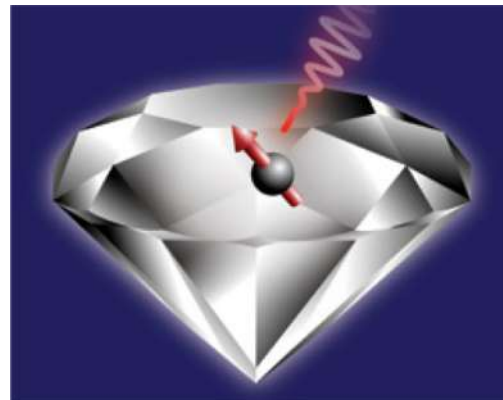


Microscopie magnétique à spin unique

Vincent JACQUES

Laboratoire Charles Coulomb UMR 5221, Université Montpellier, and CNRS

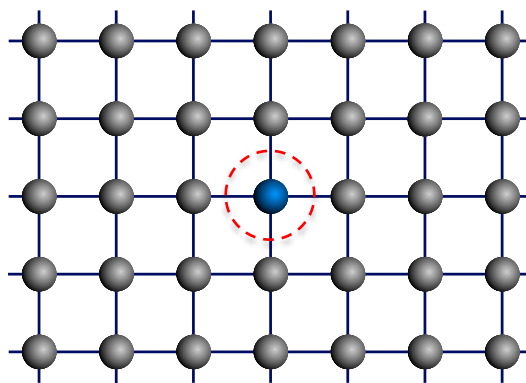


Optically active point defects in wide bandgap materials

Single Photon Source

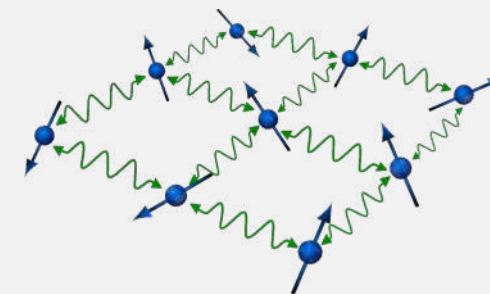


- Quantum cryptography
- Single photon interference



Solid-state "artificial atom"

Quantum Information

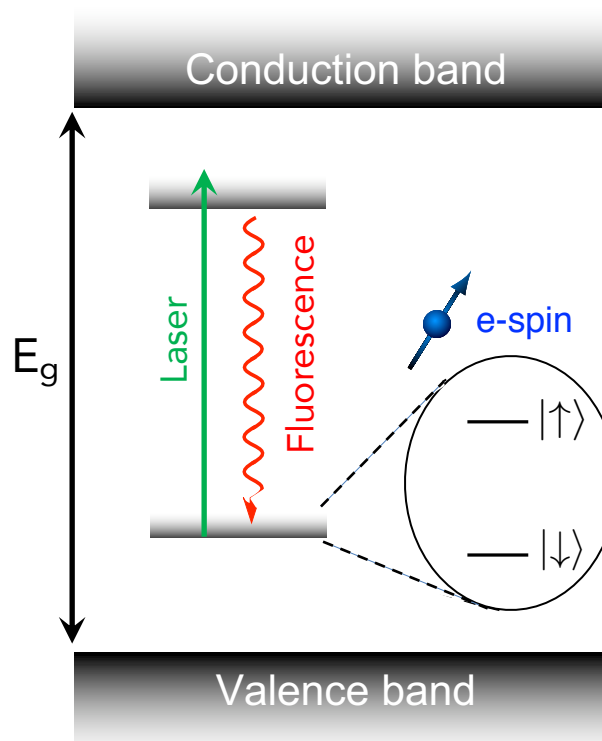


- Spin physics
- Spin/photon interface

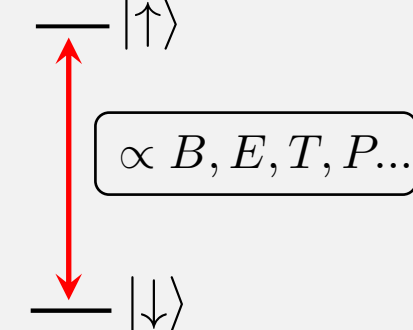
Fluorescent Biomarkers



Mohan, *Nano Lett.* 10, 3692 (2010)



Quantum sensing

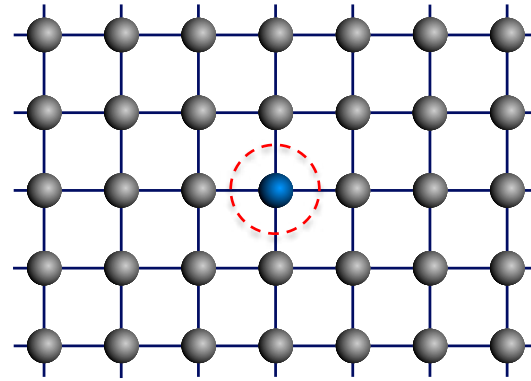


Optically active point defects in wide bandgap materials

Single Photon Source

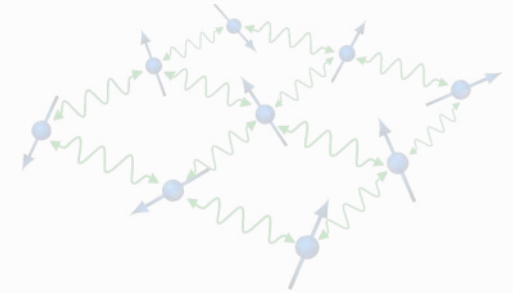


- Quantum cryptography
- Single photon interference



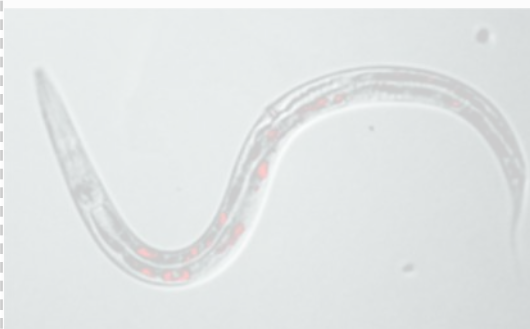
Solid-state "artificial atom"

Quantum Information



- Spin physics
- Spin/photon interface

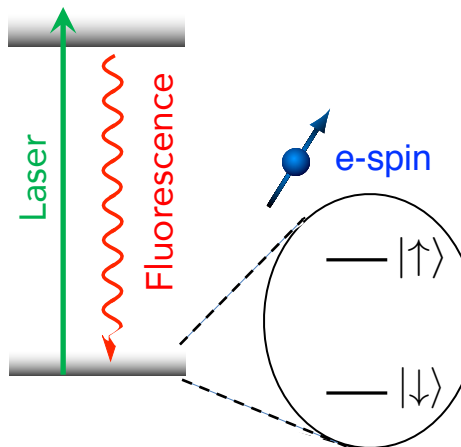
Fluorescent Biomarkers



Mohan, *Nano Lett.* 10, 3692 (2010)

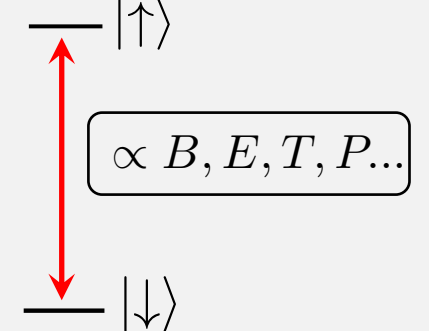
Conduction band

E_g



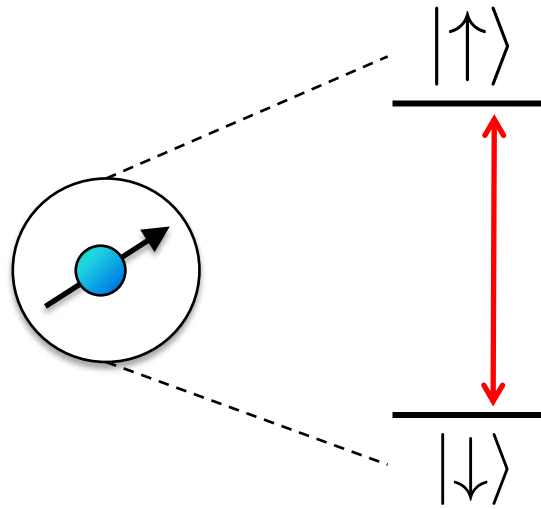
Valence band

Quantum sensing

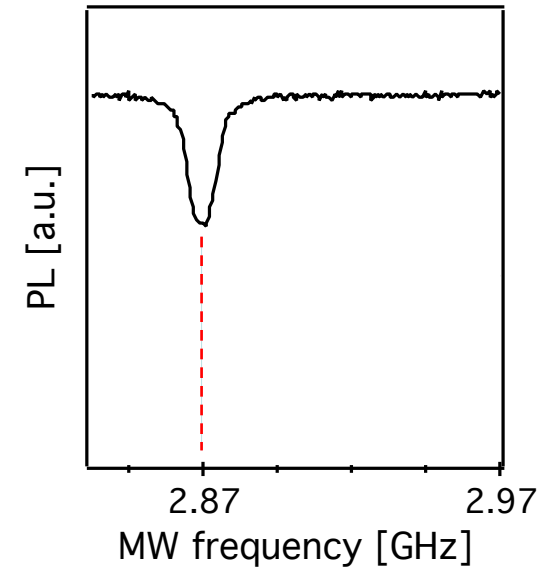


Magnetic field sensing with a single spin

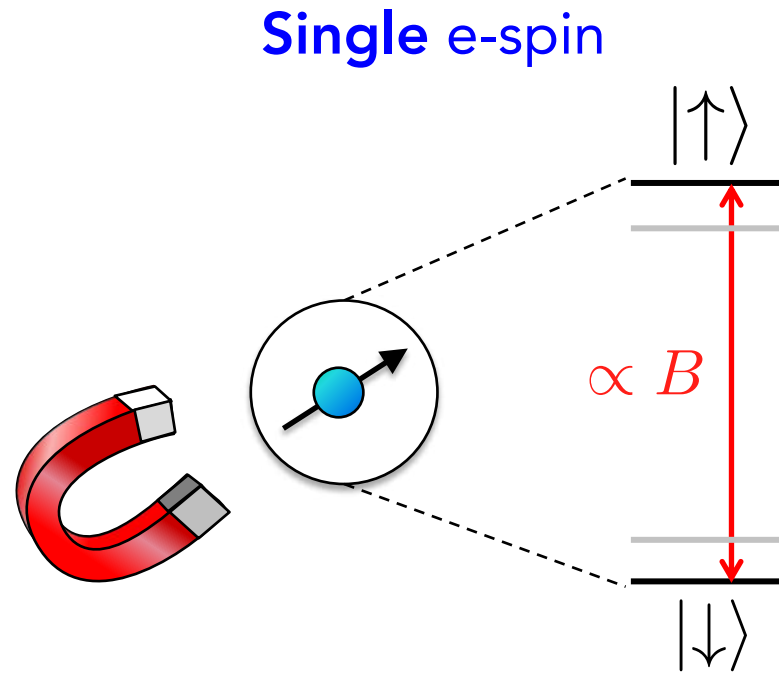
Single e-spin



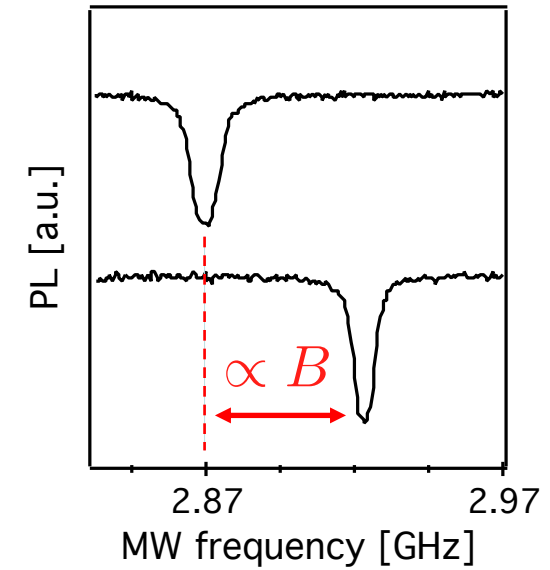
Electron Spin Resonance (ESR)



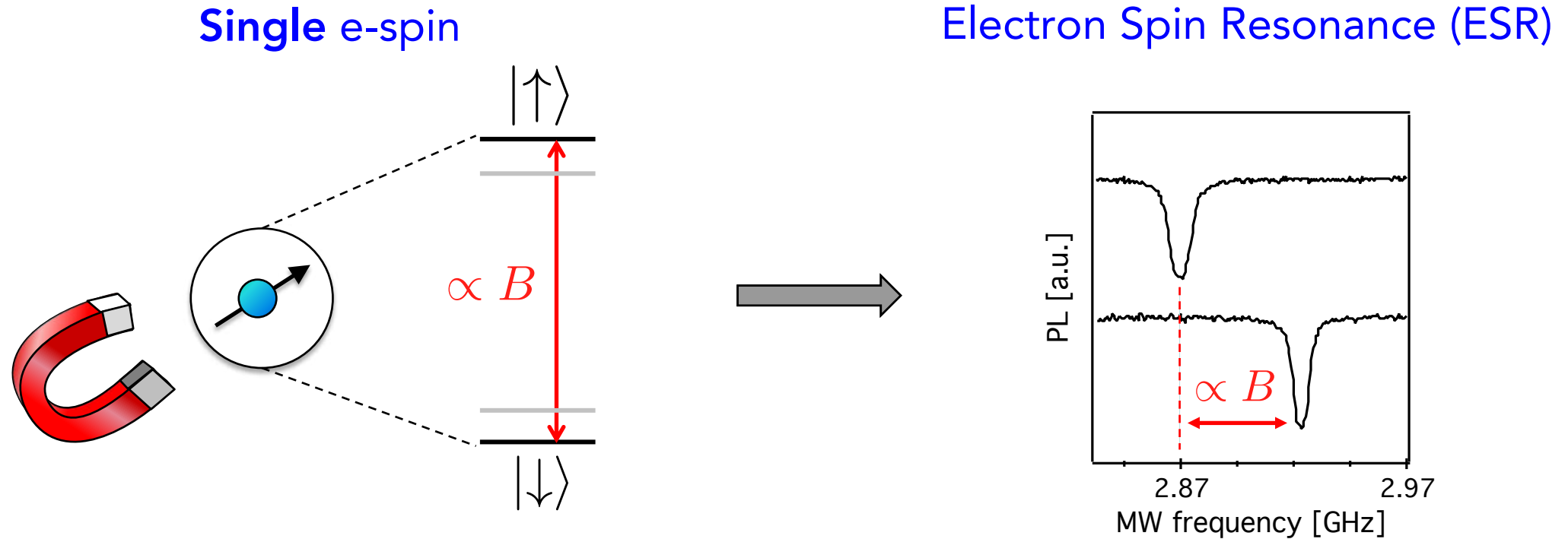
Magnetic field sensing with a single spin



Electron Spin Resonance (ESR)

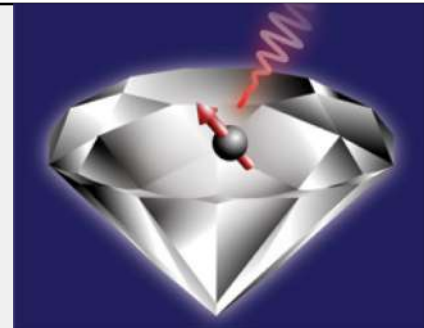


Magnetic field sensing with a single spin

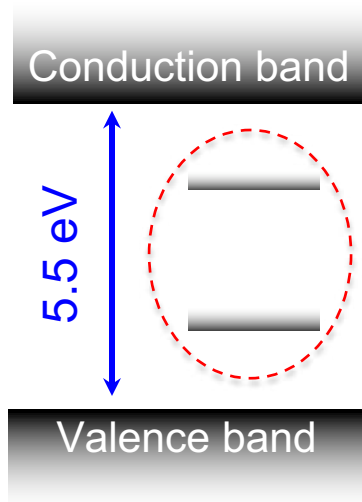


Can be realized with **NV defects in diamond**

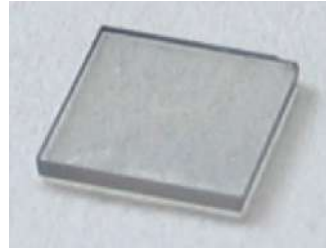
Maze, *Nature* (2008), Degen, *APL* (2008)
Balasubramanian, *Nature* (2008)



Point defects in diamond

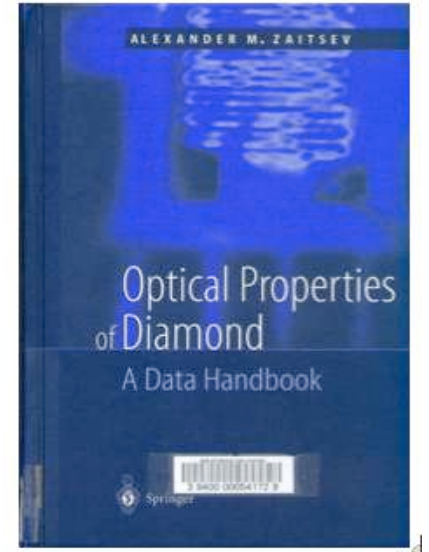


A "perfect" diamond would not absorb visible light...



... but more than 500 defects are optically active

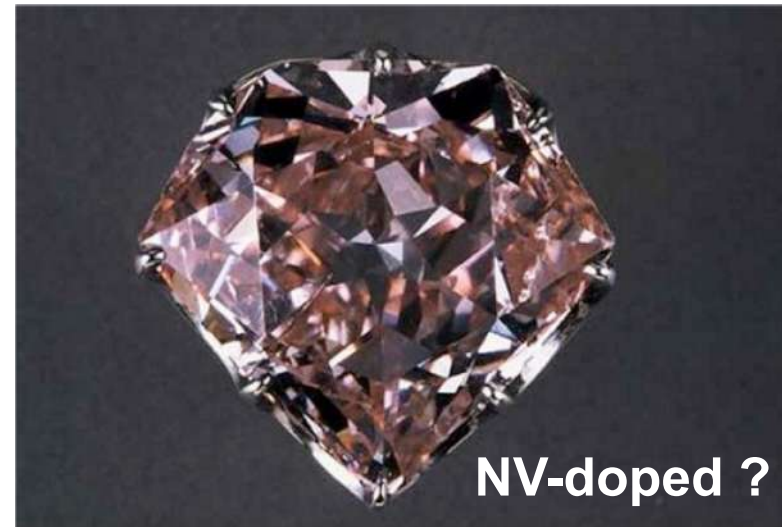
→ **Color centers**



The « Hope » diamond
(Washington)

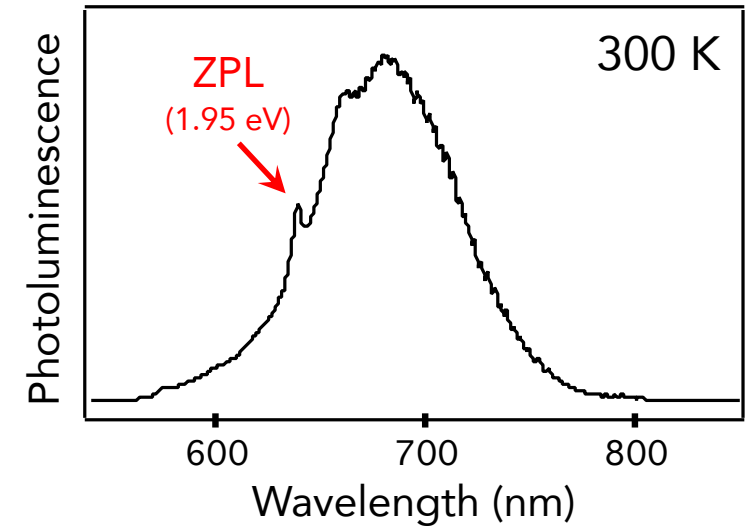
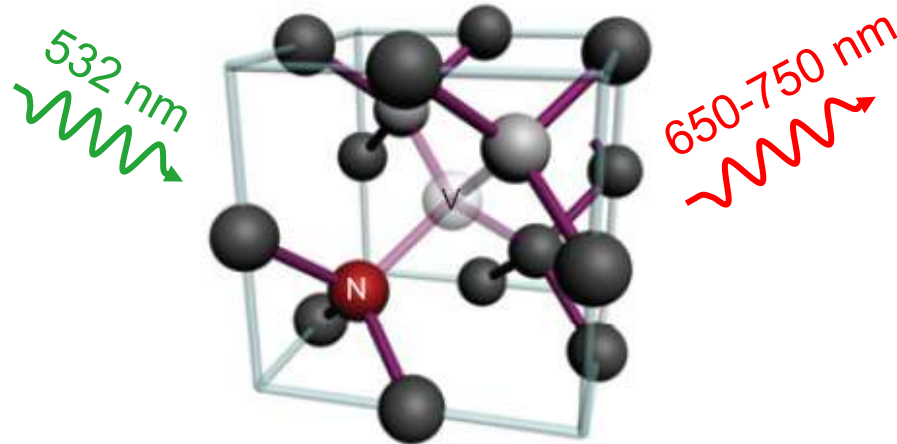
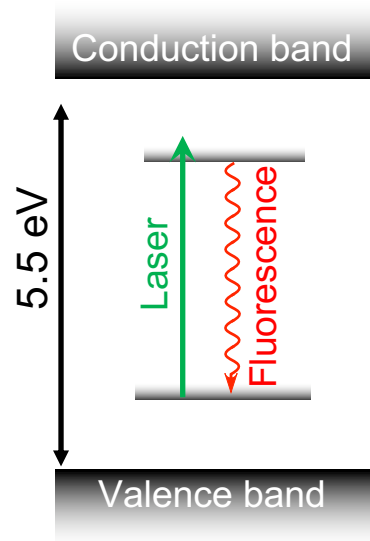


The « Hortensia » diamond
(Louvre, Paris)



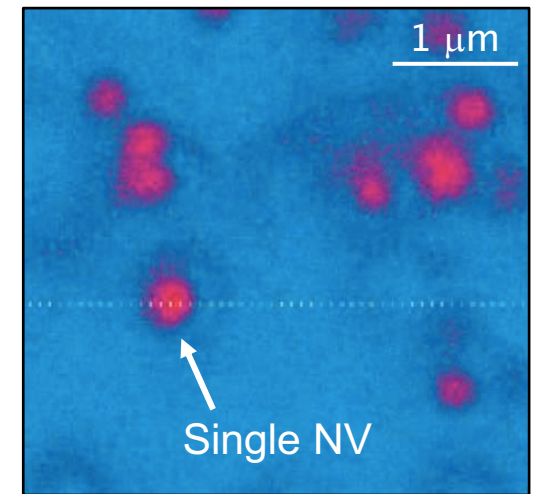
The Nitrogen-Vacancy (NV) defect in diamond

- An artificial atom "nestled" in the diamond lattice

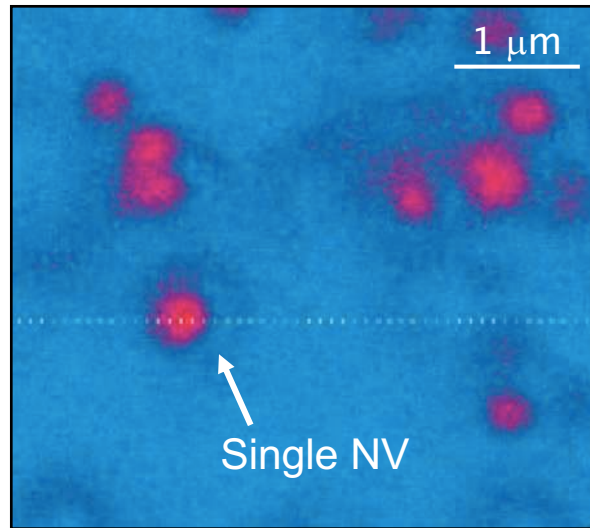


- Detection at the single emitter level at room T
(perfect photostability)

Gruber, *Science* **276**, 2012 (1997)

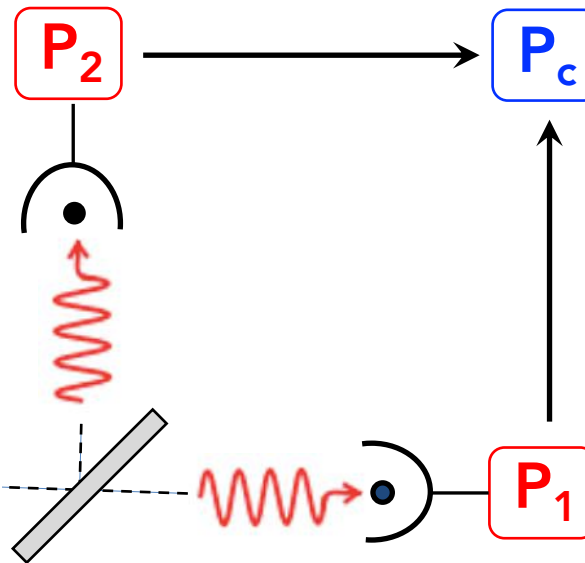
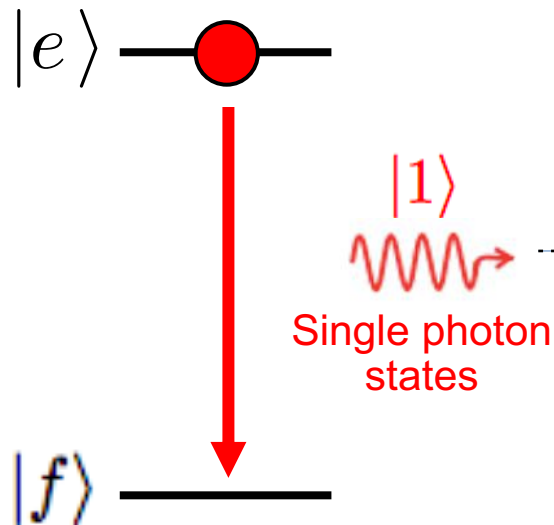


A robust single photon source



Second-order correlation function

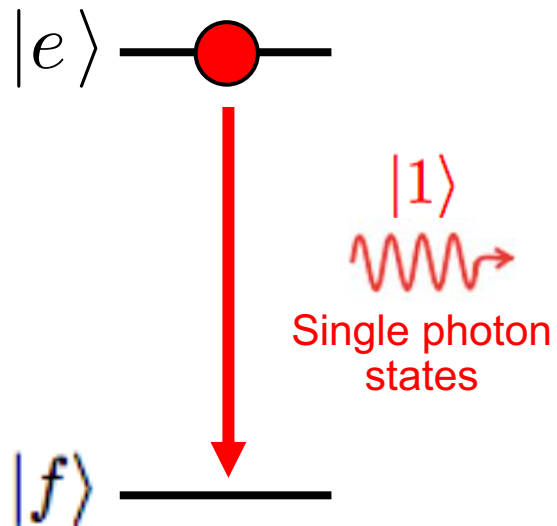
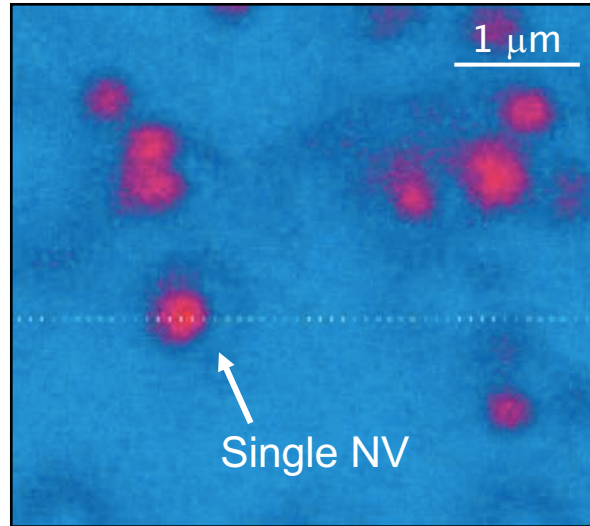
$$g^{(2)}(\tau) = \frac{\overline{\mathcal{I}(t)\mathcal{I}(t+\tau)}}}{\overline{\mathcal{I}(t)} \times \overline{\mathcal{I}(t+\tau)}} \quad \longrightarrow \quad g^{(2)}(0) = \frac{P_c}{P_1 \times P_2}$$



single-photon source, $P_c=0$

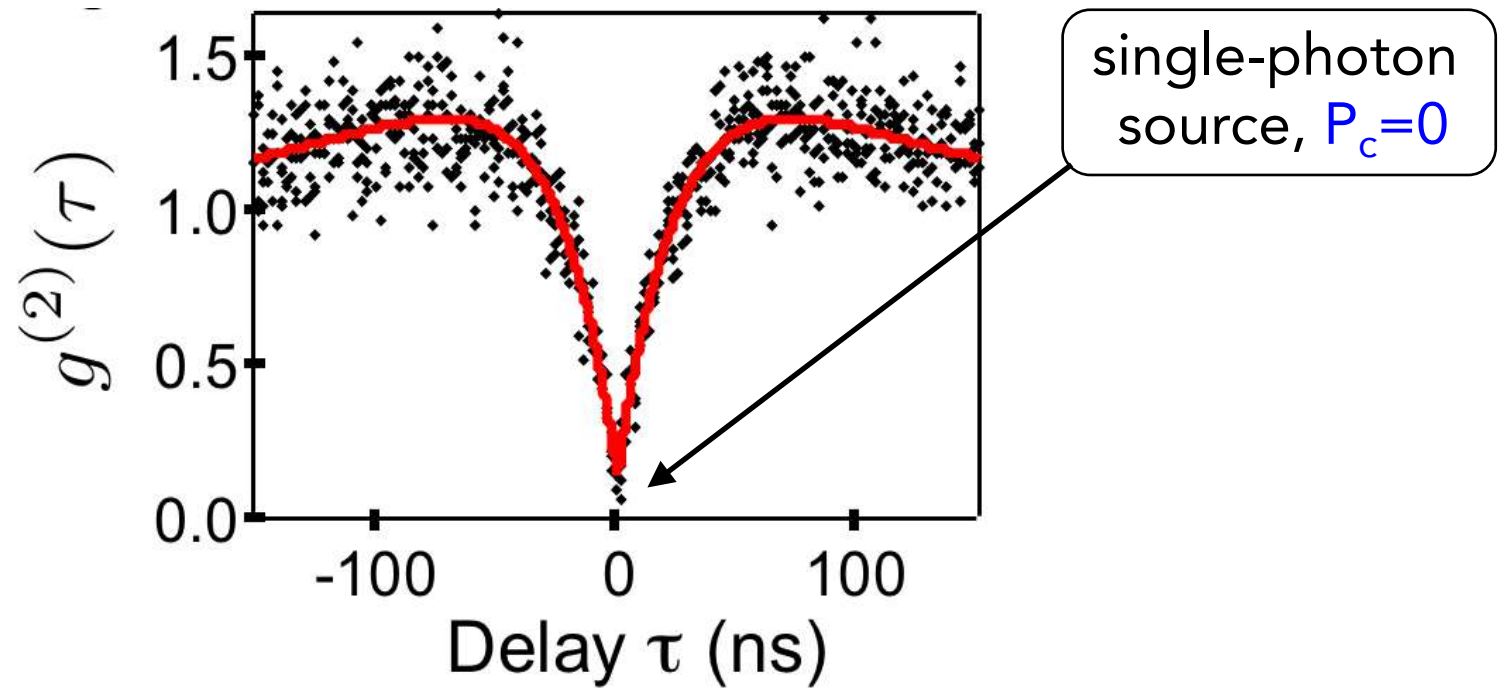
Hanbury Brown and Twiss interferometer

A robust single photon source

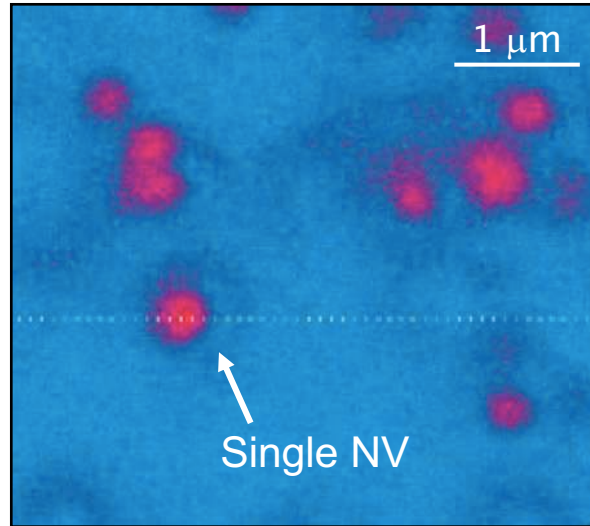


Second-order correlation function

$$g^{(2)}(\tau) = \frac{\overline{\mathcal{I}(t)\mathcal{I}(t+\tau)}}}{\overline{\mathcal{I}(t)} \times \overline{\mathcal{I}(t+\tau)}} \quad \longrightarrow \quad g^{(2)}(0) = \frac{P_c}{P_1 \times P_2}$$

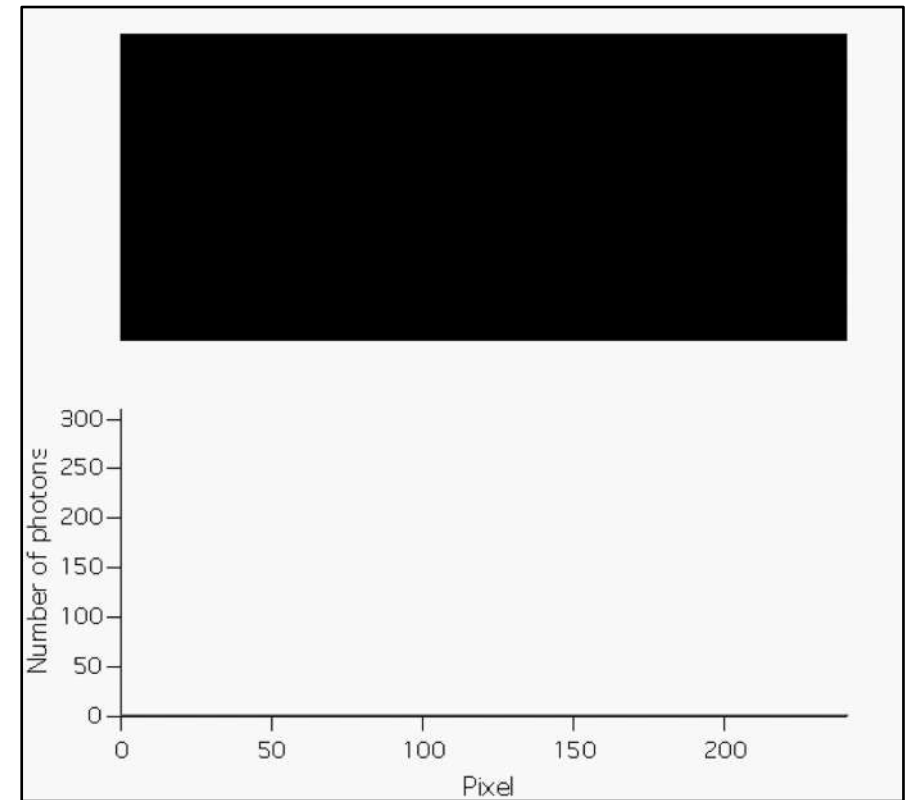
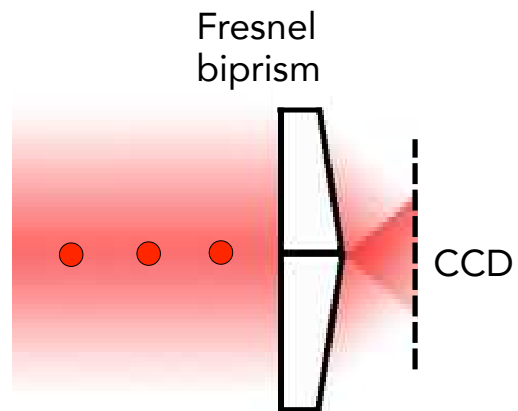
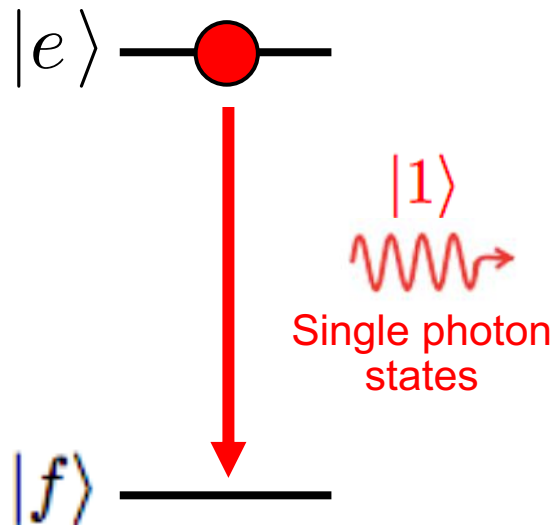


A robust single photon source



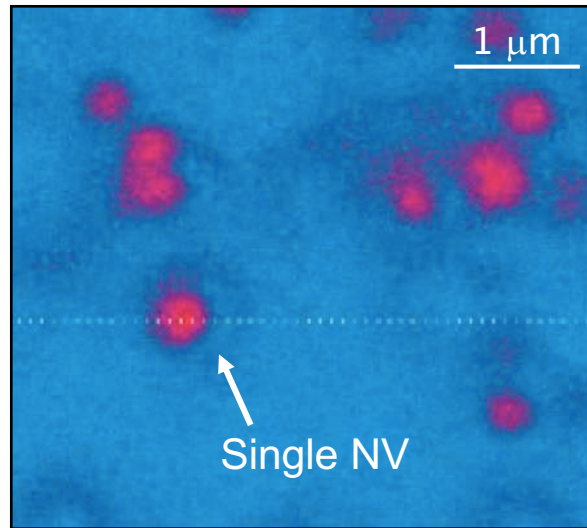
Single photon interference

Jacques, *EPJD* (2005)

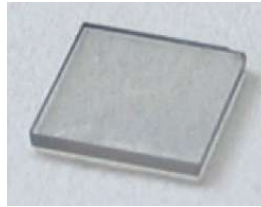


Engineering NV defects in diamond

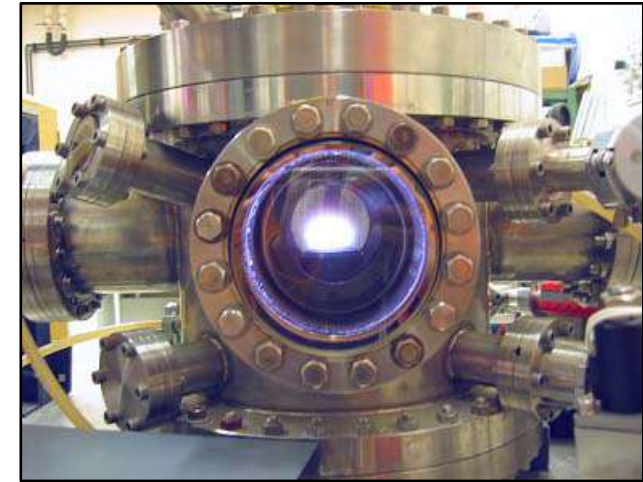
1997



High purity diamond using
CVD growth



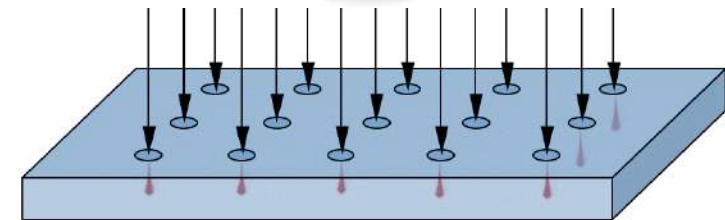
A. Tallaire and J. Achard (Villetaneuse)



nanoscale ion implantation



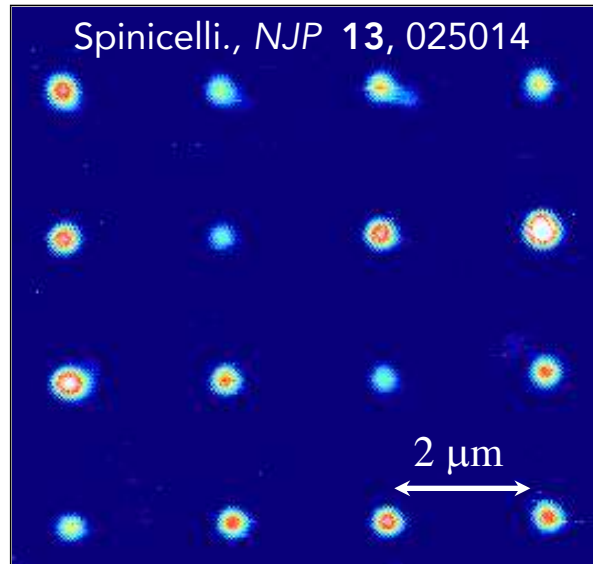
N⁺ ions



J. Meijer and S. Pezzagna (Leipzig)

2012

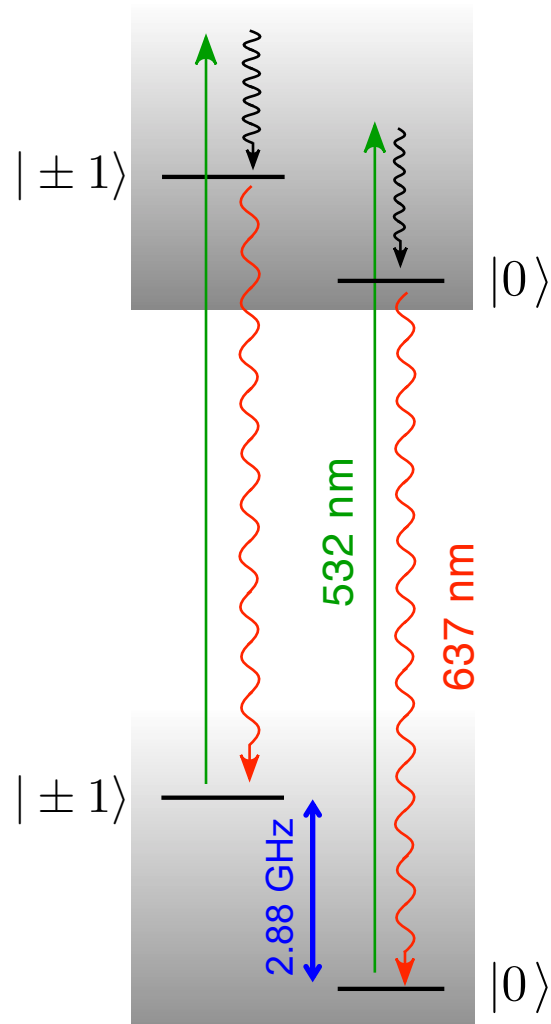
Spinicelli., *NJP* 13, 025014



Array of NV defects

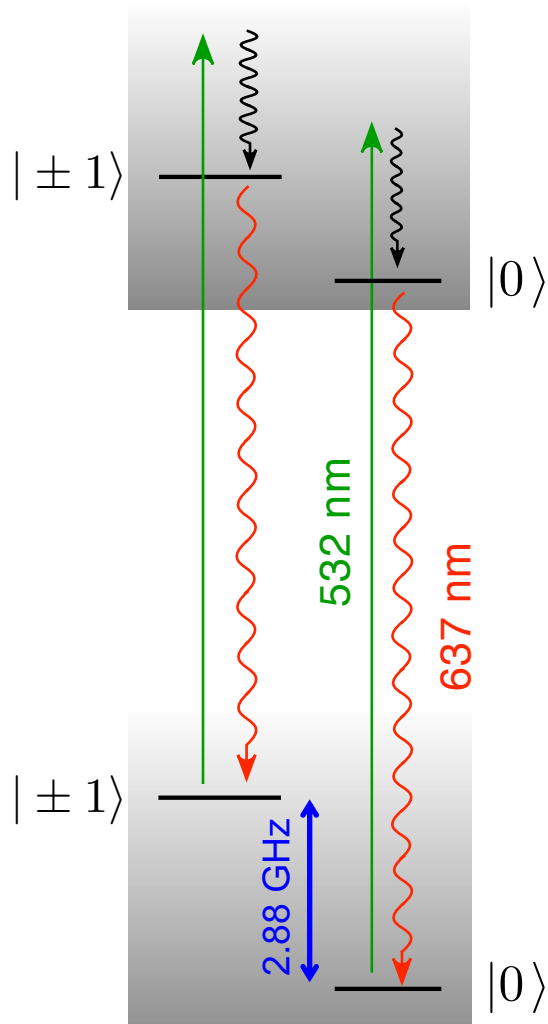
Spin properties

- Artificial atom with a spin triplet ($S=1$) ground state



Spin properties

- Artificial atom with a spin triplet ($S=1$) ground state

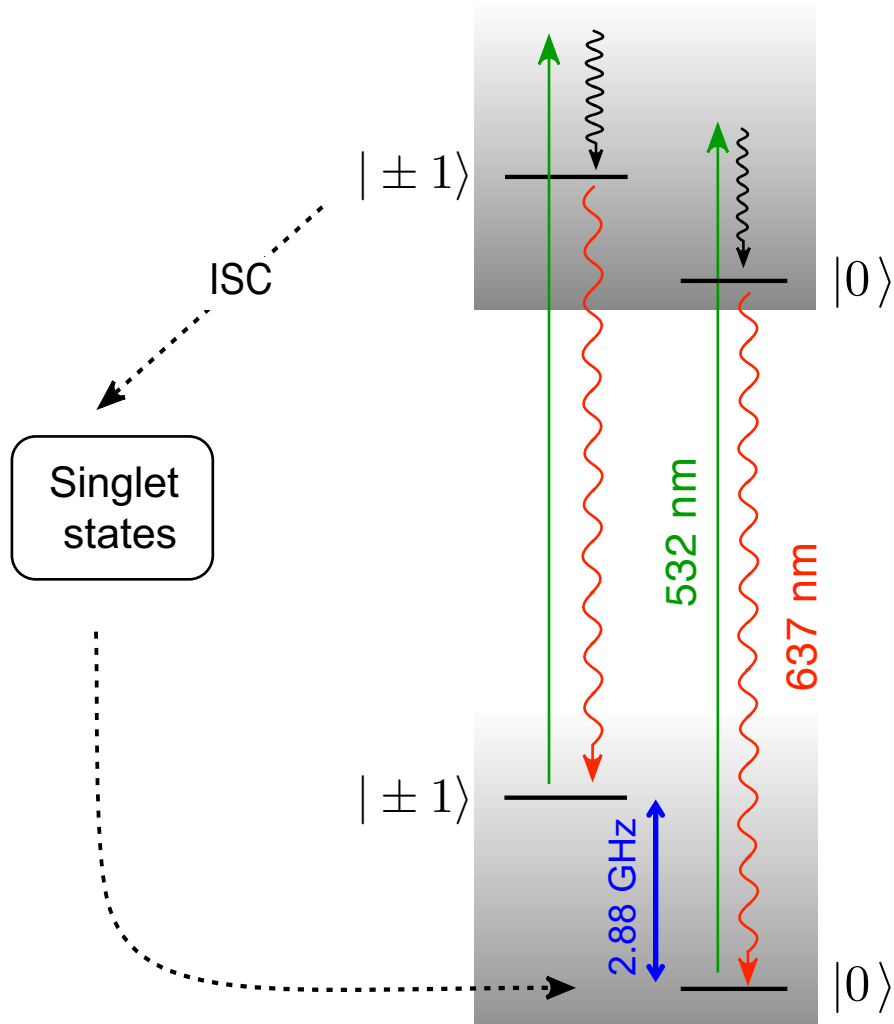


Important properties

- Spin-conserving optical transition $\Delta m_s=0$.

Spin properties

- Artificial atom with a spin triplet ($S=1$) ground state

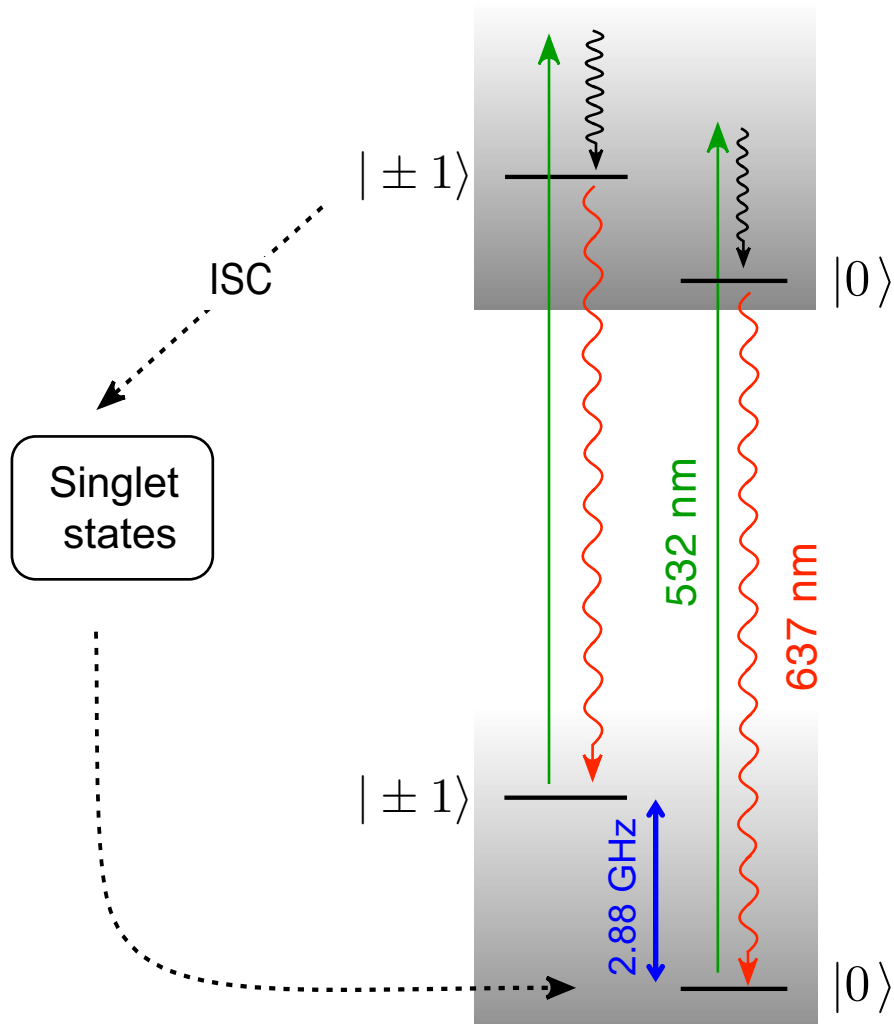


Important properties

- Spin-conserving optical transition $\Delta m_s=0$.
- Spin-dependent ISC to singlet states.

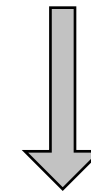
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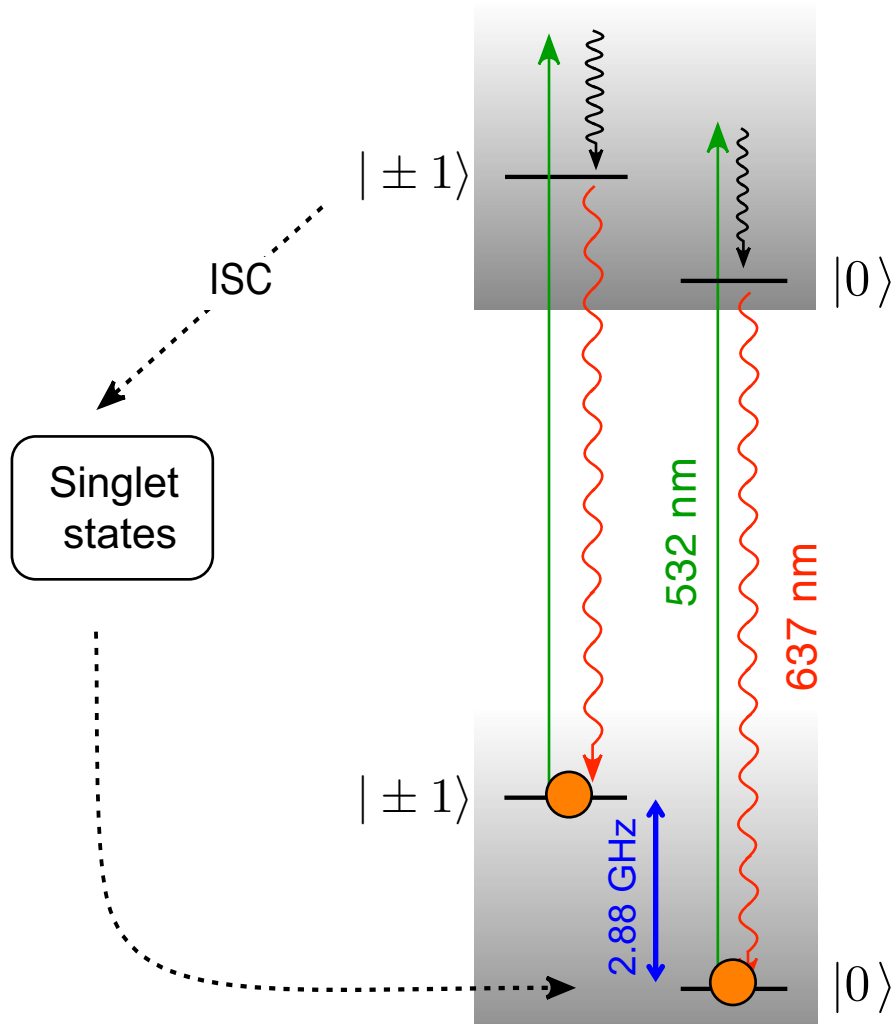


Consequences

- Polarization in $m_s = 0$ by optical pumping.

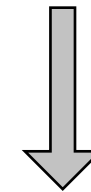
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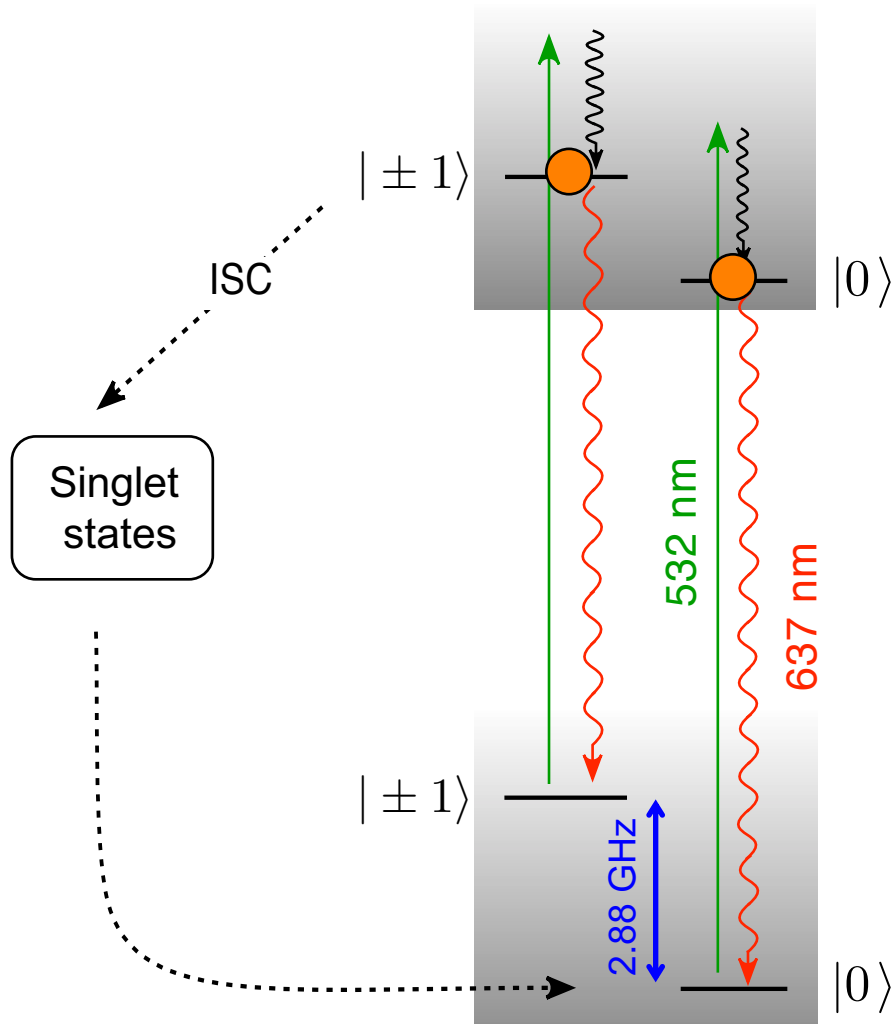


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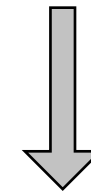
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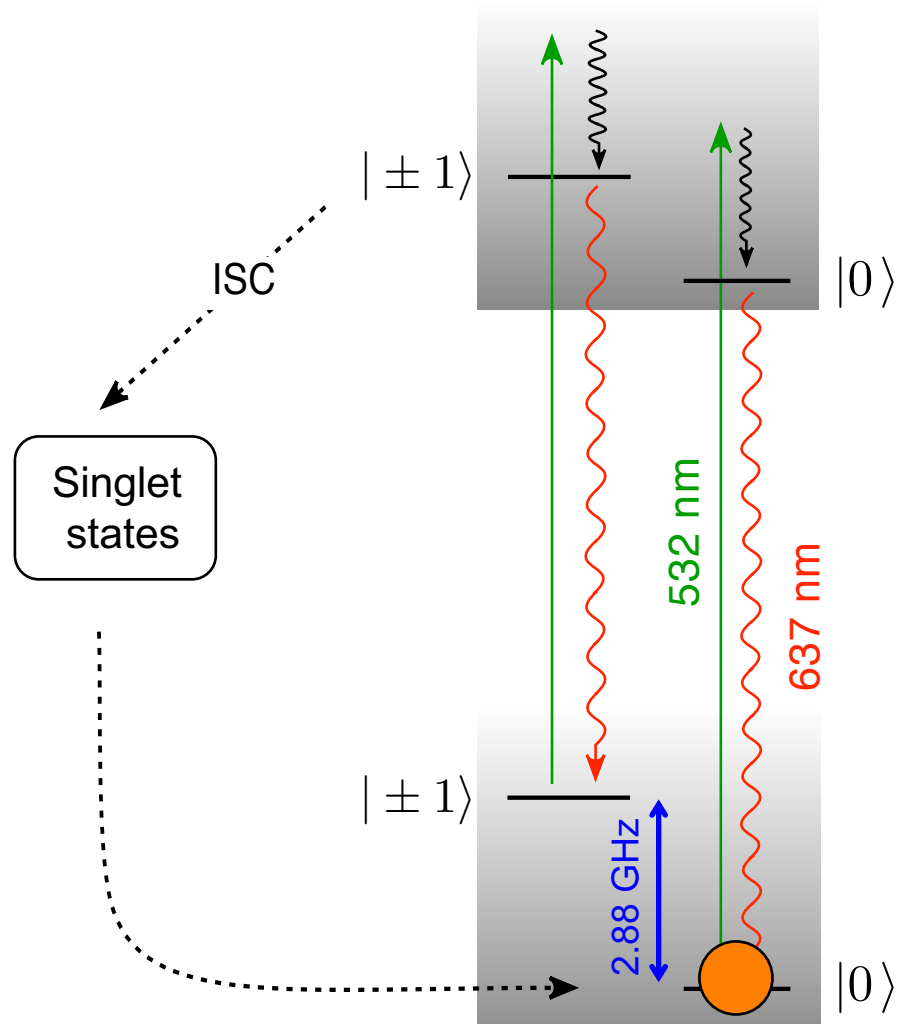


Consequences

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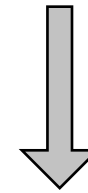
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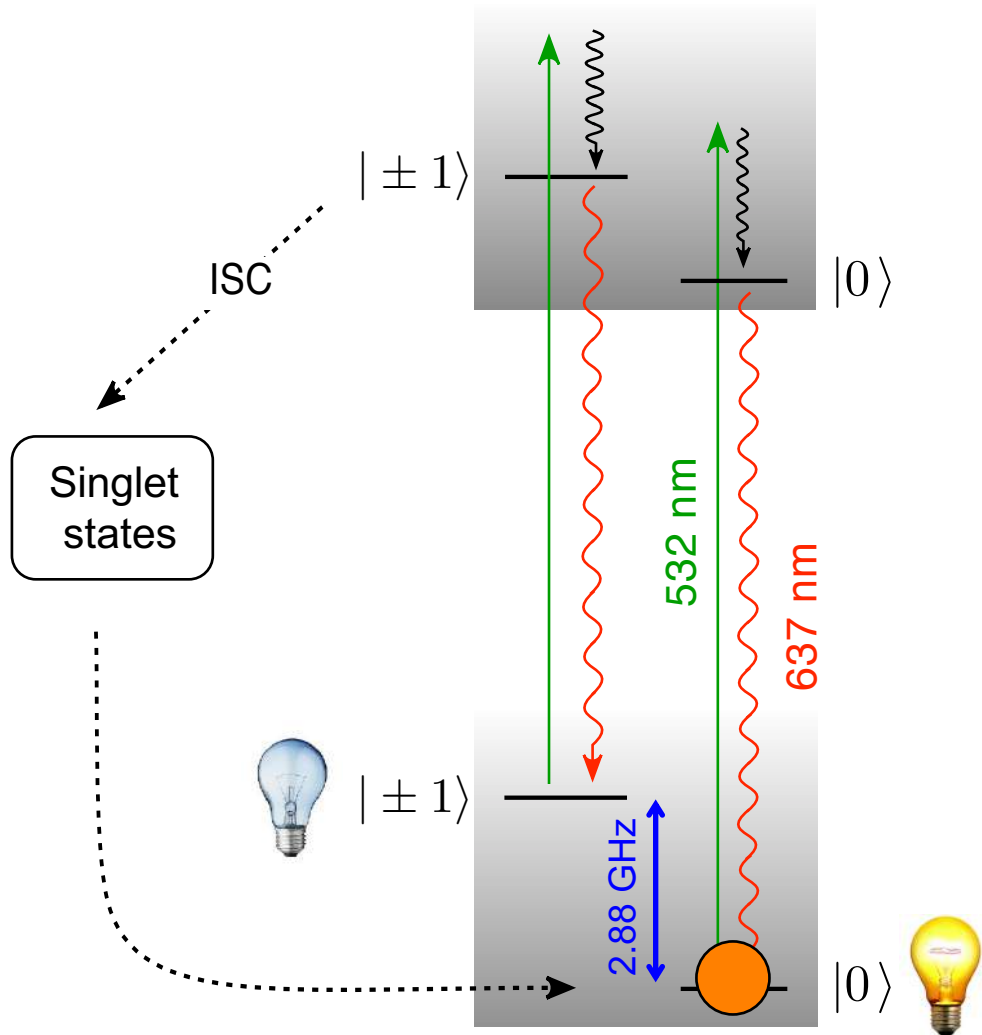


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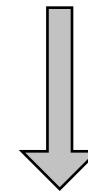
Spin properties

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Important properties

- Spin-conserving optical transition $\Delta m_s=0$.
- Spin-dependent ISC to singlet states.

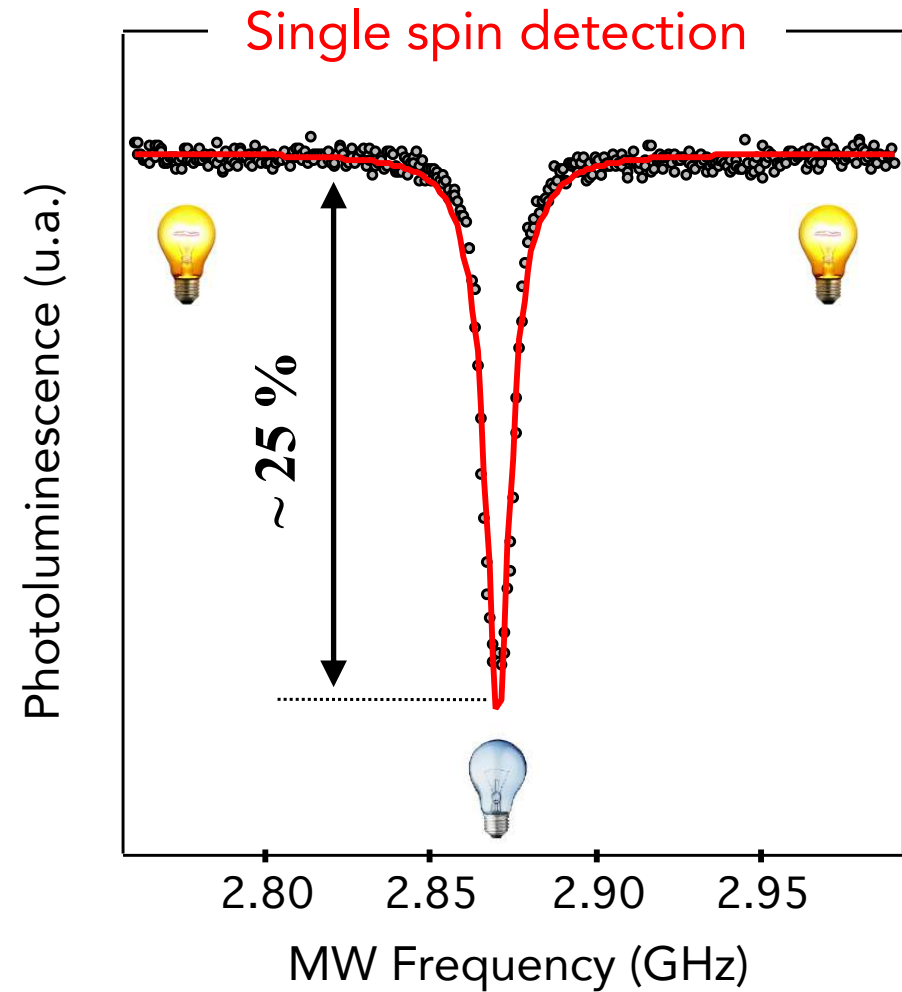
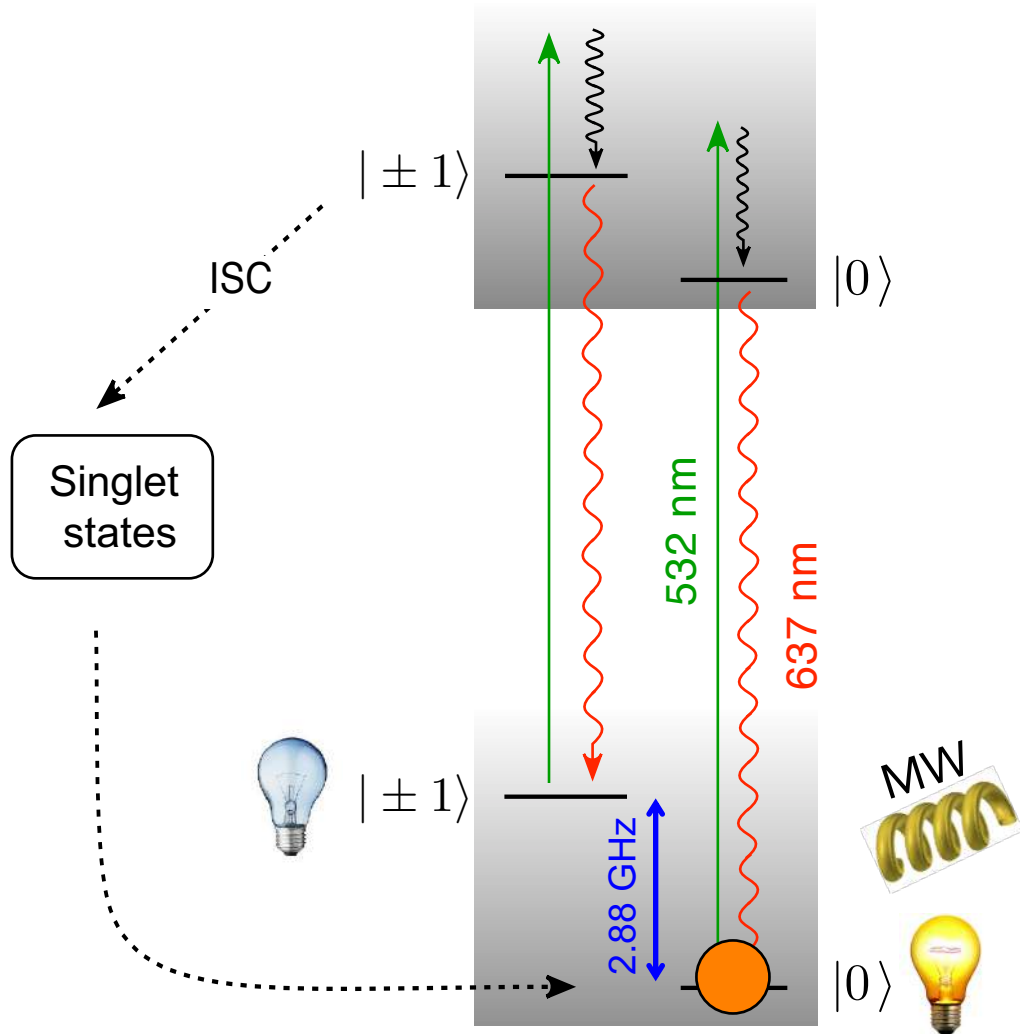


Consequences

- Polarization in $m_s=0$ by optical pumping.
- Spin-dependent fluorescence signal

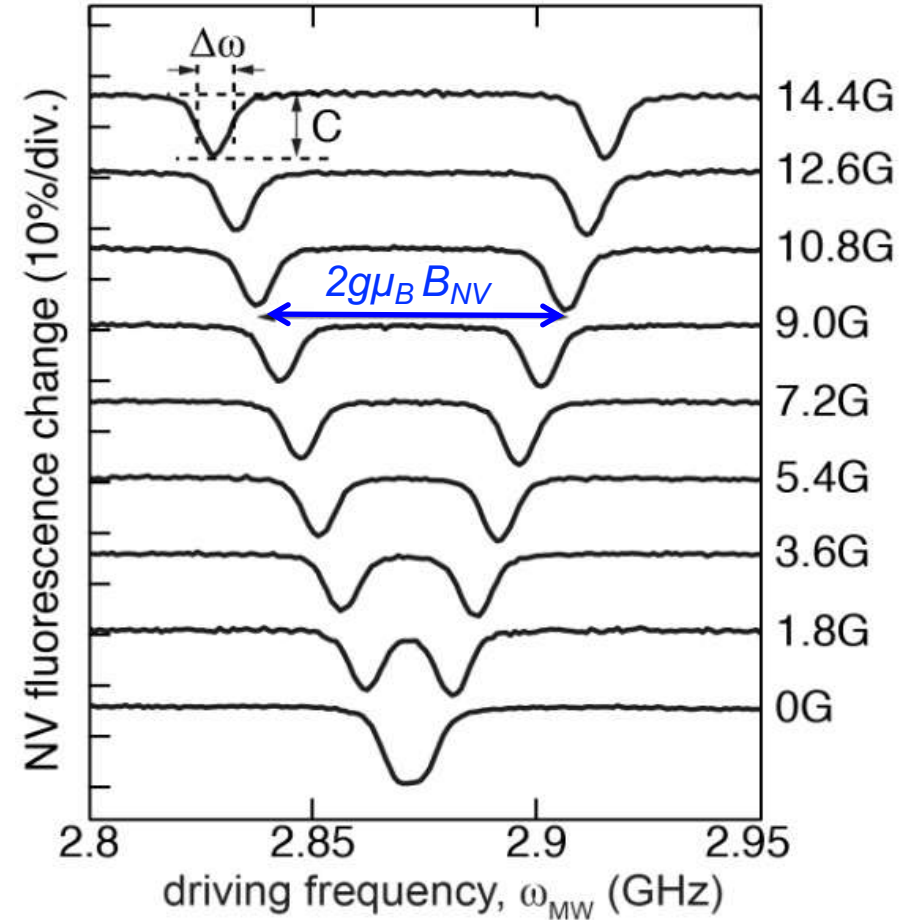
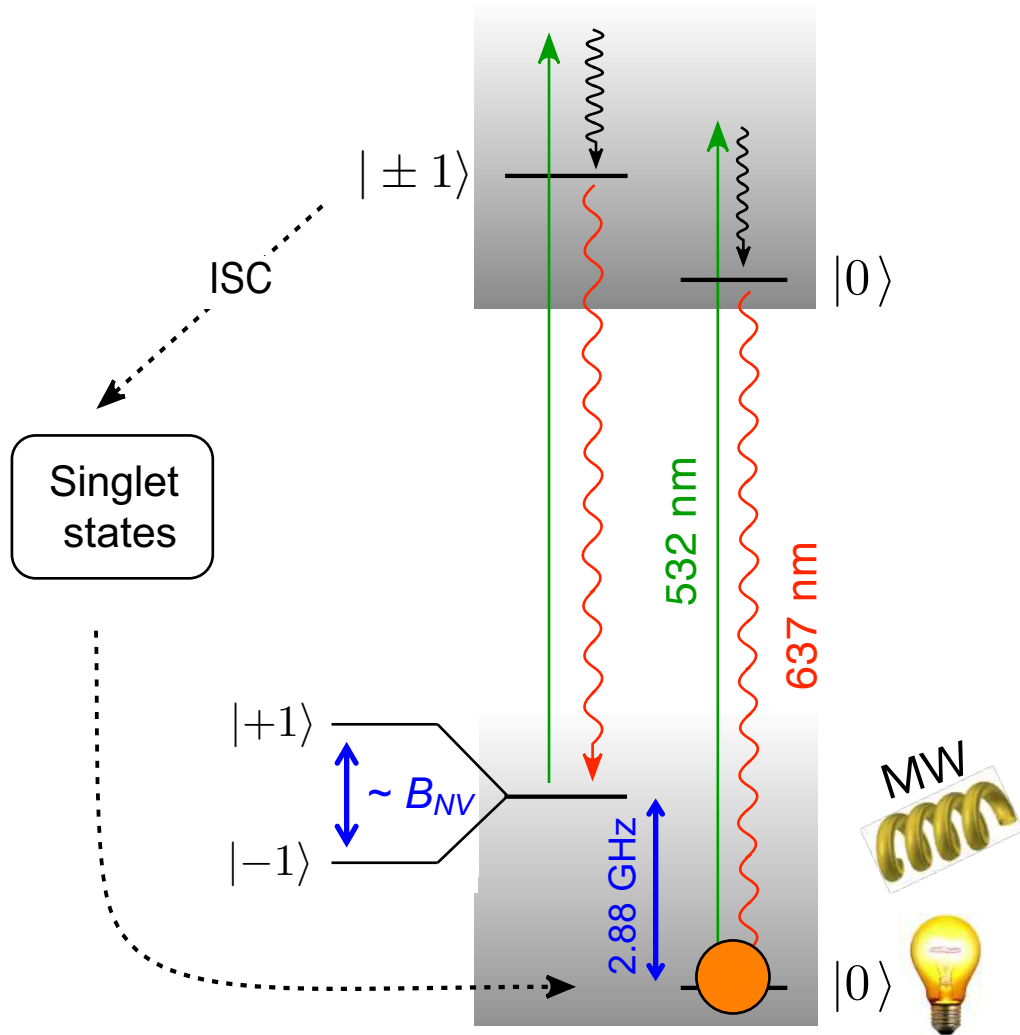
Spin properties

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Spin properties

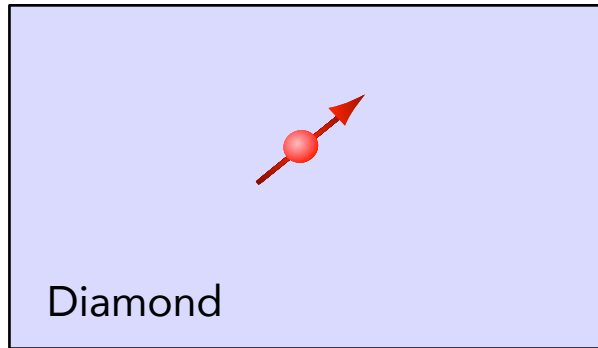
- Artificial atom with a spin triplet ($S=1$) ground state



NV = magnetometer

Magnetic sensing with NV defects

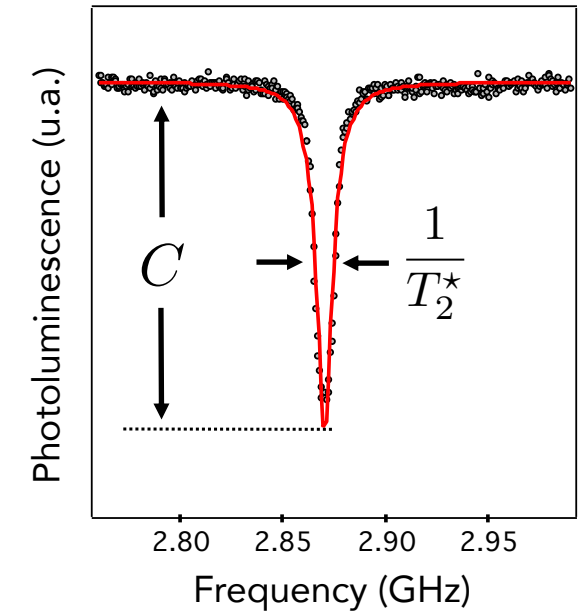
Single NV defect



Field sensitivity

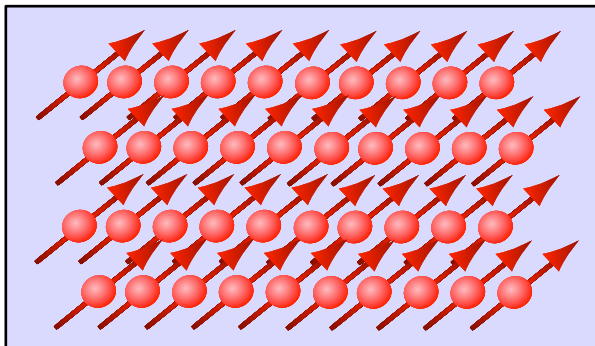
$$\eta \propto \frac{1}{C \sqrt{RT_2^*}}$$

ESR contrast Collection efficiency Coherence time



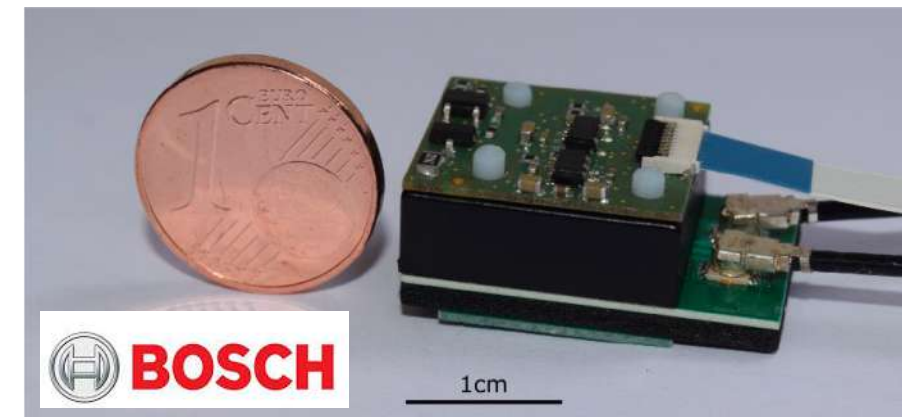
Barry *et al.*, *Rev. Mod. Phys.* **92**, 015004 (2020)

Ensemble of NV defects



$$\eta \propto \frac{1}{C \sqrt{RT_2^*}} \times \frac{1}{\sqrt{N}}$$

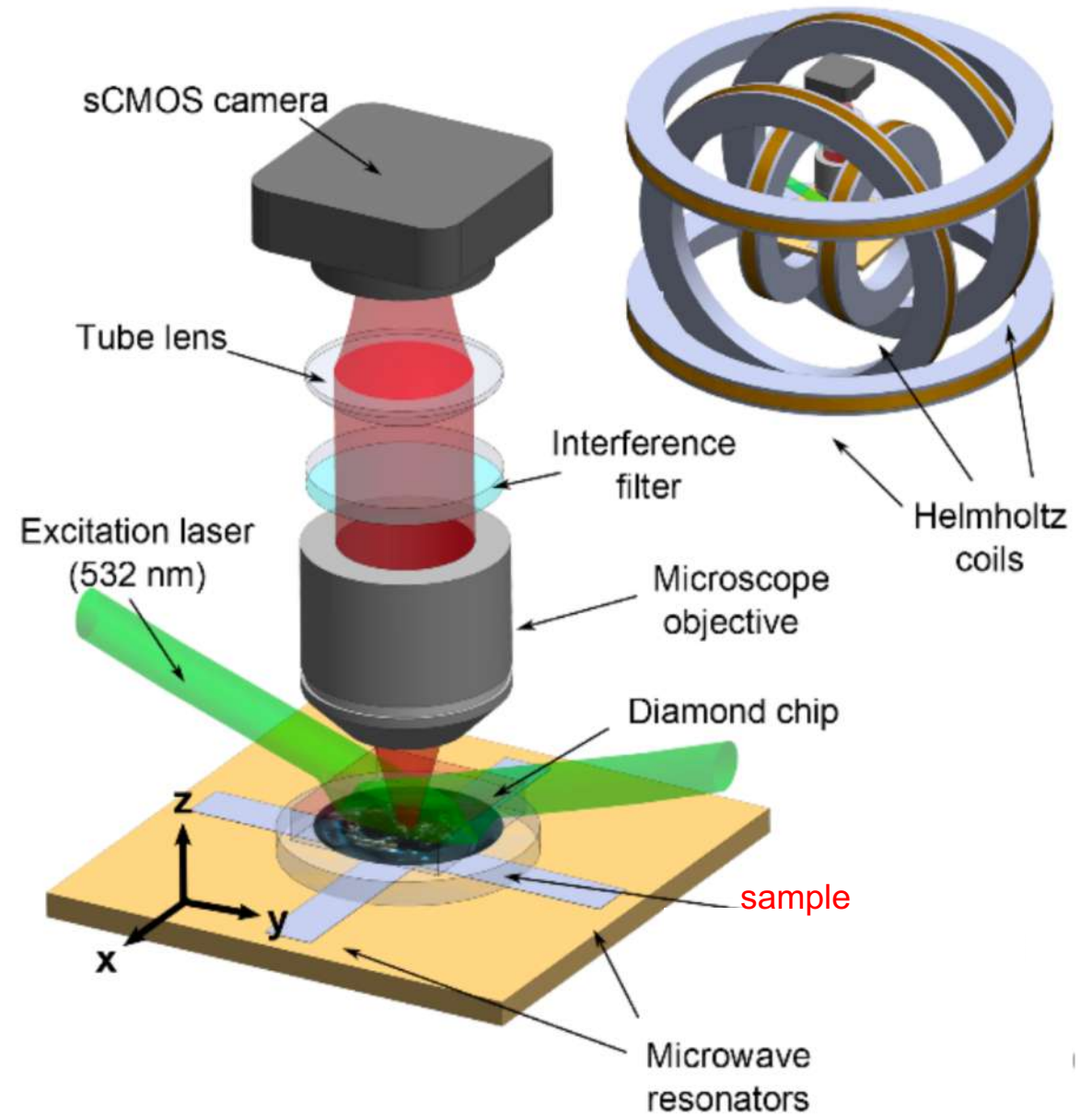
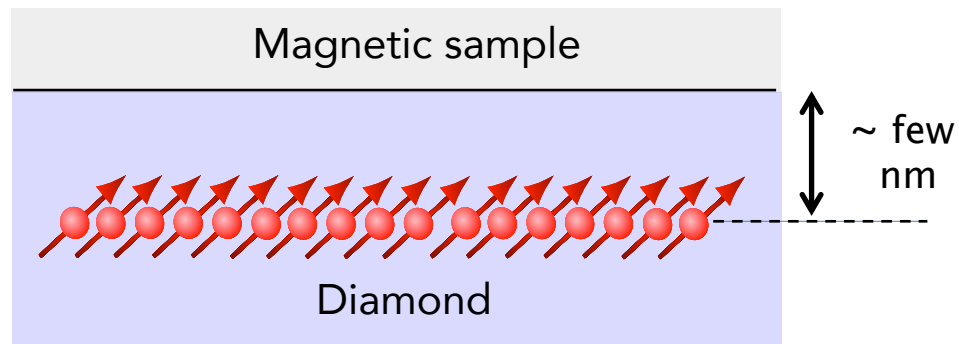
Number of NVs



Sensitivity down to few nT.Hz^{-1/2}

Magnetic imaging with an ensemble of NV defects

Using NV-doped layers close to the surface



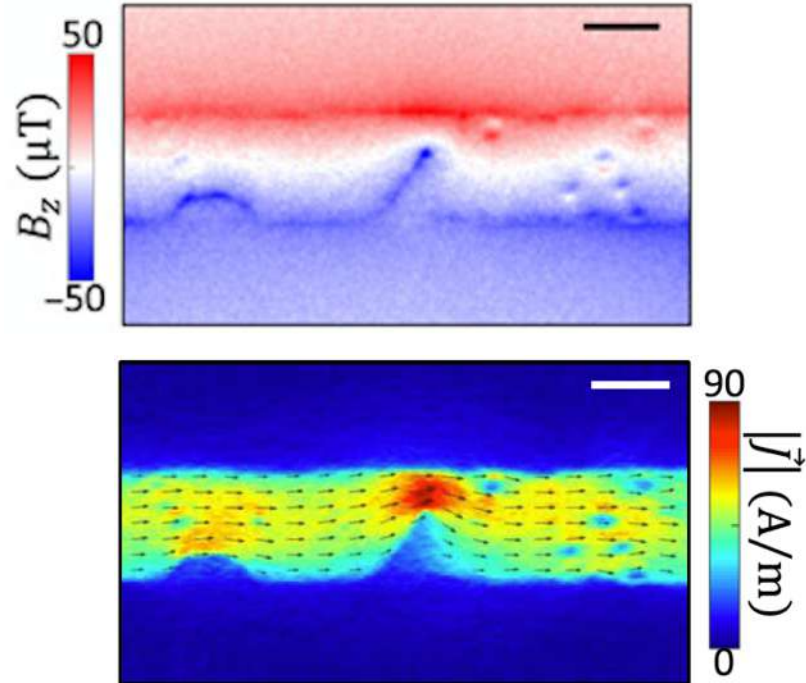
Glenn *et al.*, *Nat. Methods* **12**, 736 (2015)

Levine *et al.*, *Nanophotonics* **8**, 1945 (2019)

Magnetic imaging with an ensemble of NV defects

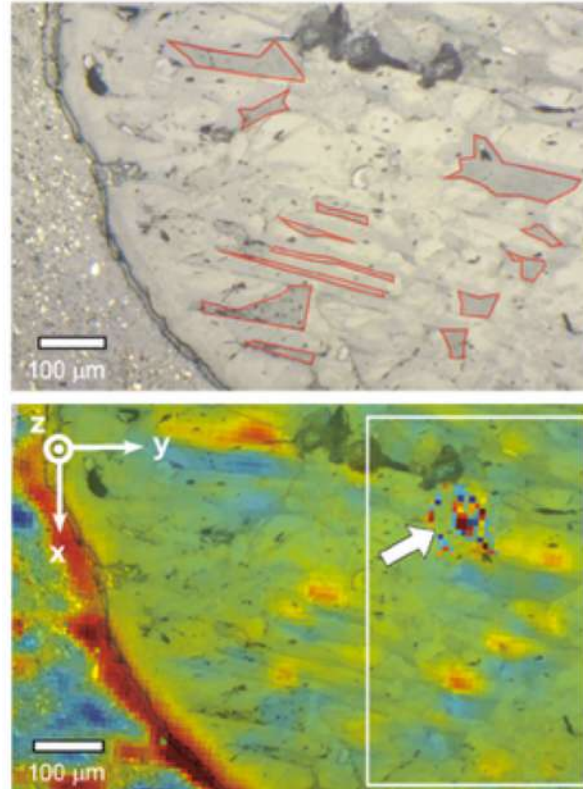
→ Condensed matter physics

current flow in graphene



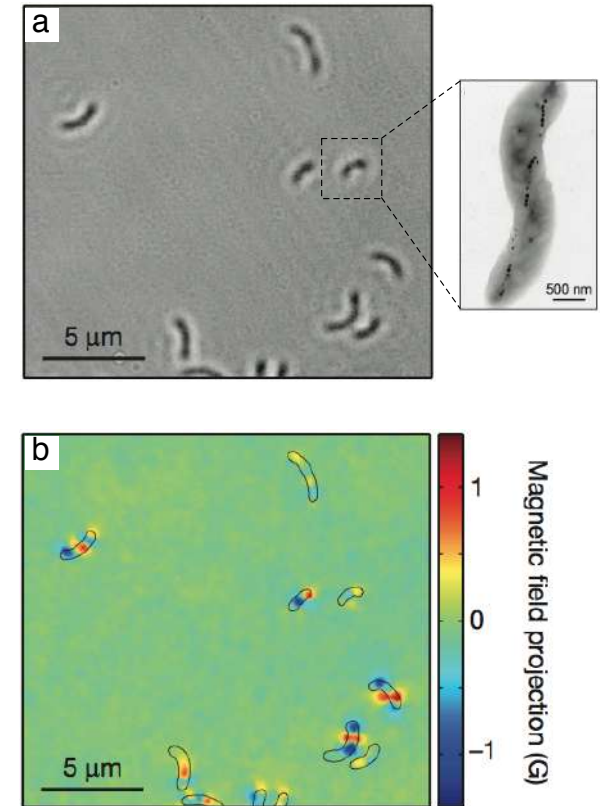
Tetienne, *Sci. Adv.* (2017)

→ Paleomagnetism



Glenn, *Geochem. GeoPhys.* (2017)

→ Biomagnetism

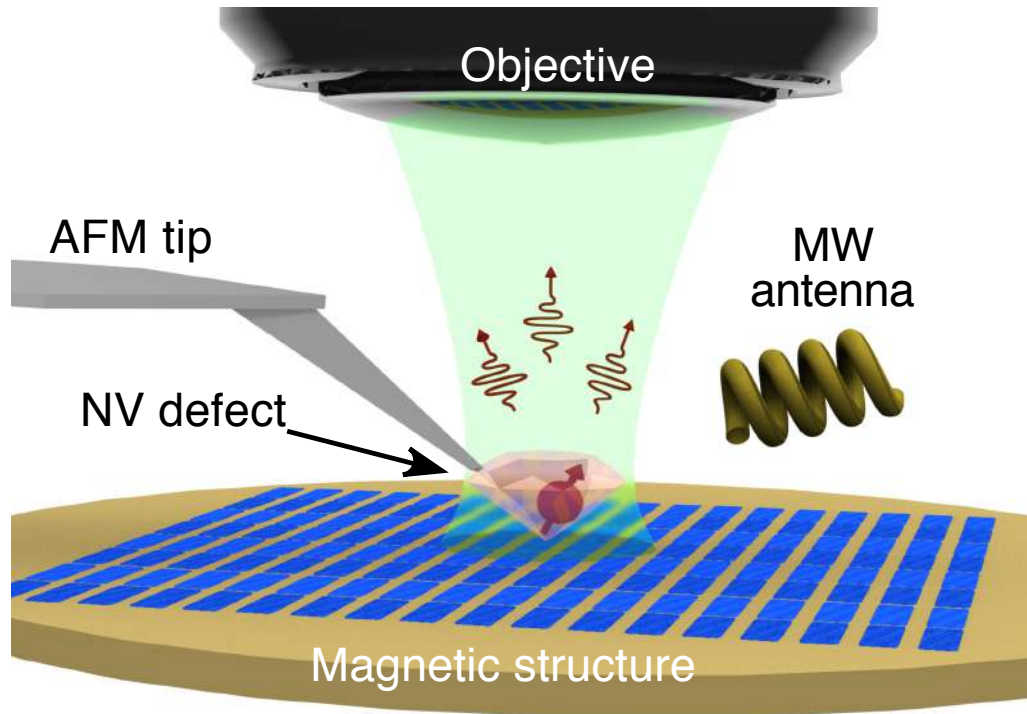


Le Sage, *Nature* (2013)

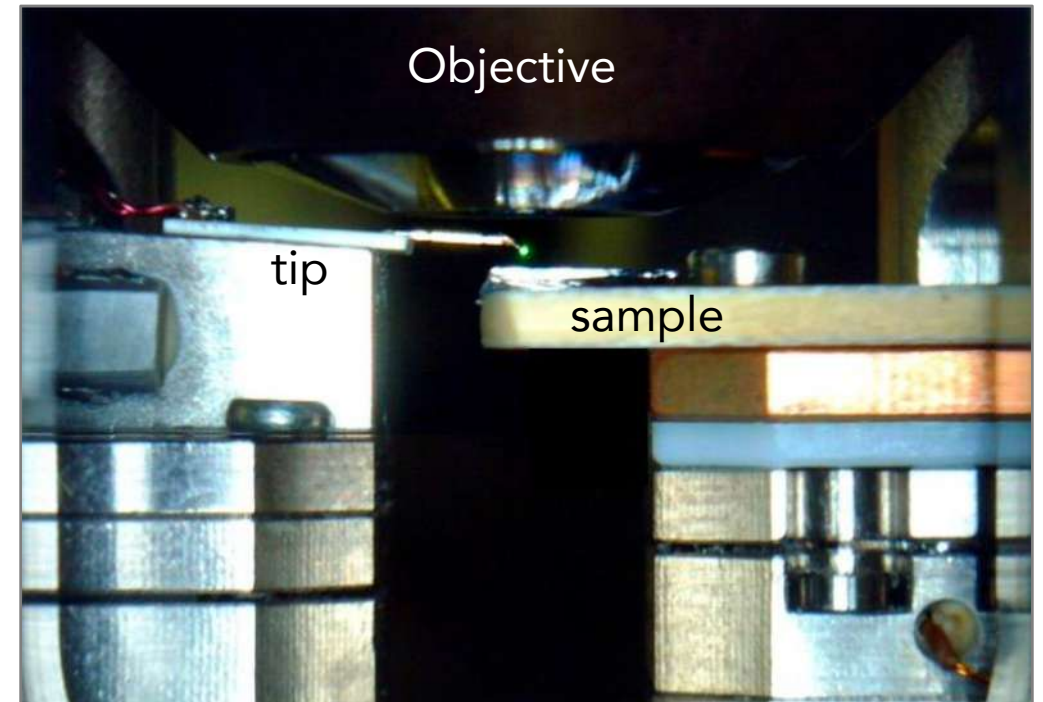
Spatial resolution limited by diffraction ($\sim 500 \text{ nm}$)

Magnetic imaging with a single NV defect

Scanning-NV magnetometry



Experimental setup

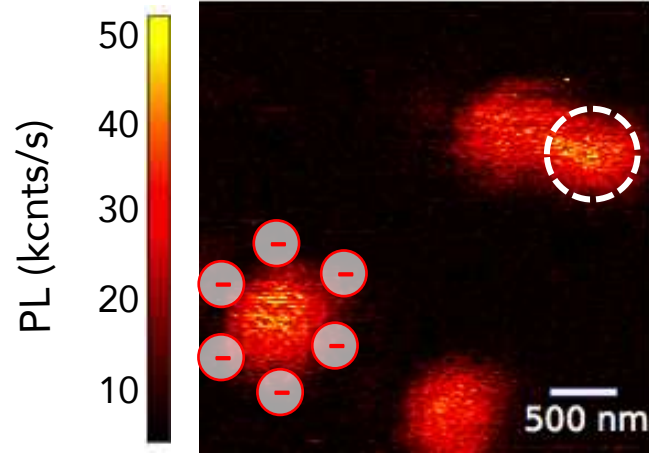


- ★ Quantitative/vectorial (sensitivity - $1 \mu\text{T}/\text{Hz}^{-1/2}$)
- ★ No magnetic back-action, operation from 4K to 300K
- ★ Atomic-size detection volume

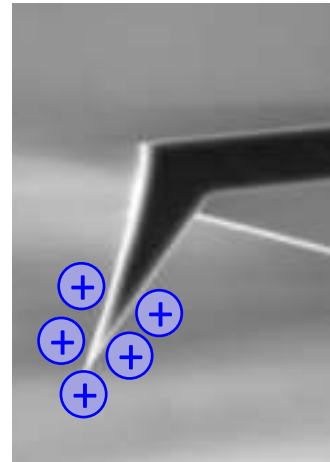
Rondin, [Appl. Phys. Lett.](#) (2012)

Engineering the NV-based sensor

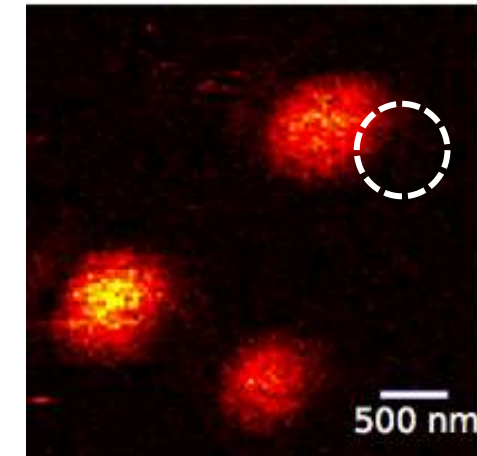
PL map of 20-40 nm diamond nanocrystal



Tip covered with a cationic polymer



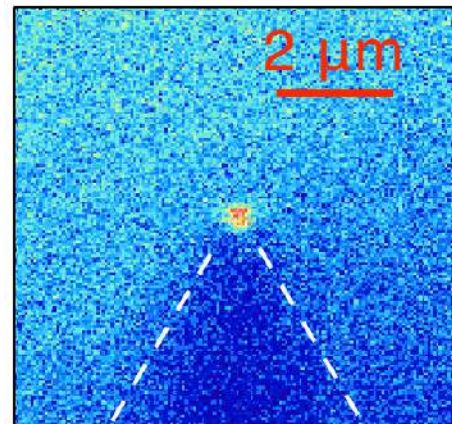
Grafting by electrostatic interaction



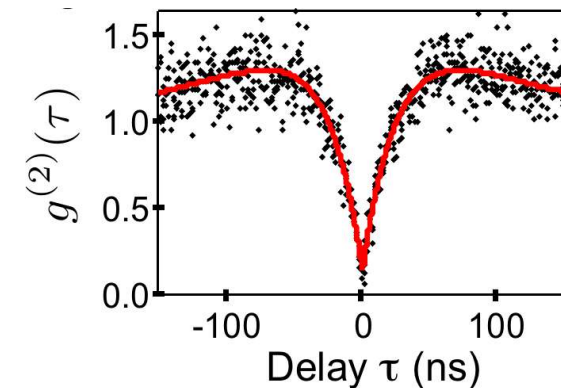
Cuche, *Opt. Exp.* 17, 19969 (2009)

➔ Photoluminescence raster scan of the AFM tip after grafting

Rondin, *Appl. Phys. Lett.* (2012)



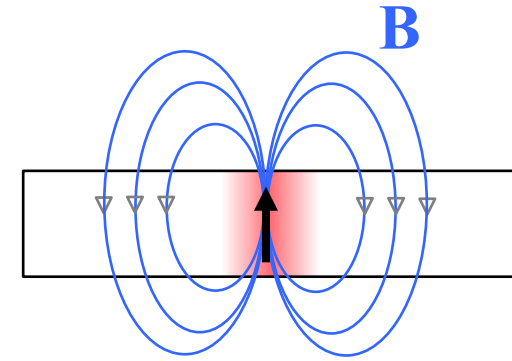
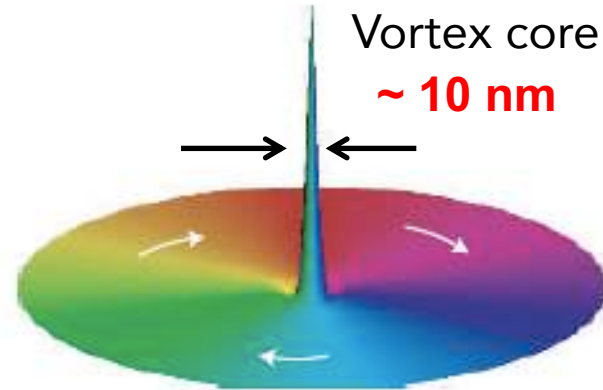
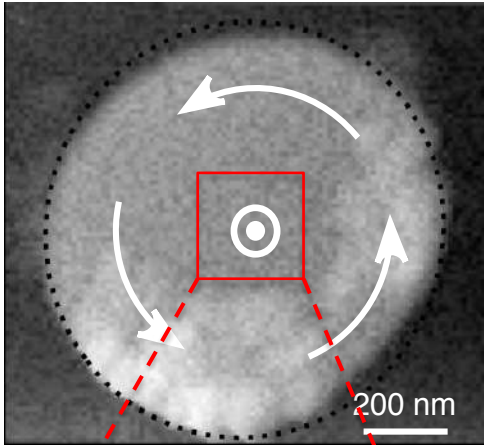
Single NV at the tip apex !



Imaging the core of a magnetic vortex

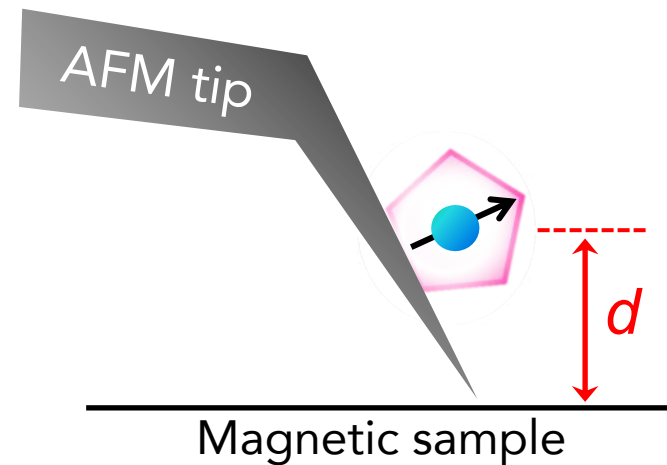
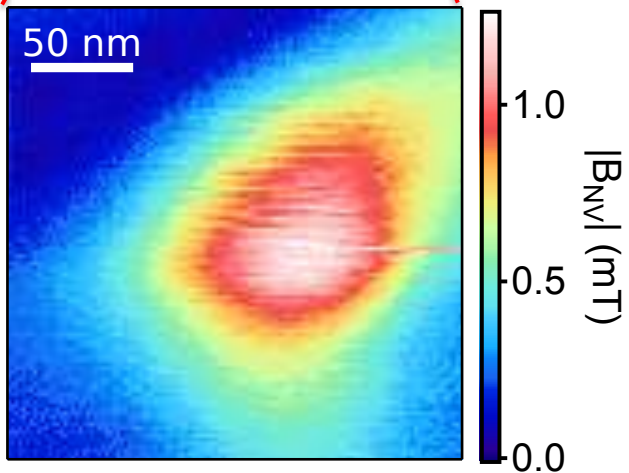
AFM image

50-nm thick disk of FeNi



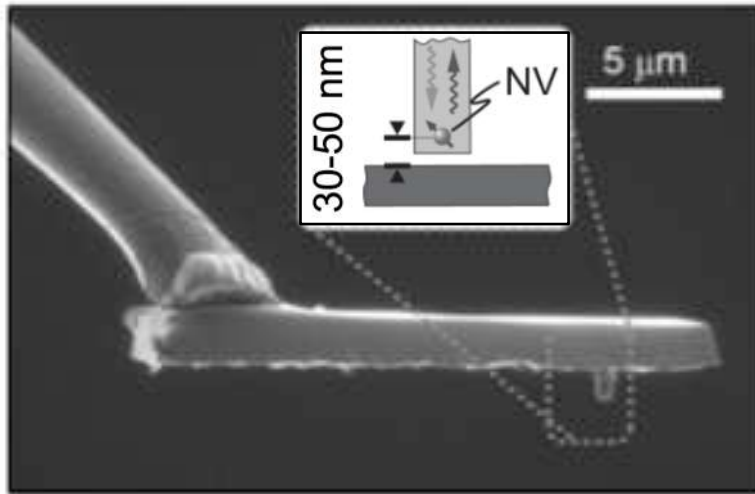
→ Resolving power $\sim 100\text{-}150\text{ nm}$
Limited by the probe-to-sample distance d

NV image



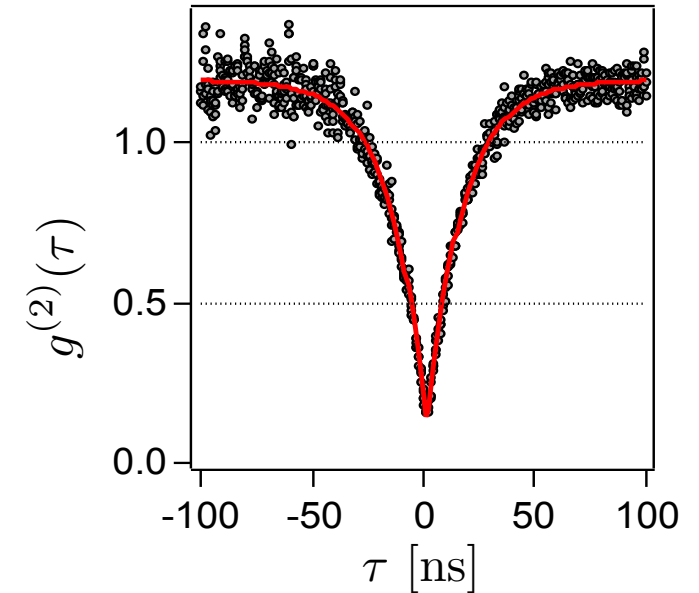
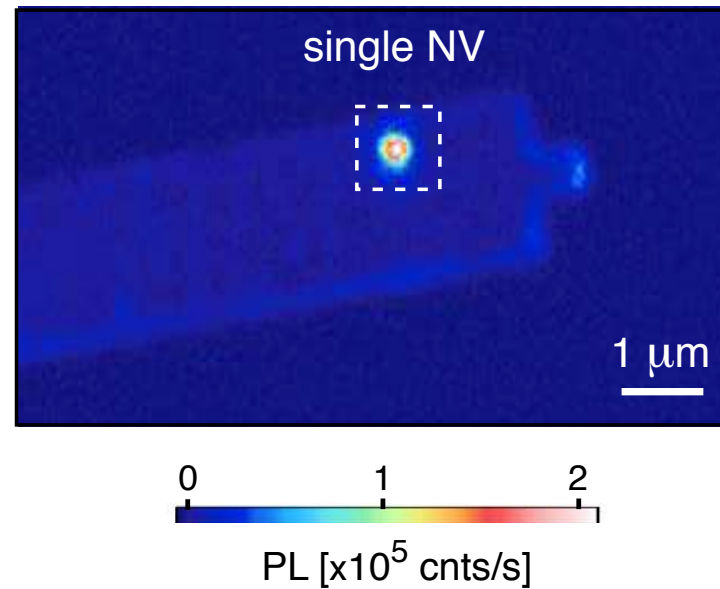
Improving the resolving power with all-diamond scanning tips

SEM image



Maletinsky, Nat. Nano. (2012)
Appel, Rev. Sci. Inst. (2016)

PL map of the diamond tip

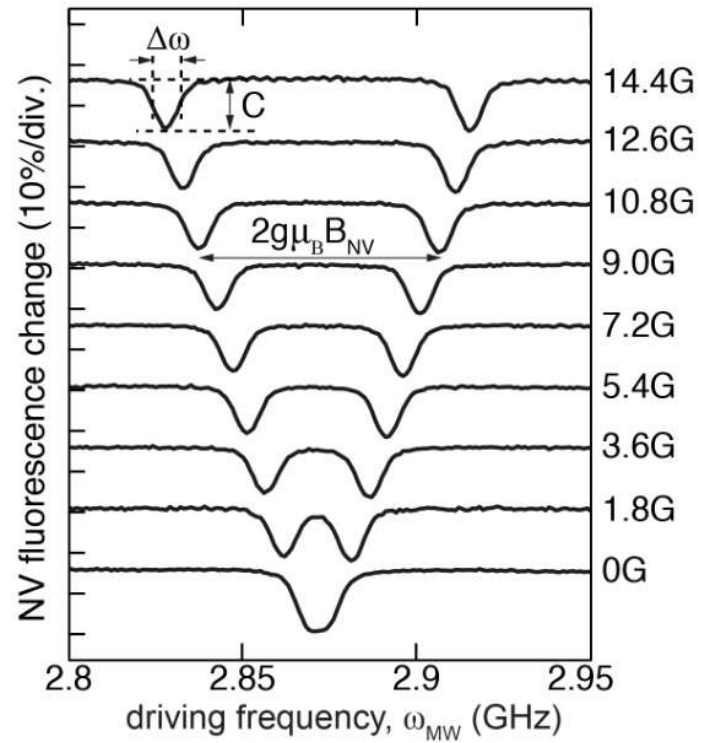
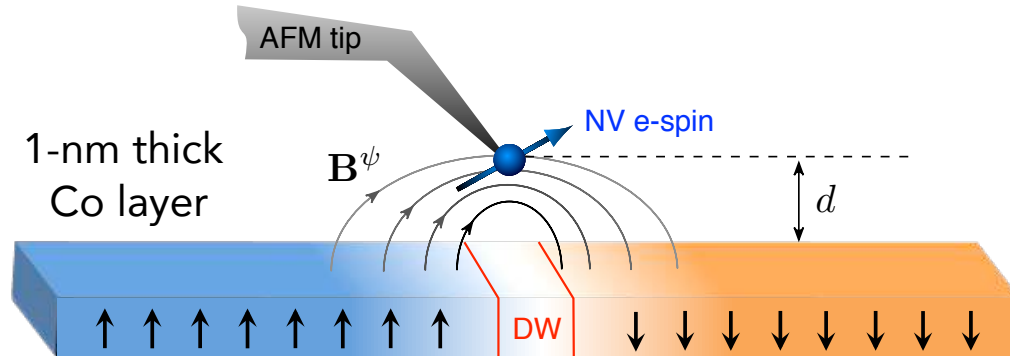


➔ Resolving power ~ 30-50 nm

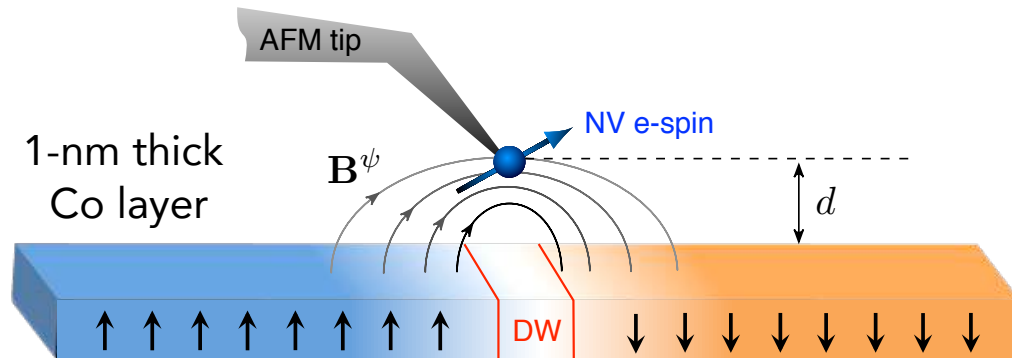
Now even commercially available !



One application: Imaging domain walls in thin ferromagnets

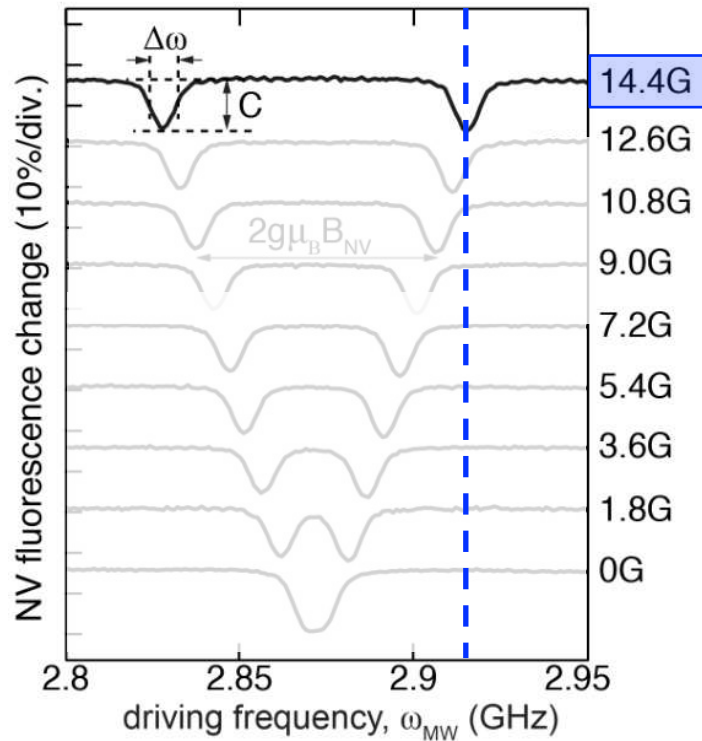
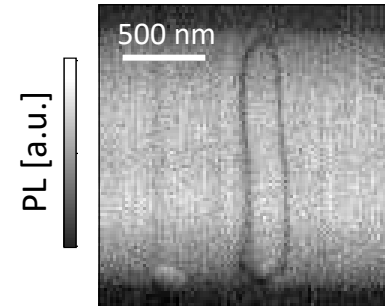


One application: Imaging domain walls in thin ferromagnets

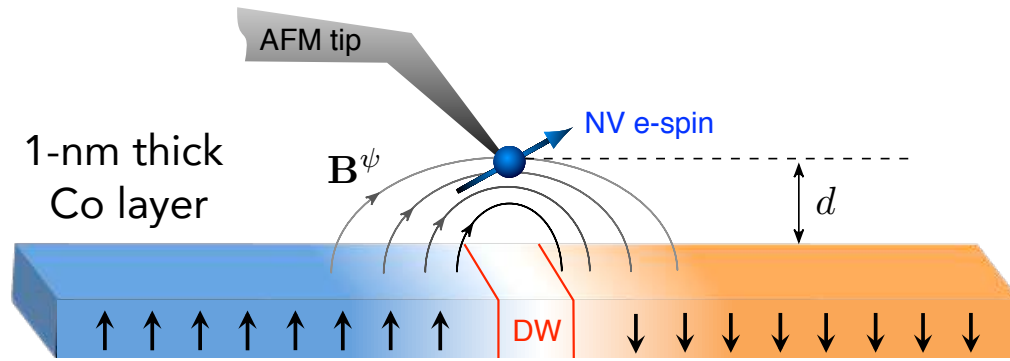


→ “Iso- B ” imaging mode

$f_0 = 2915$ MHz [14G]



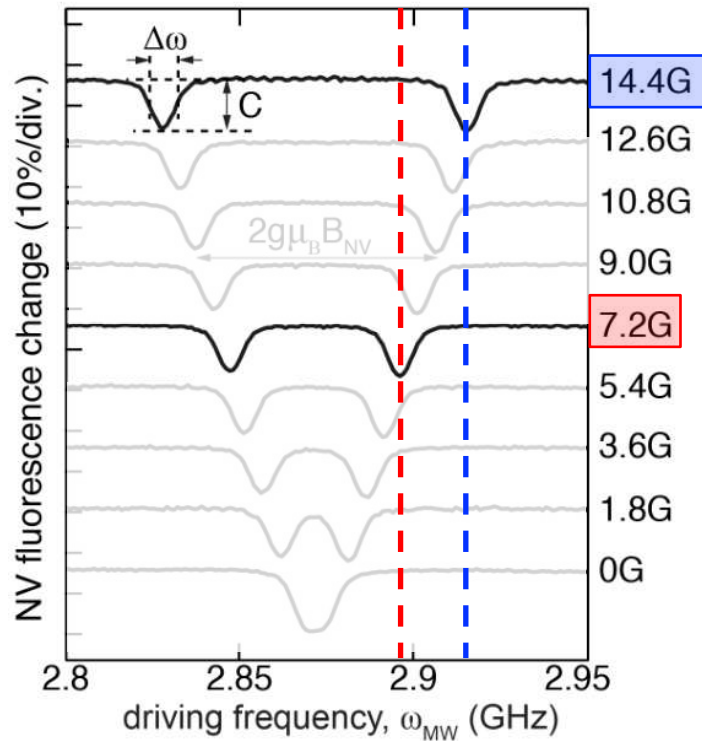
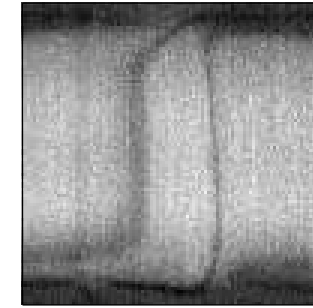
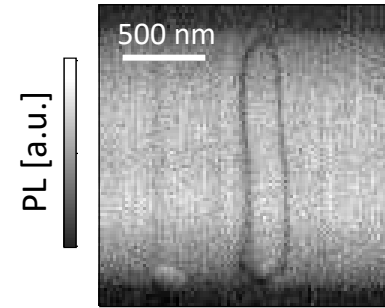
One application: Imaging domain walls in thin ferromagnets



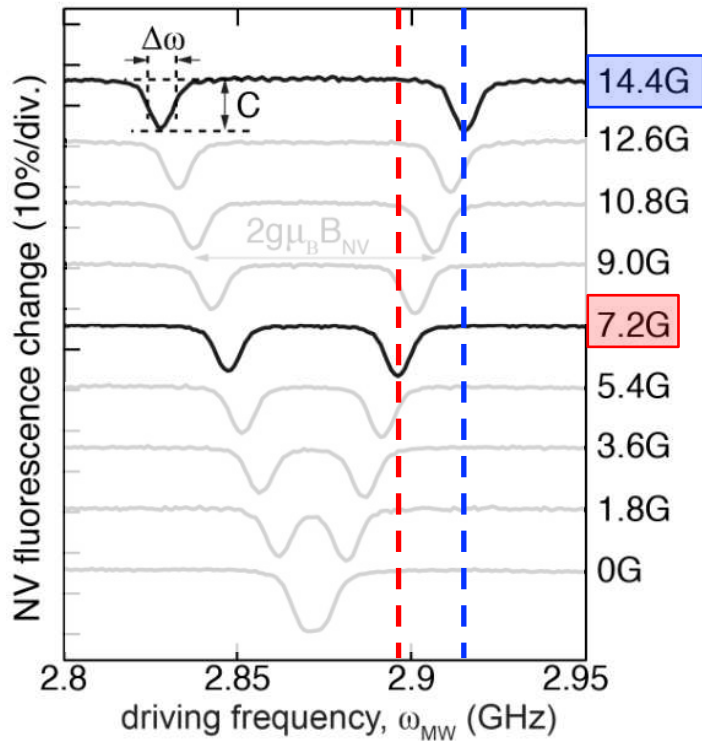
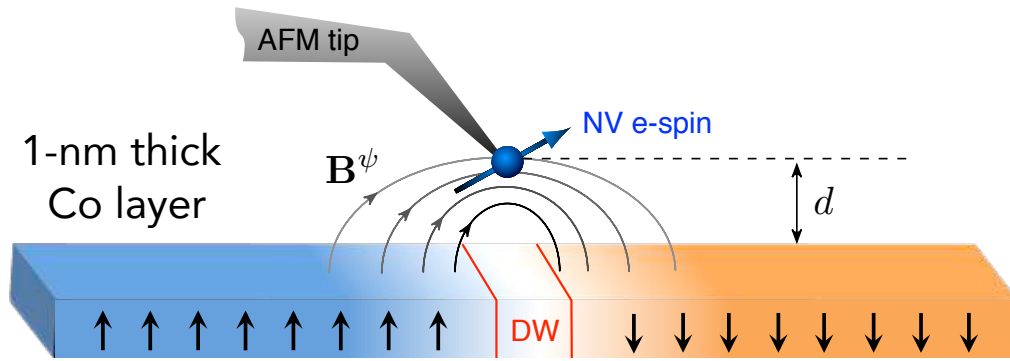
→ “Iso-B” imaging mode

$f_0 = 2915$ MHz [14G]

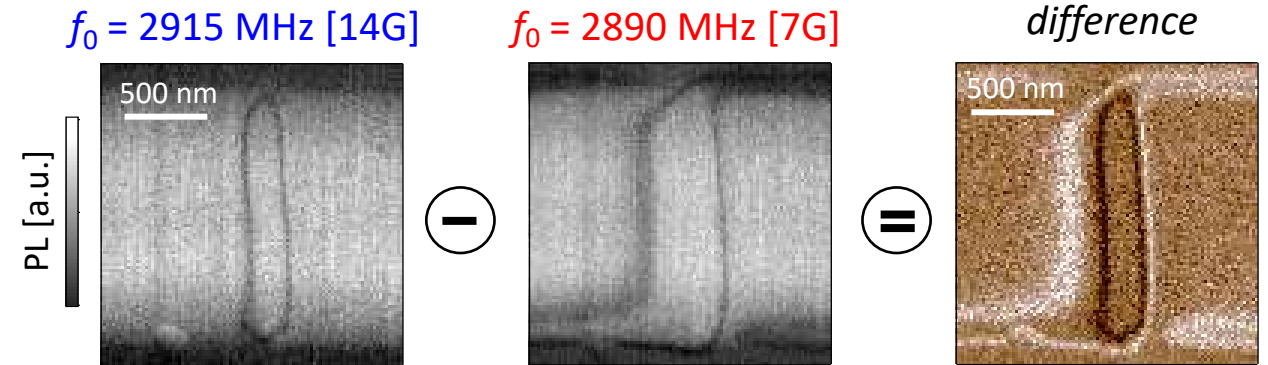
$f_0 = 2890$ MHz [7G]



One application: Imaging domain walls in thin ferromagnets

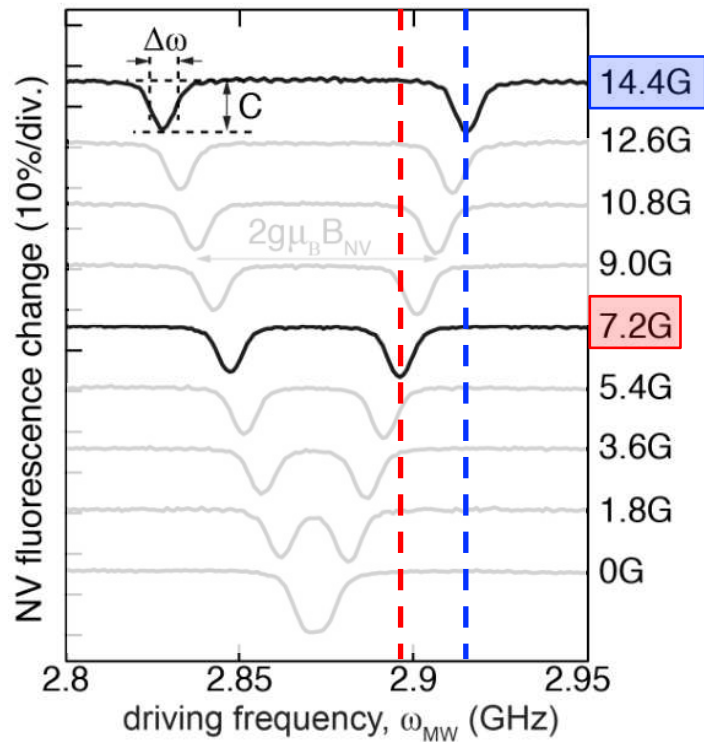
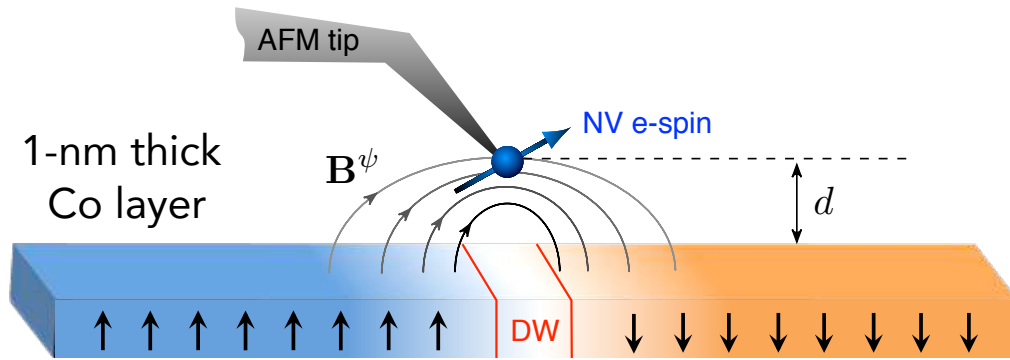


→ “Iso-B” imaging mode

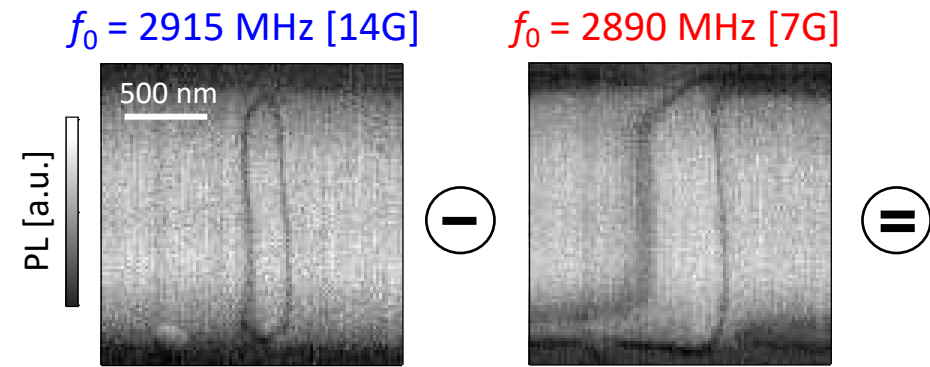


Tetienne, *Science* (2014)

One application: Imaging domain walls in thin ferromagnets



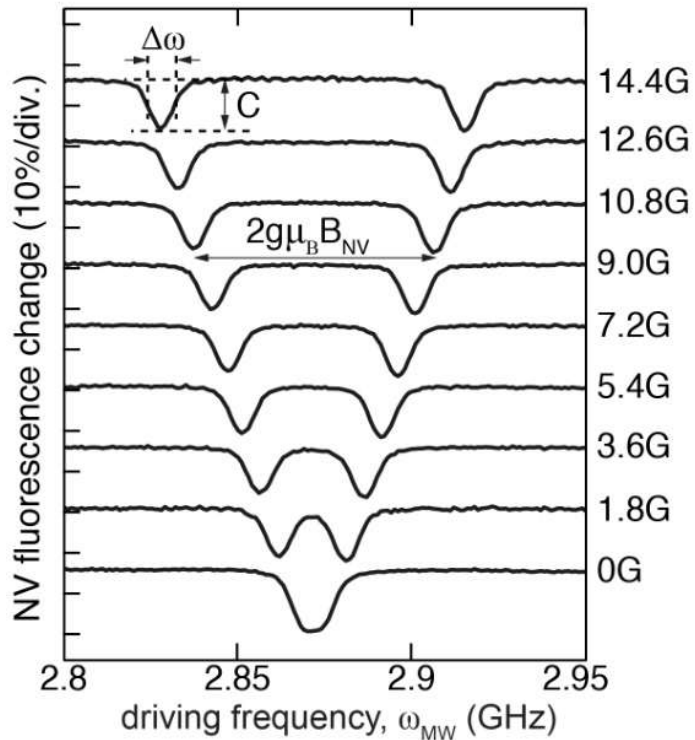
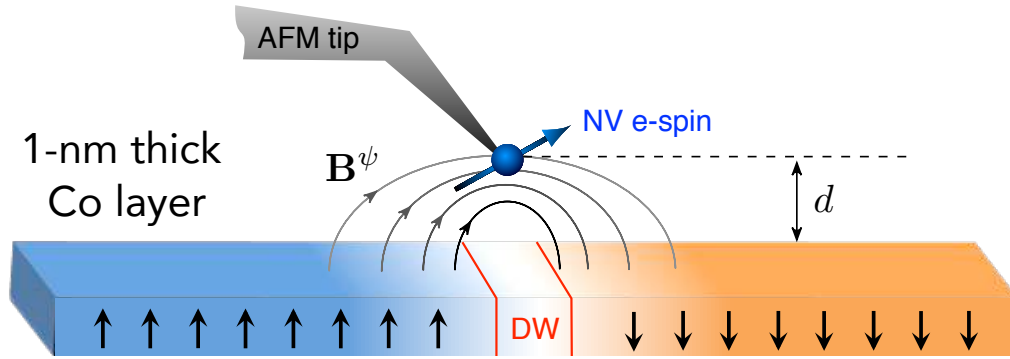
→ “Iso-B” imaging mode



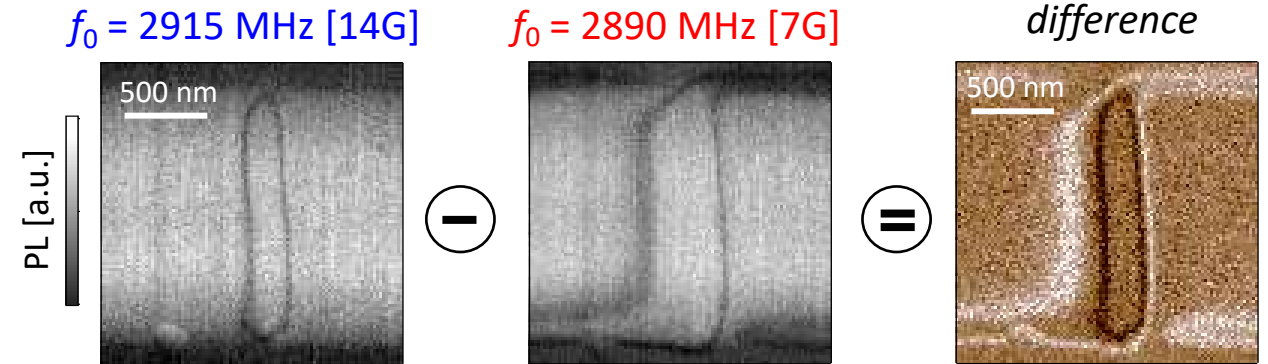
Tetienne, *Science* (2014)



One application: Imaging domain walls in thin ferromagnets

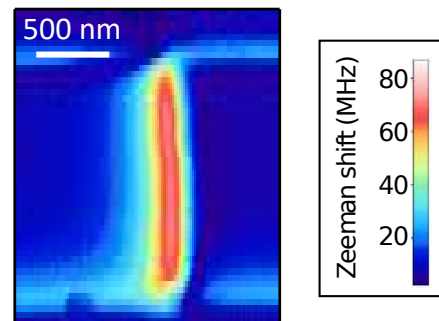


→ “Iso-B” imaging mode



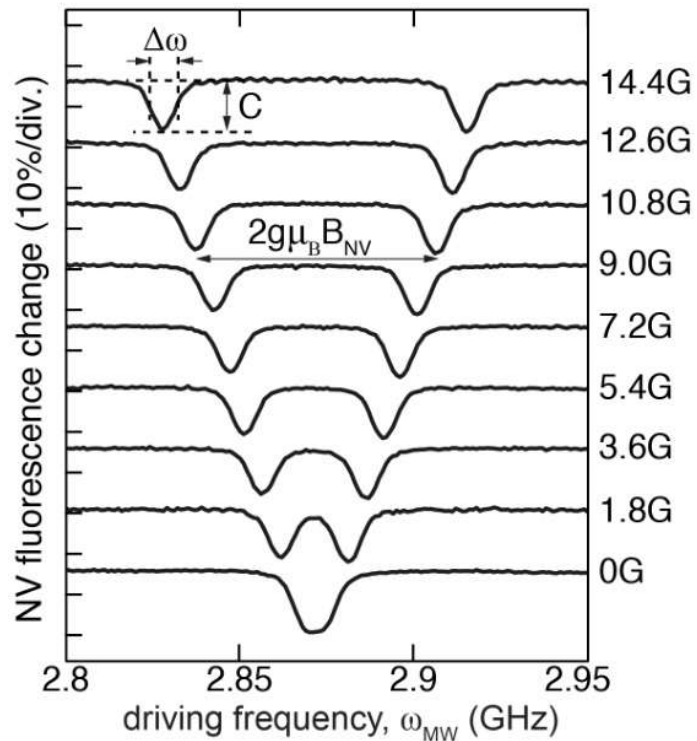
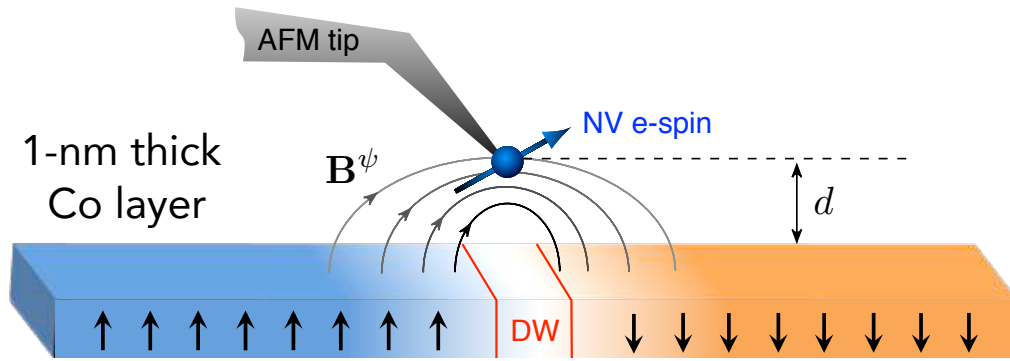
Tetienne, *Science* (2014)

→ “full-B” imaging mode

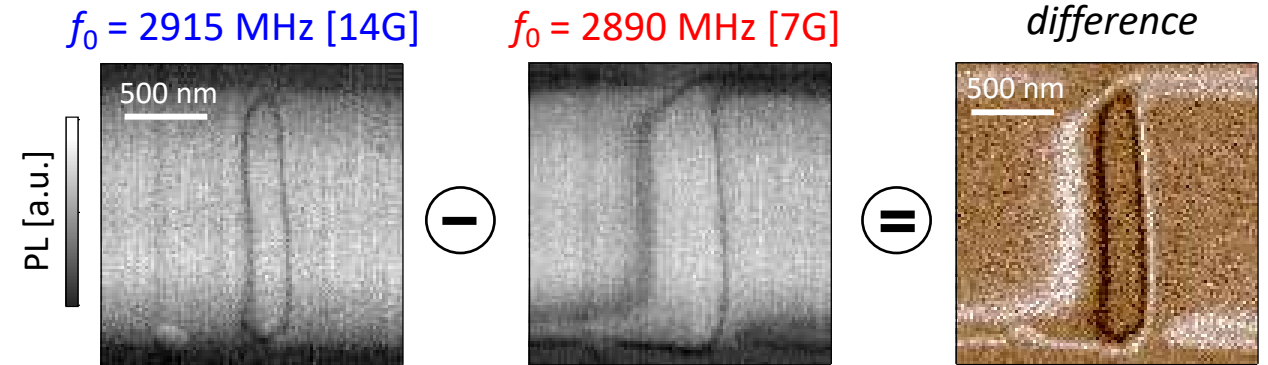


Tetienne, *Nat. Com.* (2015)

One application: Imaging domain walls in thin ferromagnets



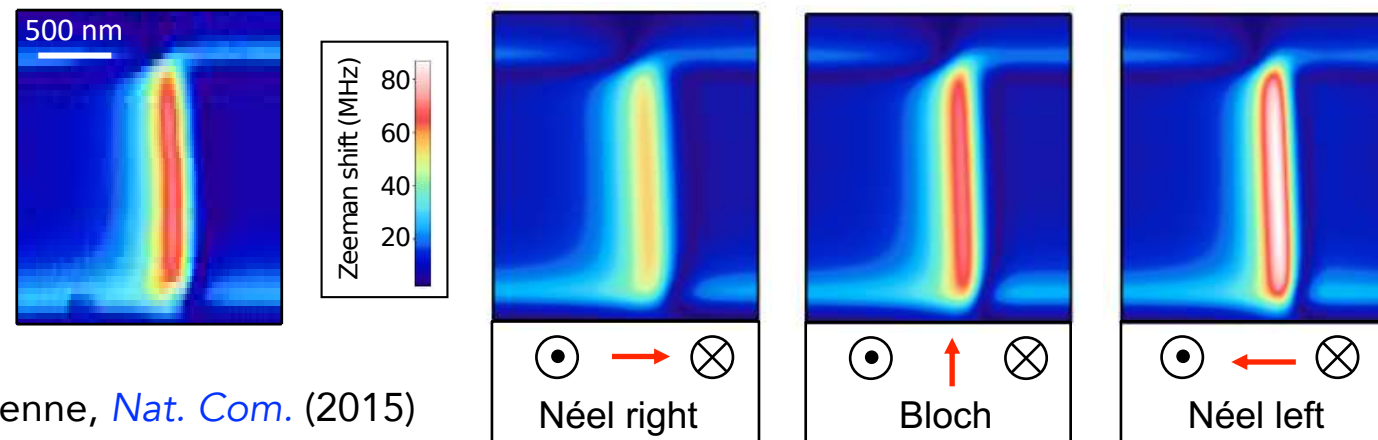
→ “Iso-B” imaging mode



Tetienne, *Science* (2014)

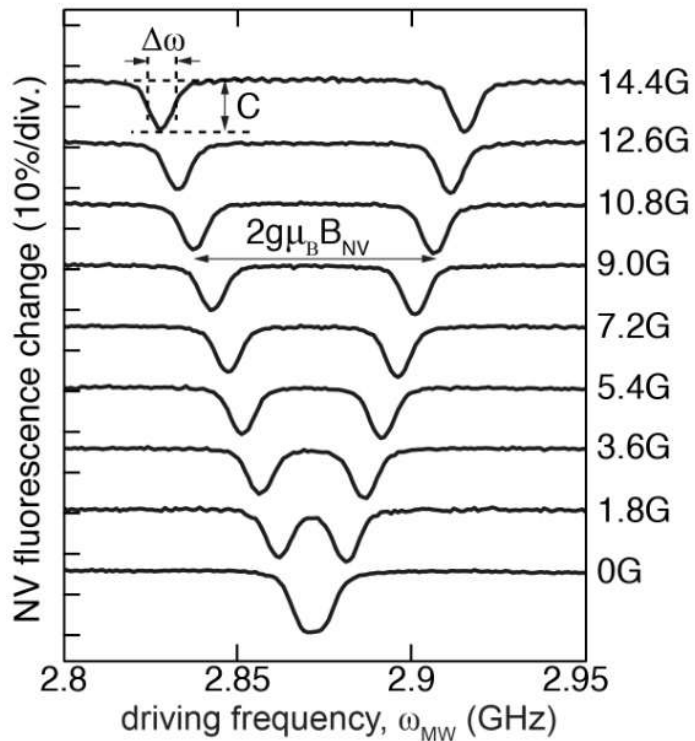
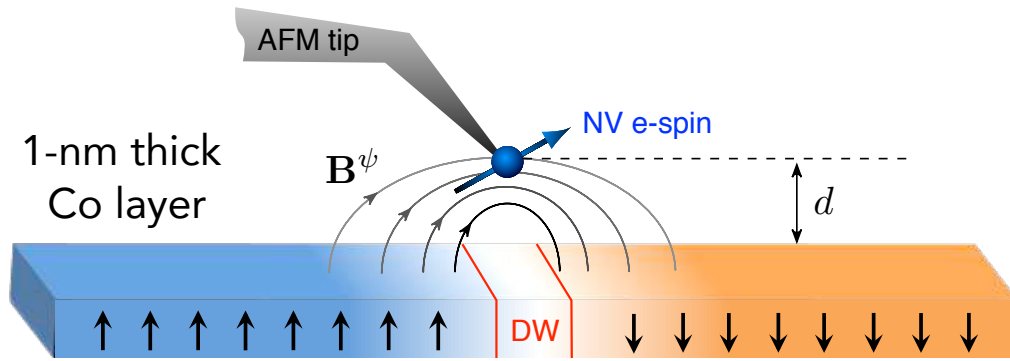
→ “full-B” imaging mode

Comparison with theoretical predictions

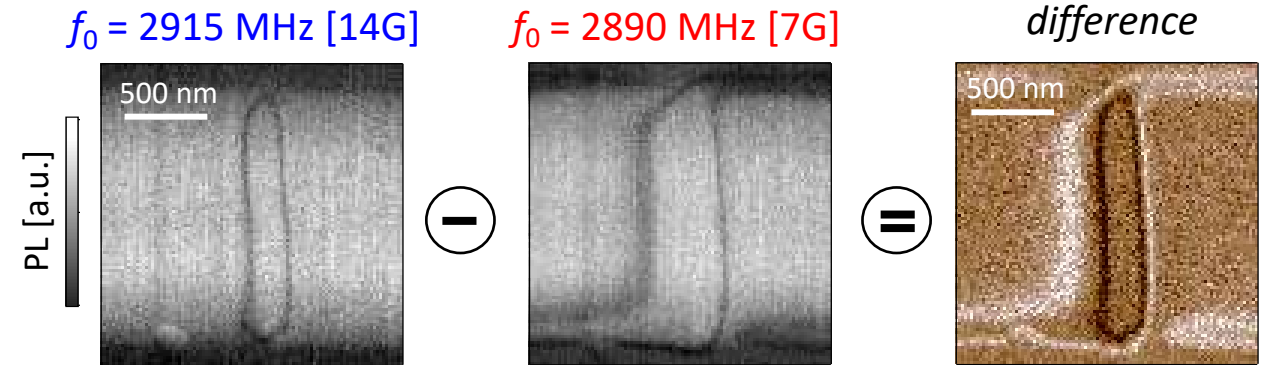


Tetienne, *Nat. Com.* (2015)

One application: Imaging domain walls in thin ferromagnets

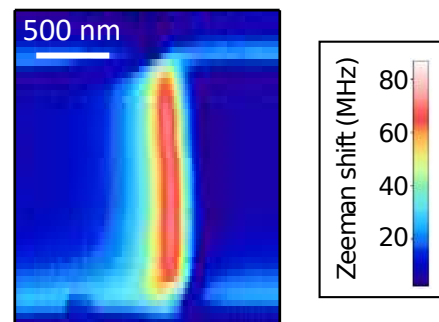


→ “Iso-B” imaging mode



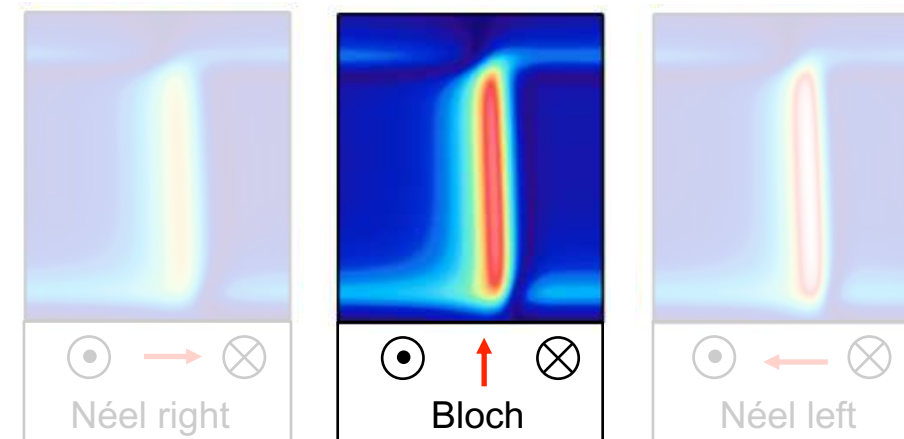
Tetienne, *Science* (2014)

→ “full-B” imaging mode

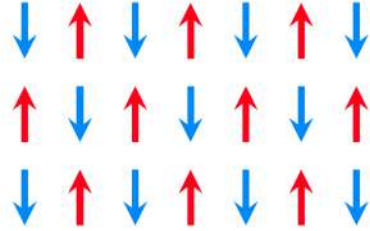


Tetienne, *Nat. Com.* (2015)

Comparison with theoretical predictions



Exploring the physics of antiferromagnetic (AF) materials

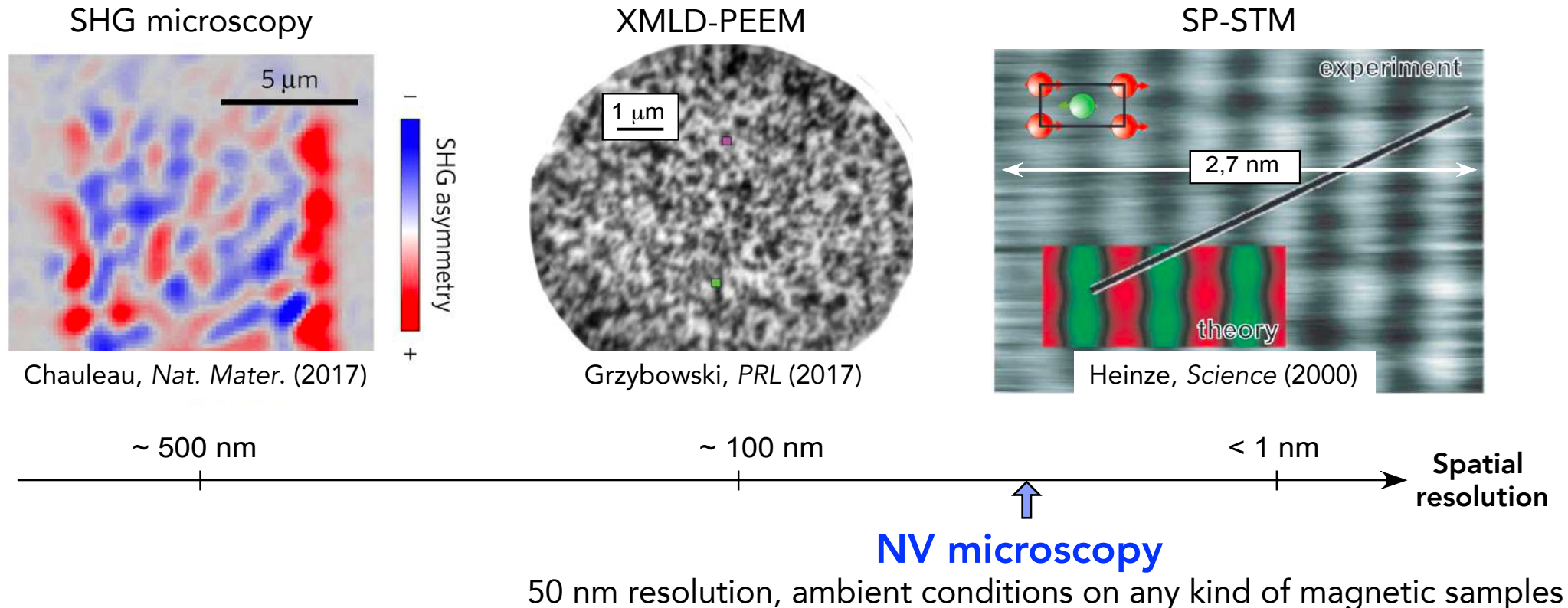


- ★ Robust against magnetic perturbations;
- ★ Ultrafast dynamics (*THz vs GHz for ferromagnets*);

→ Appealing materials for spintronics

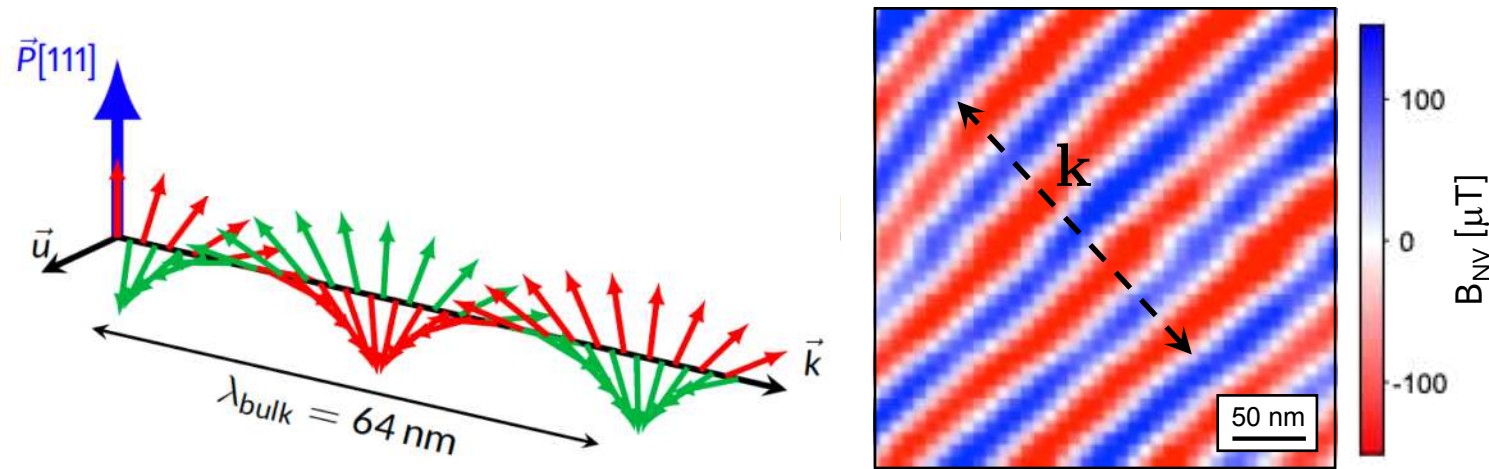
Baltz, *Rev. Prog. Phys.* (2018)

One challenge → imaging the antiferromagnetic order at the nanoscale



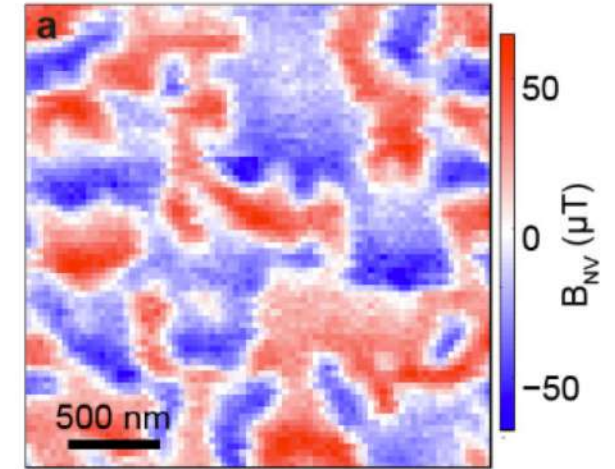
Imaging the AF order with scanning-NV magnetometry

Cycloidal AF order in BiFeO_3



Gross, *Nature* **549**, 252 (2017)
Chauleau, *Nat. Materials* **19**, 386 (2020)
Haykal, *Nat. Commun.* **11**, 1704 (2020)

AF domain walls in Cr_2O_3

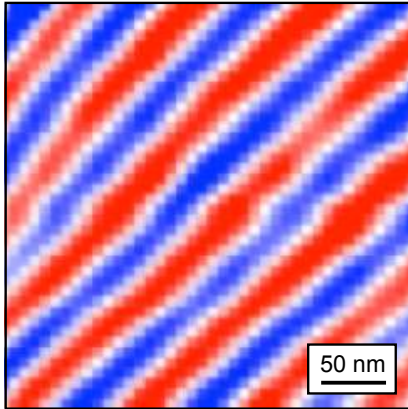


Appel, *Nano Lett.* **19**, 1682 (2019)
Hedrich, *arXiv:2009.08986*
Wornle, *arXiv:2009.09015*

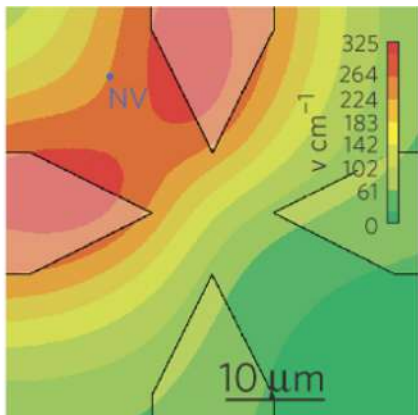
Very promising techniques to investigate the physics of antiferromagnetic materials

A multimode sensor

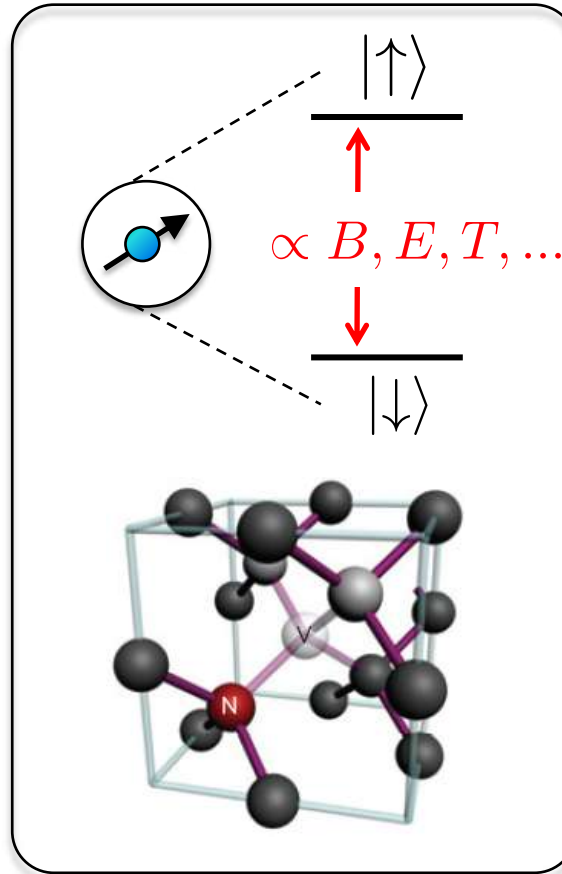
Static magnetic field



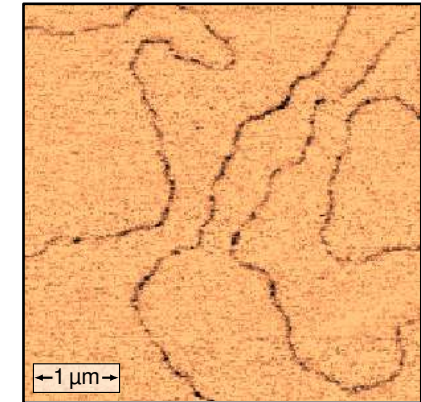
Electric field



Dolde, *Nat. Phys.* **7**, 459 (2011)

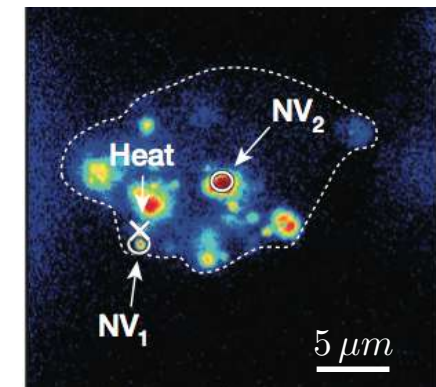


Magnetic noise



Finco, *Nat. Comm.* (2021)

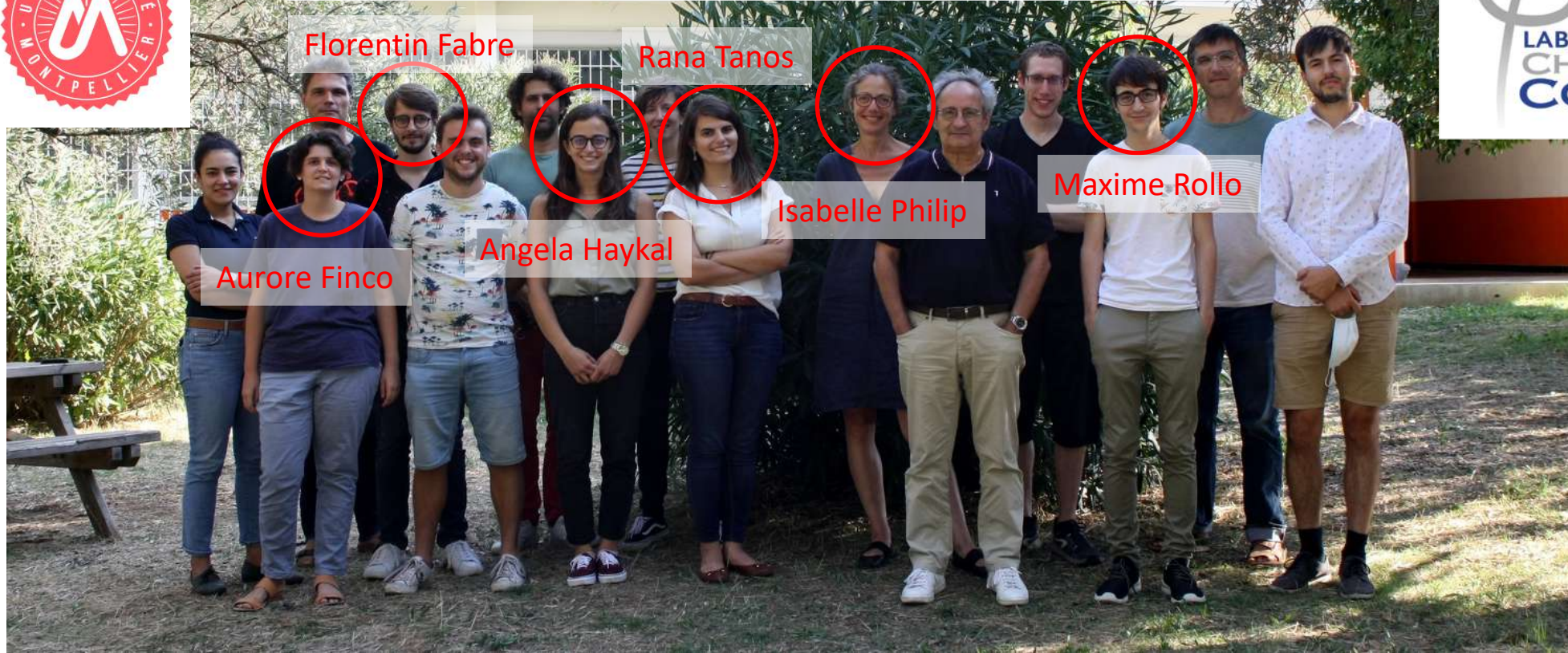
Temperature



Kucsko, *Nature* **500**, 54 (2013)



Team « Solid-State Quantum Technologies »



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- M. Viret, J.-Y. Chauleau – [CEA Saclay](#)
- J. V. Kim, T. Devolder, J.-P. Adam – [C2N](#)
- N. Jaouen - [Soleil](#)

Fundings

