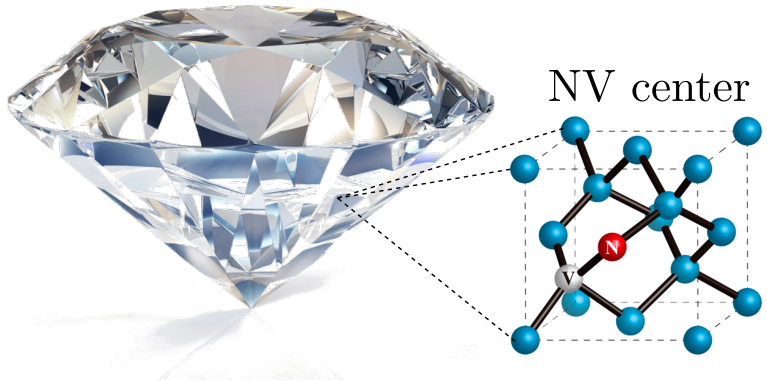
Cross-relaxation in dense ensembles of NV centers and application to magnetometry

Clément Pellet-Mary

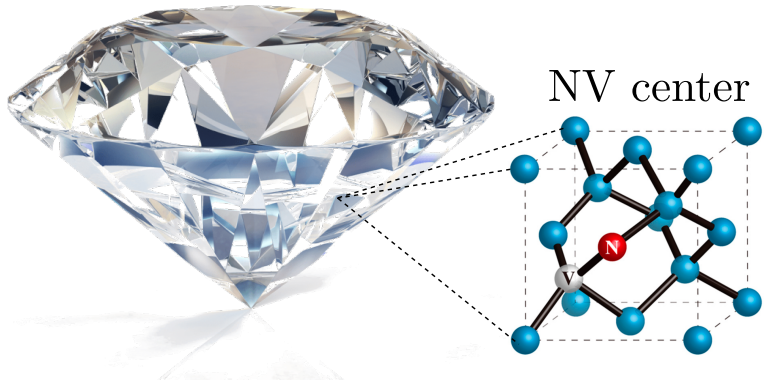
PhD Defense

Context of my PhD



Controllable spin
at room temperature

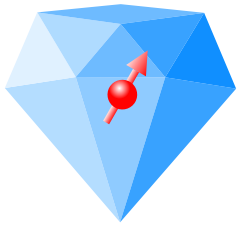
Context of my PhD



Controllable spin
at room temperature

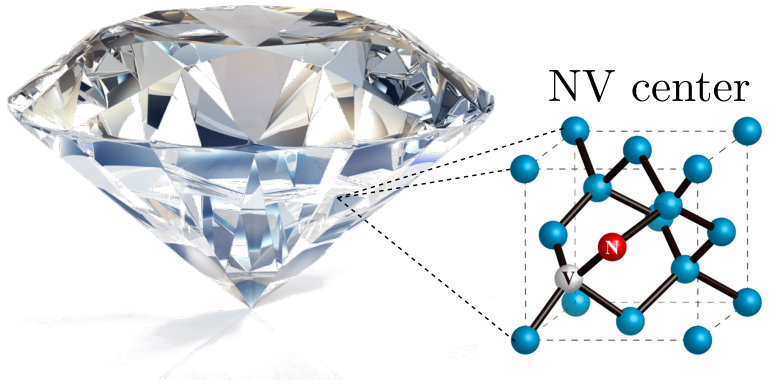
First isolated
NV center

1997



- Quantum memory
- Entanglement
- Quantum computing

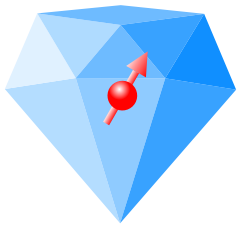
Context of my PhD



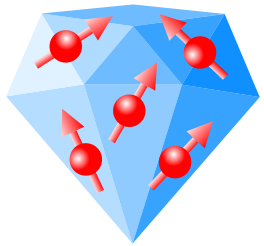
Controllable spin
at room temperature

First isolated
NV center

1997

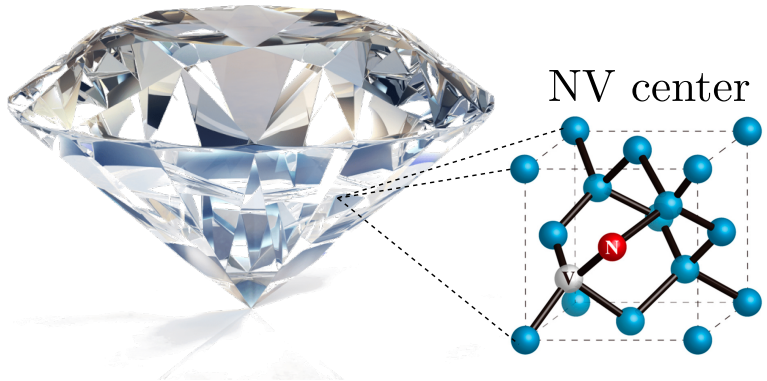


- Quantum memory
- Entanglement
- Quantum computing

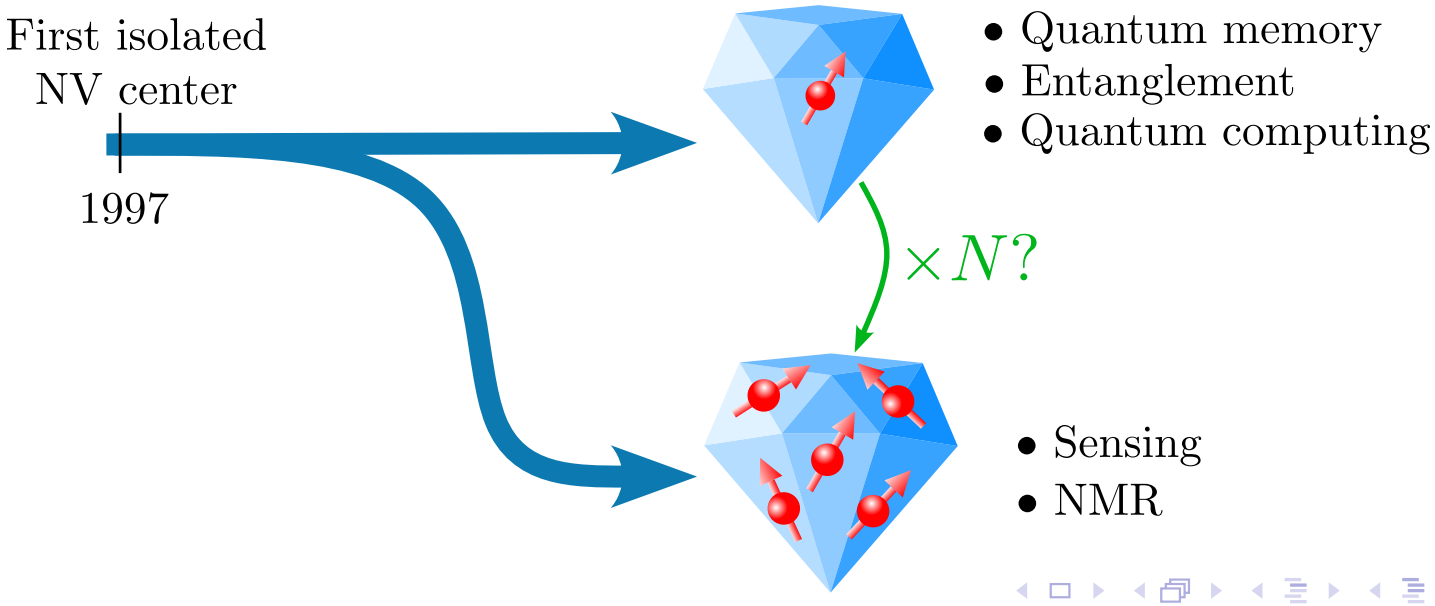


- Sensing
- NMR

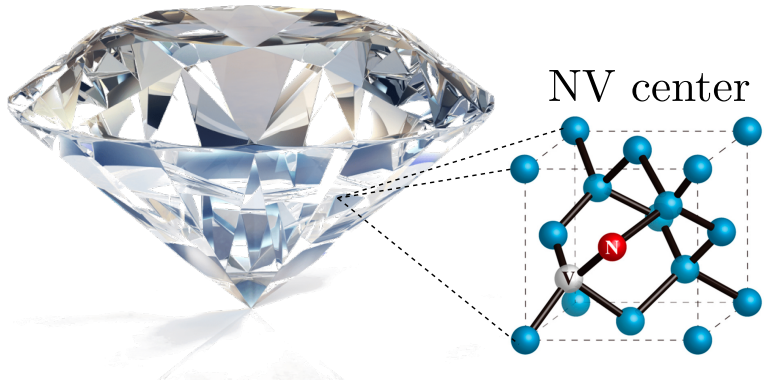
Context of my PhD



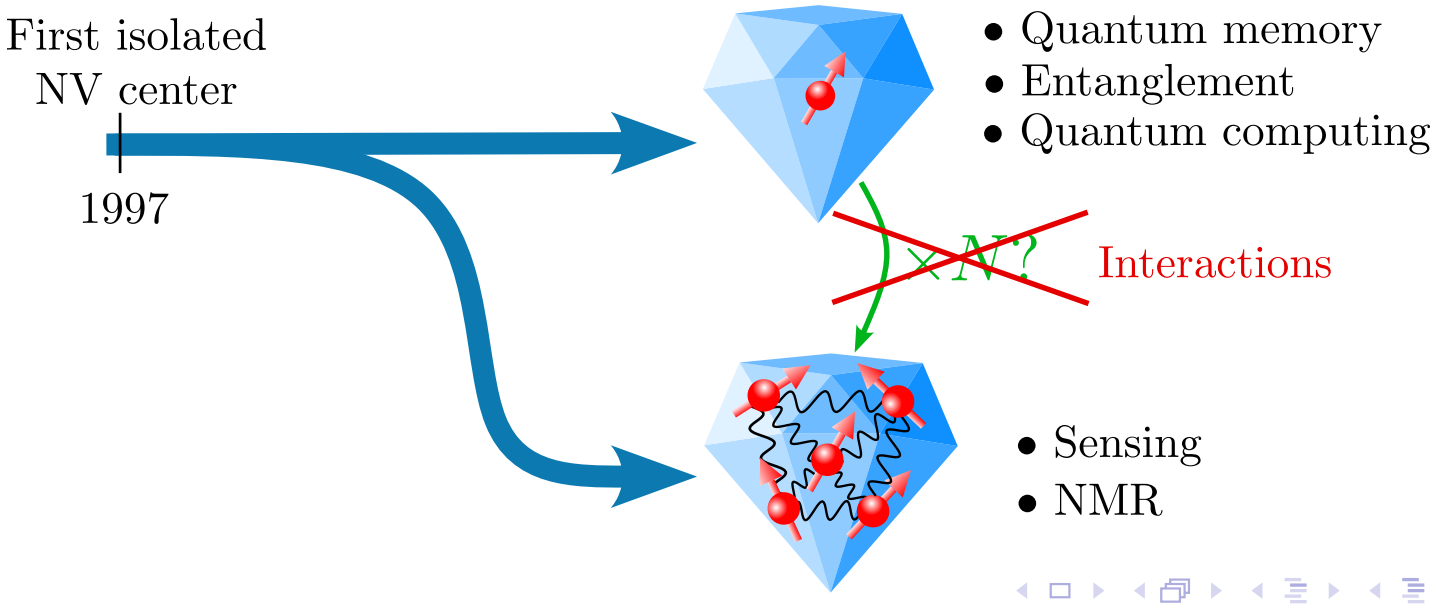
Controllable spin
at room temperature



Context of my PhD



Controllable spin
at room temperature



Outline

Outline

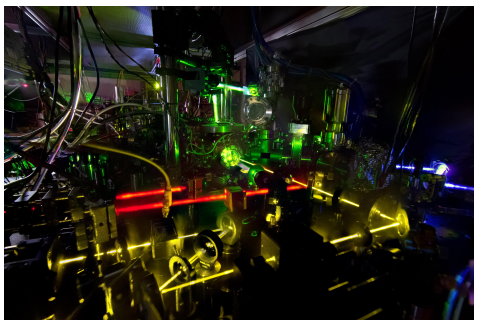
Quantum sensing and metrology

Quantum metrology:

Using quantum* properties to create more sensitive measurement protocols.

* quantum ≡ discrete energy levels

Time measurement

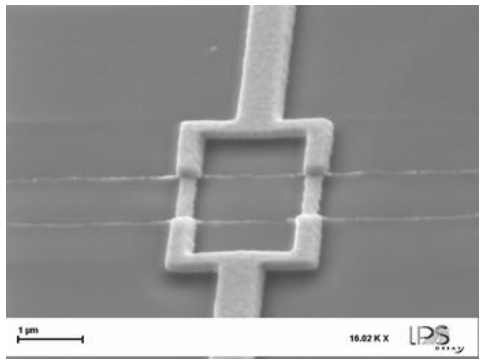


Atomic clock

Medical imaging

MRI

Magnetometry

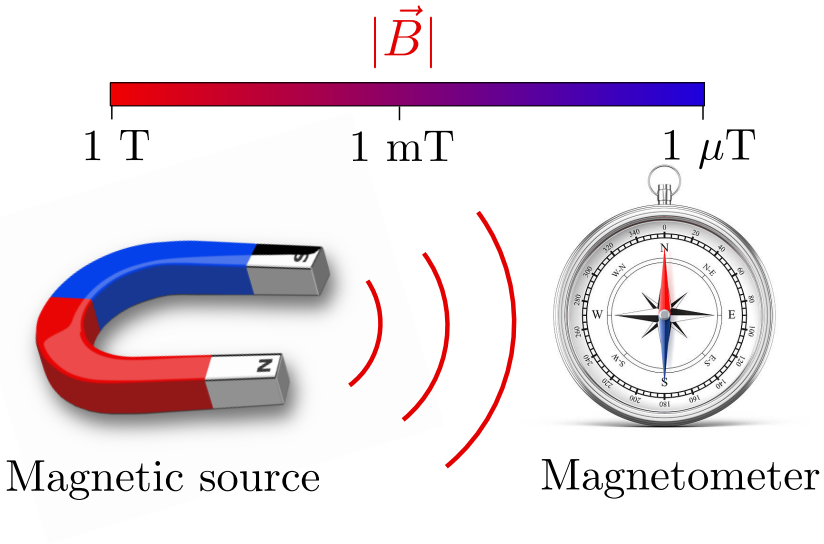


SQUIDs

Key properties of magnetometers

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

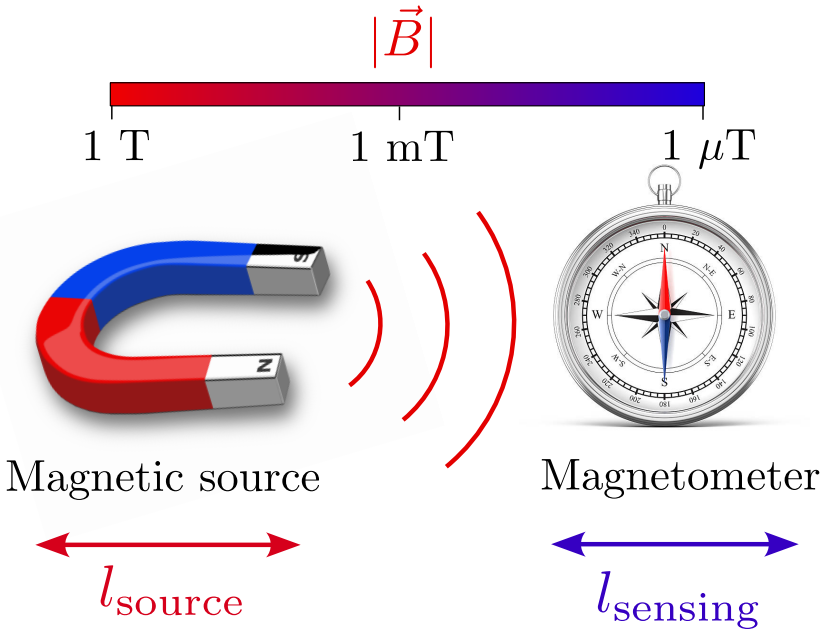
Minimum magnetic field value detectable with a signal-to-noise ratio of 1 within 1 second.



Key properties of magnetometers

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

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



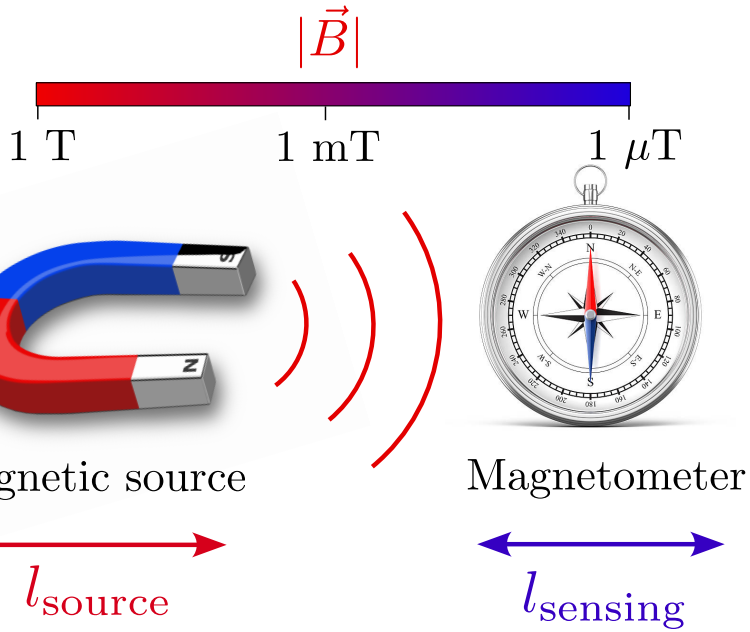
Optimum spatial resolution (imaging):

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

Key properties of magnetometers

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

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



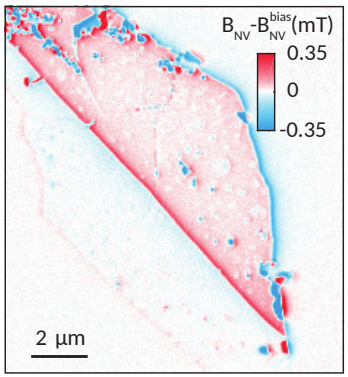
Optimum spatial resolution (imaging):

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

Magnetoencephalography

$|\vec{B}| \sim 10$ fT
$l \sim 1$ cm

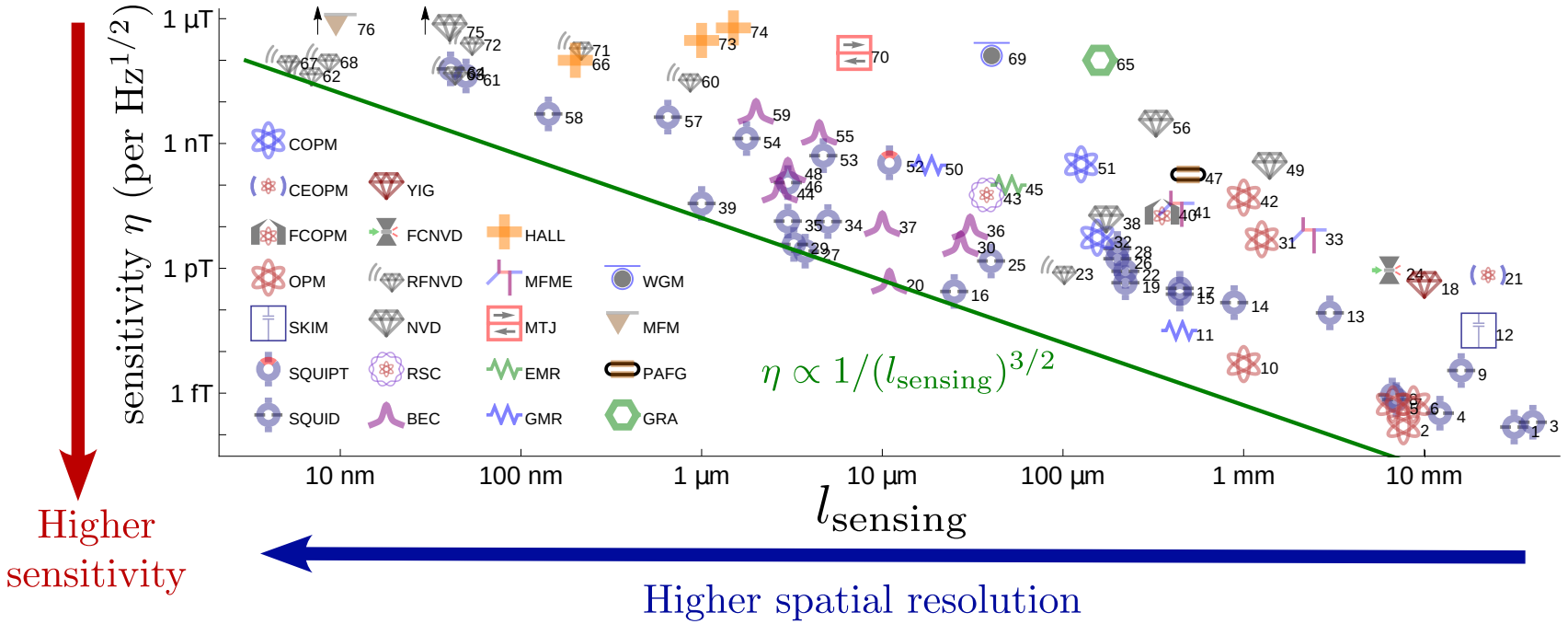
2D materials magnetism



$|\vec{B}| \sim 100$ μT
$l \sim 50$ nm

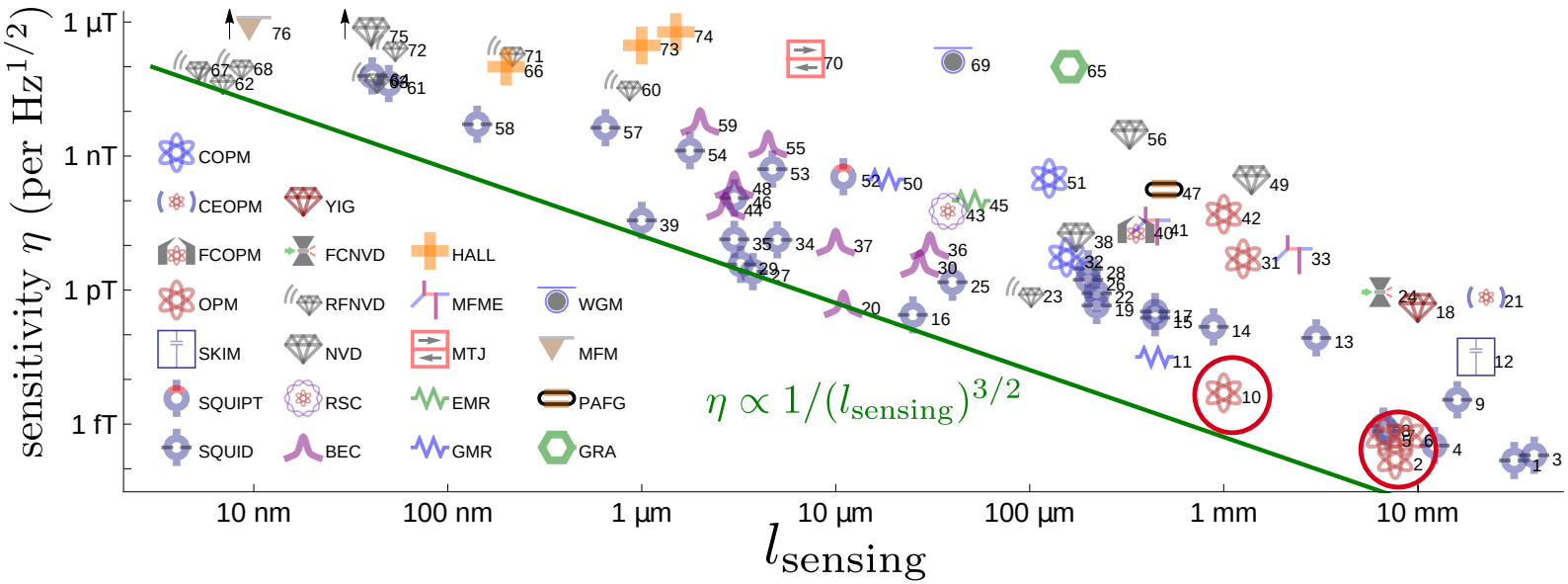
Thiel, L. et al (2019). Science, 364(6444), 973-976.

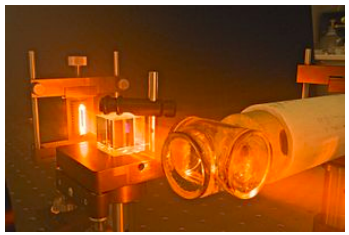
Sate of the art magnetometers



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

Sate of the art magnetometers



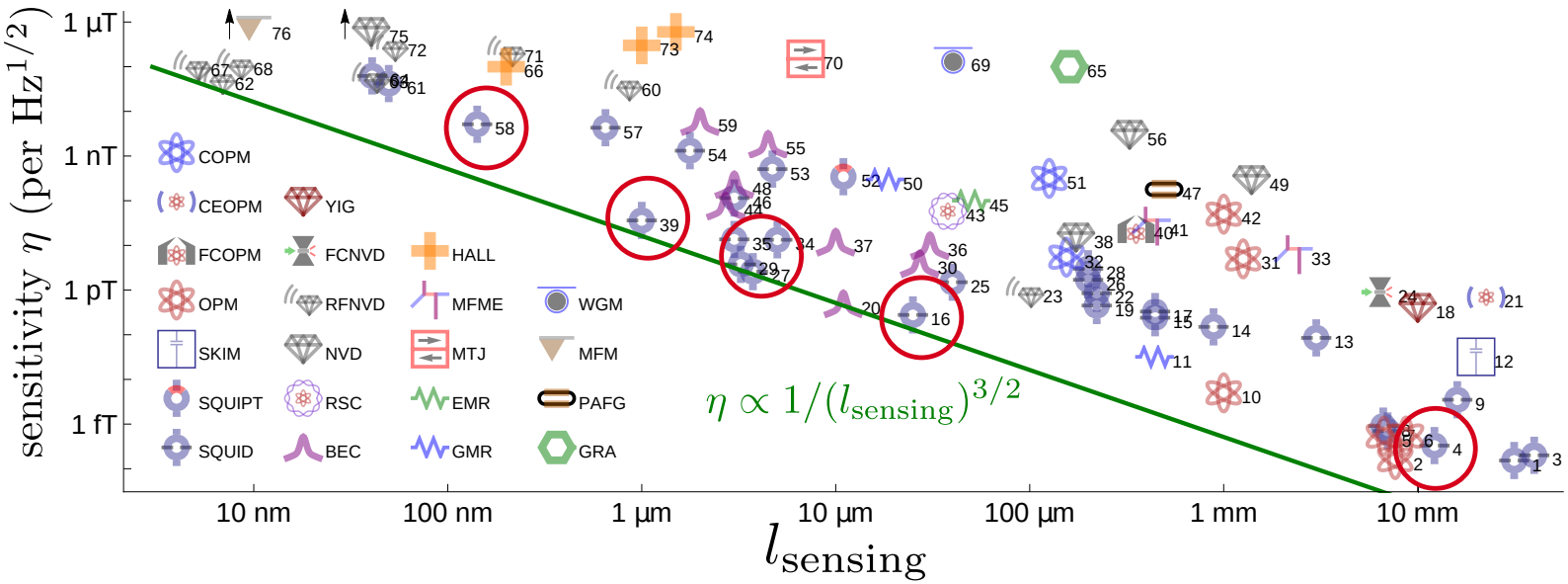


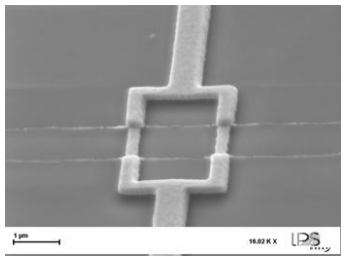
Optically pumped magnetometers (OPM)

- 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



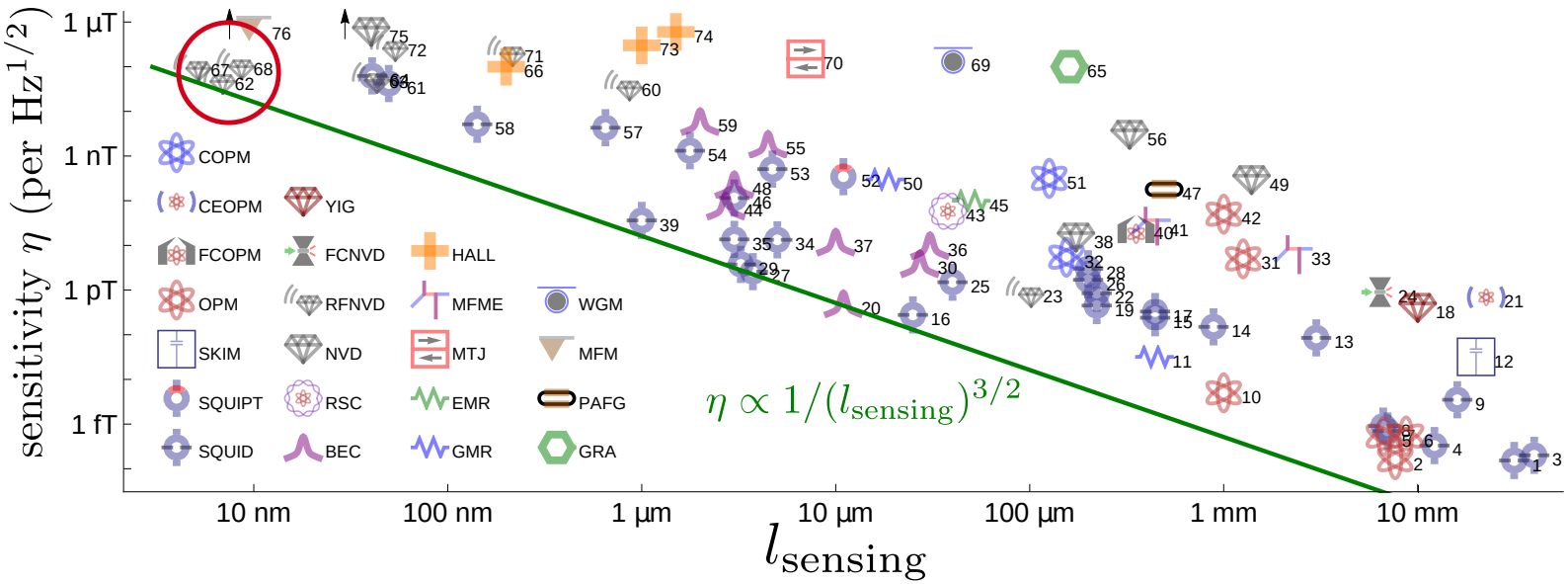


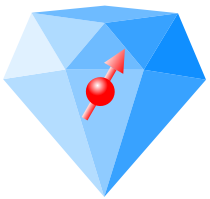
Superconducting quantum interference device (SQUID)

- 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



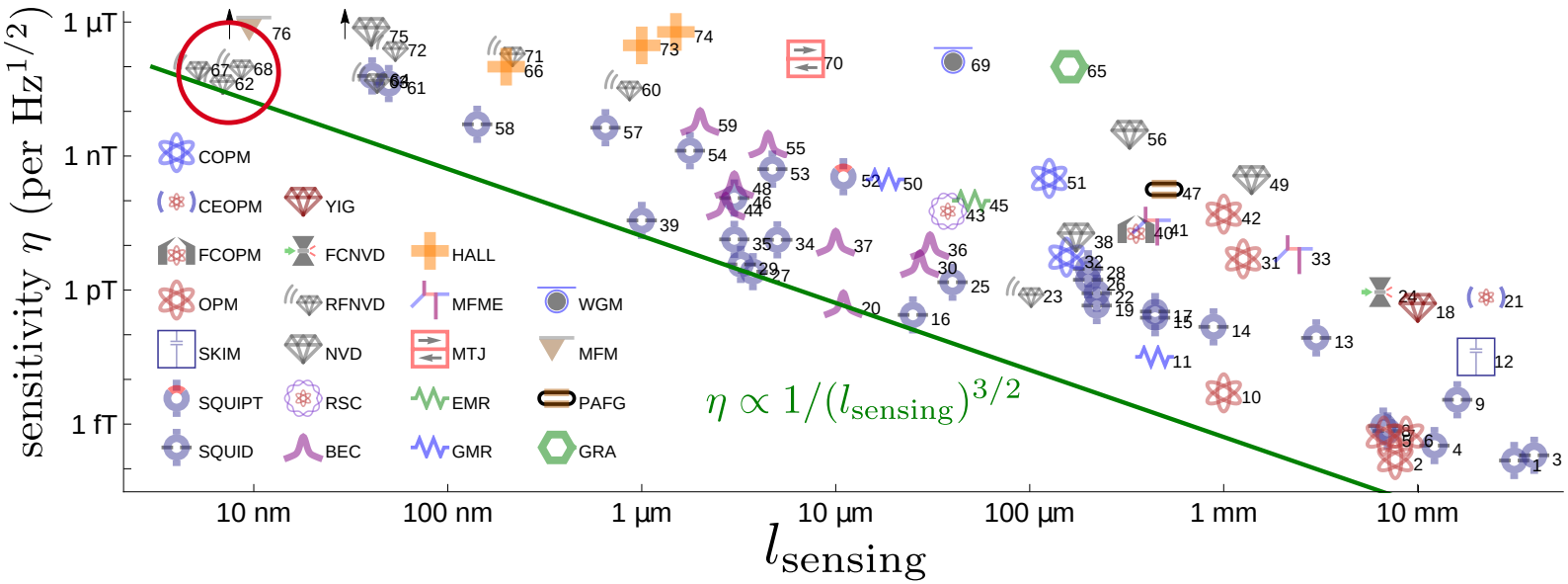


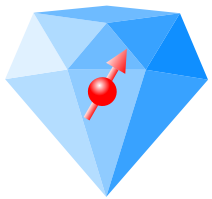
(Single) NV center

- nm resolution
- Room temperature

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

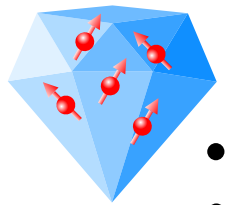
State of the art magnetometers





(Single) NV center

- nm resolution
- Room temperature

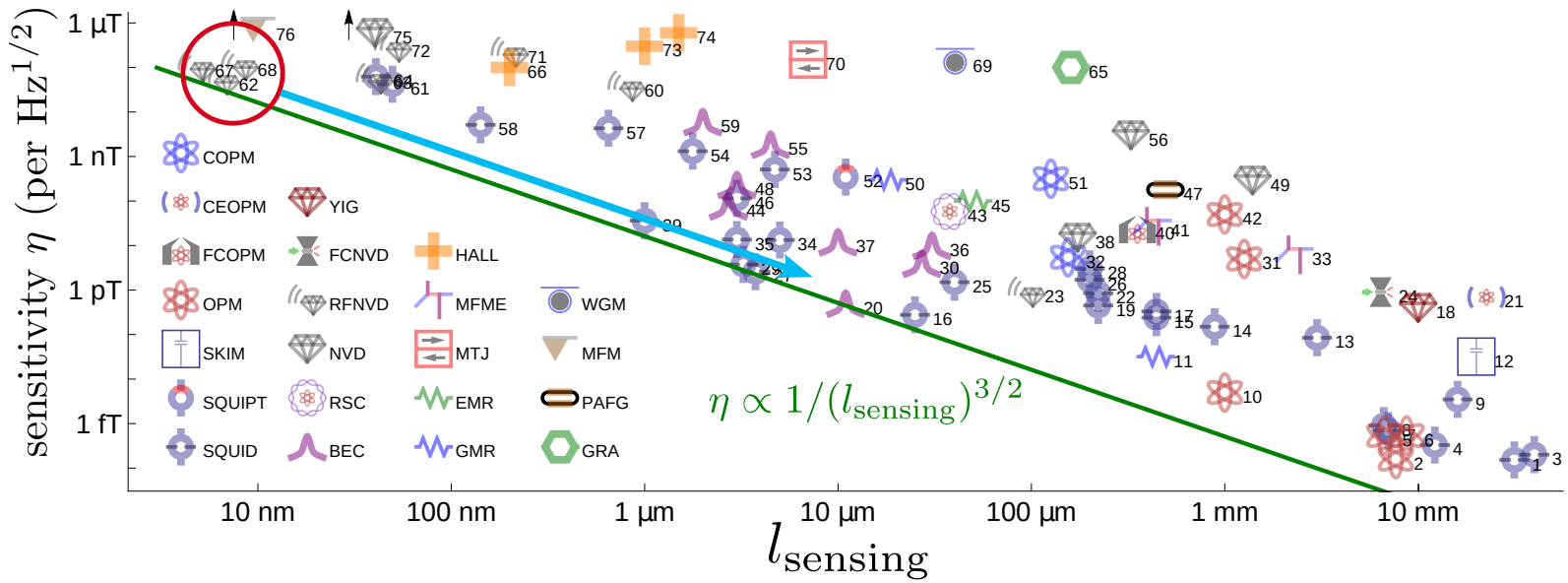


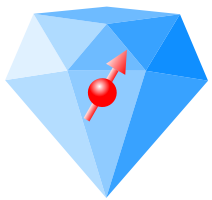
NV ensemble

- 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

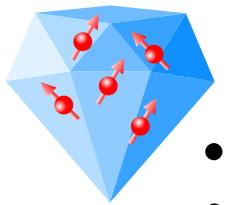




(Single) NV center

- nm resolution
- Room temperature

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

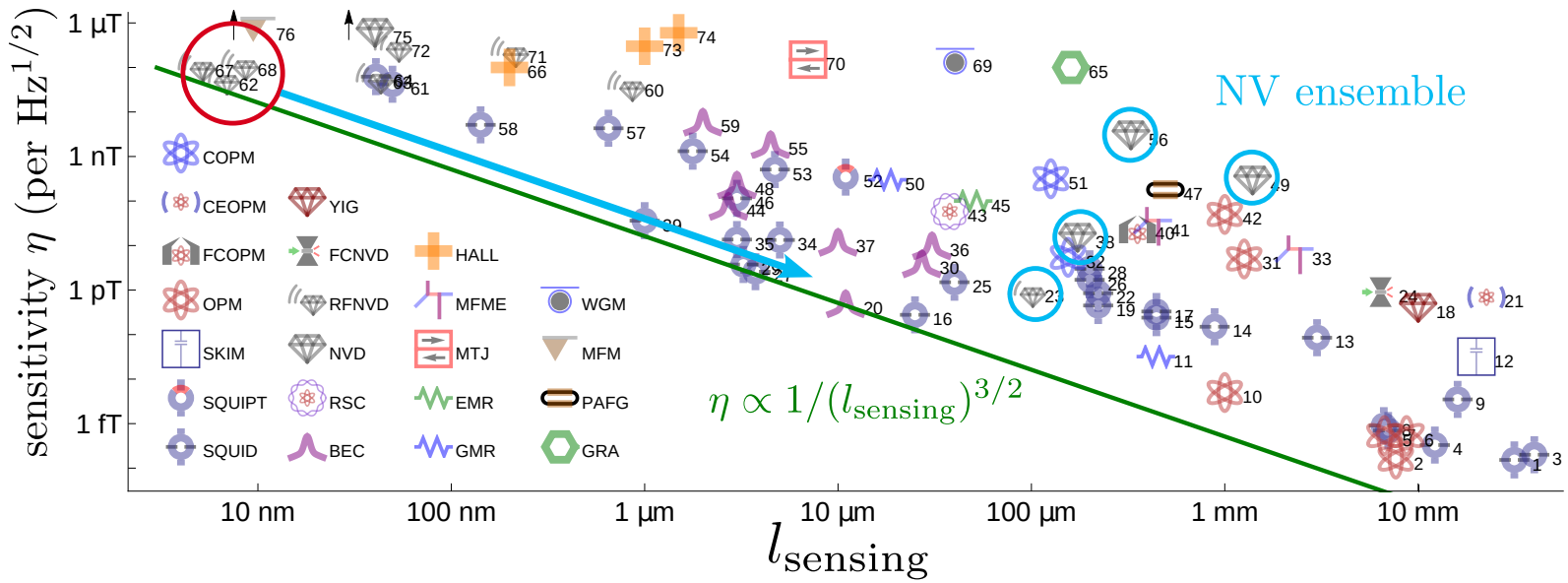


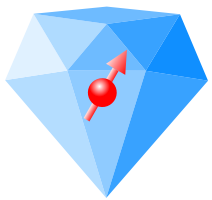
NV ensemble

- μm resolution
- Room temperature
- Higher sensitivity

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

State of the art magnetometers



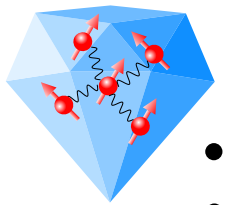


(Single) NV center

- nm resolution
- Room temperature

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

Interactions

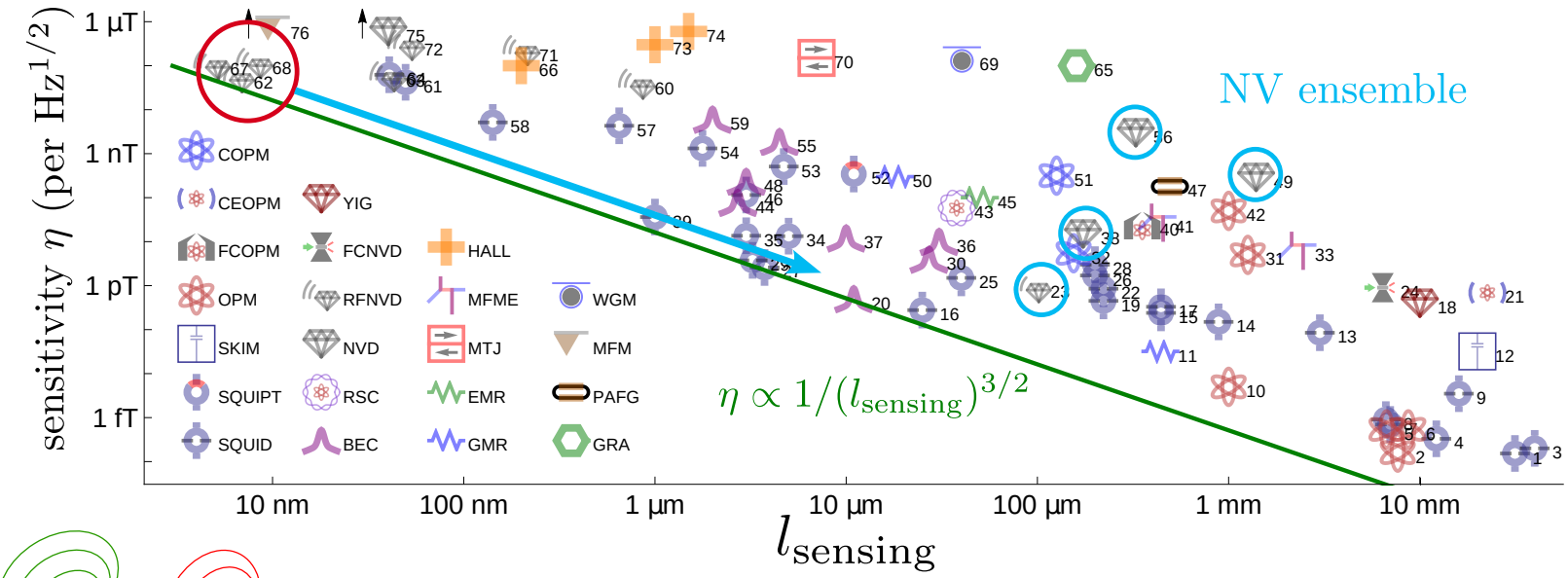


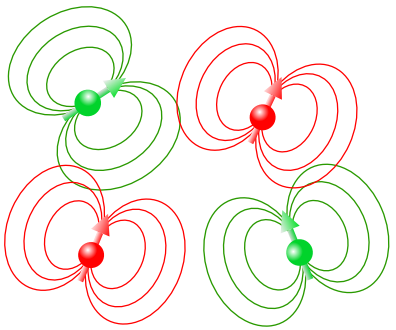
NV ensemble

- 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





= NV

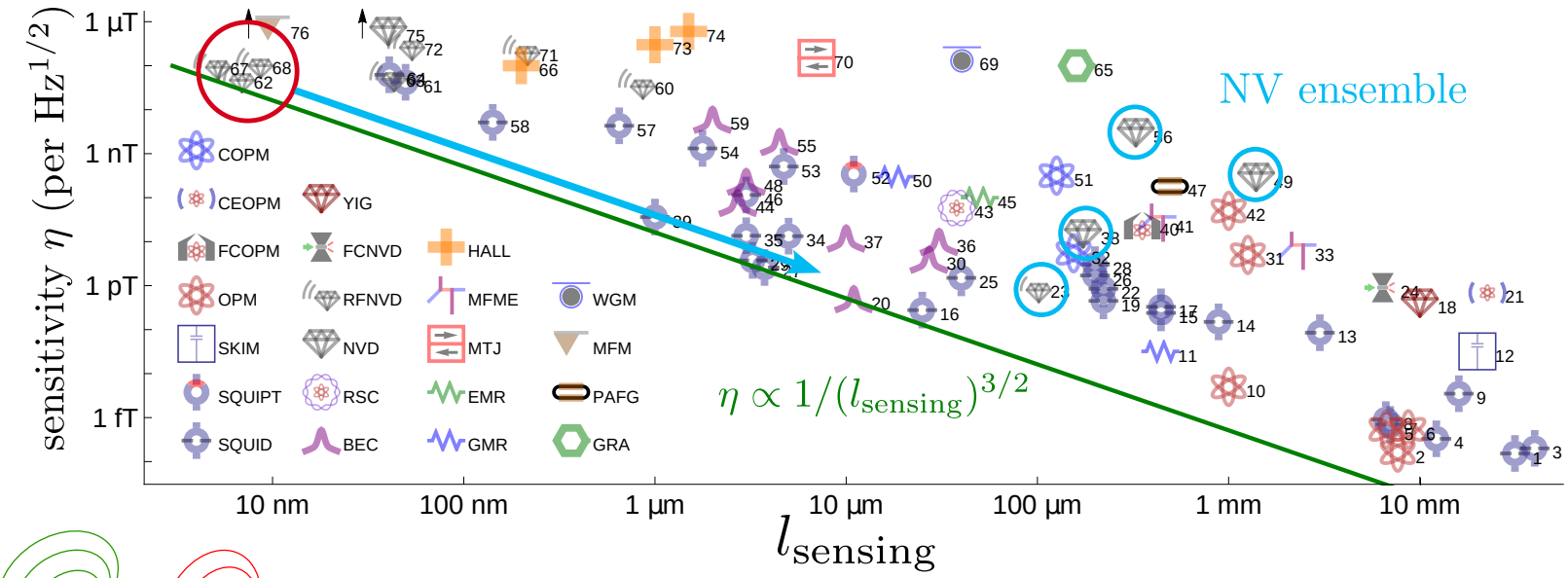
= P1

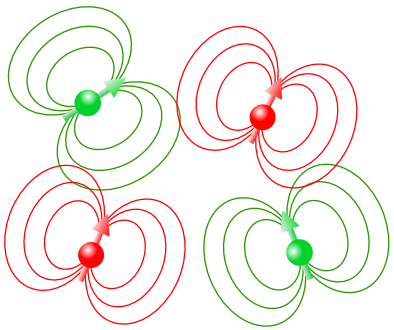
Interactions:

- Spectral broadening
- Modified spin dynamics

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

State of the art magnetometers





= NV

= P1

Interactions:

- Spectral broadening
- Modified spin dynamics

Solutions:

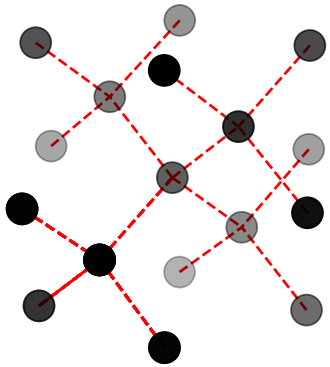
- Decoupling interactions (Hamiltonian engineering)
- Exploiting interactions

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

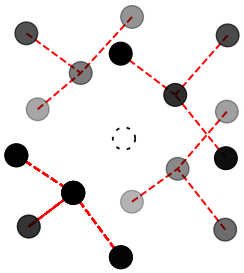
Outline

Colored centers in diamond

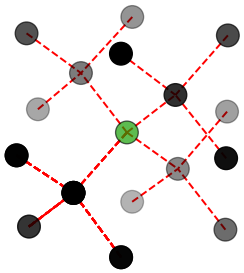
Diamond crystal lattice



Point-like defects



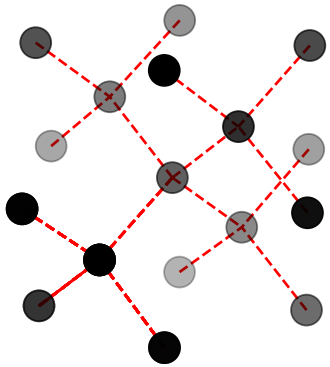
Vacancy



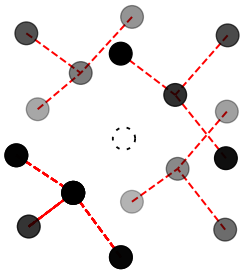
Substitution

Colored centers in diamond

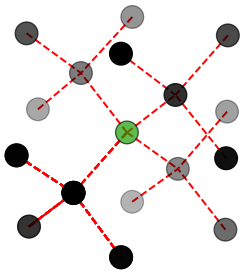
Diamond crystal lattice



Point-like defects

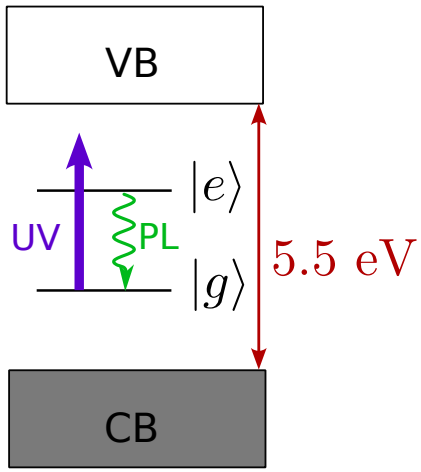


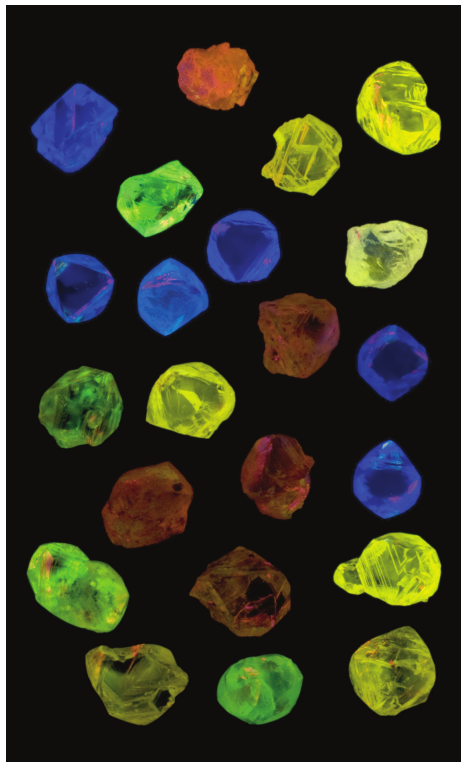
Vacancy



Substitution

Colored center fluorescence

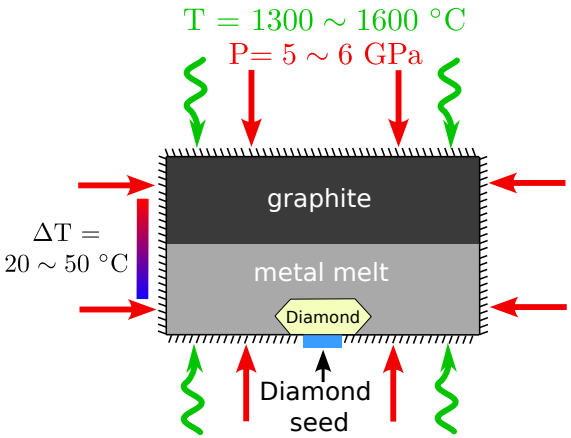


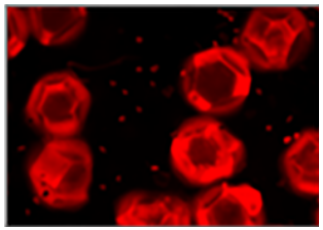


Natural diamonds fluorescence under UV light

Synthetic diamond and NV centers

Highe Pressure High Temperature (HPHT)

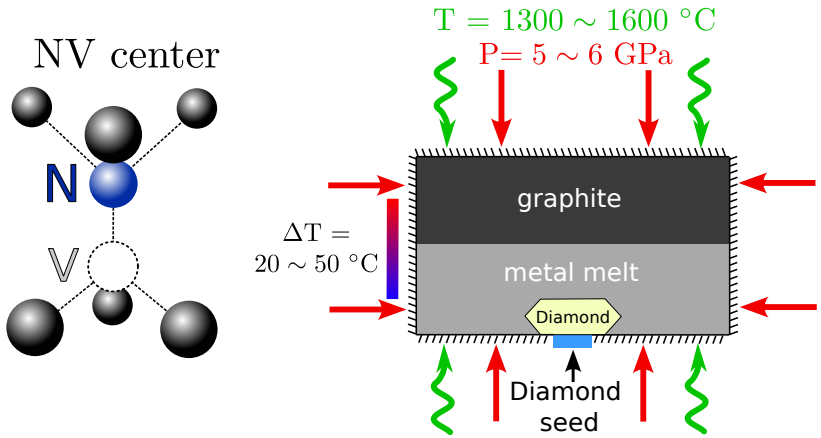


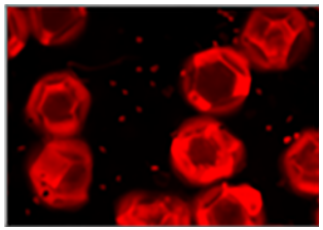


Adamas 15/150 μm

Synthetic diamond and NV centers

Highe Pressure High Temperature (HPHT)





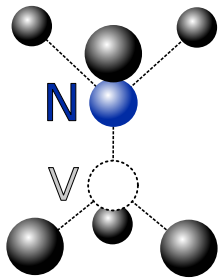
Adamas 15/150 μm

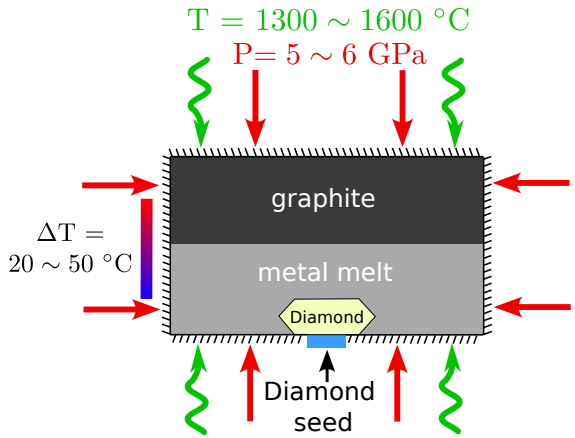
[NV] $\approx$ 3 ppm

Synthetic diamond and NV centers

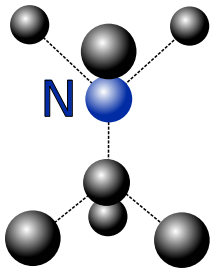
Highe Pressure High Temperature (HPHT)

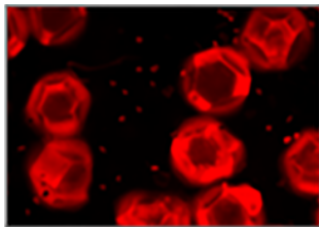
NV center





N_s (P1 center)





Adamas 15/150 μm

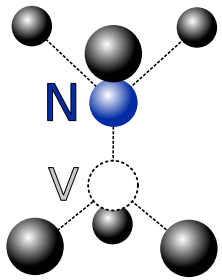
[NV] $\approx$ 3 ppm

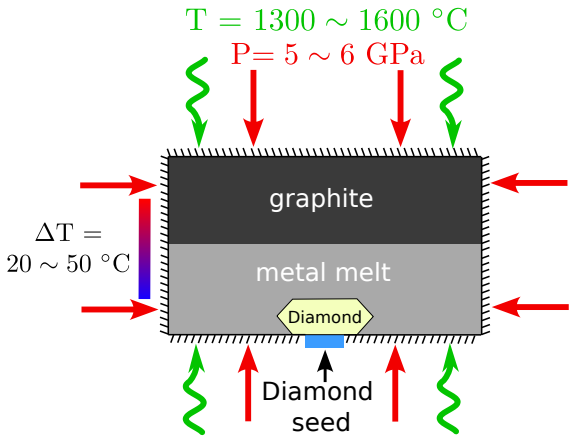
[P1] $\approx$ 100 ppm

Synthetic diamond and NV centers

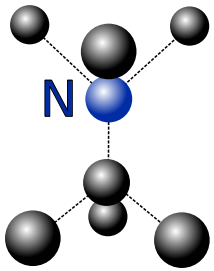
Highe Pressure High Temperature (HPHT)

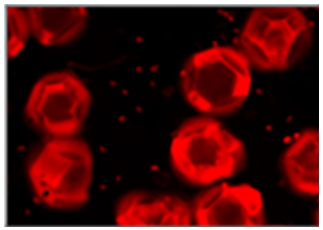
NV center





N_s (P1 center)



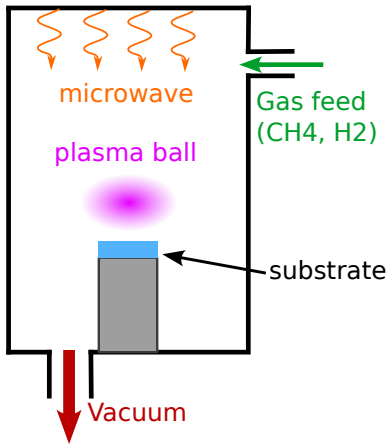


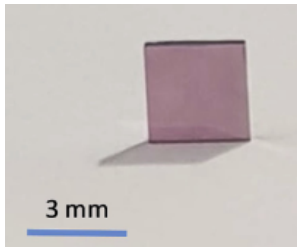
Adamas 15/150 μm

[NV] $\approx$ 3 ppm

[P1] $\approx$ 100 ppm

Chemical Vapour Deposition (CVD)





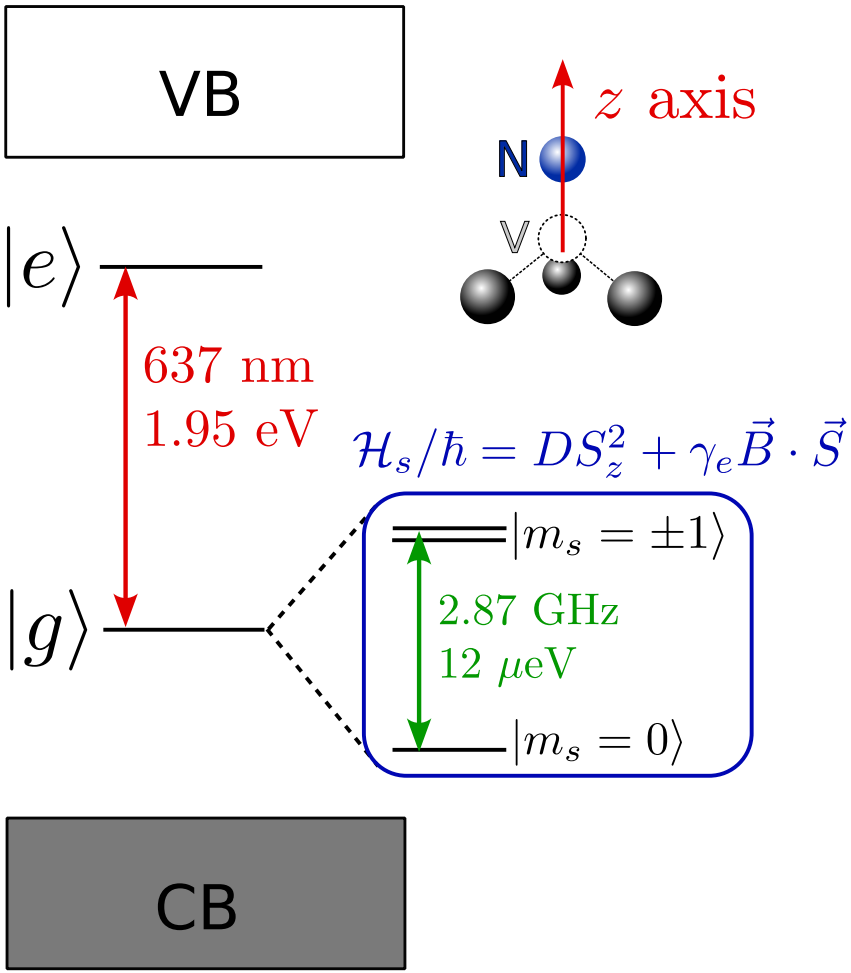
IRCP-LSPM

[NV] $\approx$ 4.5 ppm

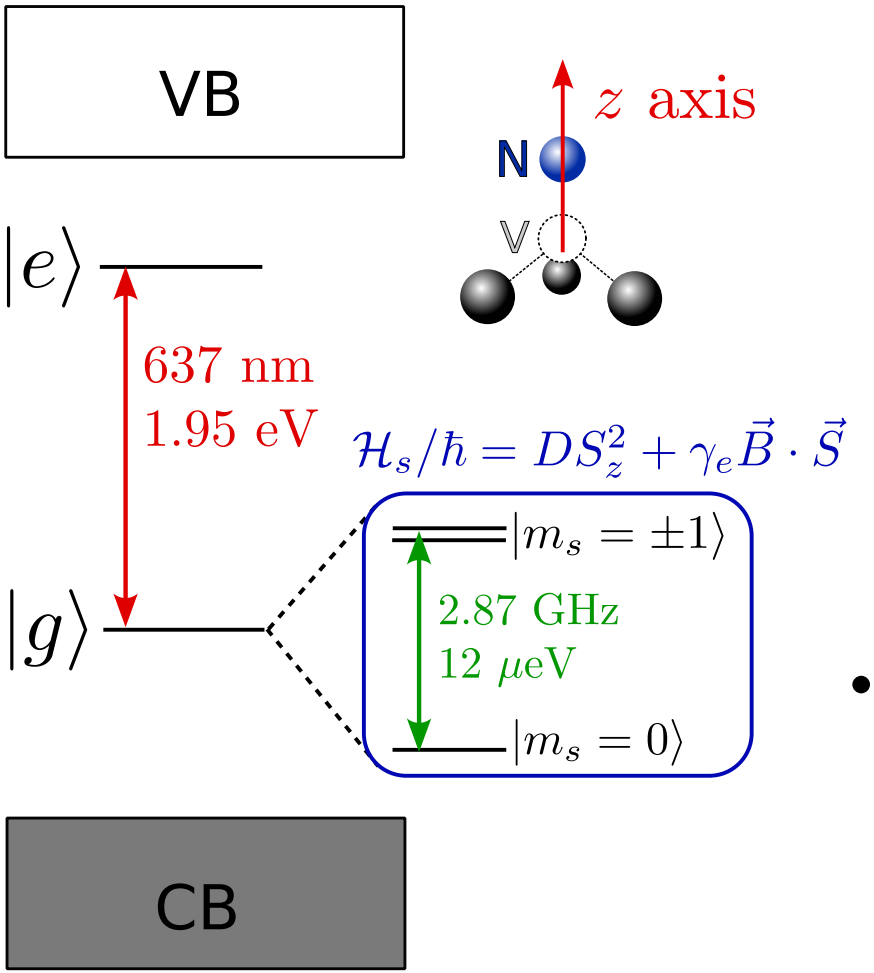
[P1] $\approx$ 25 ppm

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

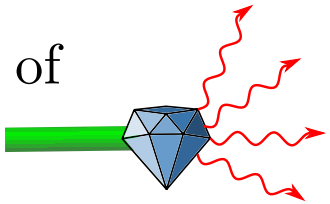
The NV center energy levels

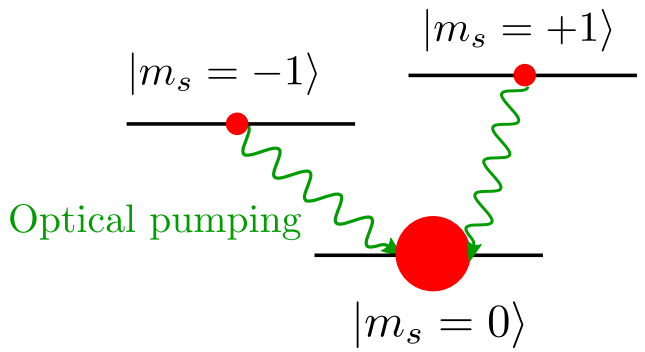


The NV center energy levels



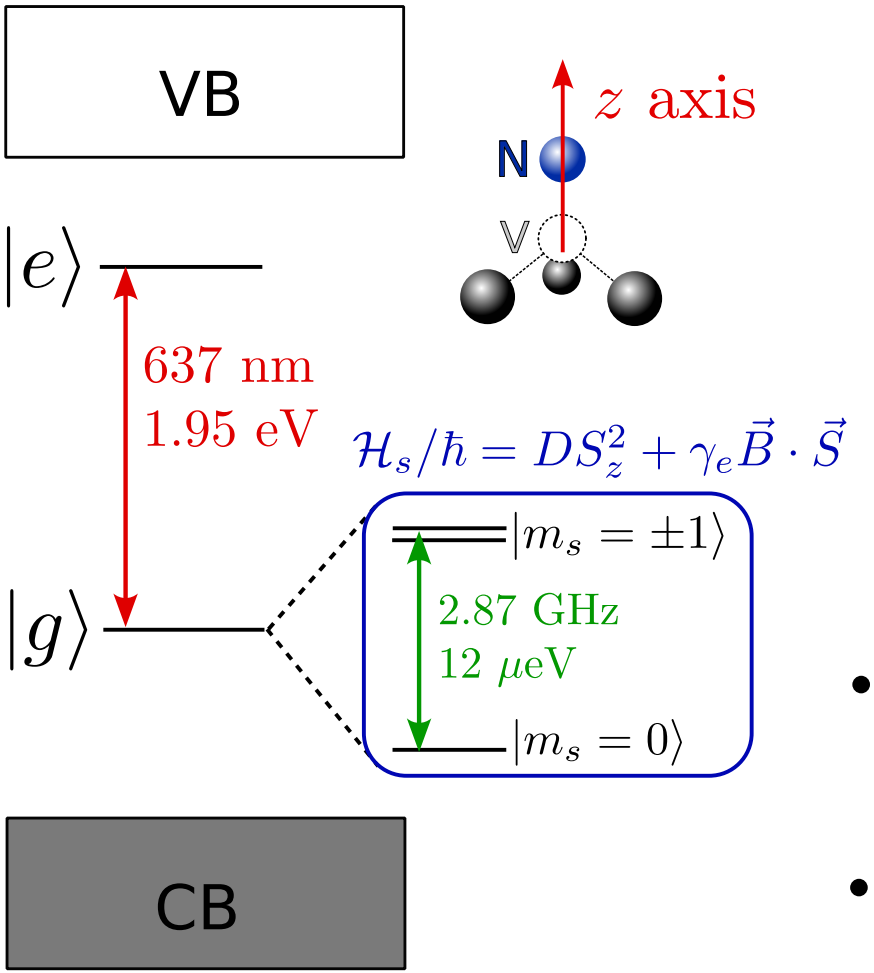
Optical pumping of the spin states

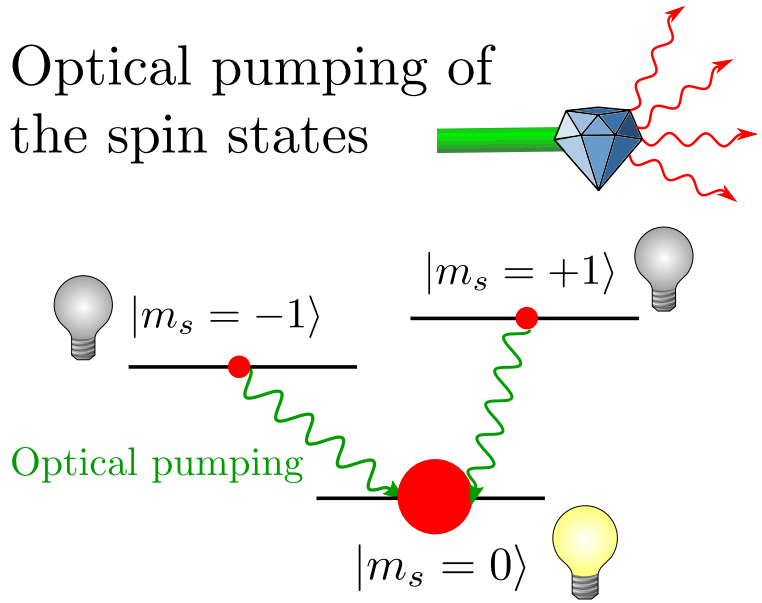




- Population accumulation in the $|0\rangle$ state
  - ↳ Initialization of the spin state

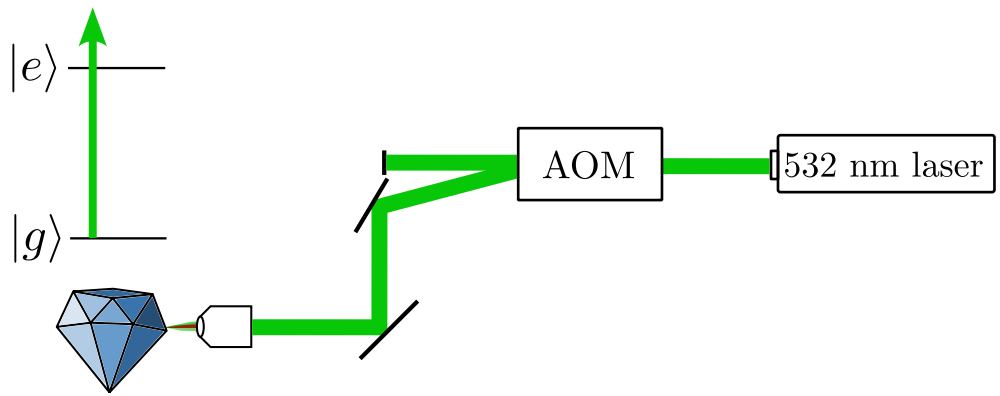
The NV center energy levels



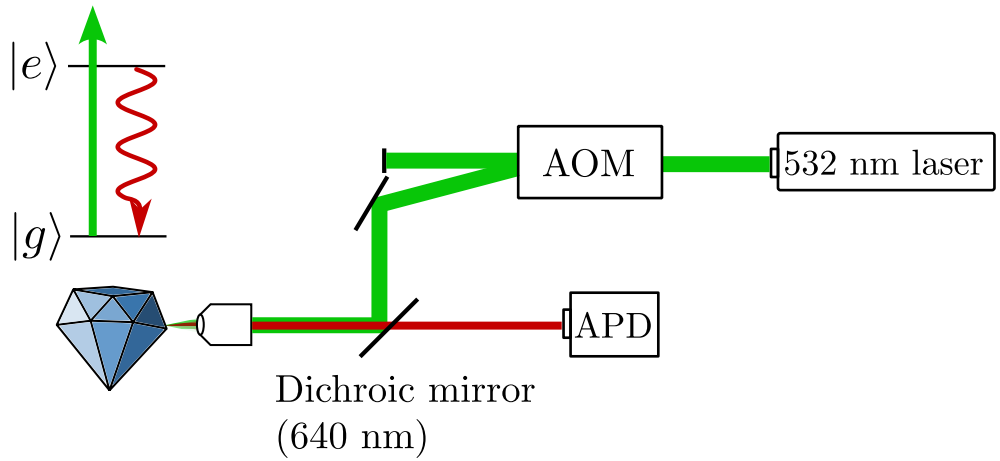


- Population accumulation in the $|0\rangle$ state
  - → Initialization of the spin state
- $|0\rangle$ sate brighter than $|\pm 1\rangle$ states
  - → Optical readout of the spin state

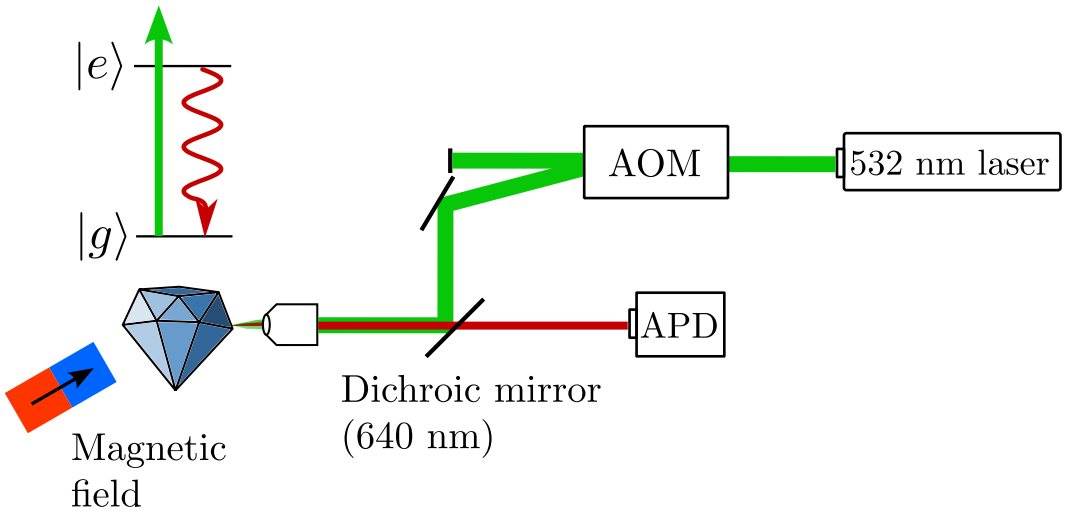
Experimental setup



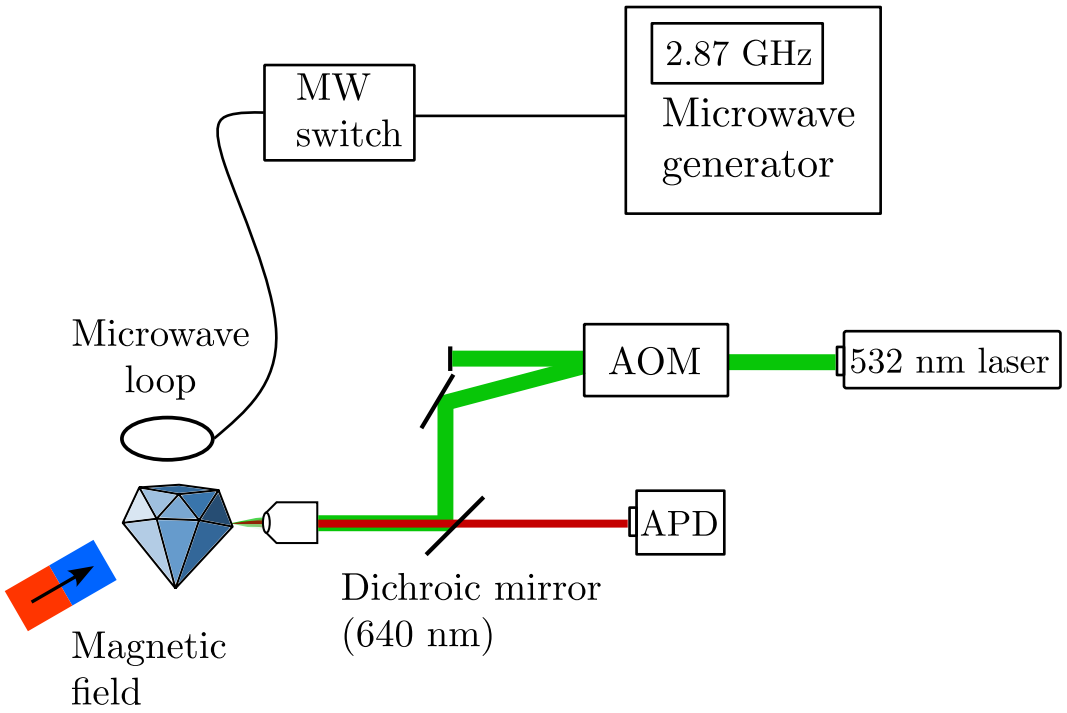
Experimental setup



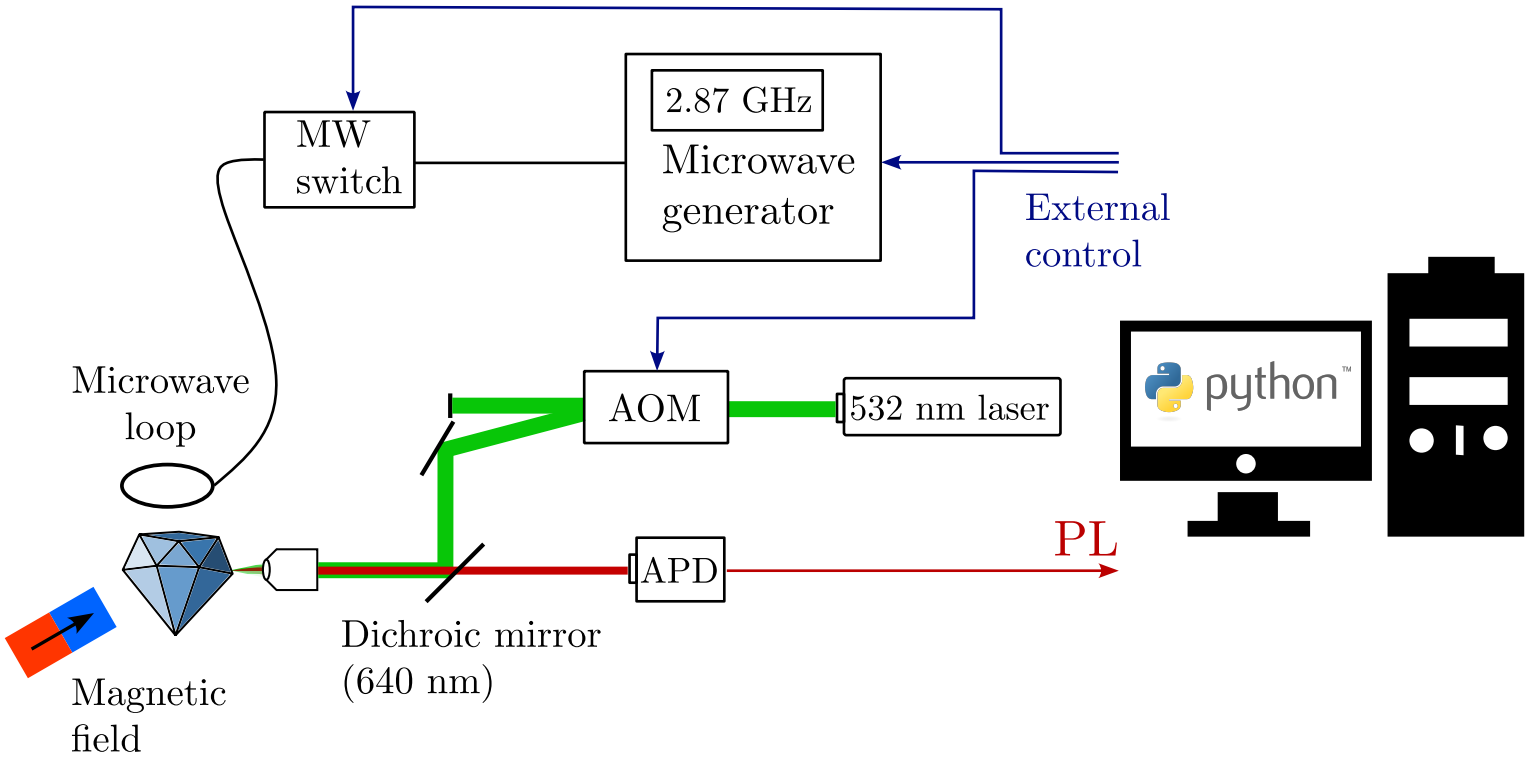
Experimental setup



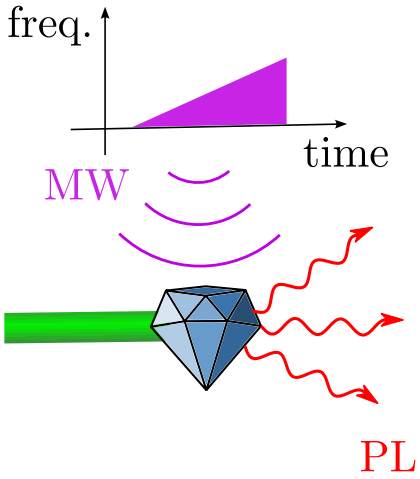
Experimental setup

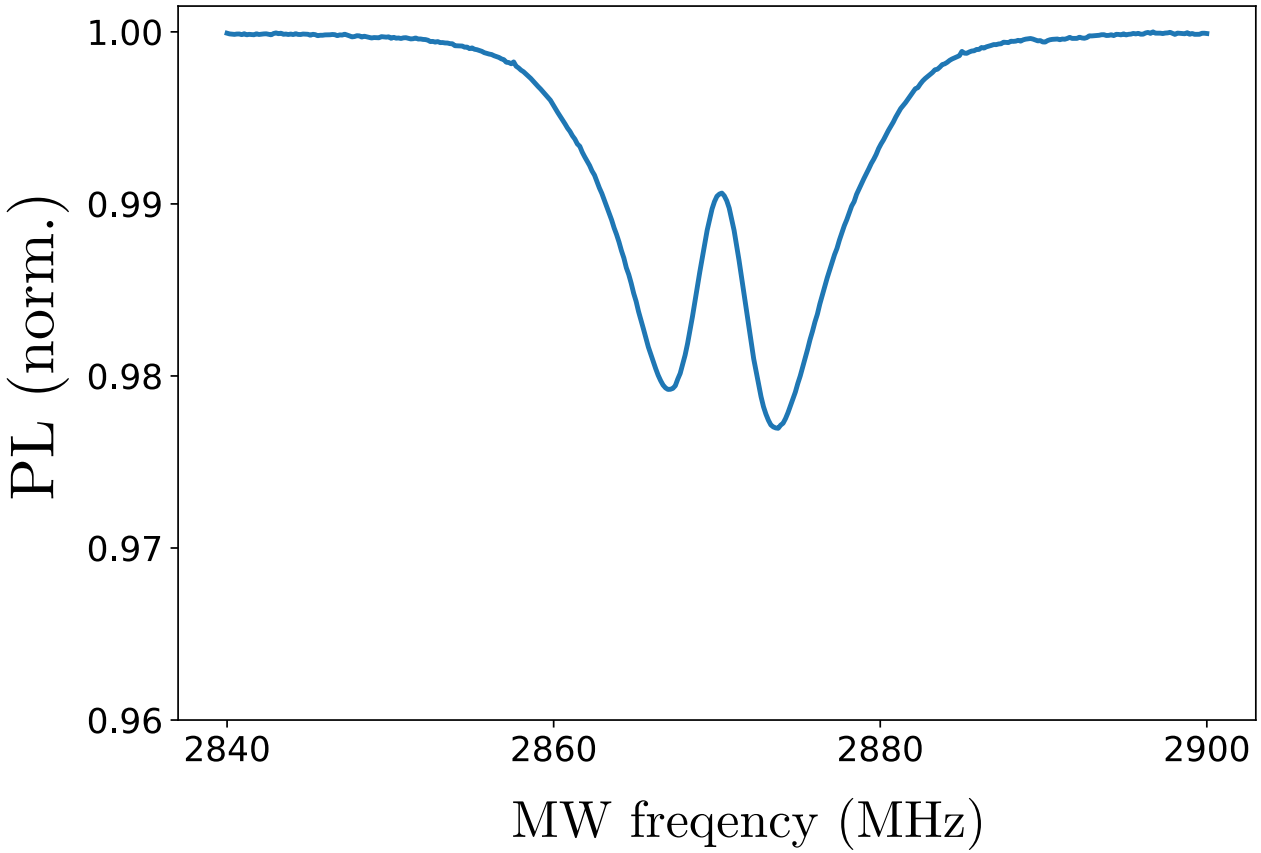


Experimental setup

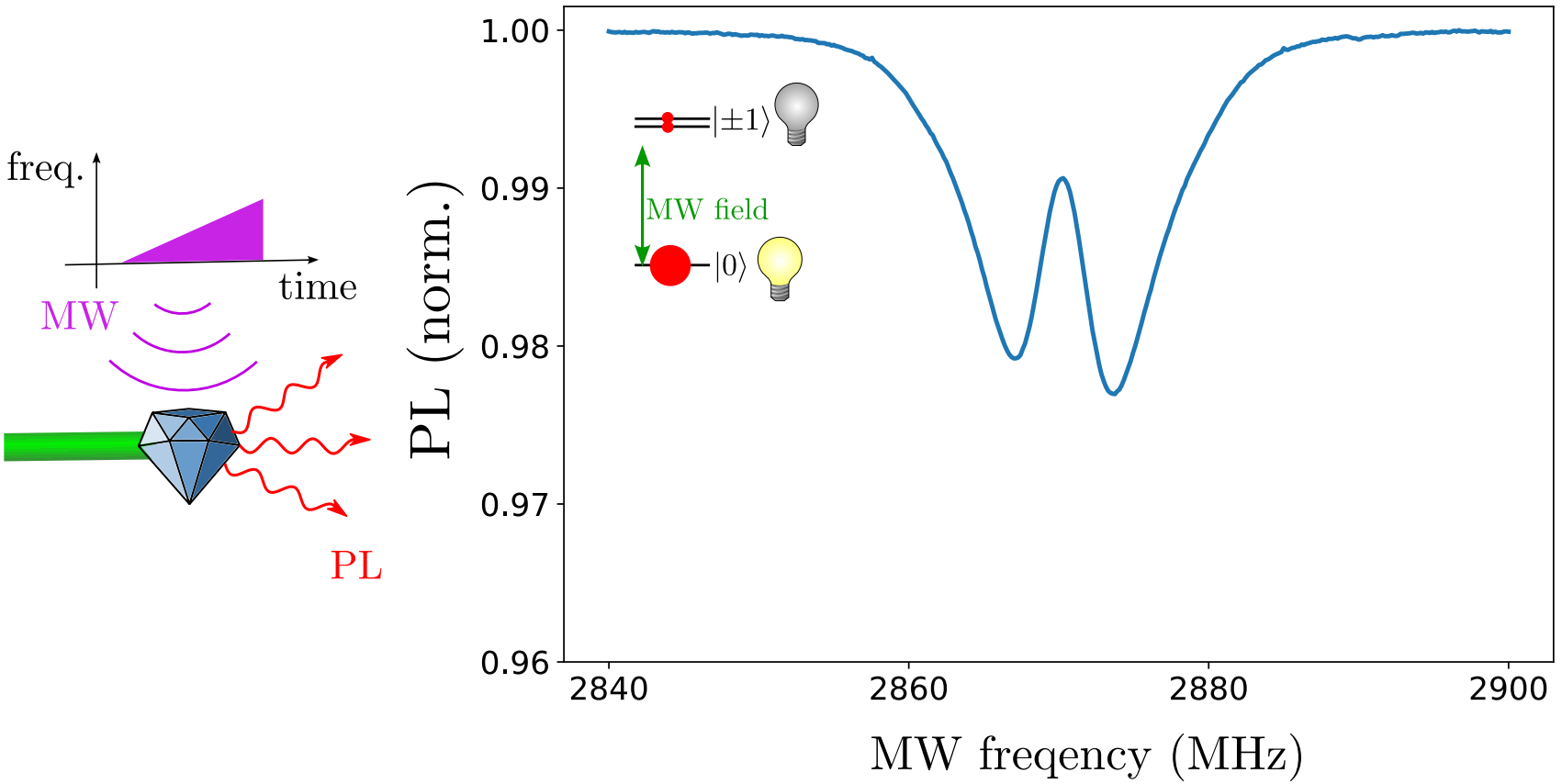


Optically detected magnetic resonance (ODMR)

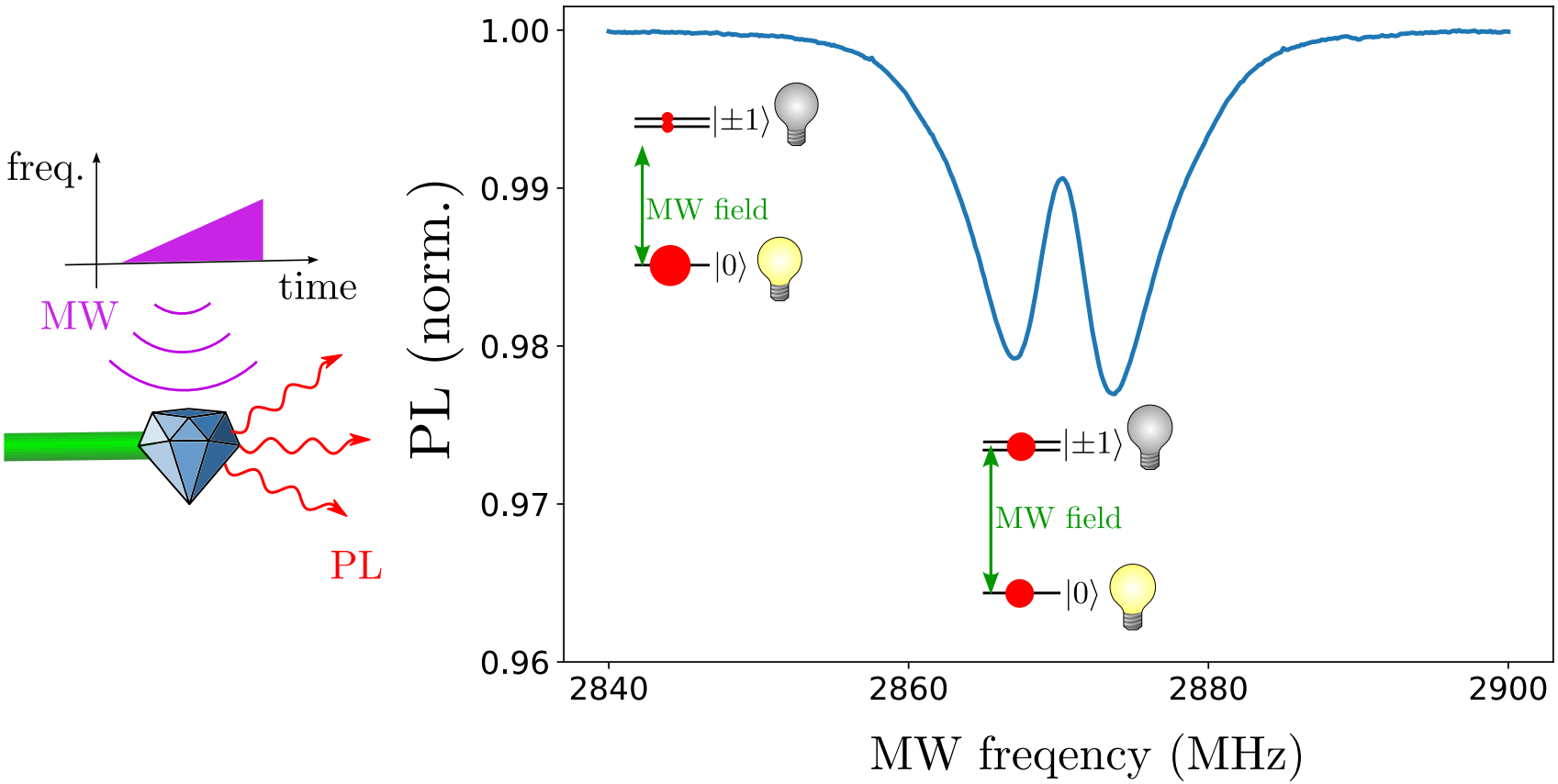




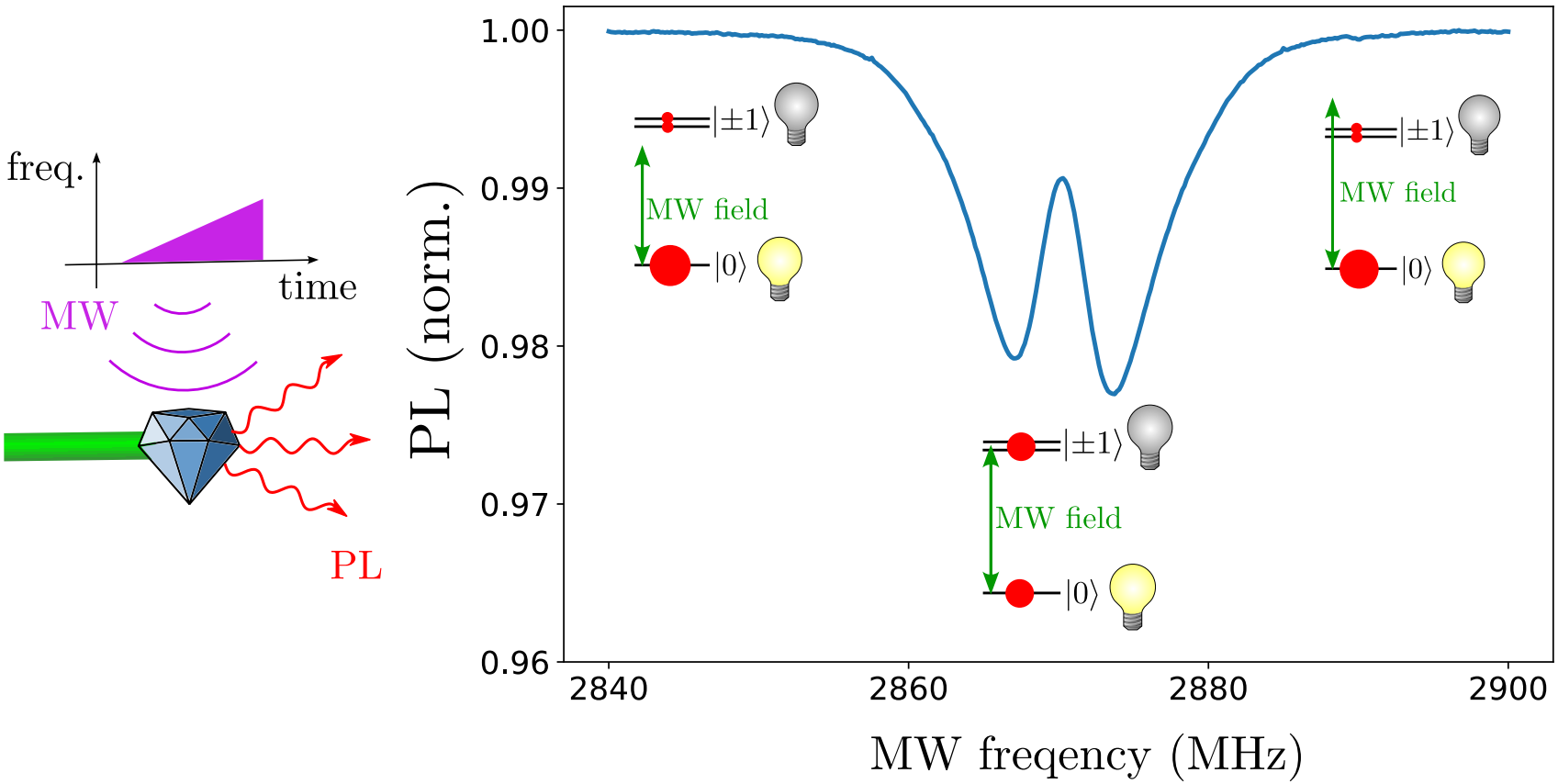
Optically detected magnetic resonance (ODMR)



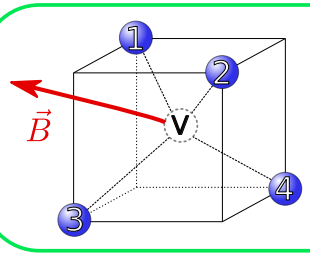
Optically detected magnetic resonance (ODMR)



Optically detected magnetic resonance (ODMR)

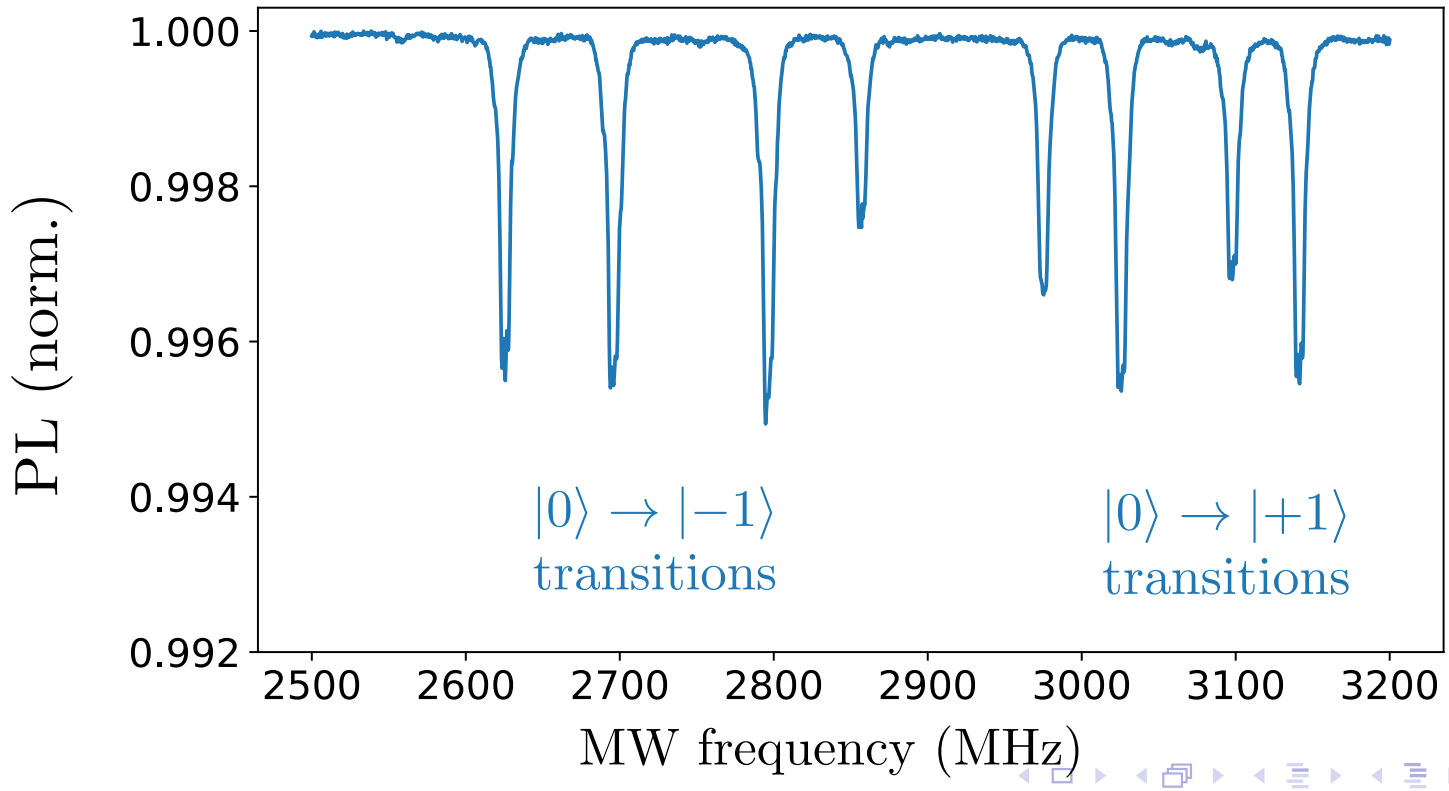


ODMR with NV ensemble: the 4 classes

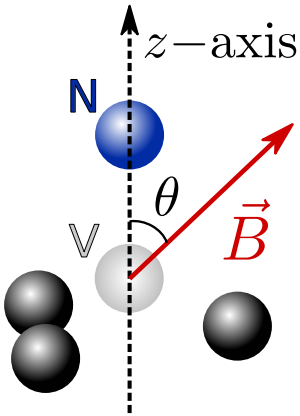


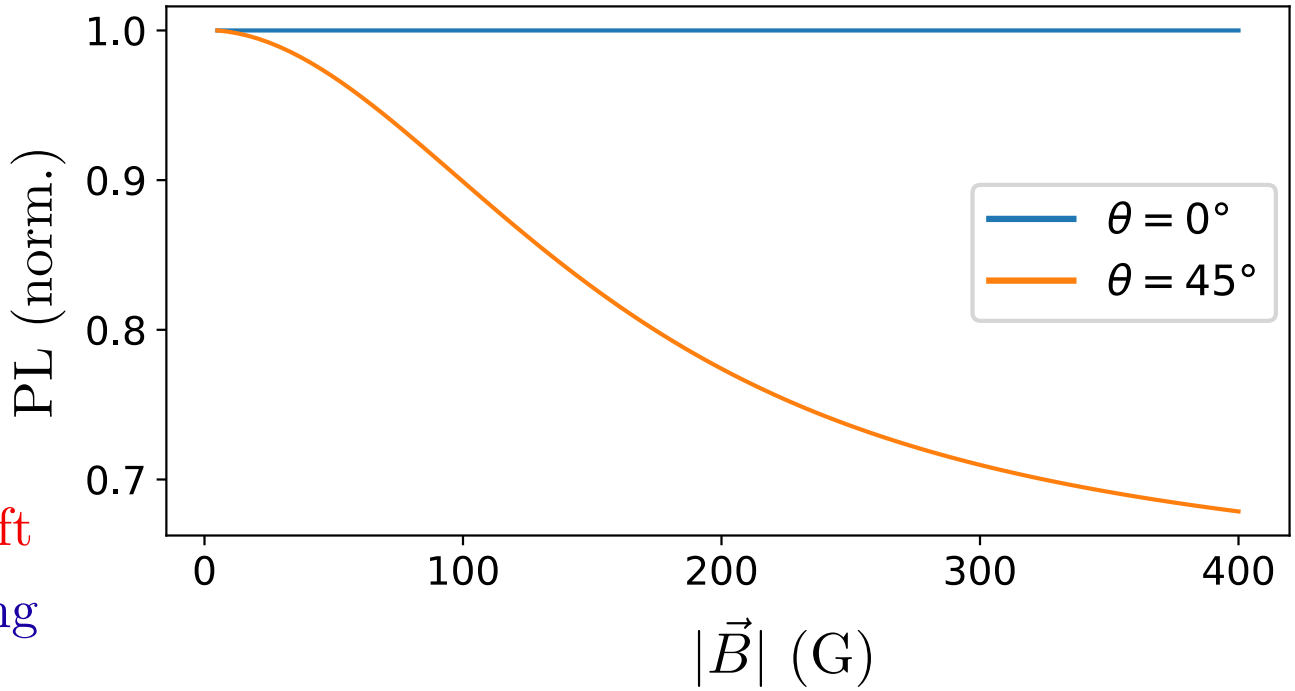
4 different projections of $\vec{B}$ over the 4 possible NV axes
$\rightarrow$ 4 classes of resonances

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



Transverse magnetic field effect





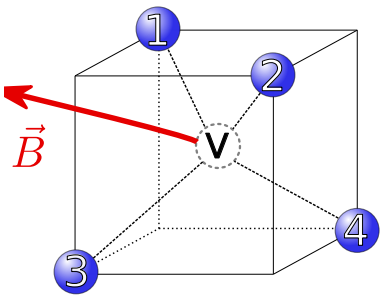
$B_{\parallel}$= Zeeman shift

$B_{\perp}$= State mixing

↳ Loss of polarization, PL decrease

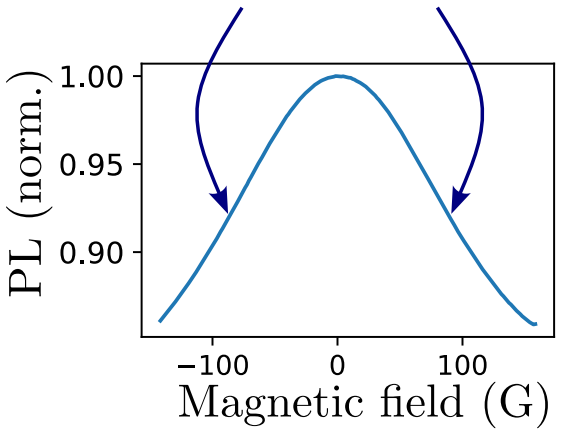
Outline

Depolarization of dense NV ensemble at low magnetic field

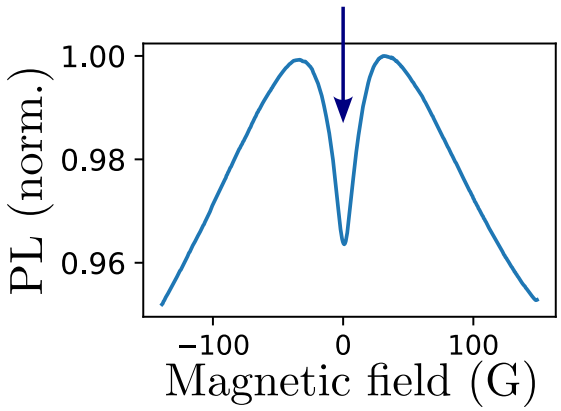


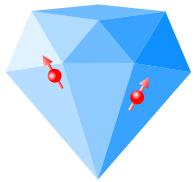
Non-zero transverse magnetic field on all 4 classes

Transverse field

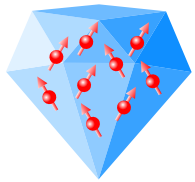


Dipole-dipole spin relaxation





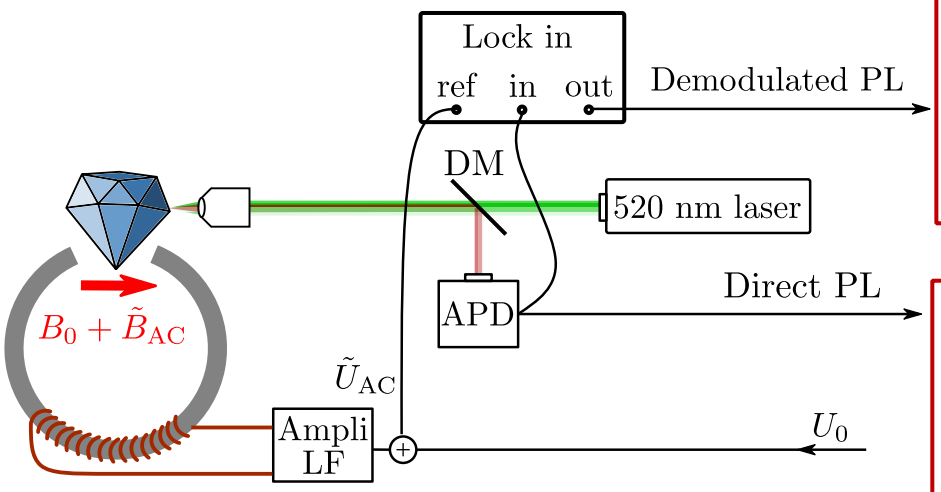
Low NV density
[NV] ≤ 100 ppb

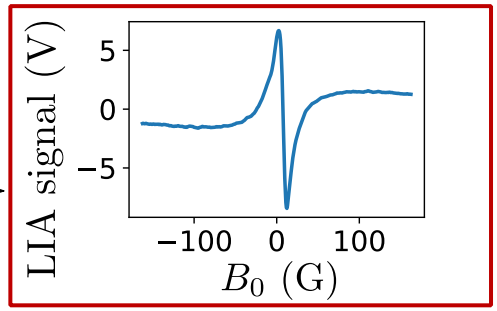


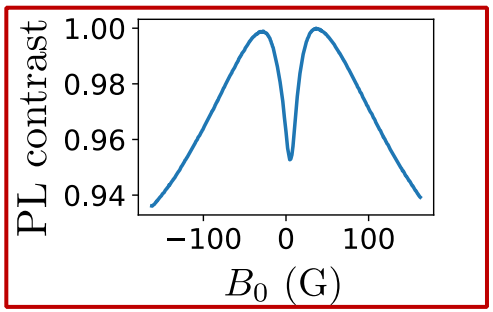
High NV density
[NV] ≥ 1 ppm

LFDM experimental setup

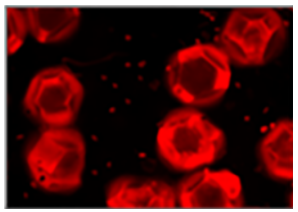
Experimental setup







Samples

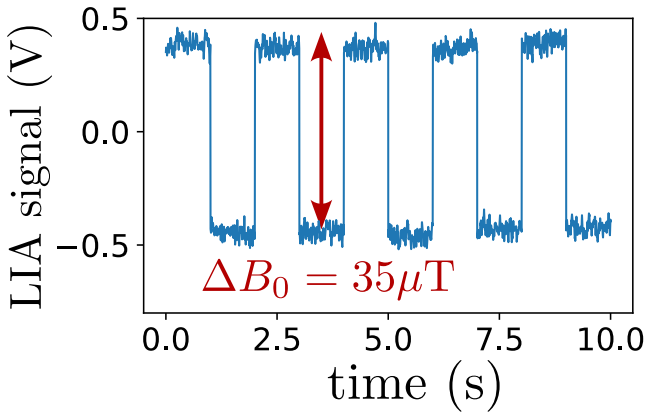


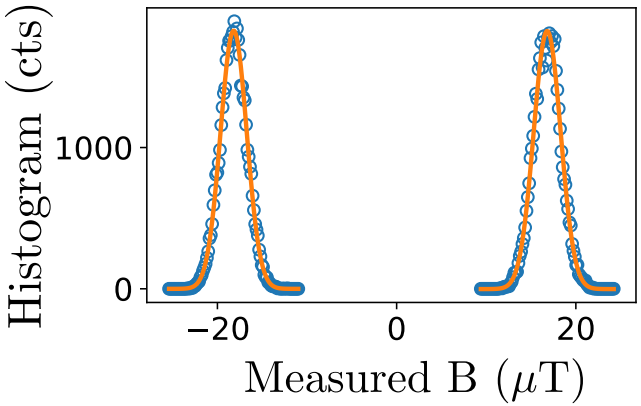
Adamas 15 μm

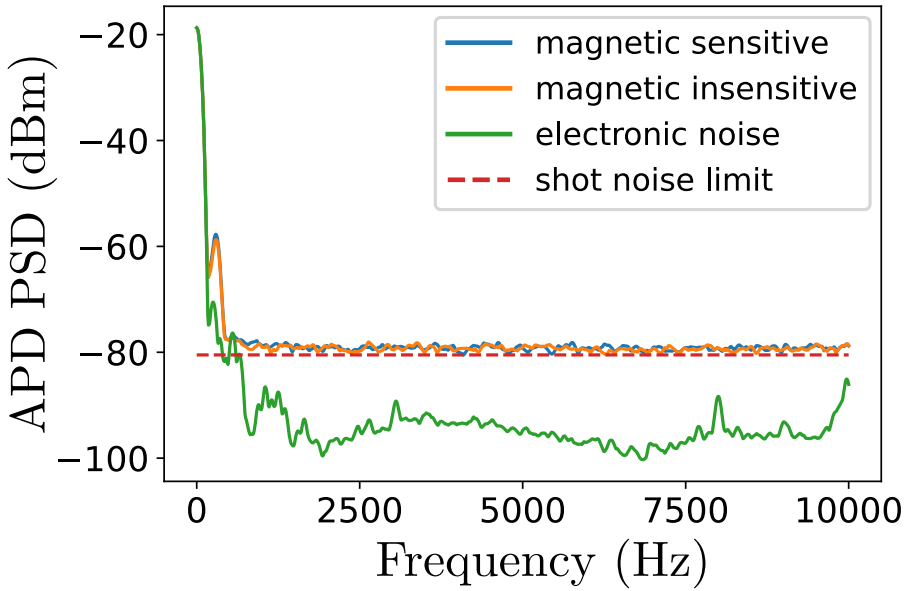
Experimental parameters

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

Sensitivity of LFDM







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

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

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

Comparison with the state of the art

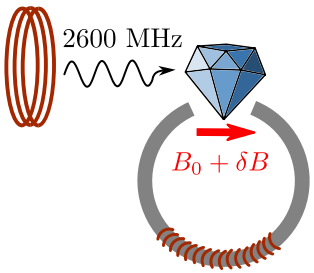
Sensitivity comparison

	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

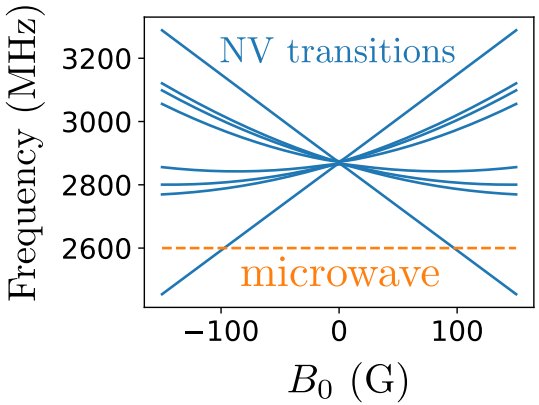
[1] Zheng, H.[...] Budker, D. (2020). Physical Review Applied, 13(4), 044023.
[2] Barry, J. F. [...] Walsworth, R. L (2016). PNAS, 113(49), 14133-14138.

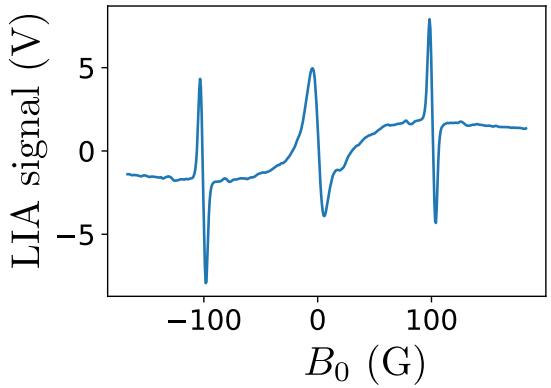
	ODMR	GSLAC	LFDM
Microwave free	✗	✓	✓
Low magnetic field (<10 G)	✓	✗	✓
Robust to T° and B-field inhomogeneities	✗	✗	✓
Orientation free (polycrystalline, powder)	✗	✗	✓

Comparison with CW ODMR

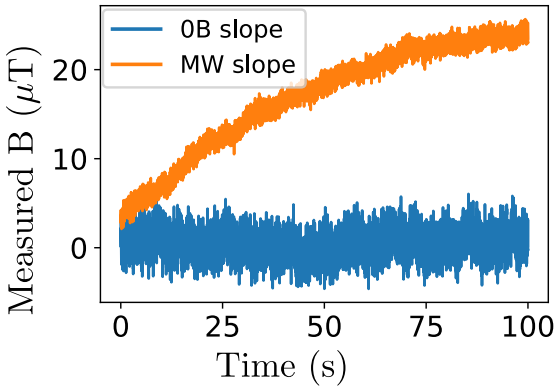


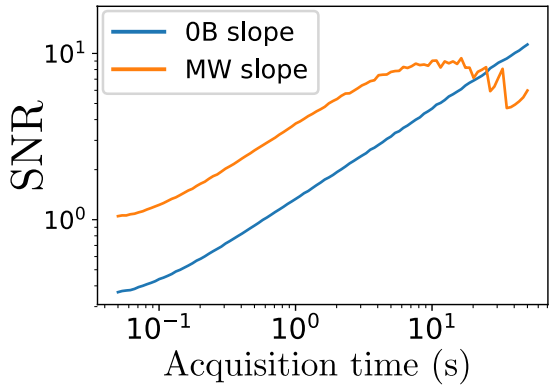
Adding a fixed microwave tone





Temporal stability



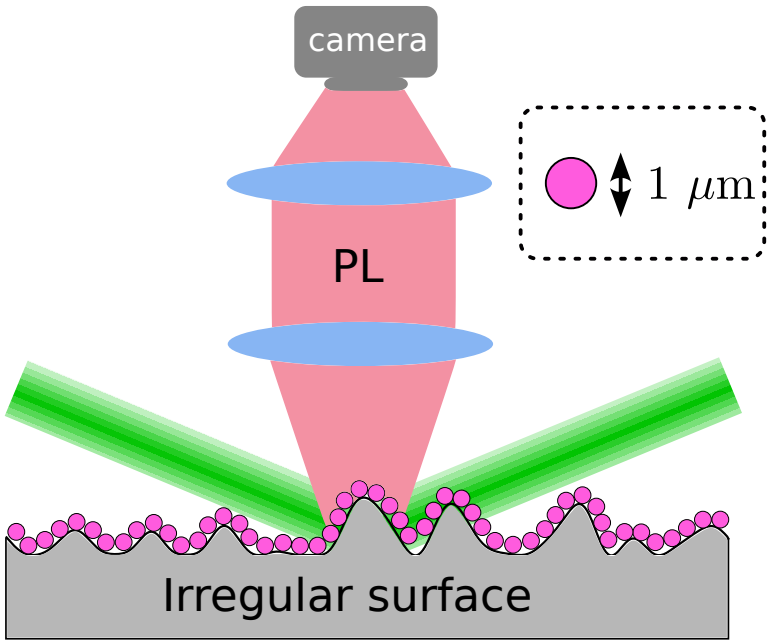


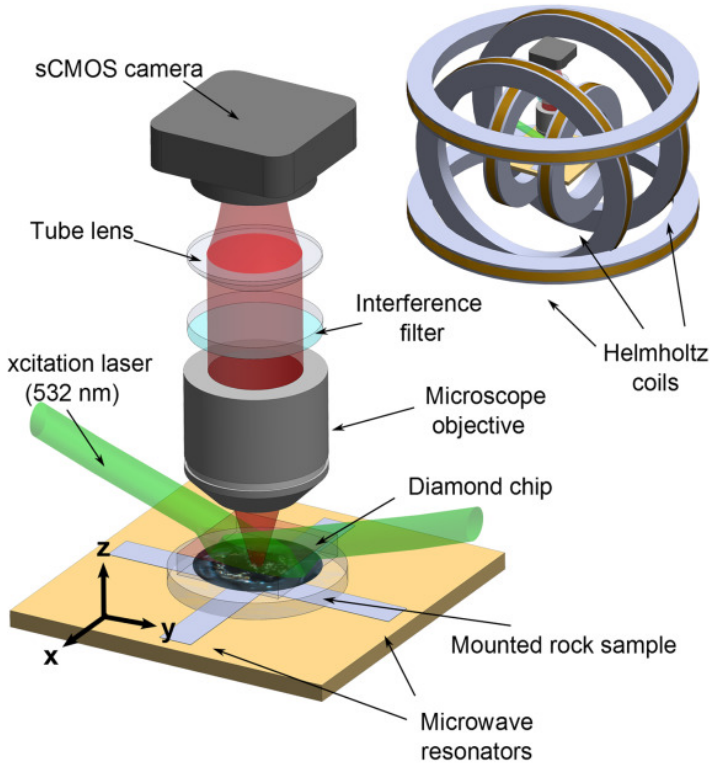
MW slope sensitivity: $\eta \approx 40$ nT/$\sqrt{\text{Hz}}$

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

Application: wide-field magnetometry on irregular surfaces

(Commercially available 1 μm diamonds)





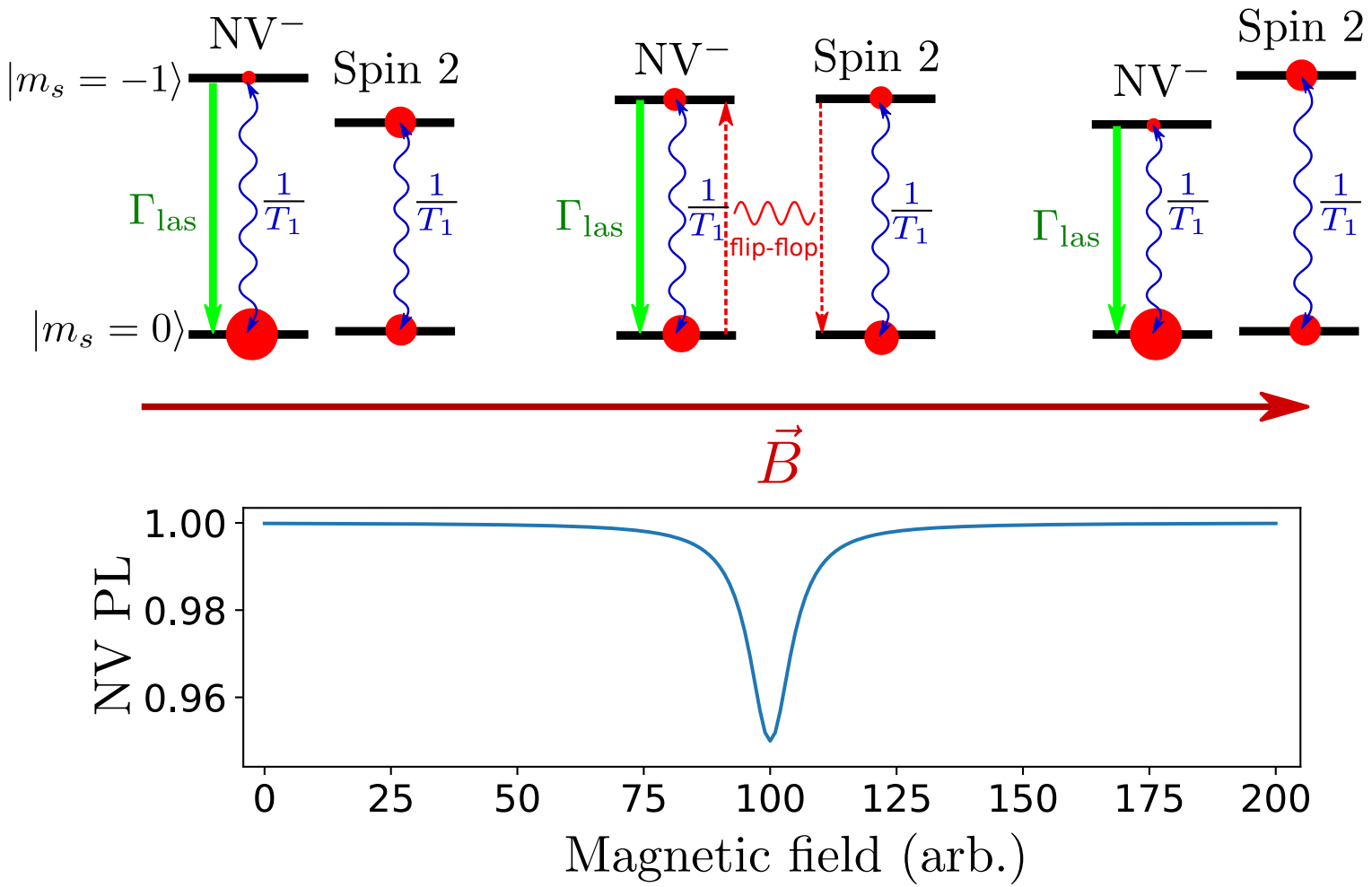
Glenn, D. R. [...] Walsworth, R. L. (2017)
Geochemistry, Geophysics, Geosystems, 18(8), 3254-3267.

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

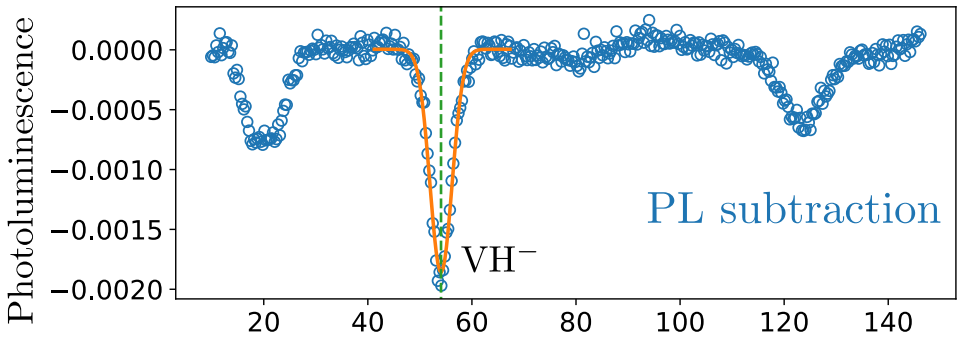
Area normalized sensitivity:
$\eta_S \approx 20\ \mu\mathrm{T} \cdot \mu\mathrm{m}/\sqrt{\mathrm{Hz}}$

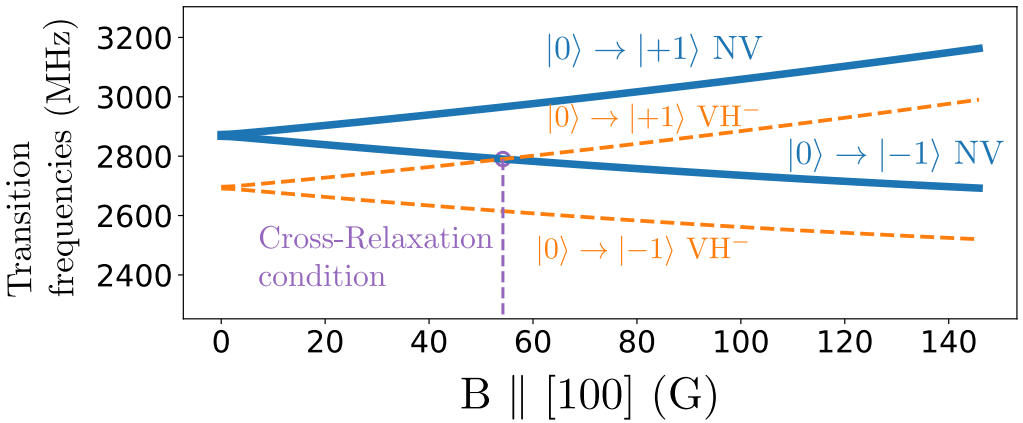
Outline

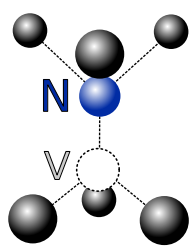
Principle of cross-relaxation with NV centers



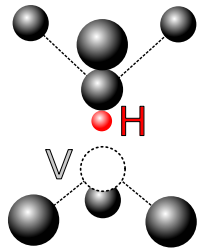
Example: Cross-relaxation between NV centers and VH^-



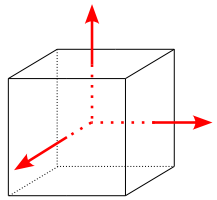




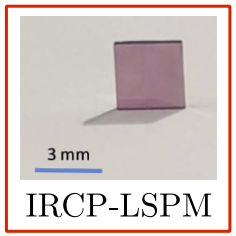
NV center



VH center



Sample



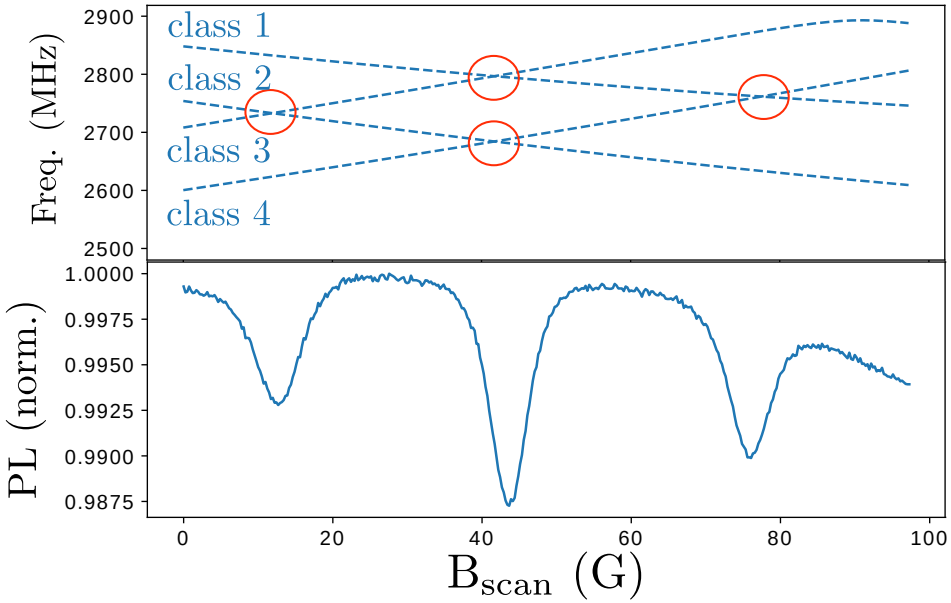
IRCP-LSPM

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

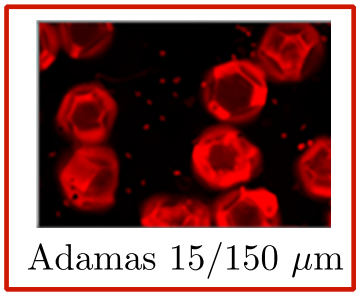
C. Pellet-Mary, P. Huillery, M. Perdriat, A. Tallaire, and G. Hétet
Phys. Rev. B **103**, L100411 – Published 24 March 2021

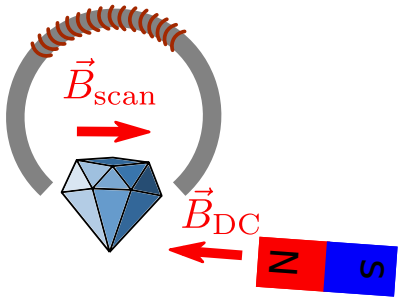
Cross-relaxation between NV centers and NV centers

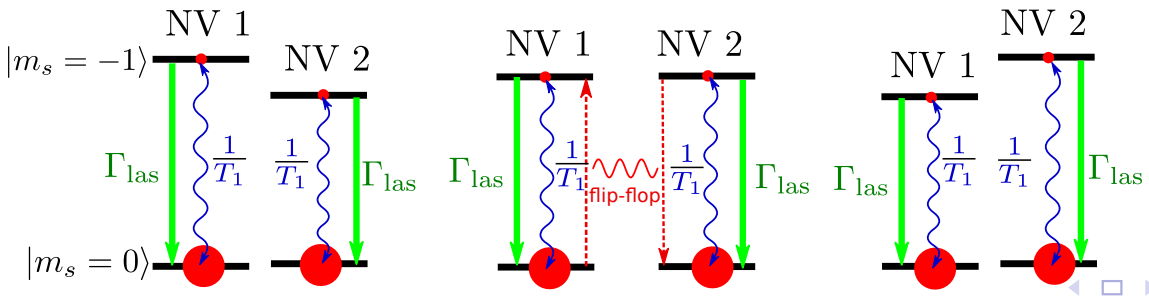
$|0\rangle \rightarrow |-1\rangle$ frequency transition for all four classes of NV



Sample

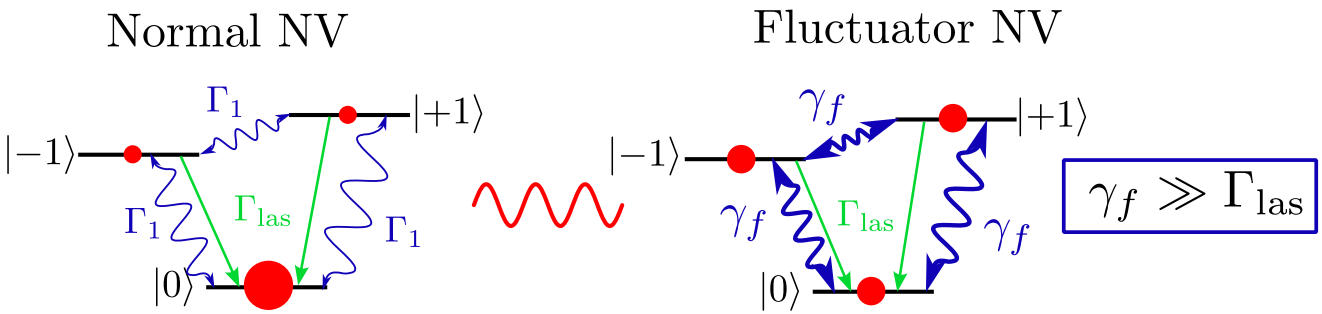






No cross-relaxation for equally polarized spins

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

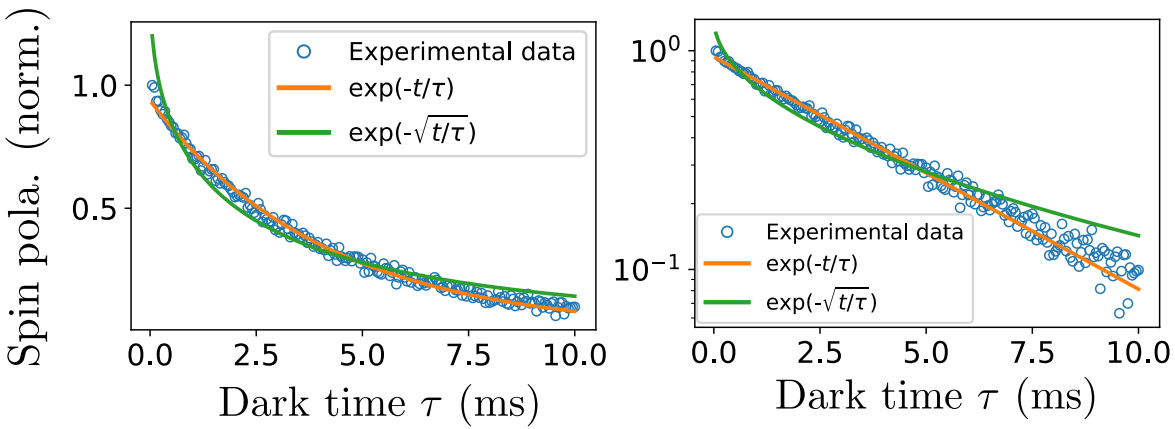
Precedents in:
- P-doped Si
- solid-state NMR
- FRET

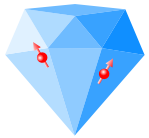
Possible microscopic explanation:
- charge tunneling
- modulation of J-coupling

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

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

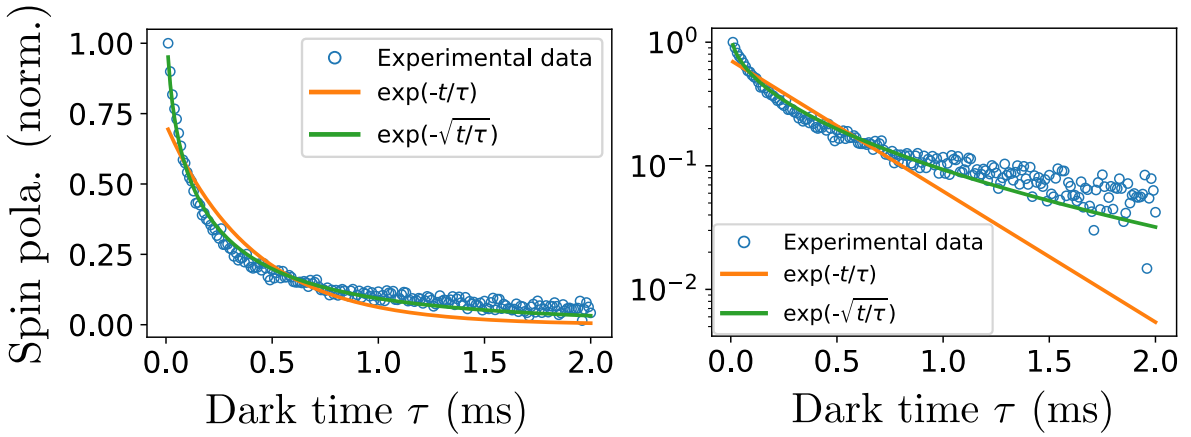
Stretched exponential decay profile

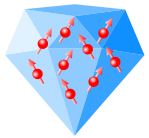




Low NV density

- Exponential profile
- $T_1 \sim 5$ ms



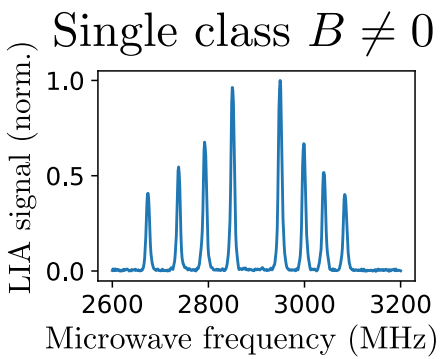


High NV density

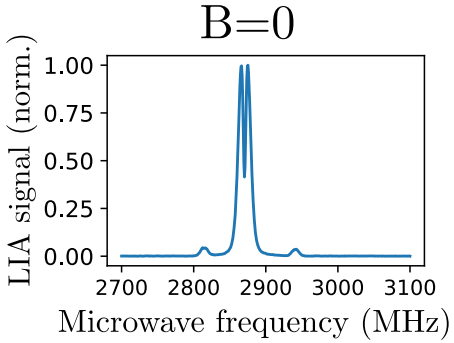
- Stretched exp. profile
- $T_1 \sim 0.5$ ms

Zero field depolarization sources (theory)

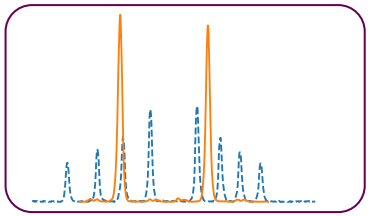
$\Gamma_1 = \Gamma_0^{\text{th}}$

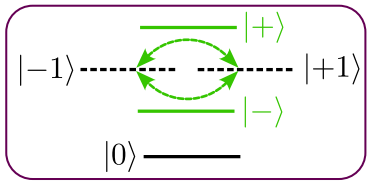


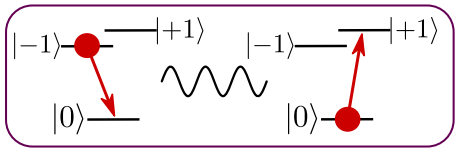
$\Gamma_1 = 102\,\Gamma_0^{\text{th}}$

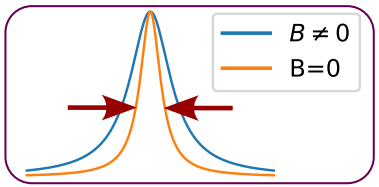


- 4-classes degeneracy
  $\Gamma_1 = \Gamma_0^{\text{th}} \rightarrow 43\,\Gamma_0^{\text{th}}$
- Eigenbasis change
  $\Gamma_1 = 43\,\Gamma_0^{\text{th}} \rightarrow 51\,\Gamma_0^{\text{th}}$
- Double flips
  $\Gamma_1 = 51\,\Gamma_0^{\text{th}} \rightarrow 94\,\Gamma_0^{\text{th}}$
- T_2^* increase
  $\Gamma_1 = 94\,\Gamma_0^{\text{th}} \rightarrow 102\,\Gamma_0^{\text{th}}$

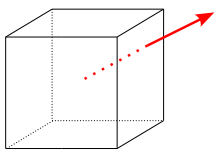






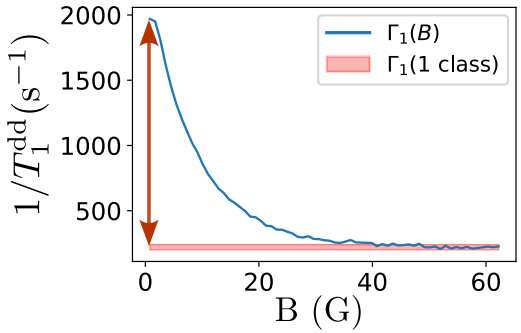


Summary of the experimental observations

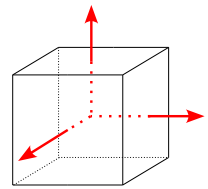


Random $\vec{B}$

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

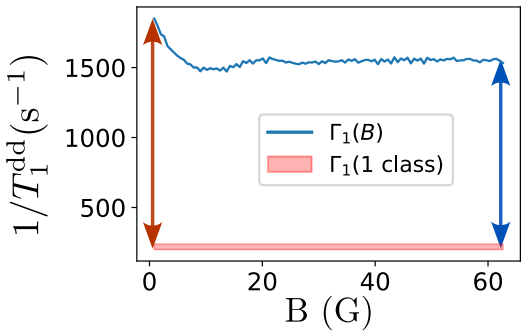


- ~~4-classes degeneracy~~
- ~~Eigenbasis change~~
- ~~Double-flips~~
- ~~T_2^* change~~

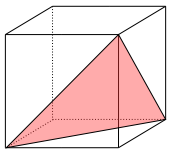


$\vec{B} \parallel \langle 100 \rangle$

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

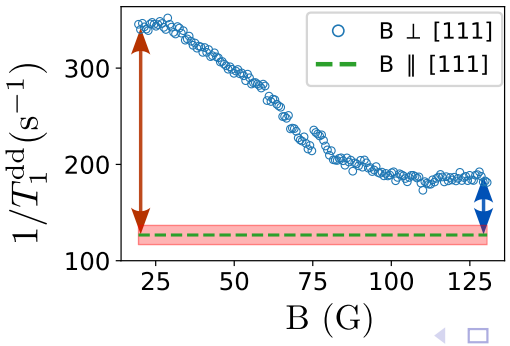


- 4-classes degeneracy
- ~~Eigenbasis change~~
- ~~Double-flips~~
- ~~T_2^* change~~



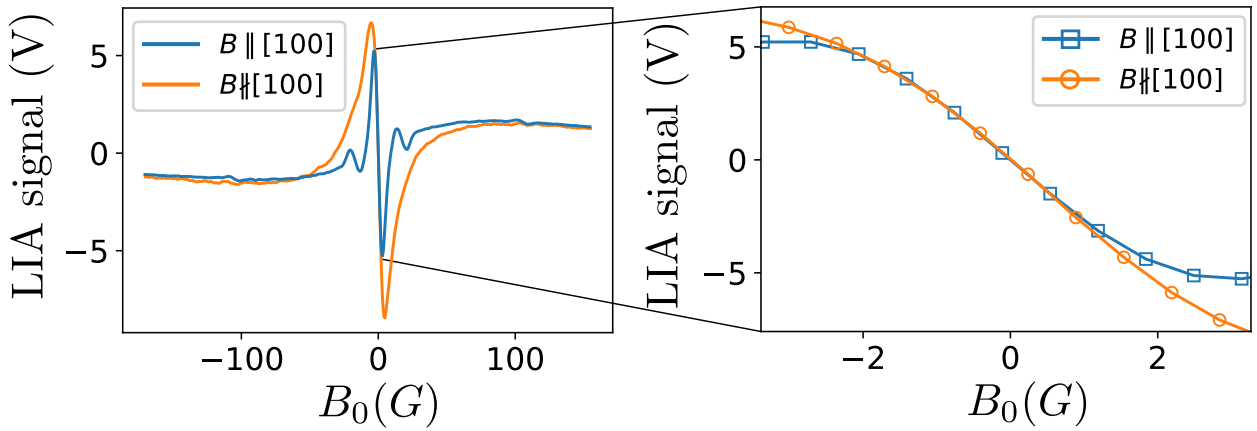
$\vec{B} \perp \langle 111 \rangle$

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

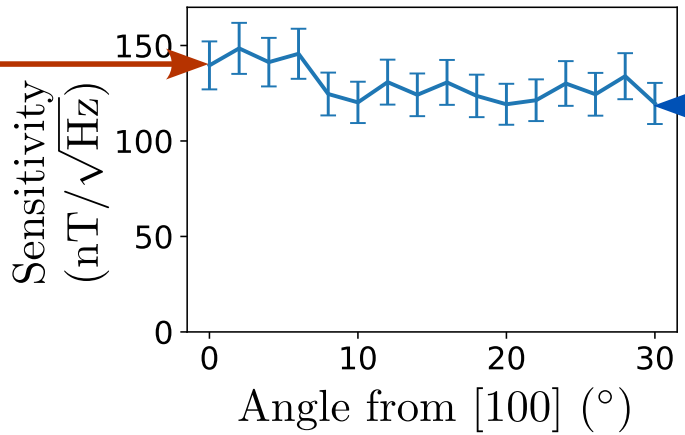


- ~~4-classes degeneracy~~
- Eigenbasis change
- ~~Double-flips~~
- T_2^* change

Angular sensitivity of LFDM



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



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

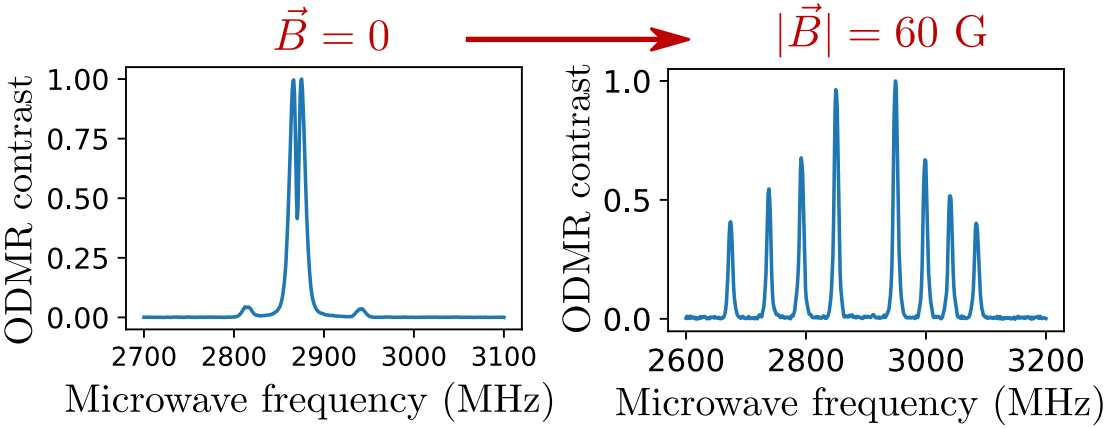
The 4-classes degeneracy is not the limiting factor of the sensitivity

Conclusion

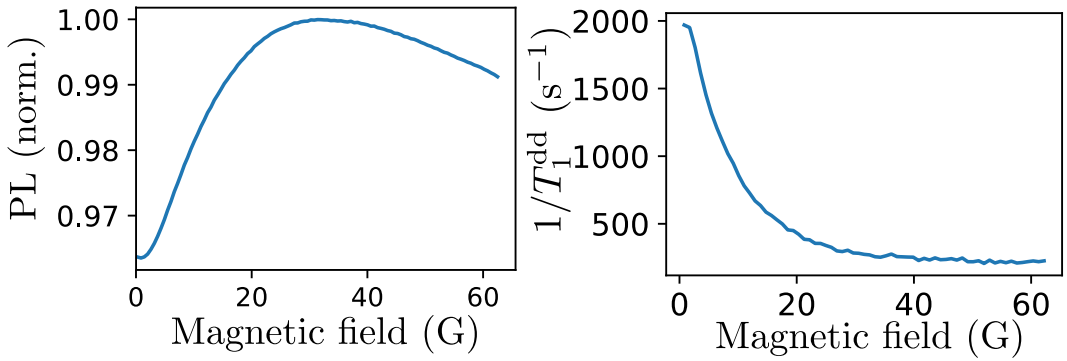
- The use of NV center ensemble for magnetometry is currently limited by the interactions between the spin defects.
- Dipole-dipole interaction within dense ensemble of NV centers result in spin depolarization which is exacerbated at low magnetic field.
- This depolarization can be used to perform microwave-free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

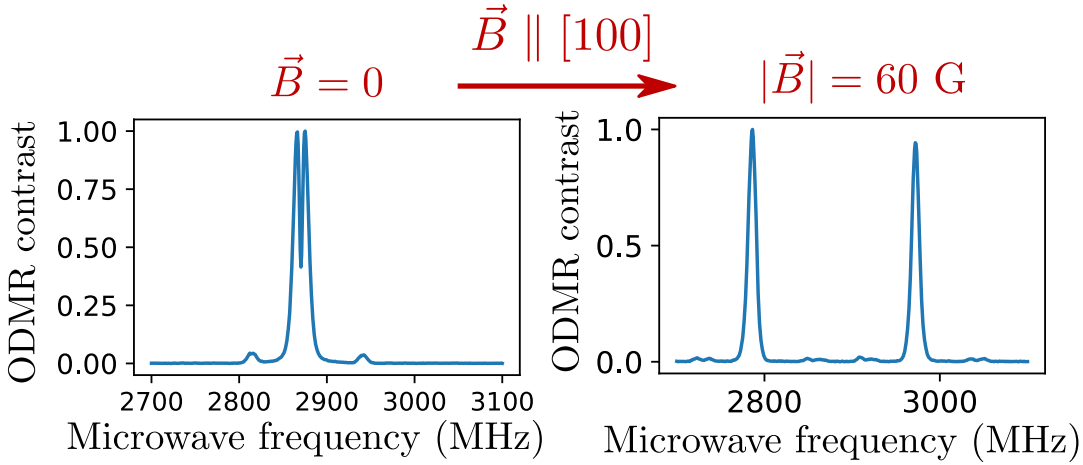


$\Gamma_1(B = 0) \approx 10\ \Gamma_1(B \neq 0)$

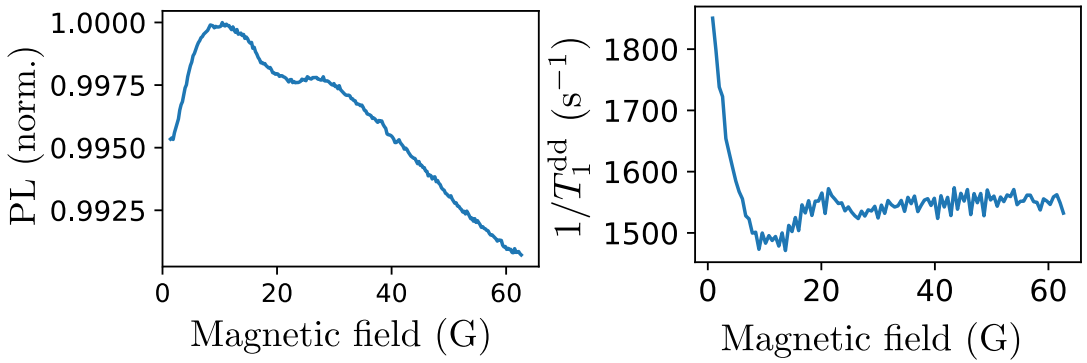
$\sim 4\%$ PL contrast

HWHM ~ 9 G

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



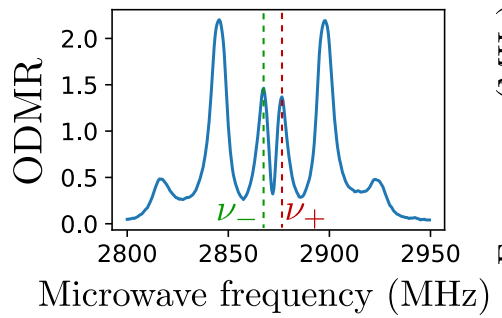
- 4-classes degeneracy
- ~~Eigenbasis change~~
- ~~Double-flips~~
- ~~T_2^* change~~

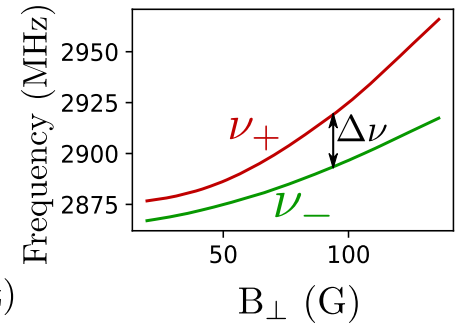


$\Gamma_1(B = 0) \approx 1.2\ \Gamma_1(B \neq 0)$
$\sim 0.5\%$ PL contrast
HWHM ~ 2 G

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

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

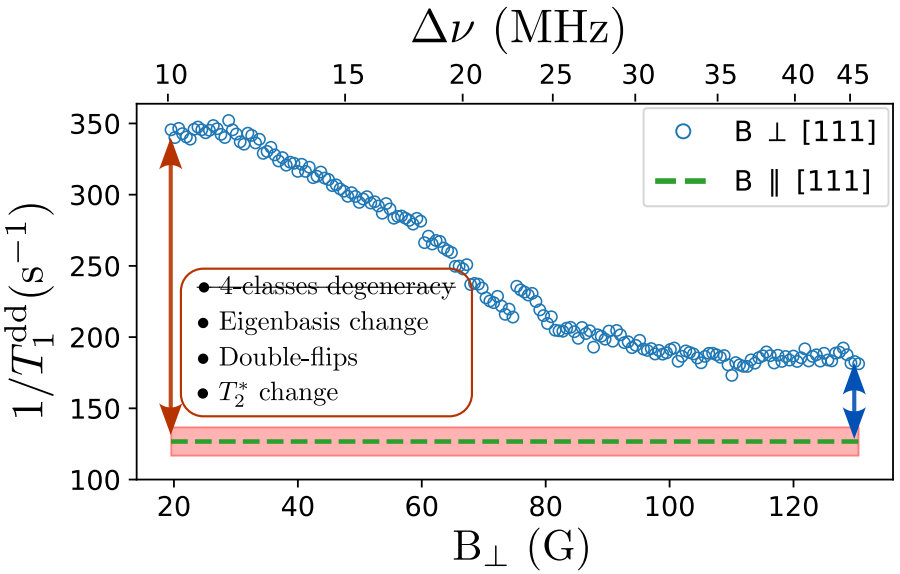




Same eigenbasis :

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

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



cancelling out double flips with transverse field

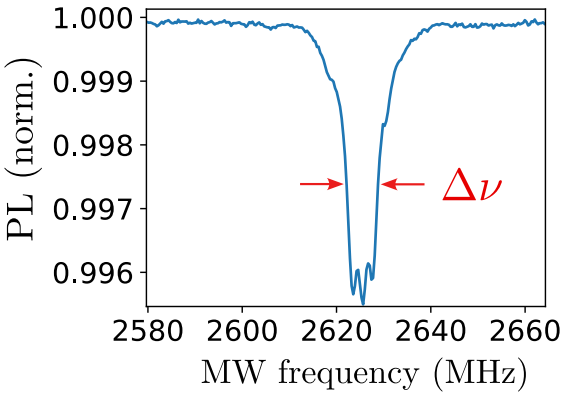
- ~~4-classes degeneracy~~
- Eigenbasis change
- ~~Double-flips~~
- T_2^* change

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

NV center magnetometry sensitivity

Ideal (DC) sensitivity for N independent NV centers:

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



- $\hbar$: 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