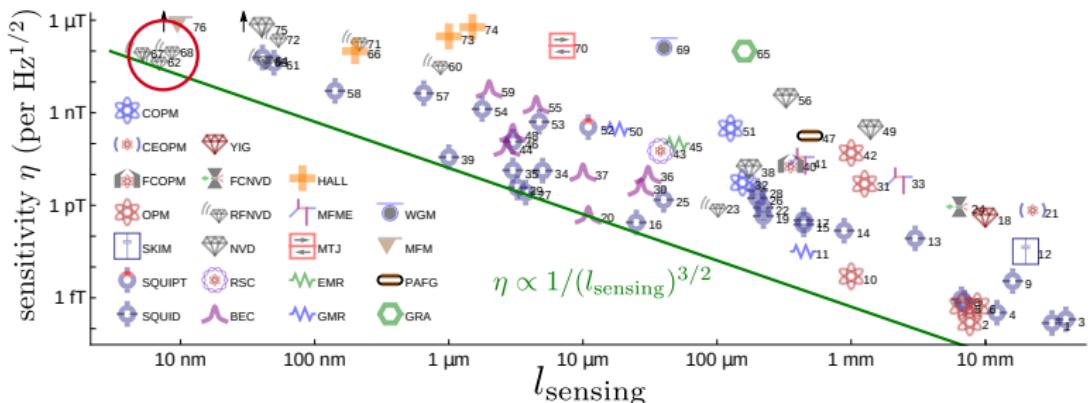


(Single) NV center

- nm resolution
- Room temperature

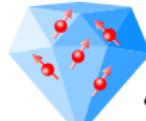
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State of the art magnetometers



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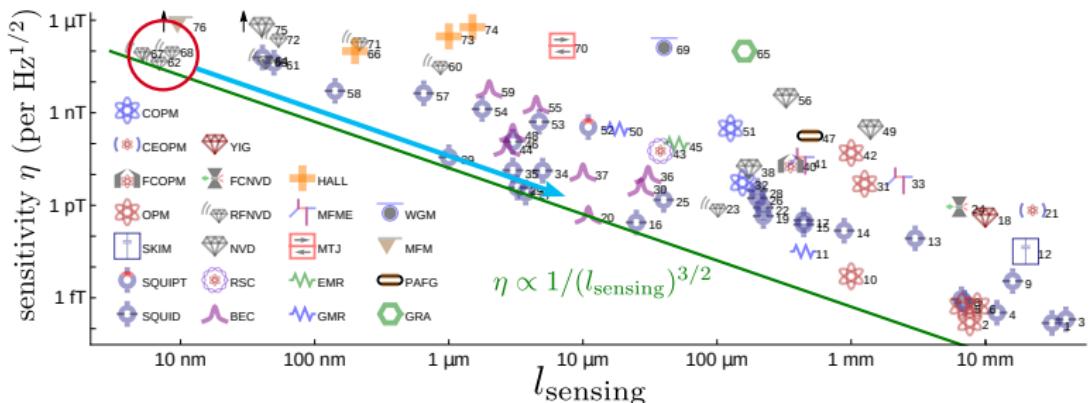


NV ensemble

- μm resolution
- Room temperature
- Higher sensitivity

Mitchell, M. W., & Alvarez, S. P. (2020). Reviews of Modern Physics, 92(2), 021001

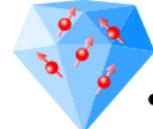
State of the art magnetometers



(Single) NV center

- nm resolution
- Room temperature

$$\eta \propto 1/\sqrt{N} \propto 1/\sqrt{V}$$

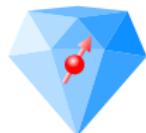
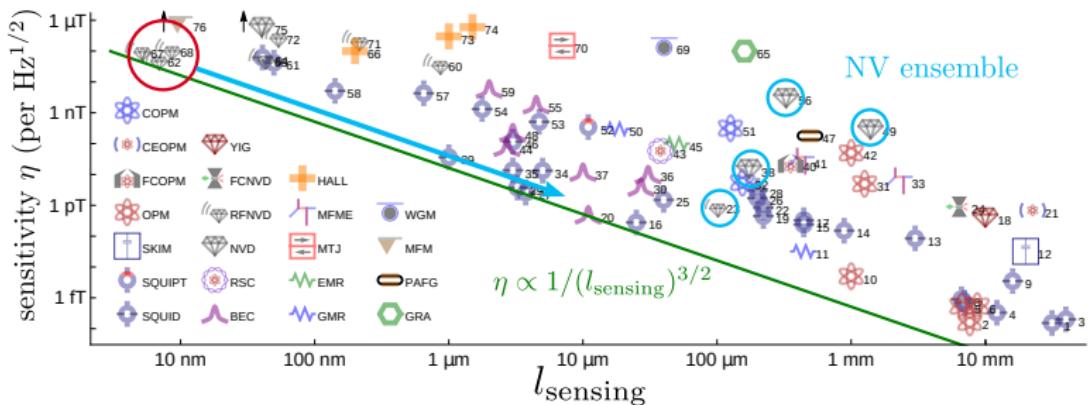


NV ensemble

- μm resolution
- Room temperature
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State of the art magnetometers

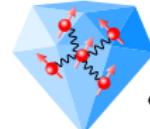


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Interactions

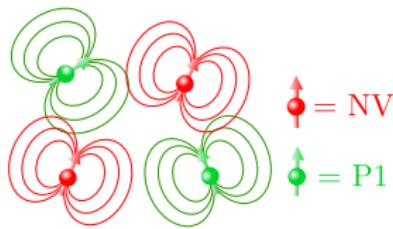
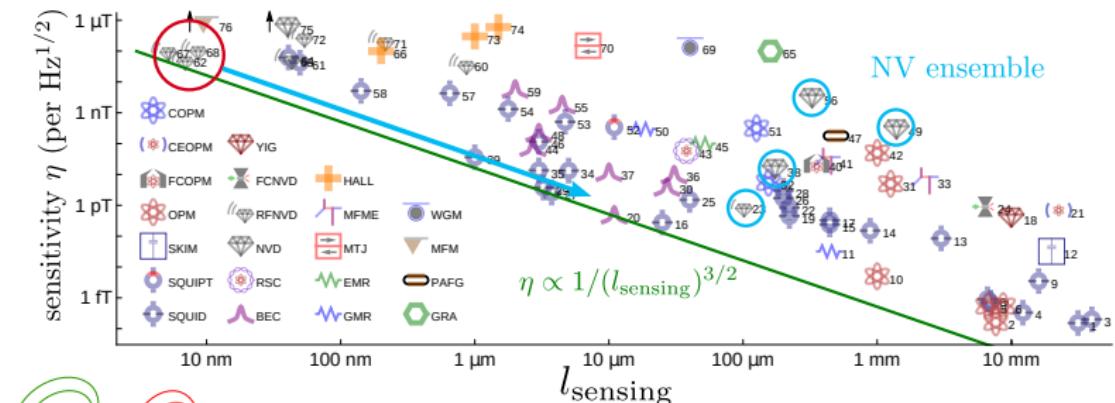


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State of the art magnetometers

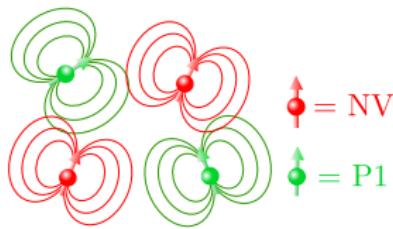
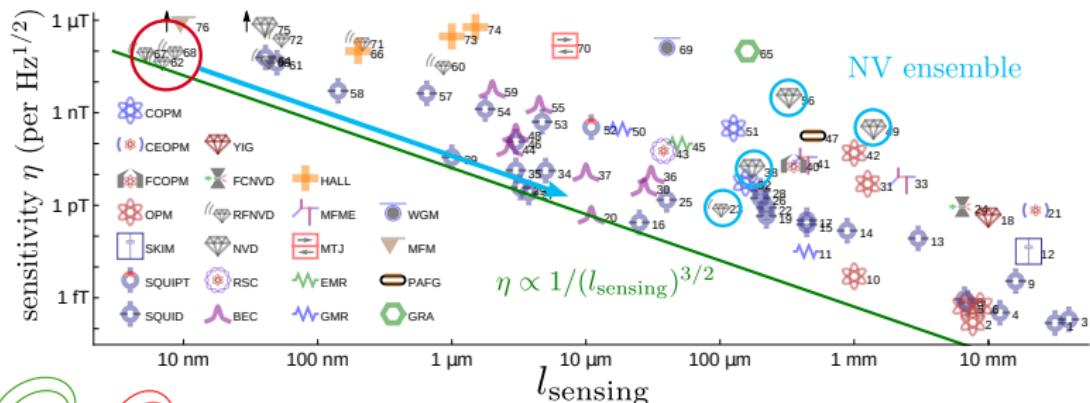


Interactions:

- Spectral broadening
- Modified spin dynamics

Mitchell, M. W., & Alvarez, S. P. (2020). *Reviews of Modern Physics*, 92(2), 021001

State of the art magnetometers



Interactions:

- Spectral broadening
- Modified spin dynamics

Solutions:

- Decoupling interactions (Hamiltonian engineering)
- Exploiting interactions

Mitchell, M. W., & Alvarez, S. P. (2020). *Reviews of Modern Physics*, 92(2), 021001

Sensing with quantum mechanics

NV centers and diamonds in practice

Low field depolarization magnetometry (LFDM)

Depolarization mechanisms in dense NV ensemble

Bonus slides

Diamond properties

NV center energy levels

Basic experiment with NV centers

Outline

Sensing with quantum mechanics

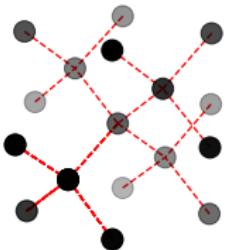
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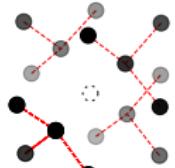
Depolarization mechanisms in dense NV ensemble

Colored centers in diamond

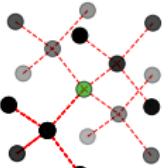
Diamond crystal lattice



Point-like defects



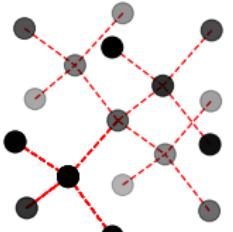
Vacancy



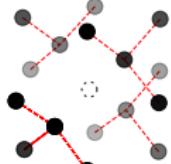
Substitution

Colored centers in diamond

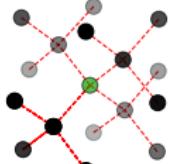
Diamond crystal lattice



Point-like defects

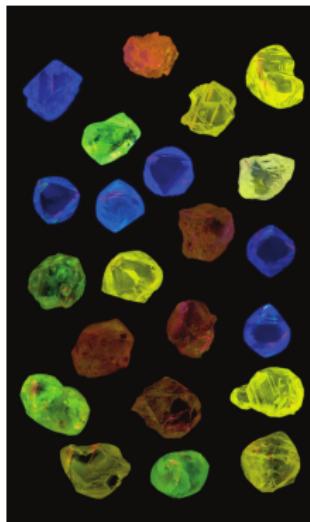
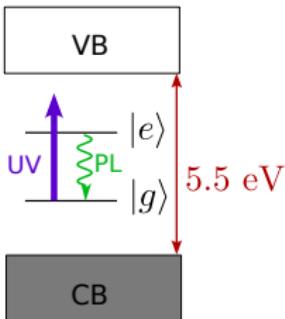


Vacancy



Substitution

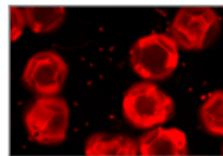
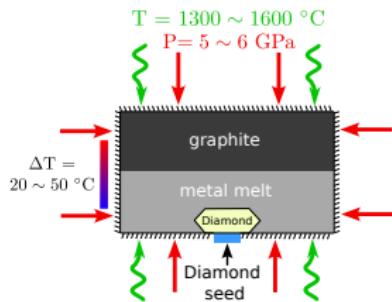
Colored center fluorescence



Natural diamonds fluorescence under UV light

Synthetic diamond and NV centers

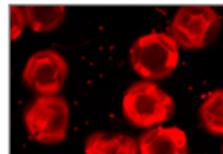
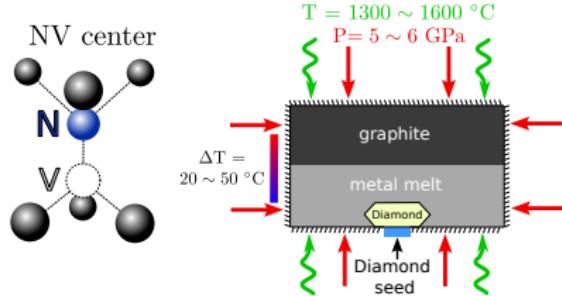
High Pressure High Temperature
(HPHT)



Adamas 15/150 μm

Synthetic diamond and NV centers

High Pressure High Temperature
(HPHT)

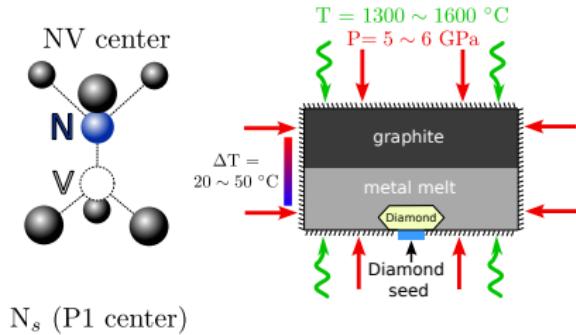


Adamas 15/150 μm

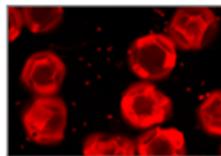
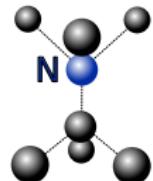
$[\text{NV}] \approx 3 \text{ ppm}$

Synthetic diamond and NV centers

High Pressure High Temperature
(HPHT)



N_s (P1 center)



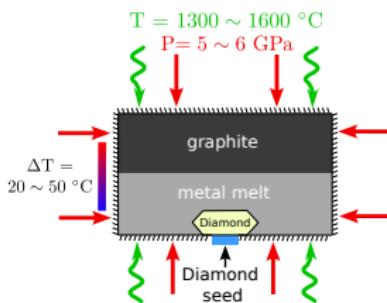
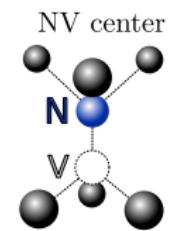
Adamas 15/150 μm

[NV] \approx 3 ppm

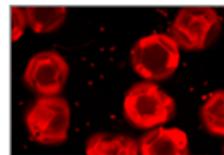
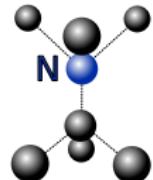
[P1] \approx 100 ppm

Synthetic diamond and NV centers

High Pressure High Temperature (HPHT)



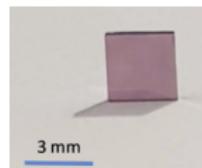
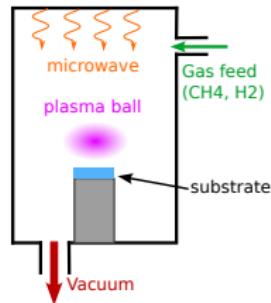
N_s (P1 center)



Adamas 15/150 μm

$[\text{NV}] \approx 3 \text{ ppm}$
 $[\text{P1}] \approx 100 \text{ ppm}$

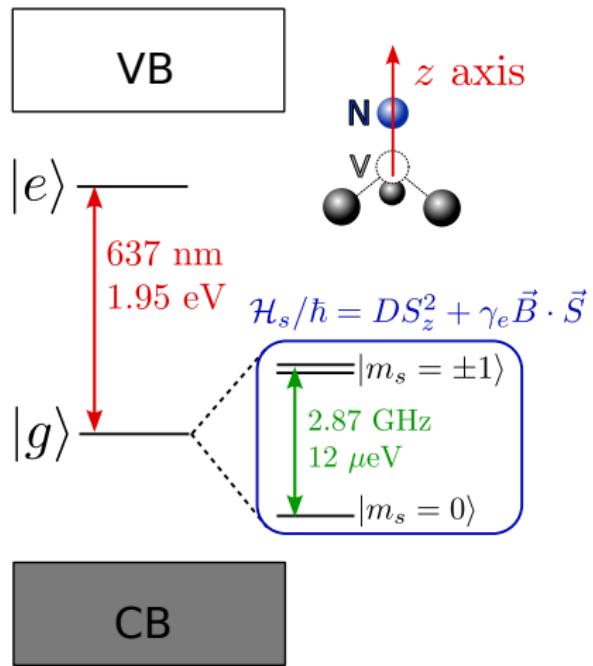
Chemical Vapour Deposition (CVD)



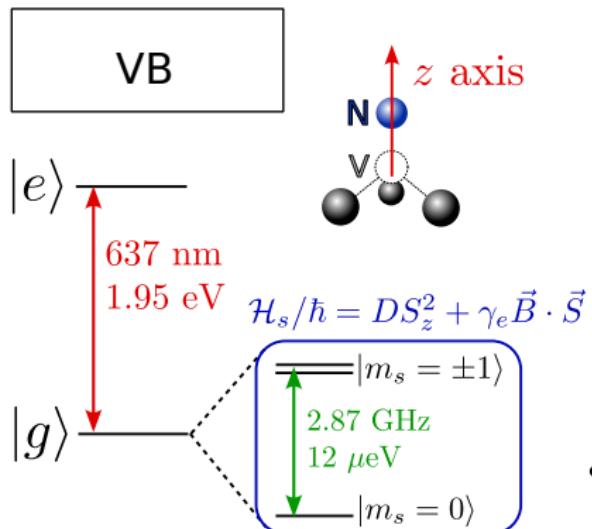
IRCP-LSPM
 $[\text{NV}] \approx 4.5 \text{ ppm}$
 $[\text{P1}] \approx 25 \text{ ppm}$

Tallaire, A., et al (2020).
 Carbon, 170, 421-429.

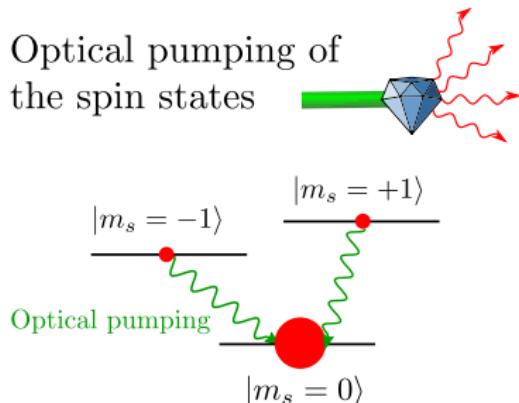
The NV center energy levels



The NV center energy levels

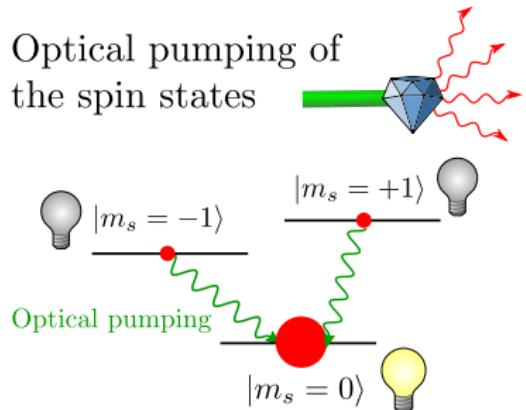
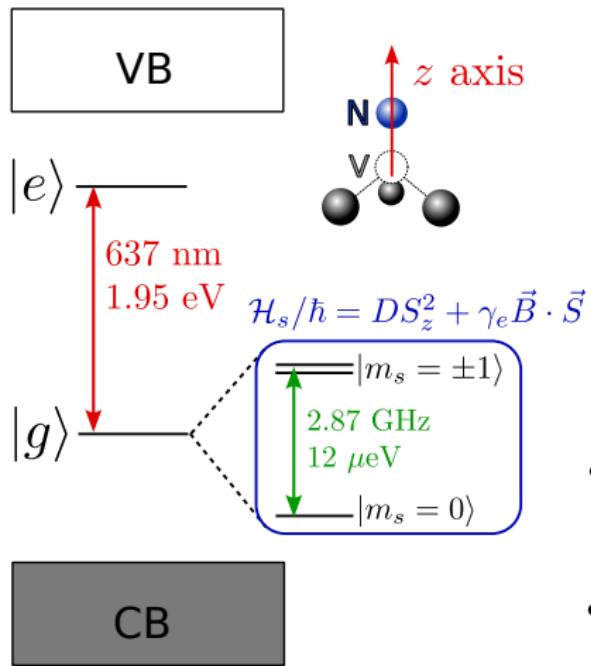


Optical pumping of the spin states



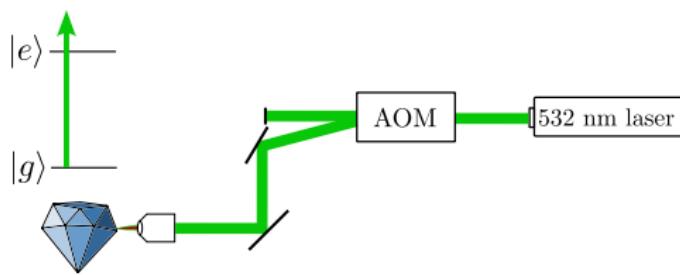
- Population accumulation in the $|0\rangle$ state
 - ↳ Initialization of the spin state

The NV center energy levels

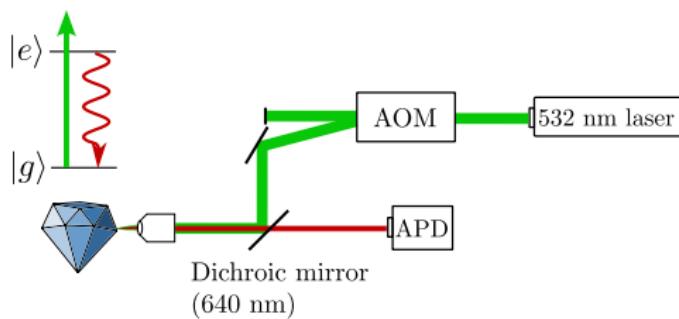


- Population accumulation in the $|0\rangle$ state
 - ↳ Initialization of the spin state
- $|0\rangle$ state brighter than $|\pm 1\rangle$ states
 - ↳ Optical readout of the spin state

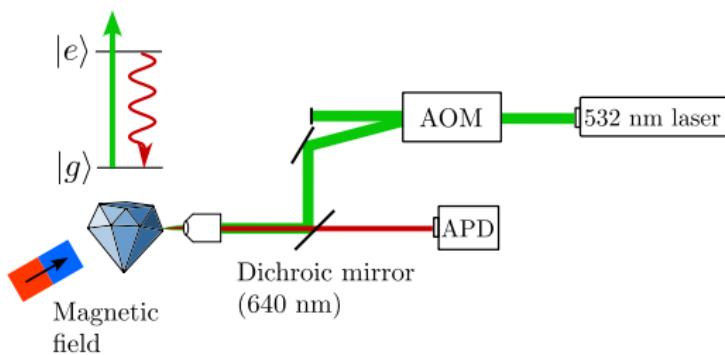
Experimental setup



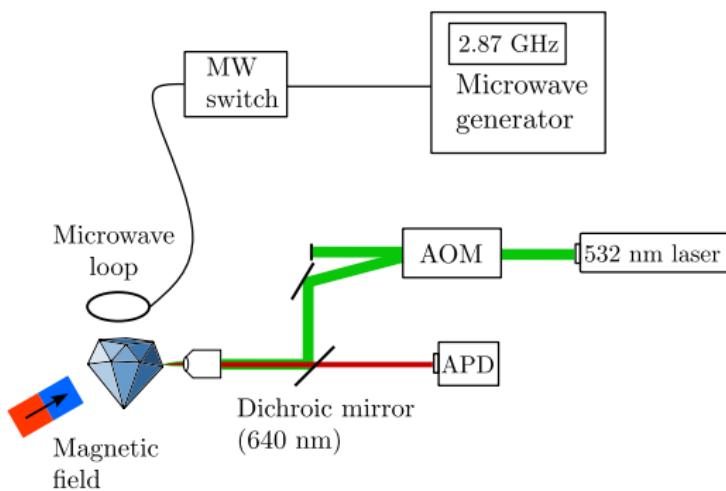
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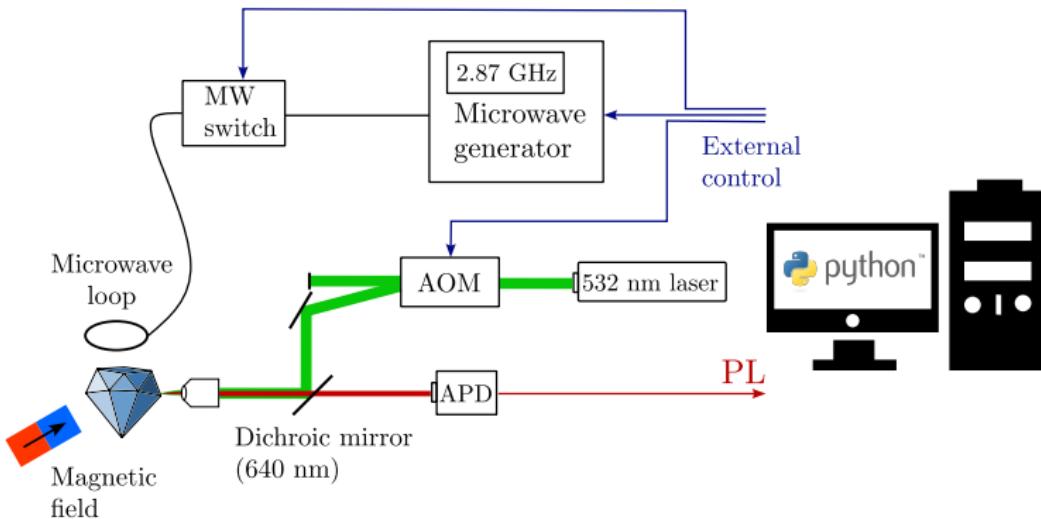
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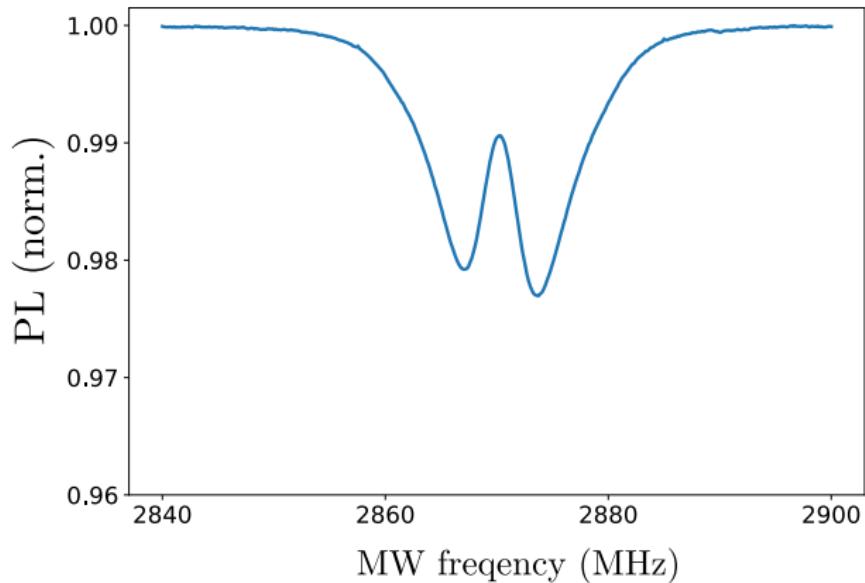
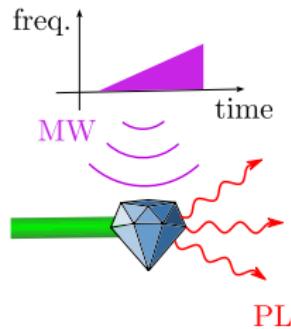
Experimental setup



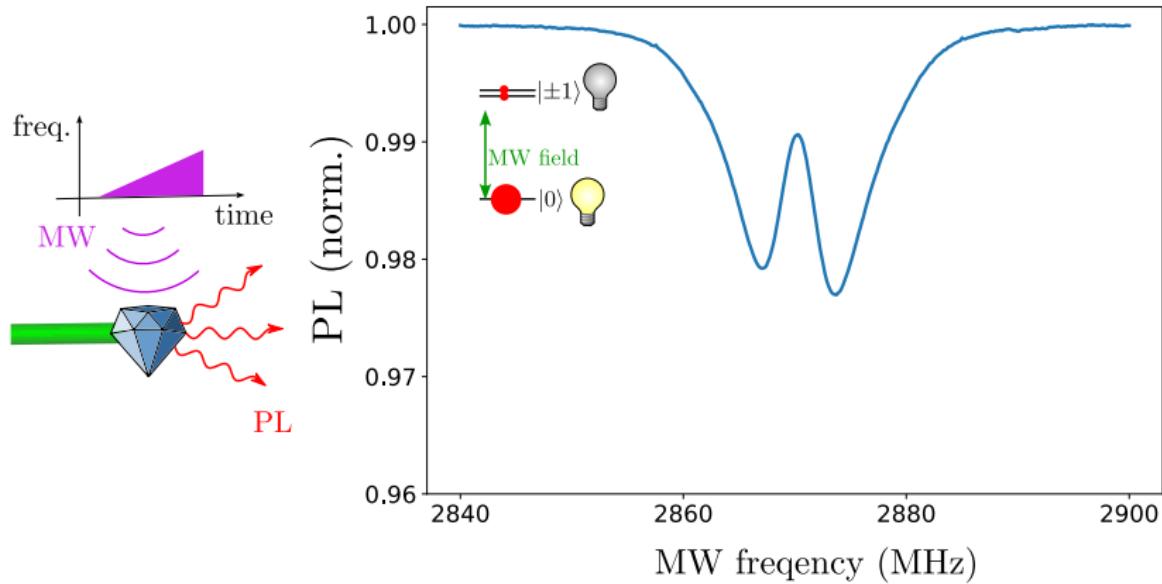
Experimental setup



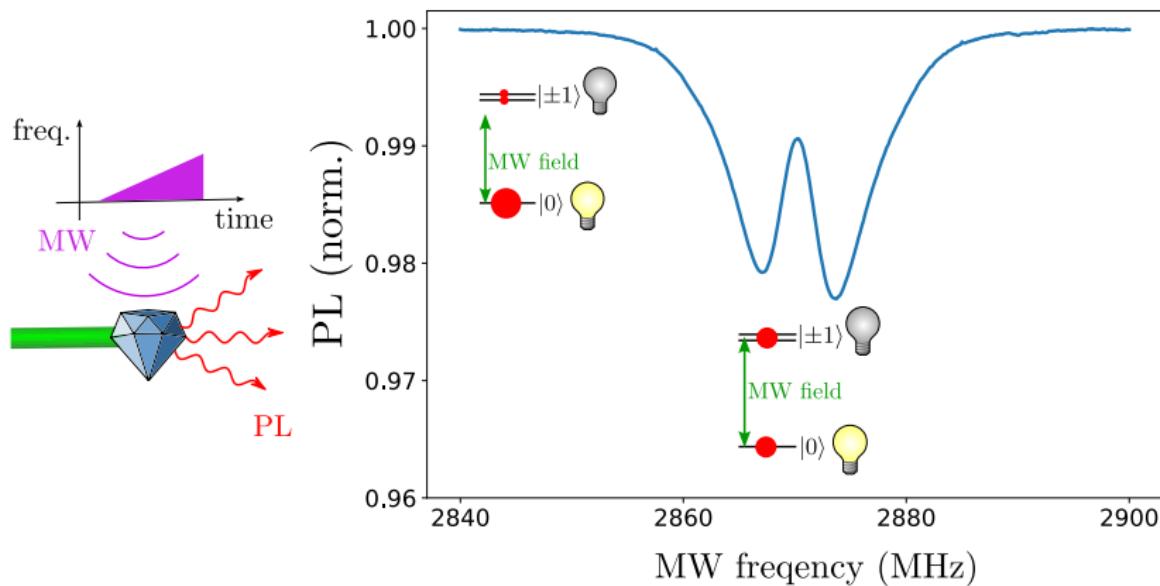
Optically detected magnetic resonance (ODMR)



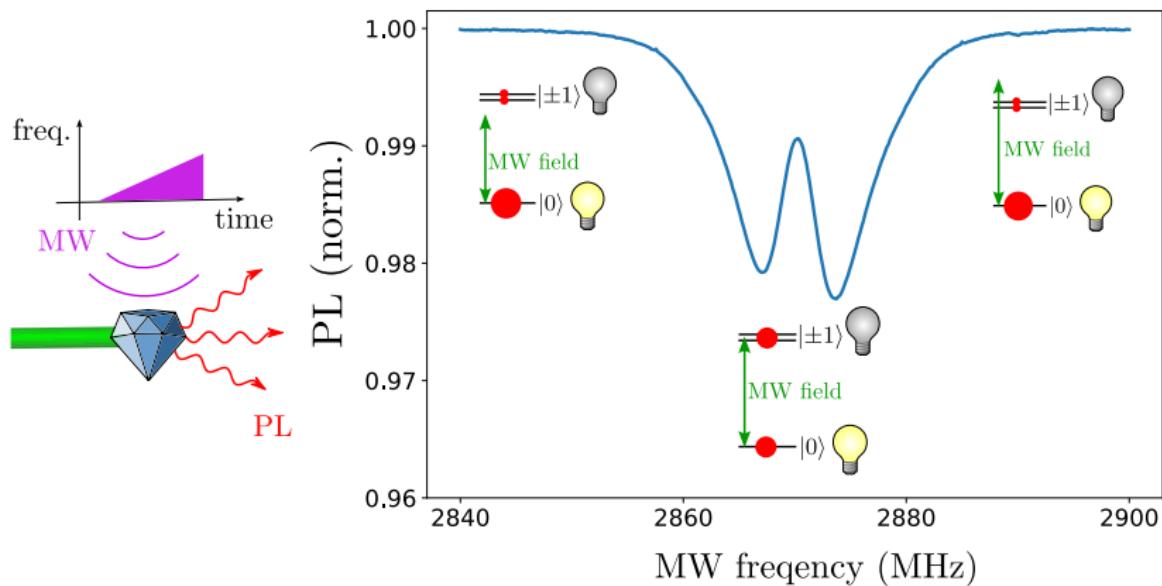
Optically detected magnetic resonance (ODMR)



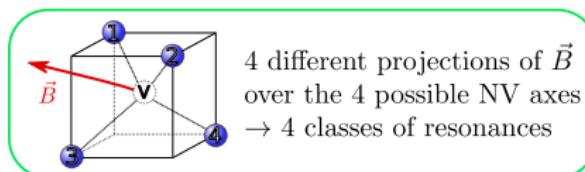
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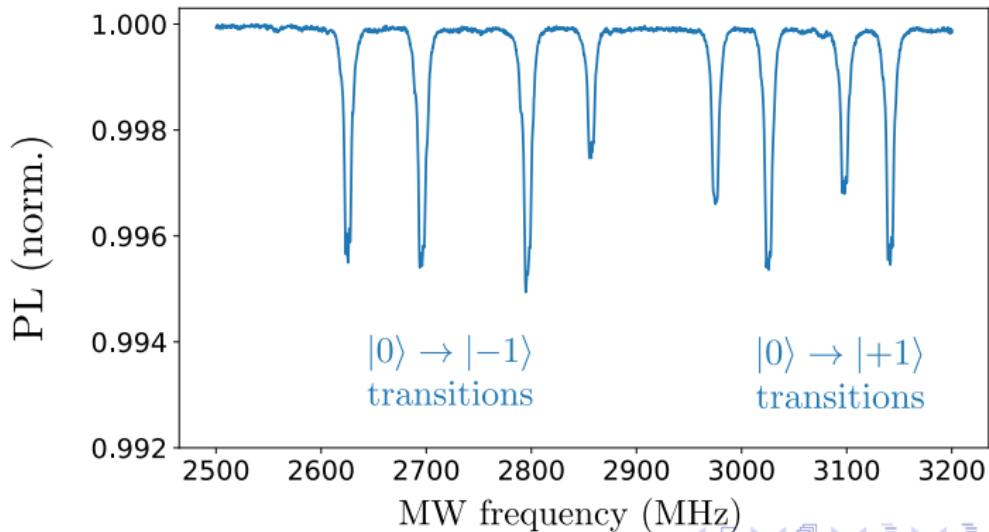
Optically detected magnetic resonance (ODMR)



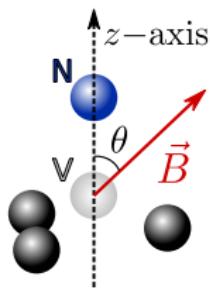
ODMR with NV ensemble: the 4 classes



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



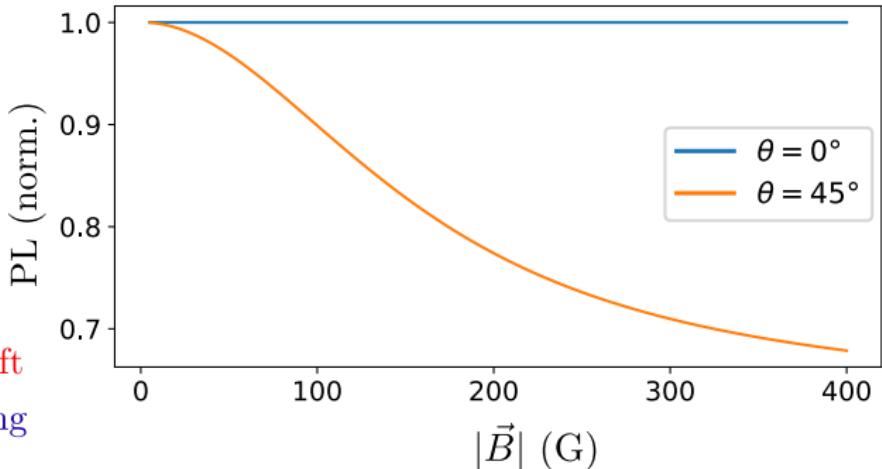
Transverse magnetic field effect



B_{\parallel} = Zeeman shift

B_{\perp} = State mixing

Loss of polarization, PL decrease



Sensing with quantum mechanics

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Bonus slides

Principle

Characterization

Applications

Outline

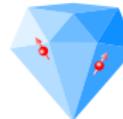
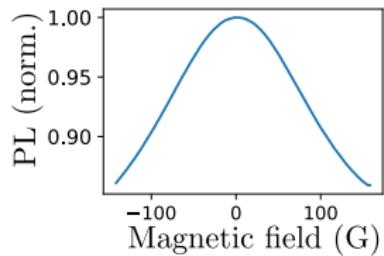
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NV centers and diamonds in practice

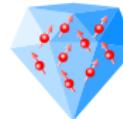
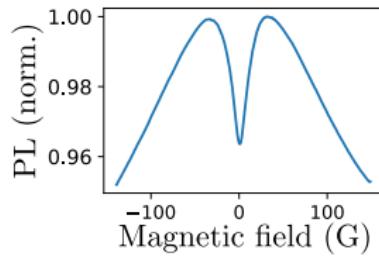
Low field depolarization magnetometry (LFDM)

Depolarization mechanisms in dense NV ensemble

Depolarization of dense NV ensemble at low magnetic field

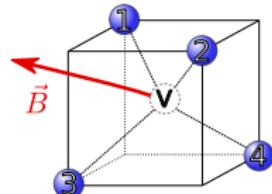


Low NV density
 $[NV] \leq 100$ ppb

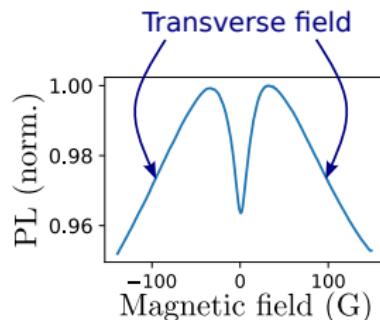
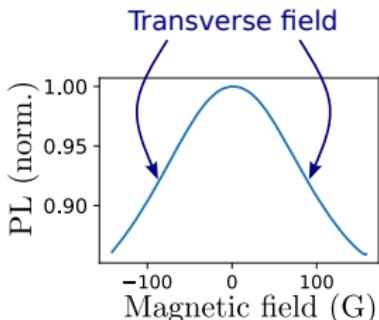


High NV density
 $[NV] \geq 1$ ppm

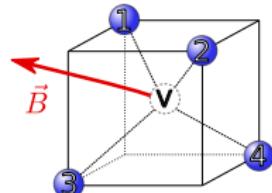
Depolarization of dense NV ensemble at low magnetic field



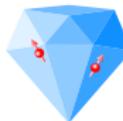
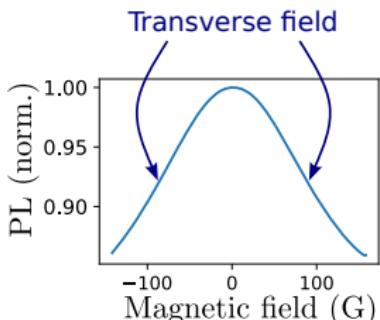
Non-zero transverse
magnetic field
on all 4 classes



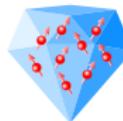
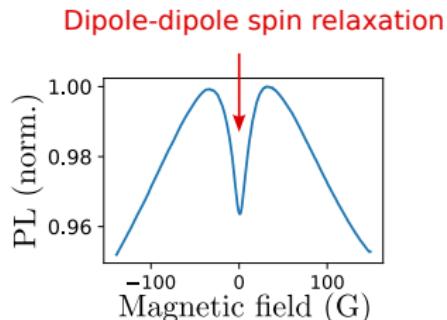
Depolarization of dense NV ensemble at low magnetic field



Non-zero transverse
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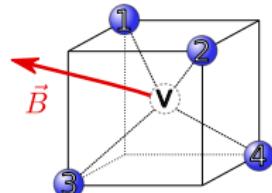


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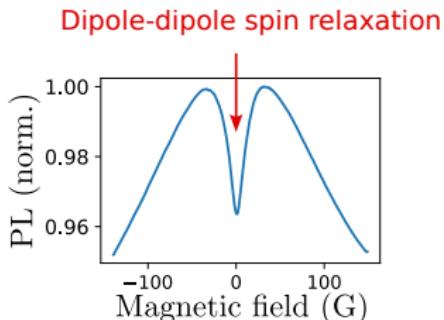
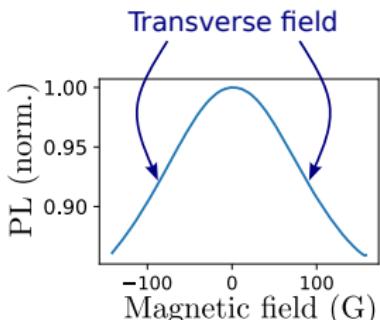


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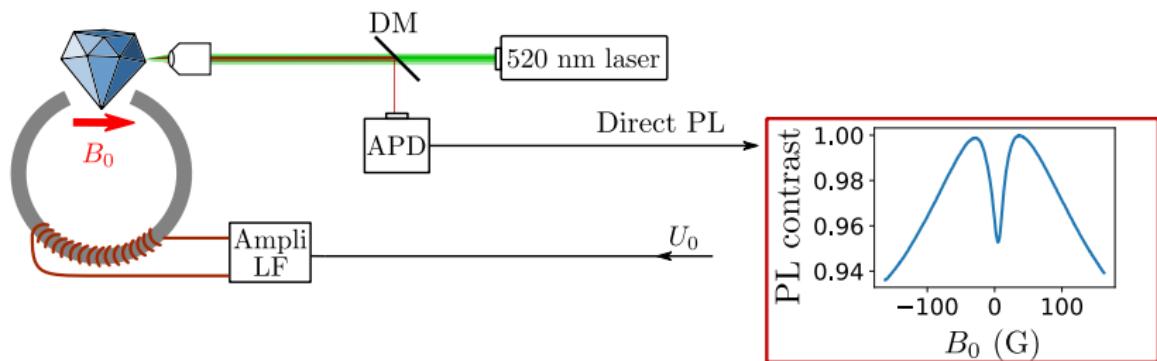


Non-zero transverse
magnetic field
on all 4 classes

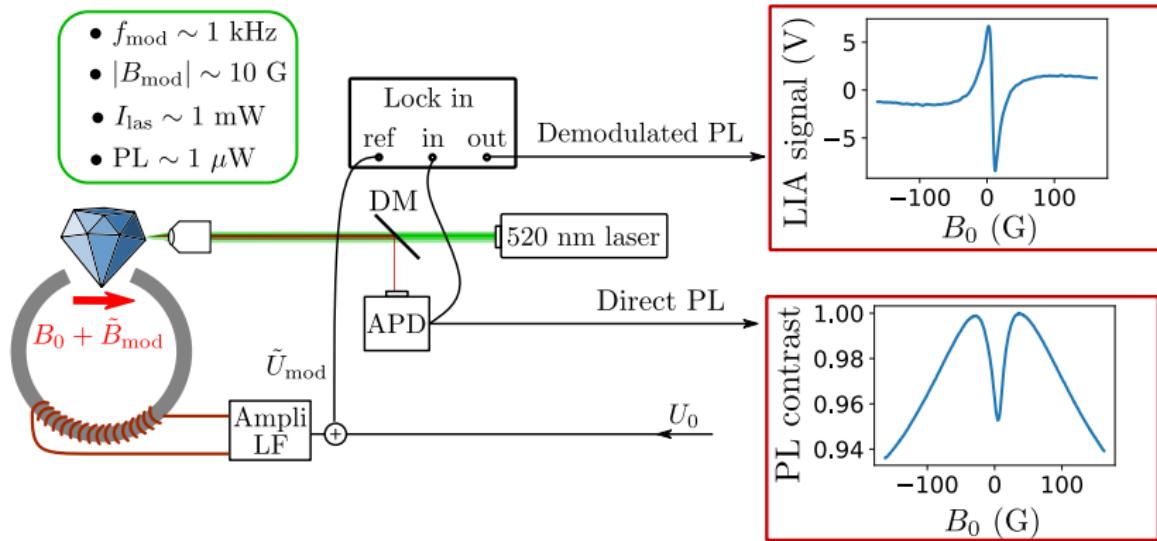


Sharp PL feature → magnetometry

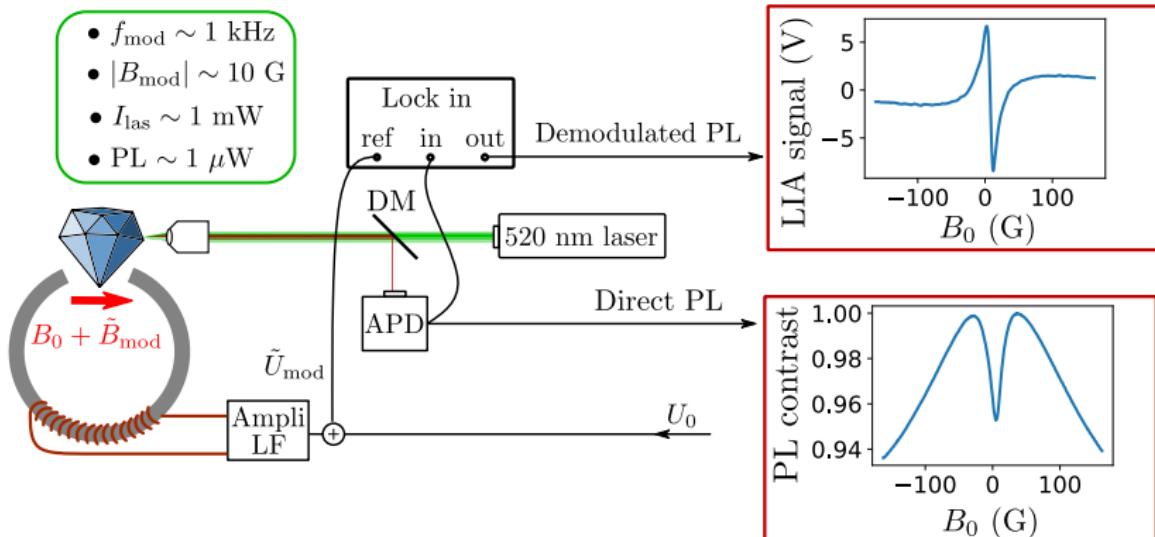
LFDM experimental setup



LFDM experimental setup



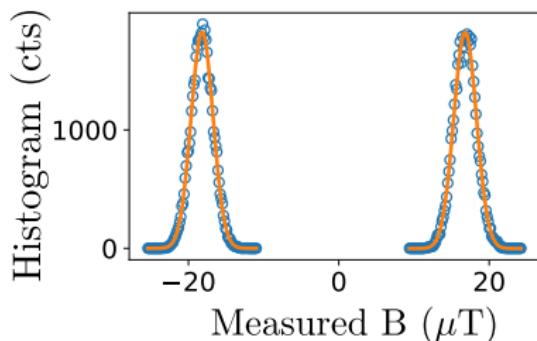
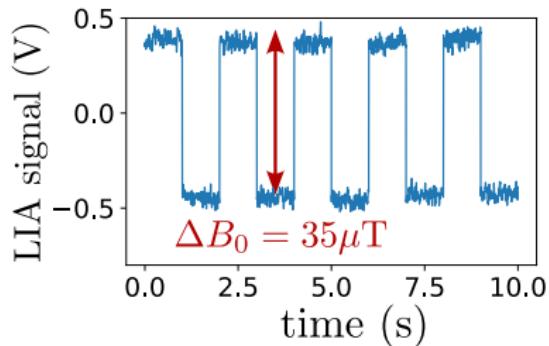
LFDM experimental setup



LIA:

- Avoid low frequency noise
- Linear dependance between U_{out} and B_0

Sensitivity of LFDM



$$\text{Measurement time } \tau = 3 \text{ ms} \quad \sqrt{\langle \delta B^2 \rangle} \approx 1.2 \mu\text{T}$$

$$\rightarrow \text{sensitivity } \eta = \sqrt{2\tau \langle \delta B^2 \rangle} \approx 120 \text{ nT}/\sqrt{\text{Hz}}$$

Comparison with the state of the art

Sensitivity comparison

	ODMR [1]	GSLAC [2]	LFDM
η (nT/ $\sqrt{\text{Hz}}$)	0.015	0.3	116

[1] Barry, J. F. [...] Walsworth, R. L (2016). PNAS, 113(49), 14133-14138.

[2] Zheng, H.[...] Budker, D. (2020). Physical Review Applied, 13(4), 044023.

Comparison with the state of the art

Sensitivity comparison			
	ODMR [1]	GSLAC [2]	LFDM
η (nT/ $\sqrt{\text{Hz}}$)	0.015	0.3	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] Barry, J. F. [...] Walsworth, R. L (2016). PNAS, 113(49), 14133-14138.

[2] Zheng, H.[...] Budker, D. (2020). Physical Review Applied, 13(4), 044023.

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	ODMR	GSLAC	LFDM
Microwave free	✗	✓	✓

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	ODMR	GSLAC	LFDM
Microwave free	✗	✓	✓
Low magnetic field (<10 G)	✓	✗	✓

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[2] Zheng, H.[...] Budker, D. (2020). Physical Review Applied, 13(4), 044023.

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

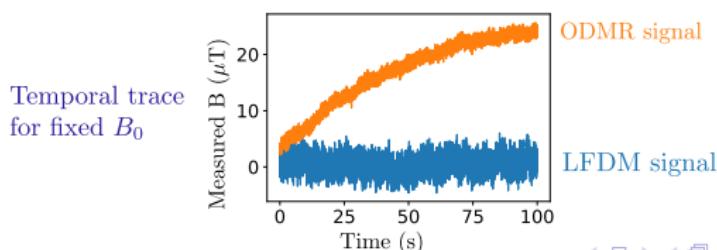
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[2] Zheng, H.[...] Budker, D. (2020). Physical Review Applied, 13(4), 044023.

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



Comparison with the state of the art

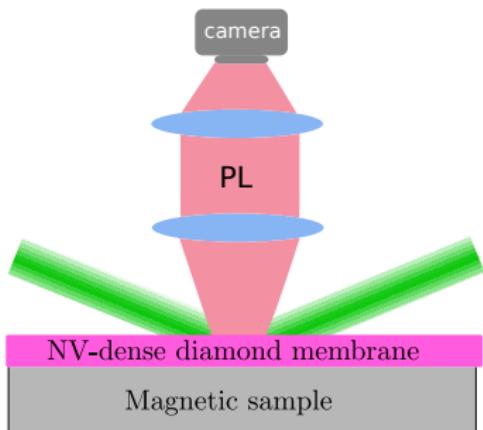
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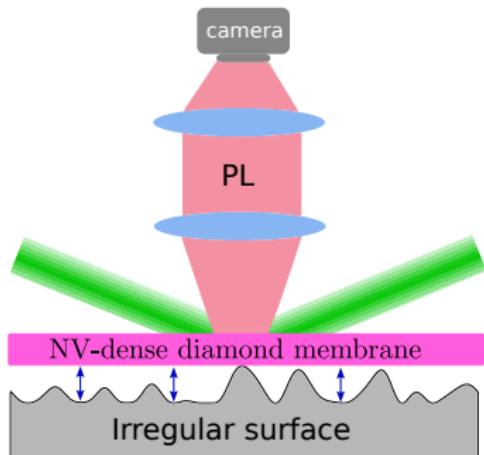
[2] Zheng, H.[...] Budker, D. (2020). Physical Review Applied, 13(4), 044023.

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

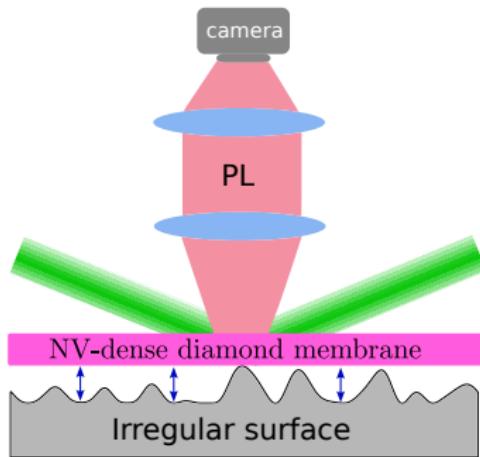
Application: wide-field magnetometry



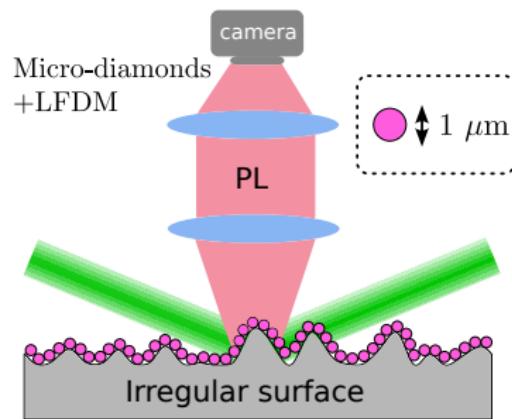
Application: wide-field magnetometry



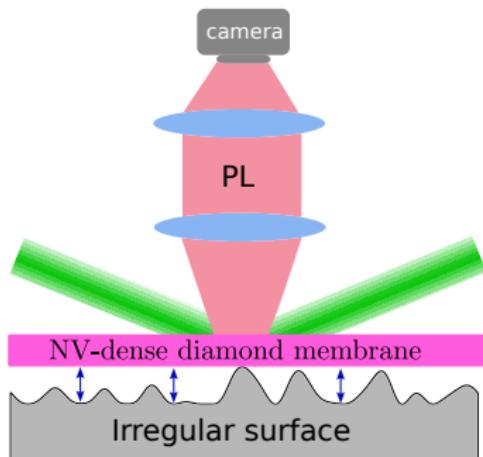
Application: wide-field magnetometry



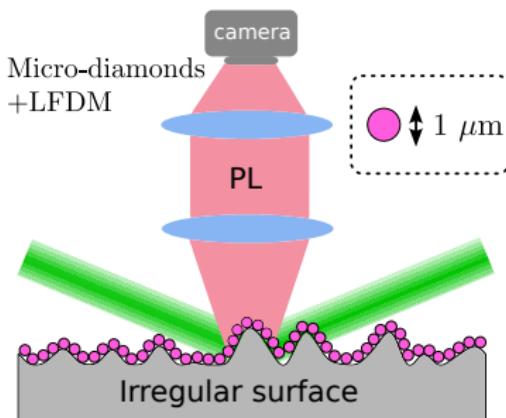
Gaps: loss of spatial resolution



Application: wide-field magnetometry



Gaps: loss of spatial resolution



State of the art [1]:

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

LFDM :

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

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

Outline

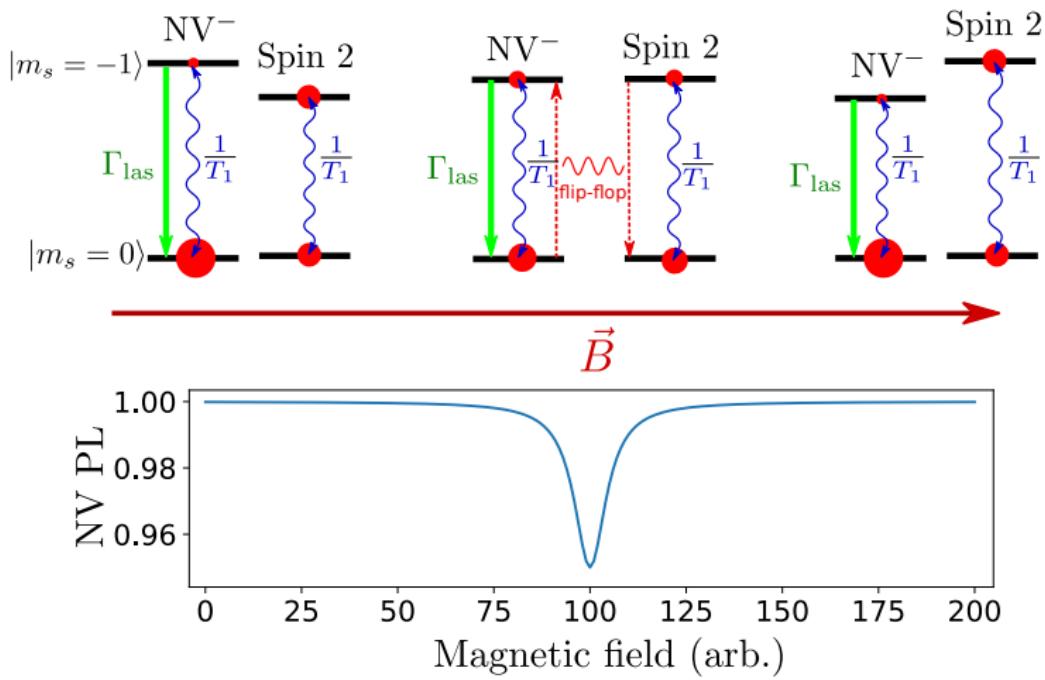
Sensing with quantum mechanics

NV centers and diamonds in practice

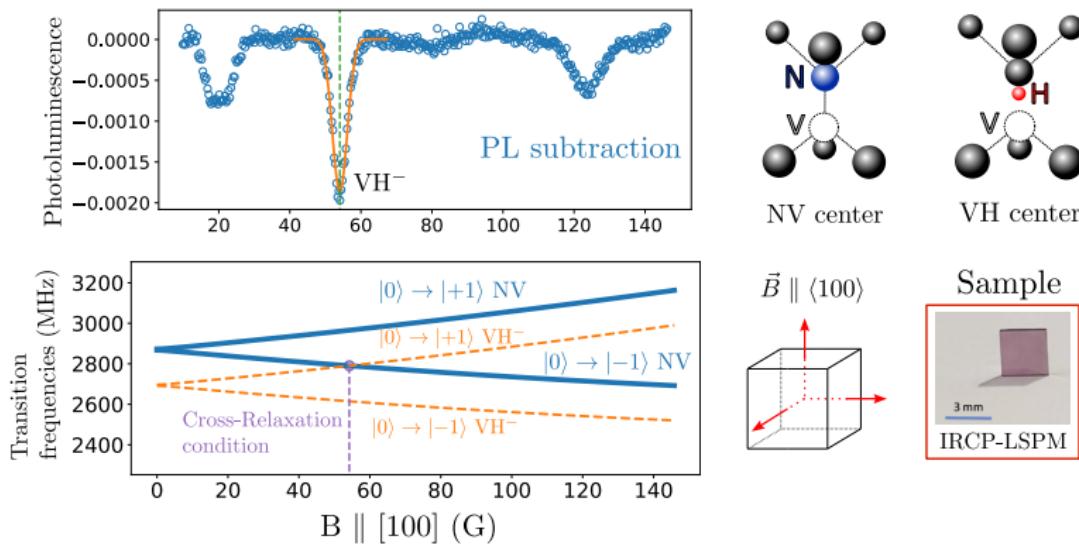
Low field depolarization magnetometry (LFDM)

Depolarization mechanisms in dense NV ensemble

Principle of cross-relaxation with NV centers



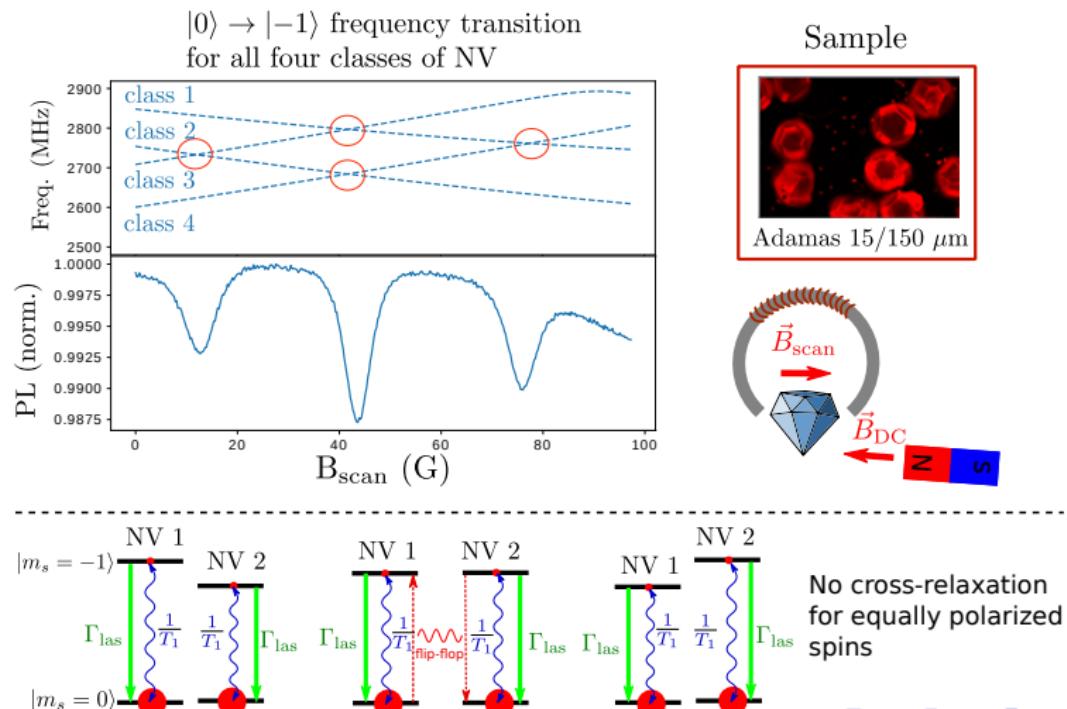
Example: Cross-relaxation between NV centers and VH⁻



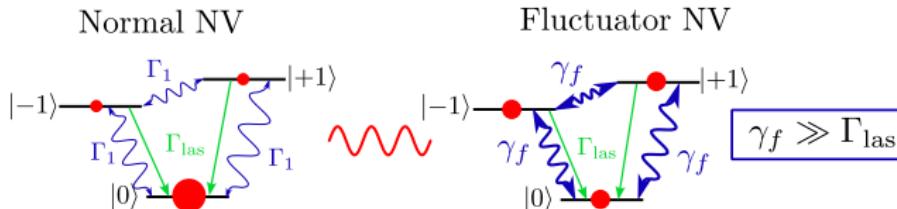
Optical detection of paramagnetic defects in diamond grown by chemical vapor deposition

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



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

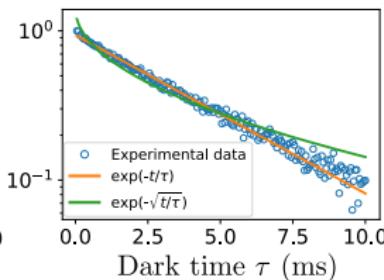
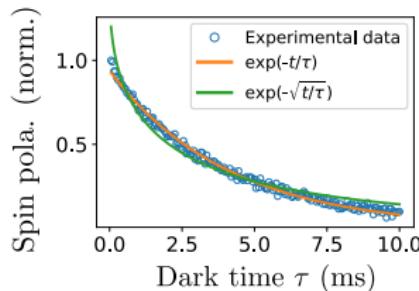
Possible microscopic explanation:

- charge tunneling
- modulation of J-coupling

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

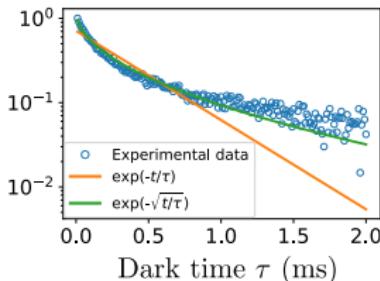
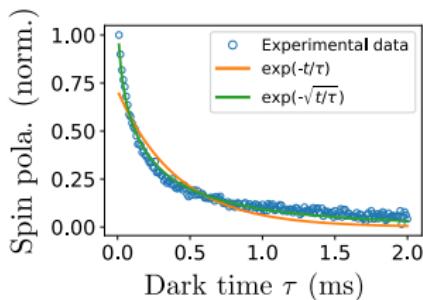
Choi, Joonhee, et al. Physical review letters 118.9 (2017): 093601.

Stretched exponential decay profile



Low NV density

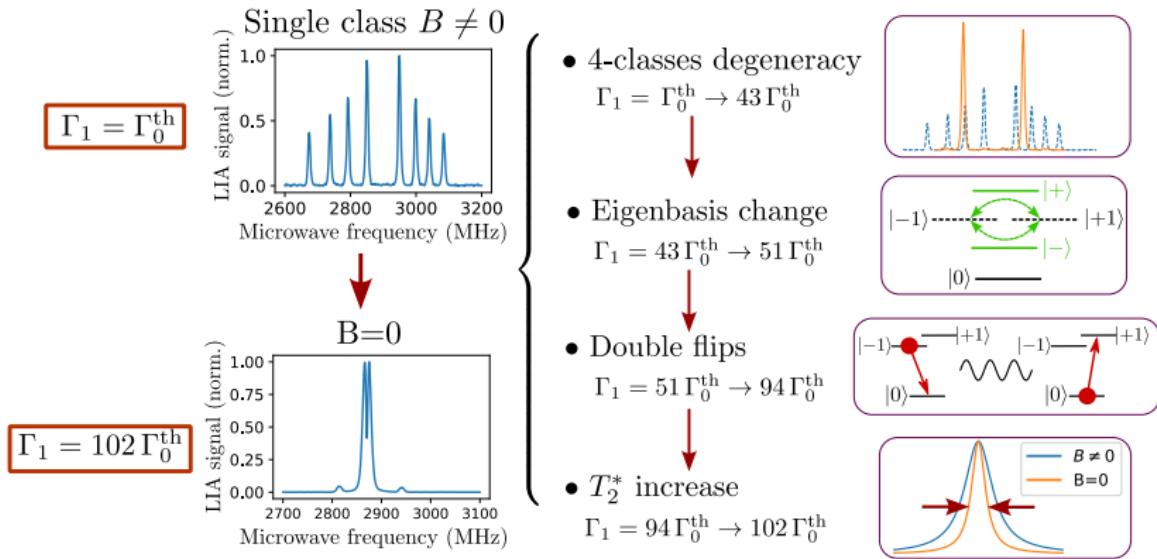
- Exponential profile
- $T_1 \sim 5$ ms



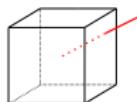
High NV density

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

Zero field depolarization sources (theory)

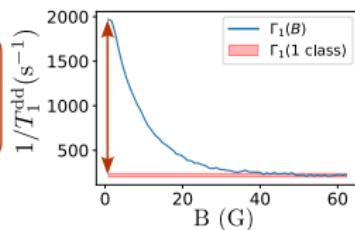


Summary of the experimental observations

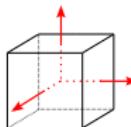


Random \vec{B}

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

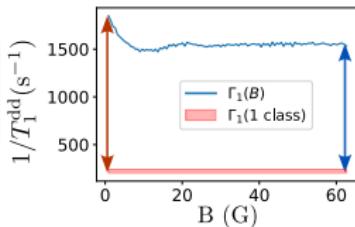


- 4-classes degeneracy
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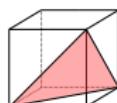


$\vec{B} \parallel \langle 100 \rangle$

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

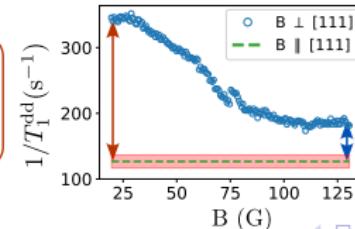


- 4-classes degeneracy
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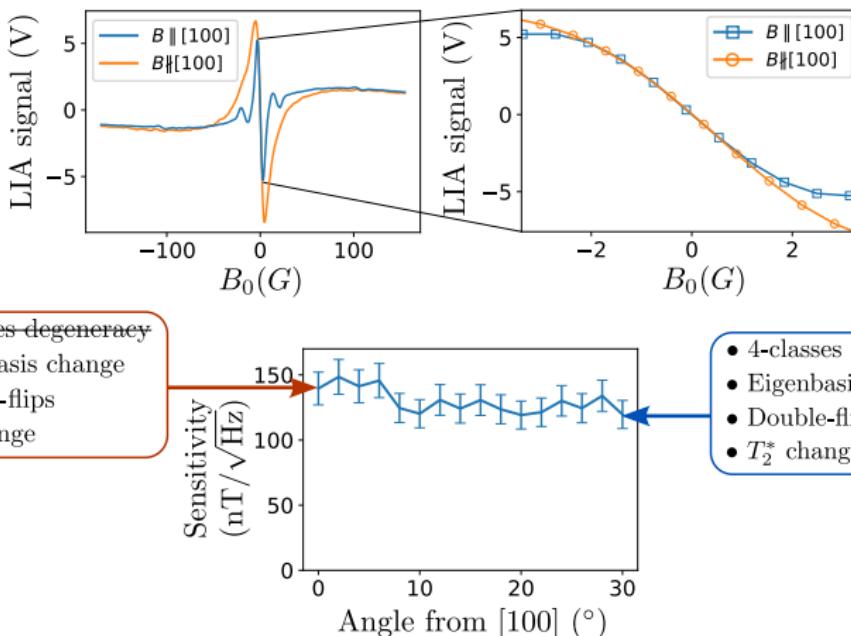
$\vec{B} \perp \langle 111 \rangle$

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



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

Angular sensitivity of LFDM



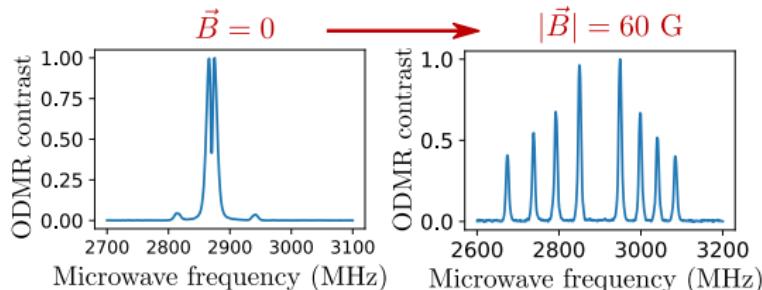
The 4-classes degeneracy is not the limiting factor of the sensitivity

Conclusion

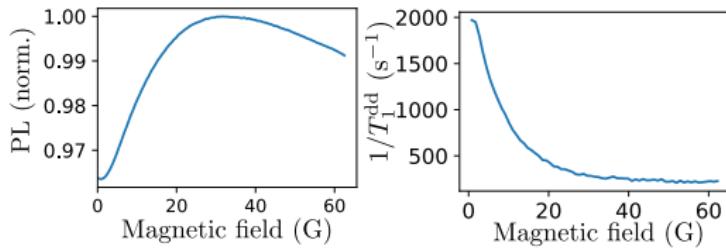
- ▶ The use of NV center ensemble for magnetometry is currently limited by the interactions between the spin defects.
- ▶ Dipole-dipole interaction within dense ensemble of NV centers result in spin depolarization which is exacerbated at low magnetic field.
- ▶ This depolarization can be used to perform microwave-free and orientation-free magnetometry at low magnetic field.
- ▶ the LFDM sensitivity seem to be determined by the double-flips.

Acknowledgments

Experiment: \vec{B} in arbitrary direction

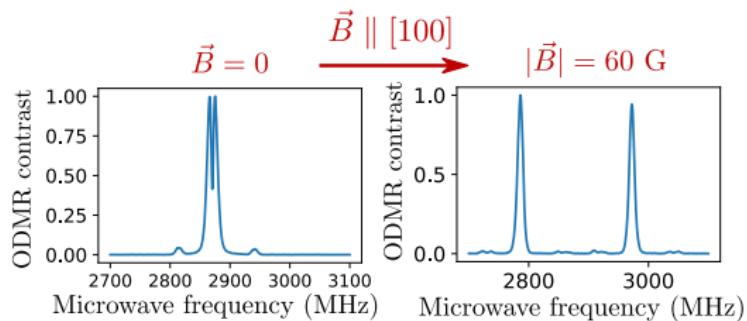


- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change

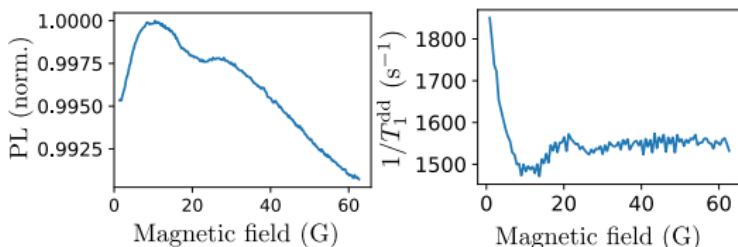


$\Gamma_1(B = 0) \approx 10 \Gamma_1(B \neq 0)$
 $\sim 4\%$ PL contrast
HWHM ~ 9 G

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



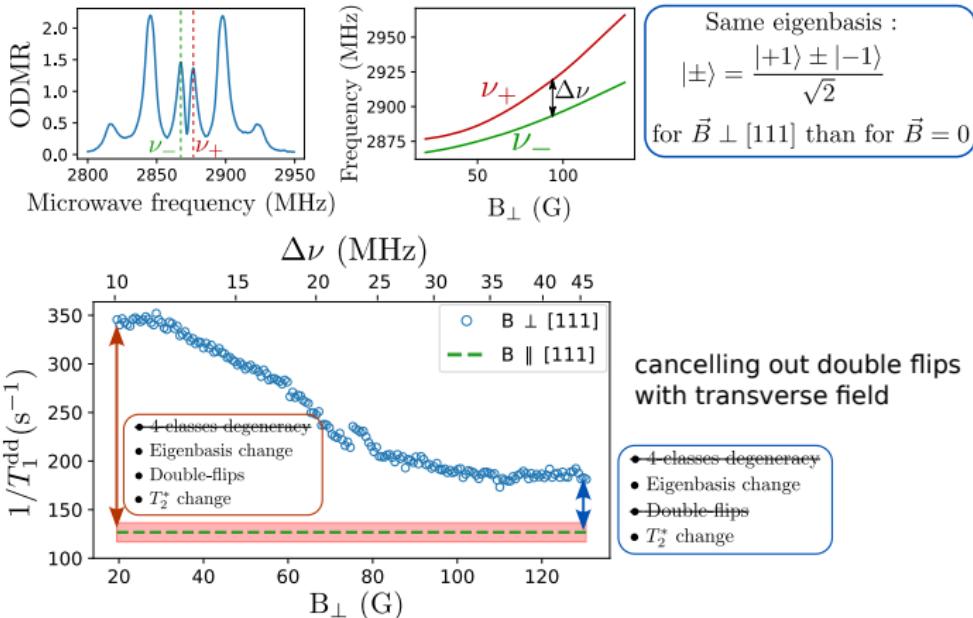
- 4-classes degeneracy
- Eigenbasis change
- Double-flips
- T_2^* change



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

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

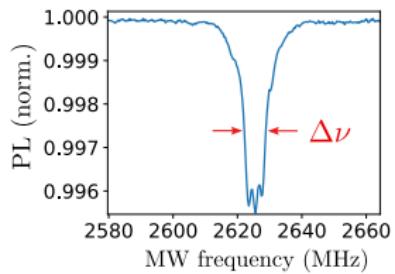
Experiment: $\vec{B} \perp [111]$



NV center magnetometry sensitivity

Ideal (DC) sensitivity for
 N independent NV centers:

$$\eta[T/\sqrt{\text{Hz}}] \approx \frac{\hbar\sqrt{\Delta\nu}}{g\mu_B C\sqrt{N}}$$



- \hbar : Planck constant
 - μ_B : Bohr magneton
 - g : NV electron Landé factor
 - C : Spin readout contrast
 - N : Number of NV centers
 - $\Delta\nu = \frac{1}{T_2^*}$: Spectral linewidth
- Constants

Experimental parameters
- Sample parameters

