The NV center in diamond & applications in nanoscale magnetometry

Anaïs Dréau

Laboratoire Charles Coulomb, CNRS & Montpellier University, France

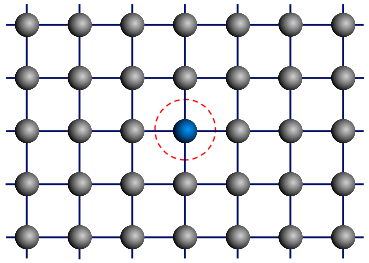
anais.dreau@umontpellier.fr

Introduction to spin defects in semiconductors

Quantum communication

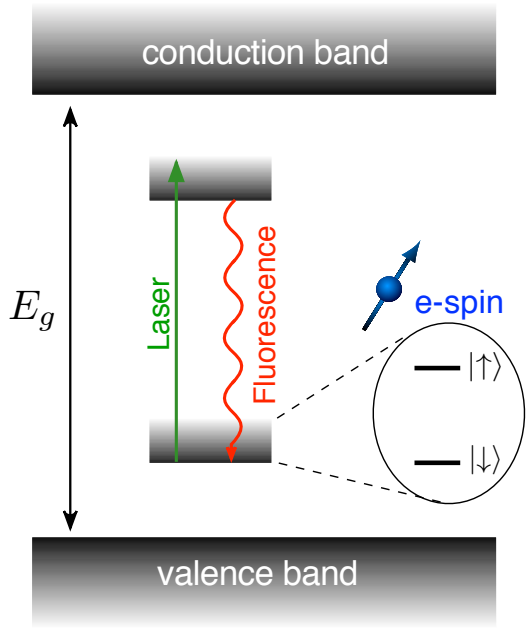
Biology imaging

Mohan et al., *Nano Lett. 10* (2010)

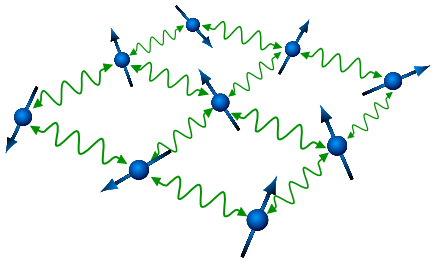


solid-state

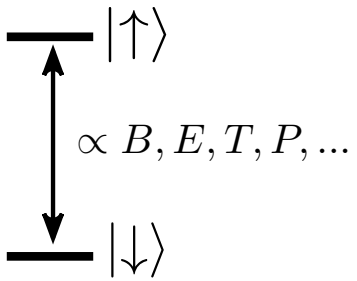
« artificial atom »



Quantum information

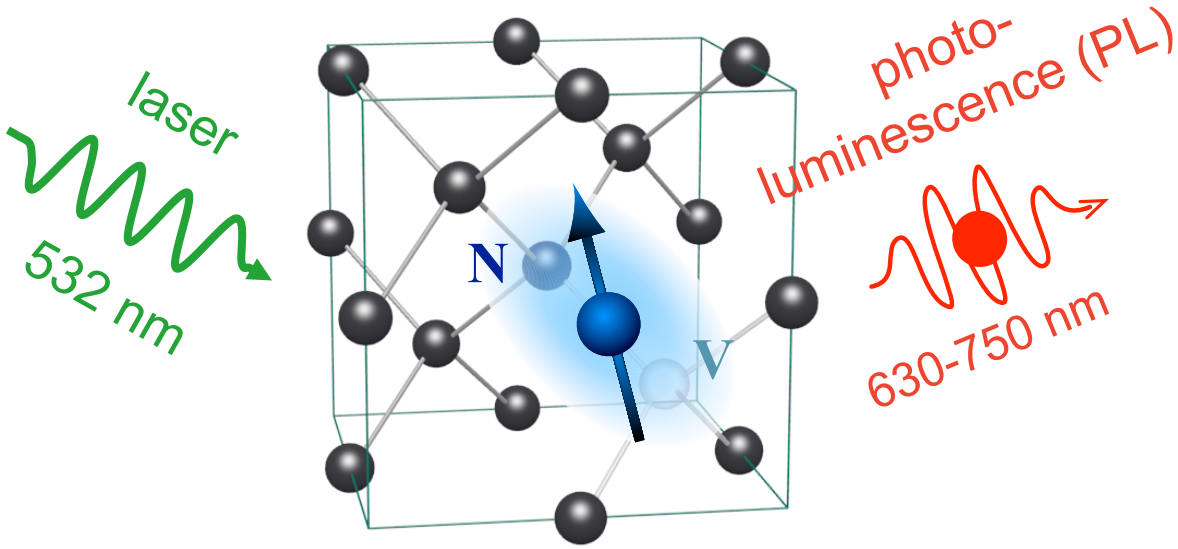


Sensing



1. The NV center in diamond

The Nitrogen-Vacancy center in diamond



- 1st isolation at single scale in 1997

  Gruber et al., *Science* ***276*** (1997)

- "native" defects or created by implantation, laser writing

  Pezzagna et al., *Phys. Stat. Sol. A* ***208*** (2011)

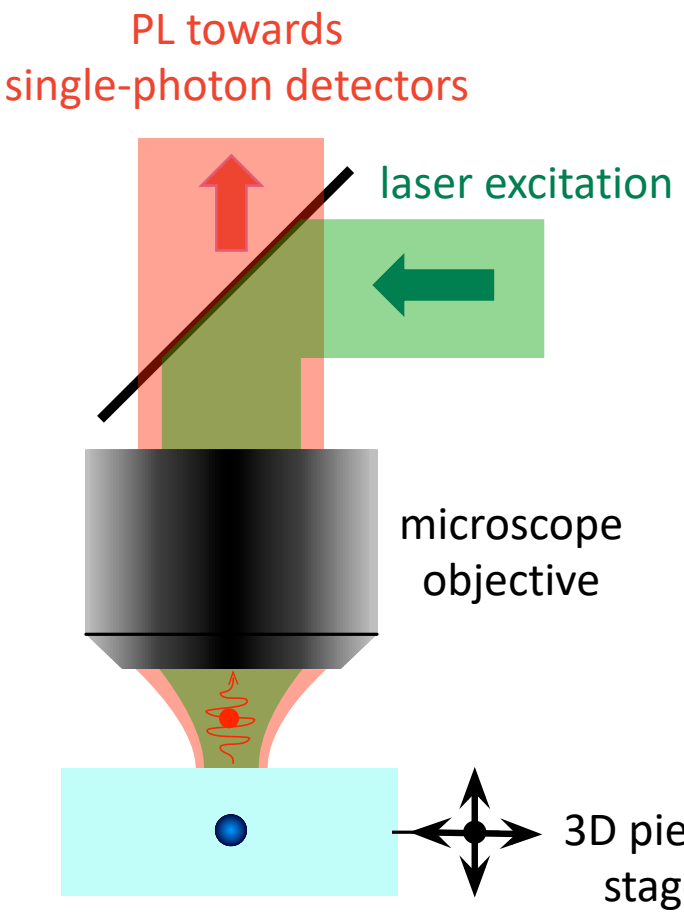
  Chen et al., *Nat. Phot.* ***11*** (2017)

- ground-state electron spin triplet S=1 controllable optically

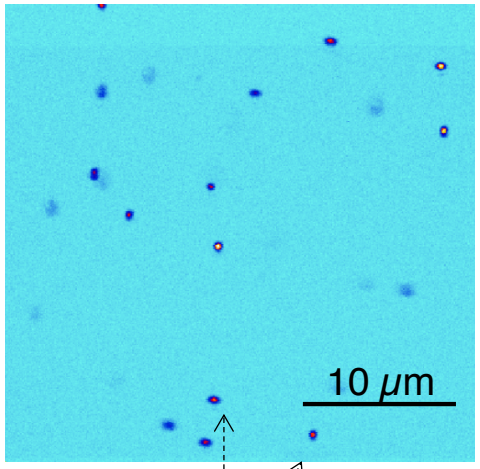
spin-qubit with **long coherence time** even at **room-temperature** **!** (~1ms)

Detection of single NV centers

- standard method:
  PL scanning confocal microscopy **@300K**

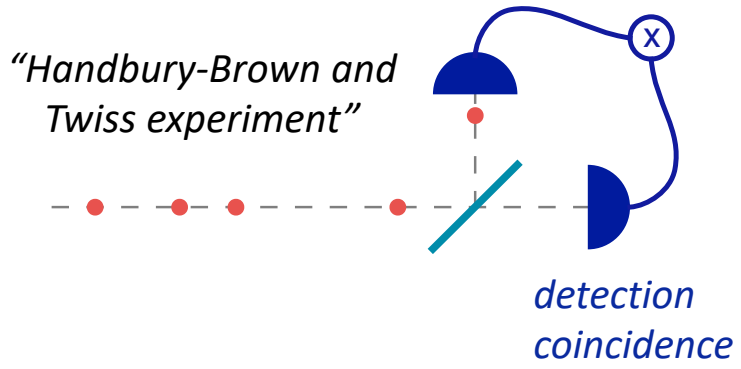


optical scan of an ultrapure bulk diamond

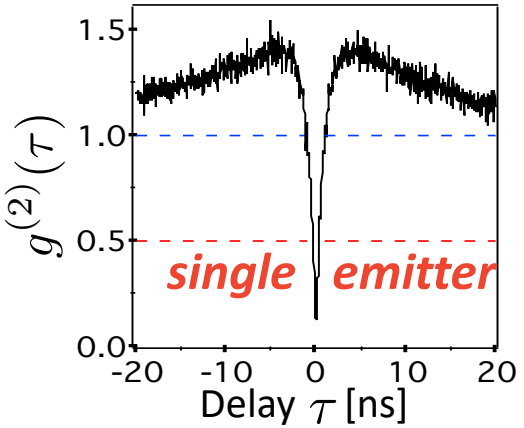


'native' single
NV defects

photon statistics analysis
in "HBT" experiment



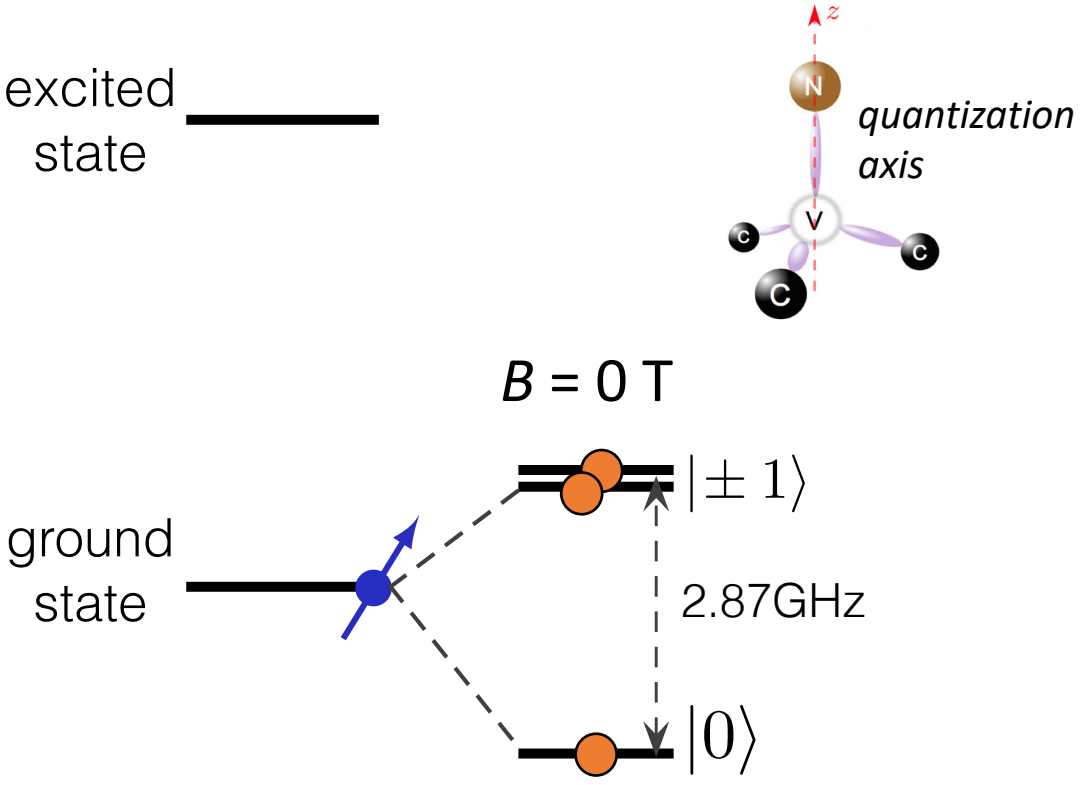
autocorrelation function



antibunching

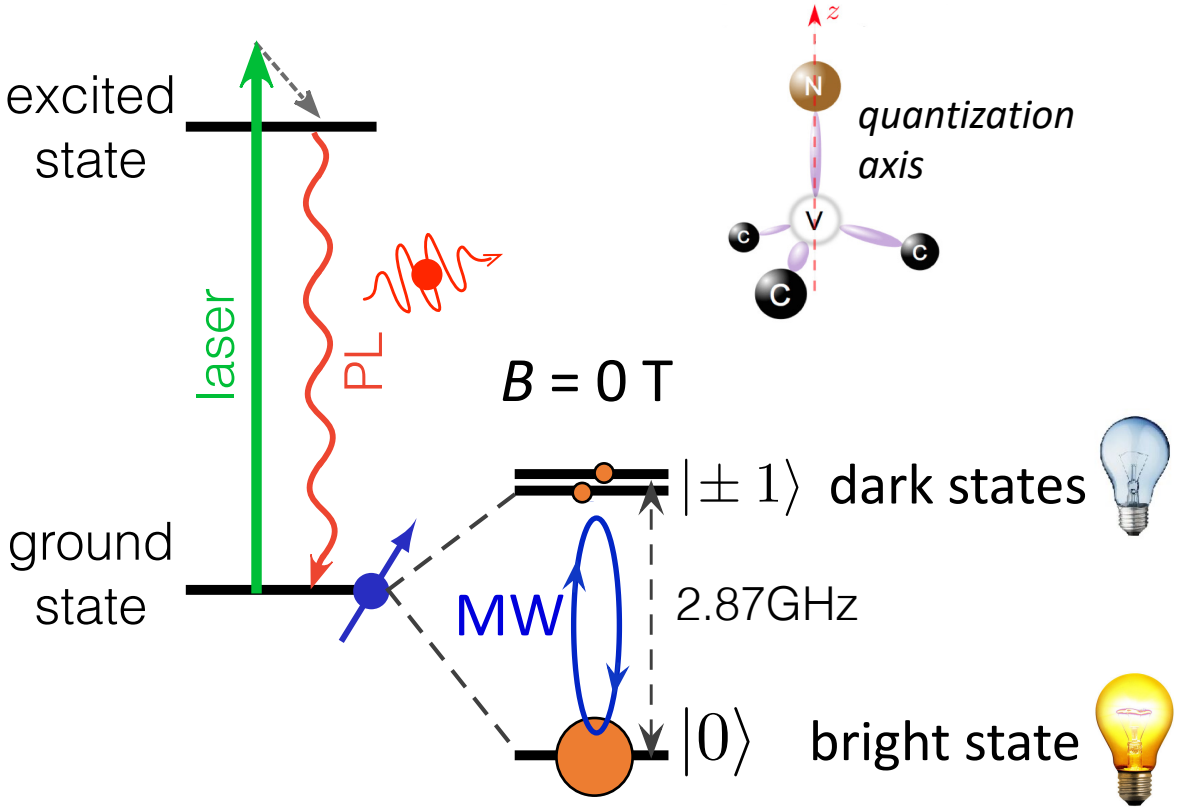
$g^{(2)}(0)<0.5$

Detecting the spin of a single NV center

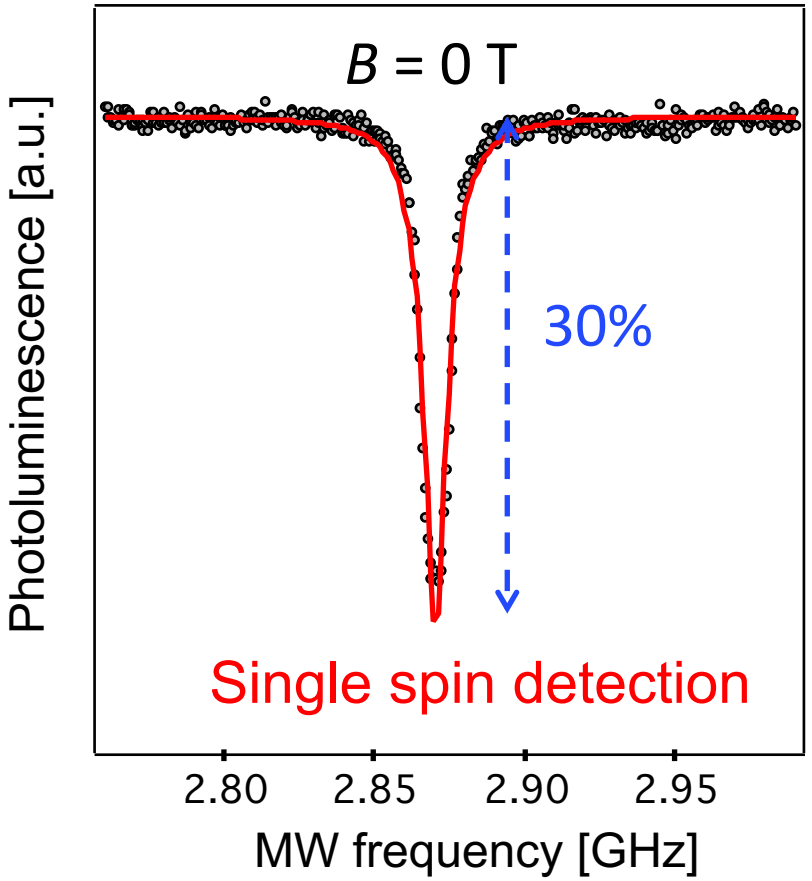


- spin state initialization?
- spin state readout?

Detecting the spin of a single NV center

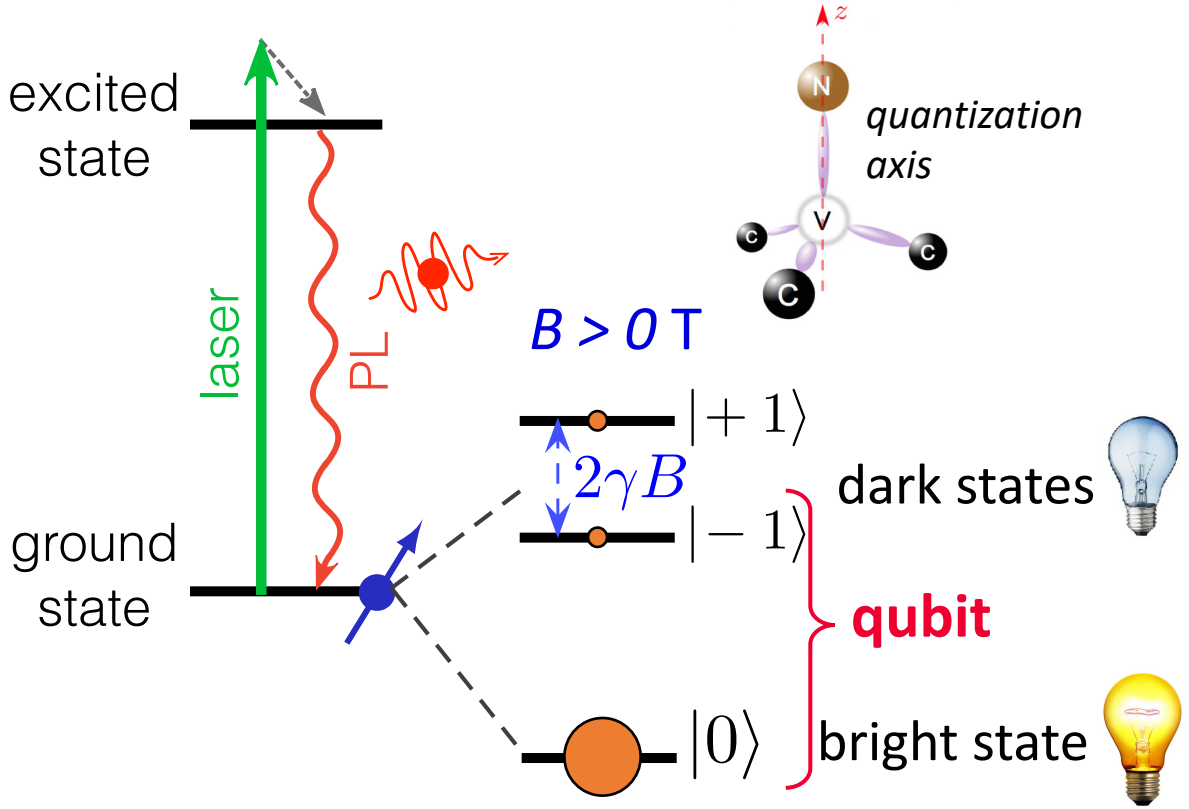


Optically detected magnetic resonance
***ODMR** spectrum*



- spin state initialization? ⇨ **optical pumping !**
- spin state readout? ⇨ **spin-dependent PL level !**

Detecting the spin of a single NV center



Optically detected magnetic resonance
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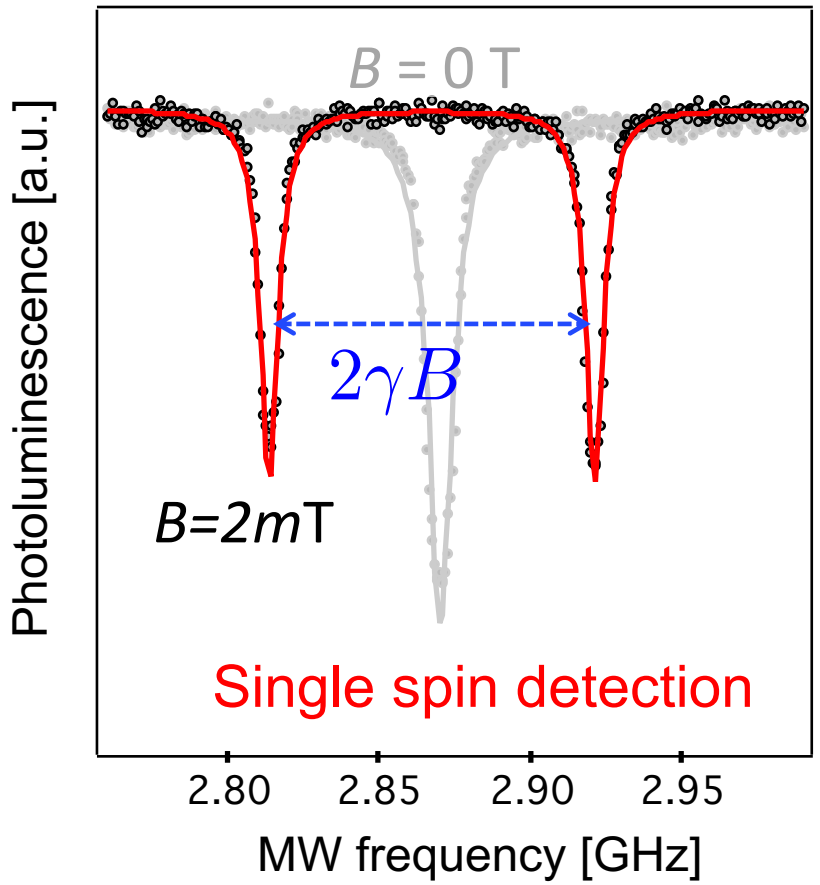
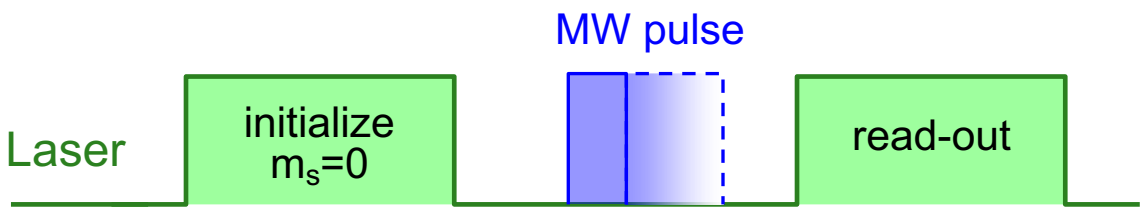
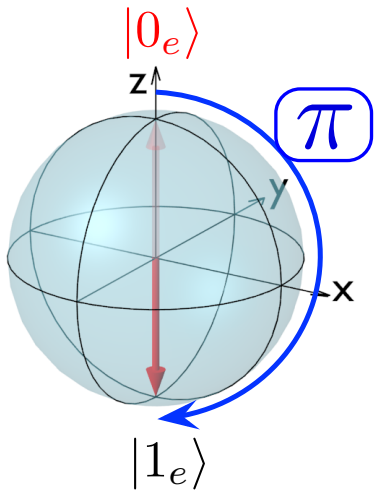


- spin state initialization? ⇨ **optical pumping !**
- spin state readout? ⇨ **spin-dependent PL level !**

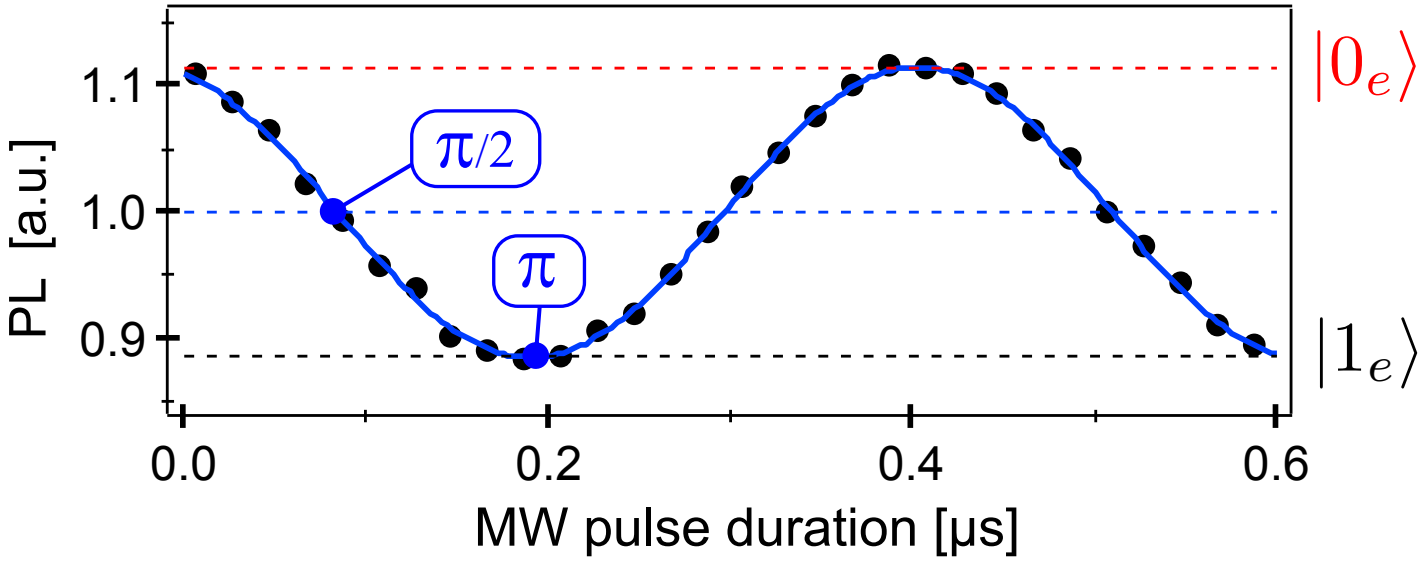
Coherent spin manipulation

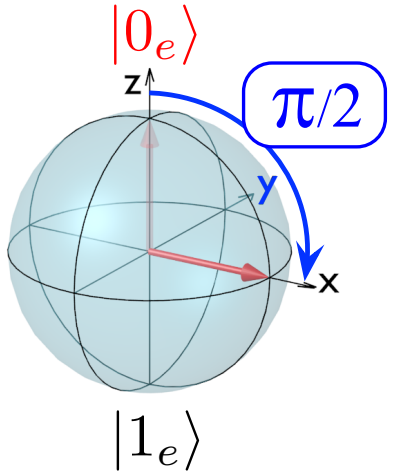




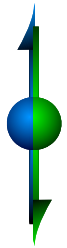
full spin-flip

➢ Rabi oscillations of a single NV spin





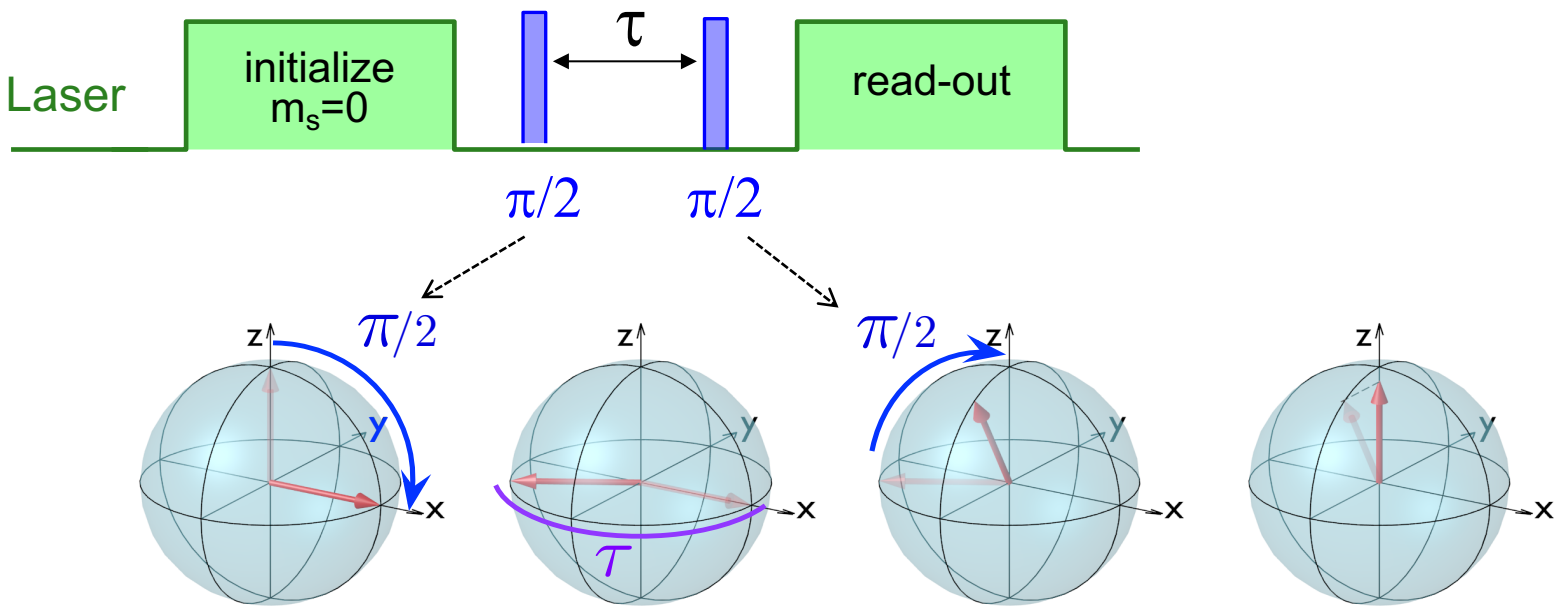
$$\frac{|0\rangle + e^{i\varphi}|1\rangle}{\sqrt{2}}$$



quantum superposition

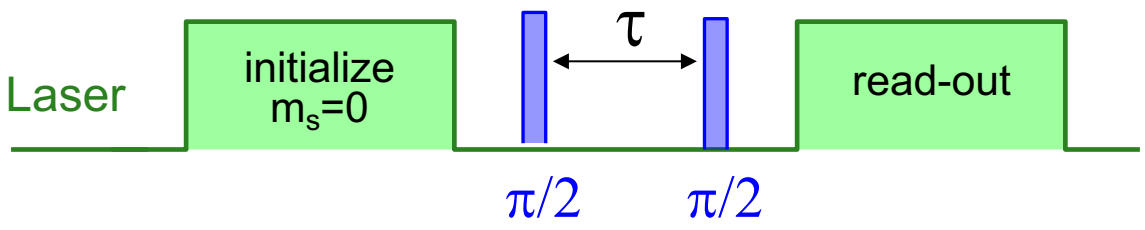
Coherence time of single NVs

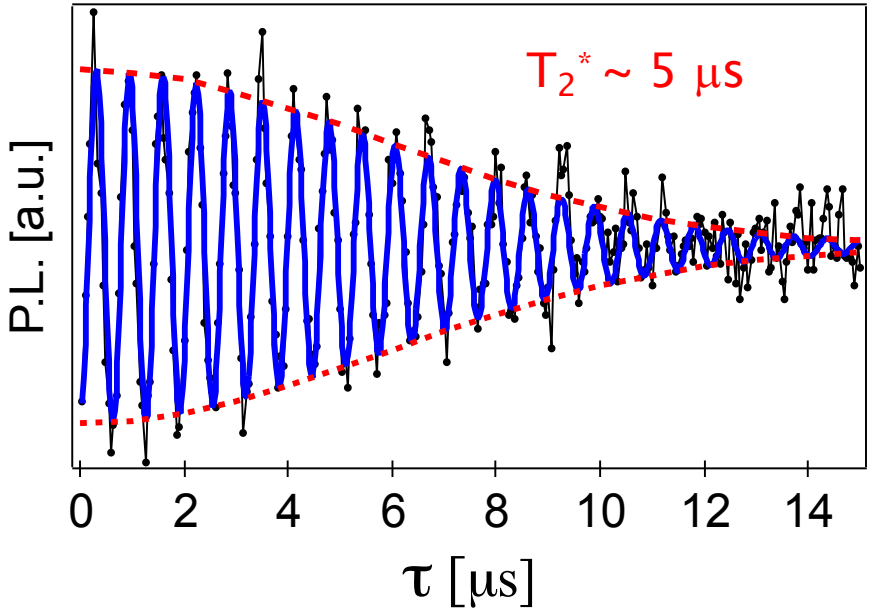
- Ramsey fringes or free-induction decay



Coherence time of single NVs

- Ramsey fringes or free-induction decay





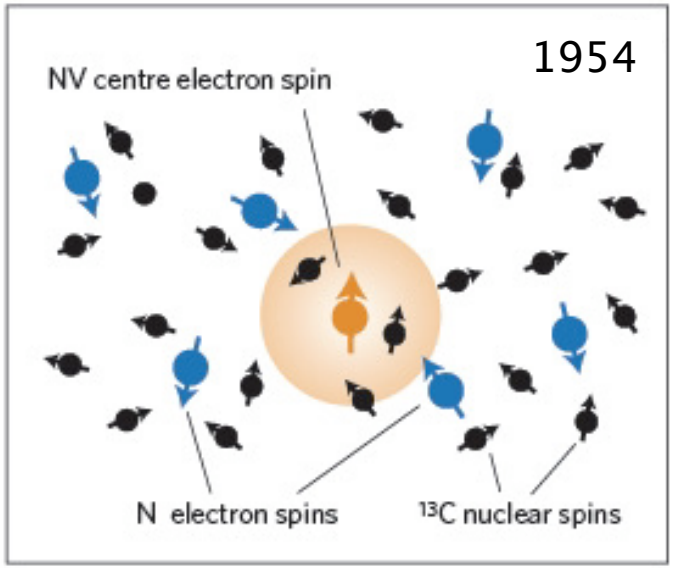
single NV in bulk diamond with natural abundance of C

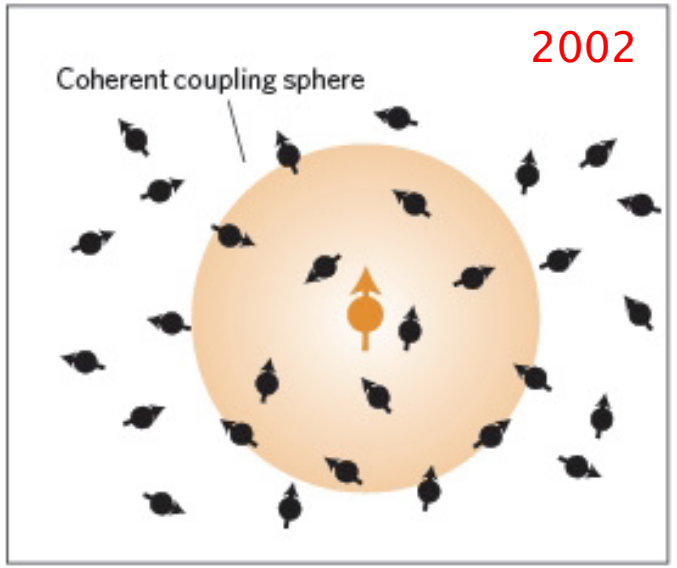
increasing the coherence time

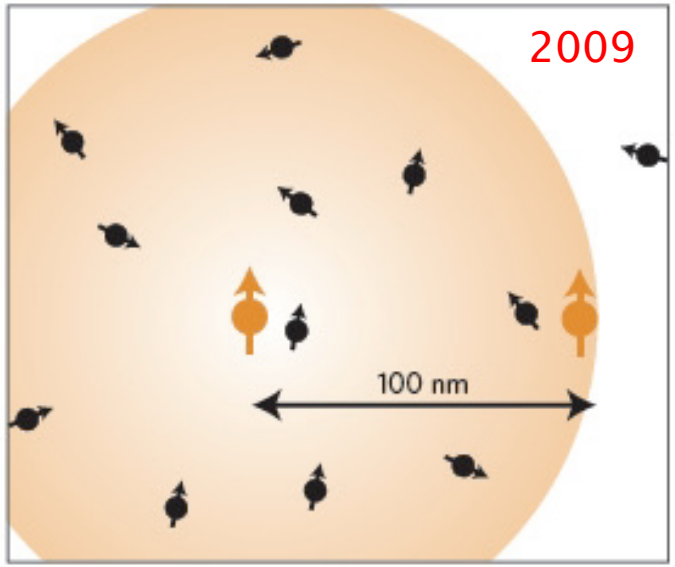
- engineering the diamond lattice
- dynamical decoupling
- clock transitions
- continuous dressing driving
- ...

Engineering the diamond lattice

Hanson, *Nat. Materials* **8** (2009)







High Pressure High Temperature
(HPHT diamond)

$$[\mathrm{N}] \simeq 100\,\mathrm{ppm}$$
$$[^{13}\mathrm{C}] \simeq 1,1\%$$

$T_2^* \sim 100$ ns

CVD diamond

$$[\mathrm{N}] \simeq 1\,\mathrm{ppb}$$
$$[^{13}\mathrm{C}] \simeq 1,1\%$$

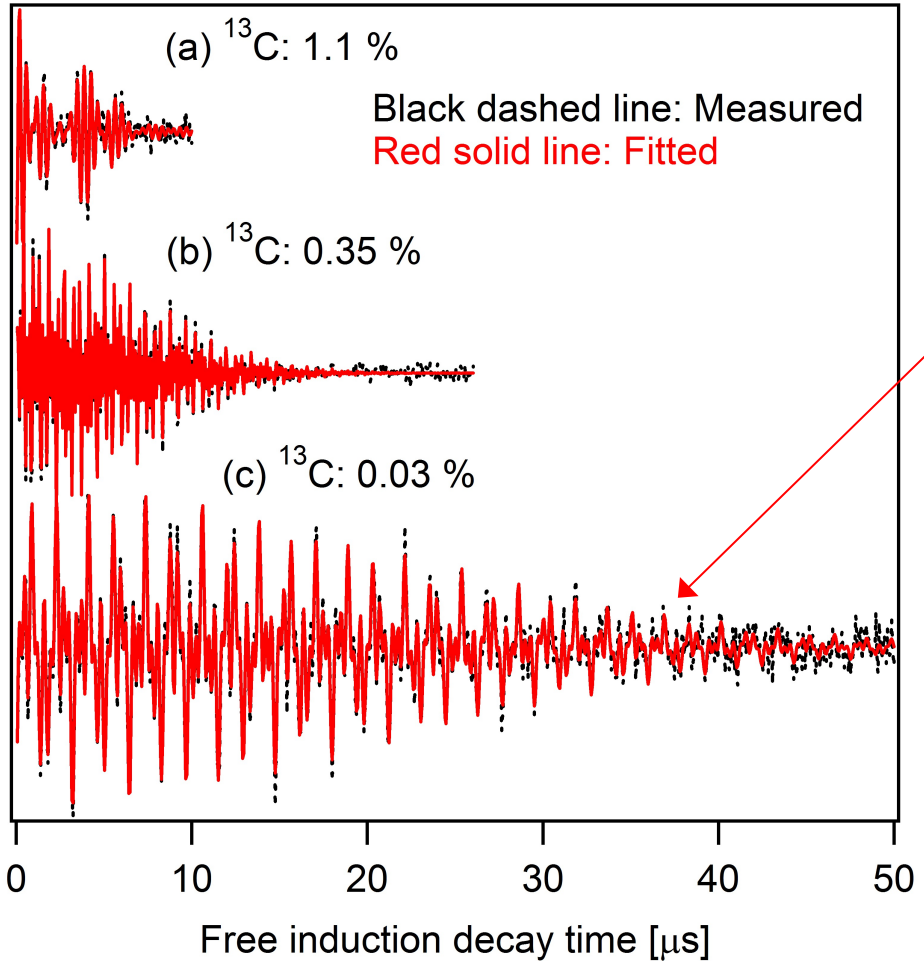
$T_2^* \sim 5$ µs

Isotopically purified
CVD-diamond

$$[\mathrm{N}] \simeq 1\,\mathrm{ppb}$$
$$[^{13}\mathrm{C}] \simeq 0,01\%$$

vacuum-like lattice

Coherence time in isotopically-purified diamonds



99,97 % ^{12}C diamond
Spin free matrix

$T_2^* \sim 50\ \mu s$

J. Wrachtrup's group, Stuttgart, Germany

- Balasubramanian *et al.*, *Nat. Mater.* **8**(2009)
- Mizuochi *et al.*, *PRB* **80** (2009)

99,999 % ^{12}C diamond

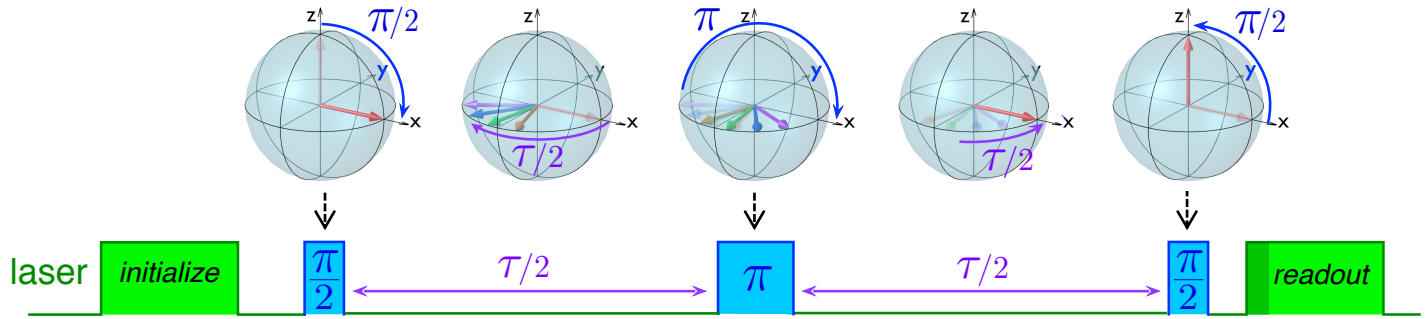
$$T_2^* \approx 500\ \mu s$$

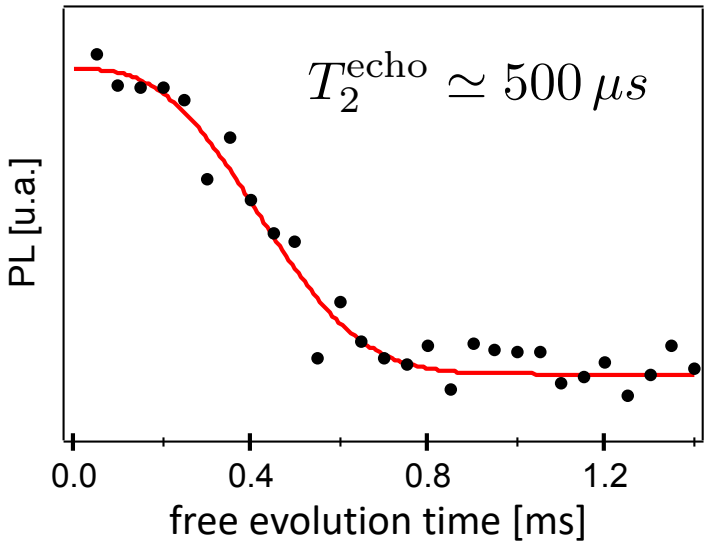
@ room temperature

Maurer *et al.*, *Science* **336** (2012)

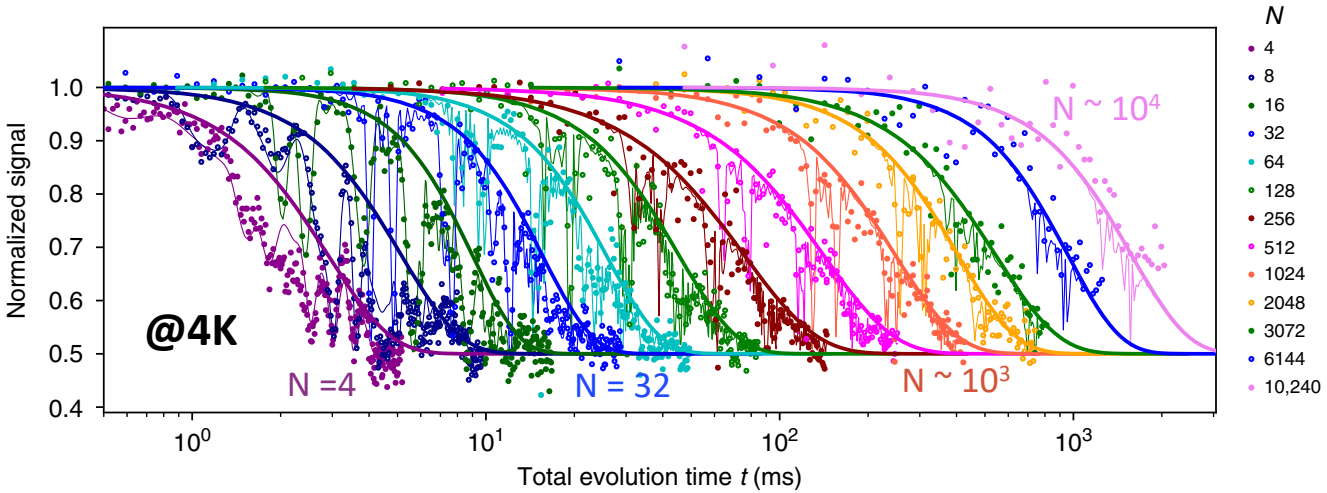
Extending T_2 by dynamical decoupling

➢ Hahn-echo or spin-echo sequence





➢ more refocusing π-pulses...



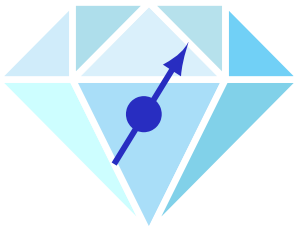
➢ T_2 reaching the T_1-limit at room-temperature

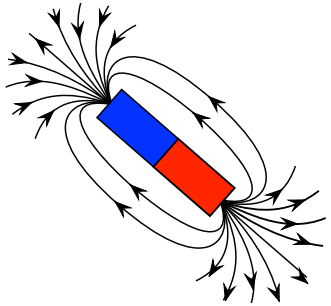
Naydenov *et al.*, *PRB* **83** (2011) **T_2 ~ 2 ms** (300K)

➢ longest T_2 => low-temperature

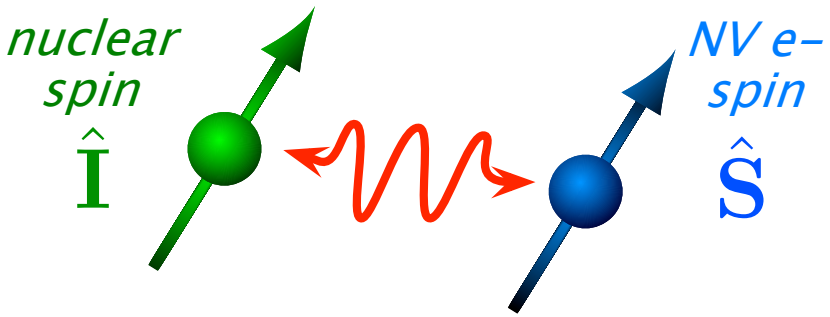
Abobeih *et al.*, *Nat. Comm.* **9** (2018) **T_2 ~ 1 s** (4K)

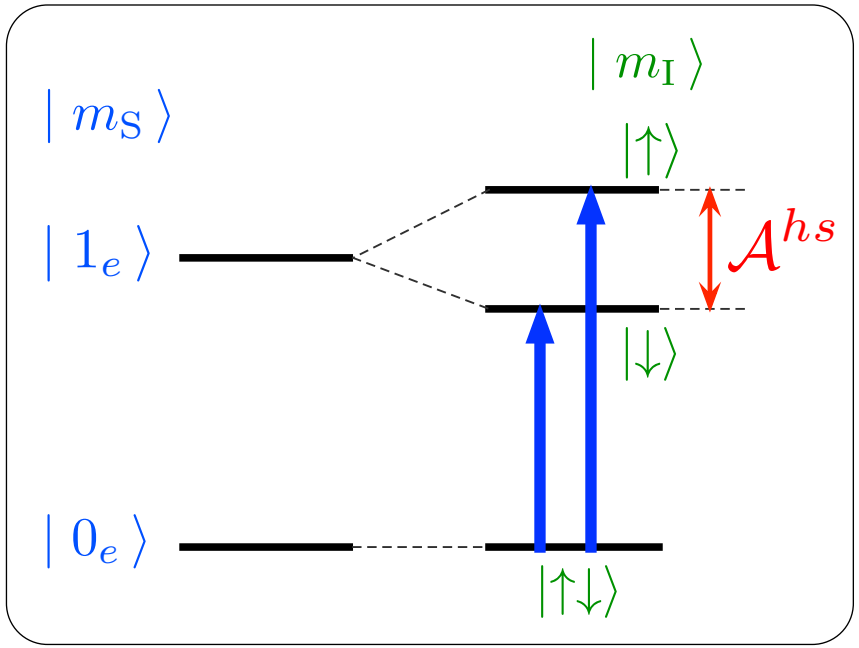
2. Nanoscale magnetometry with NV centers in diamond



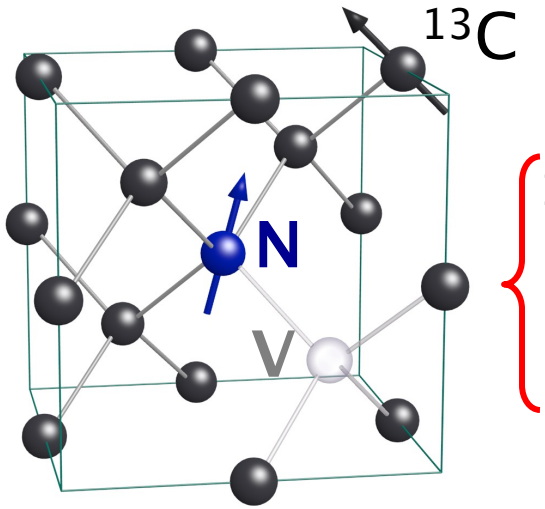


Detecting nuclear spins inside diamond





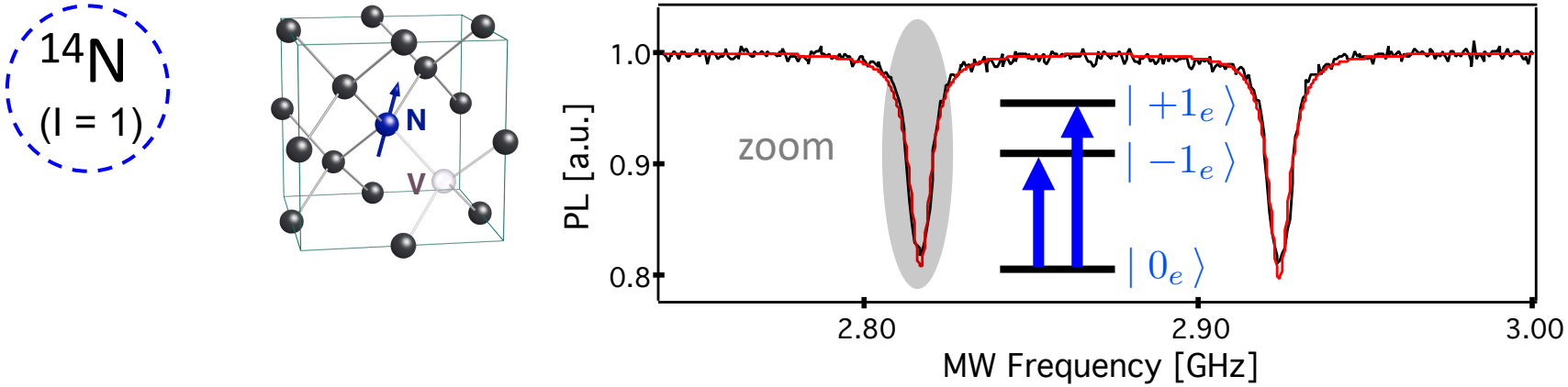
hyperfine interaction $\longrightarrow \mathcal{H}_{hi} \simeq \mathcal{A}^{hs}\, \hat{S} \cdot \hat{I}$



^{14}N $\longrightarrow$ Intrinsic nitrogen atom of the defect *(I=1)*

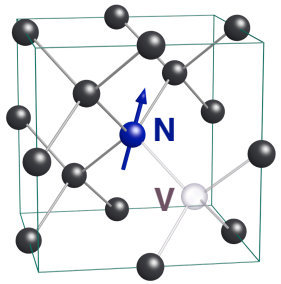
^{13}C $\longrightarrow$ Randomly placed in the lattice *(I=1/2)*

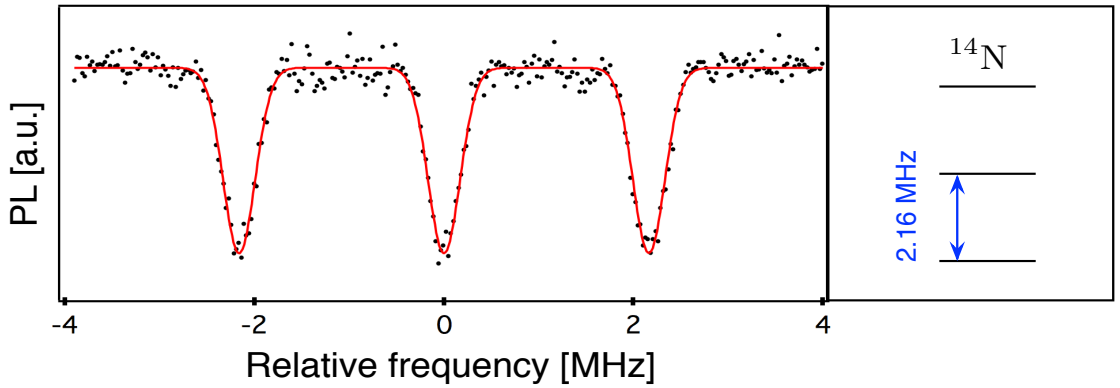
Typical hyperfine spectra



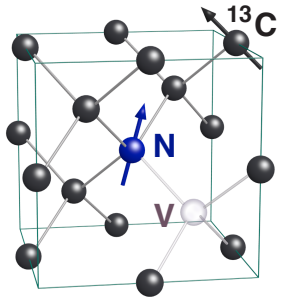
Typical hyperfine spectra

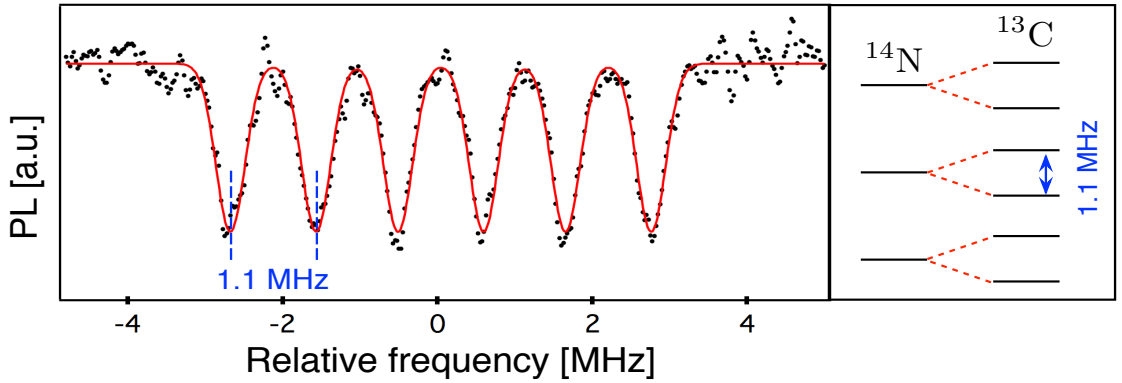
^{14}N (I = 1)



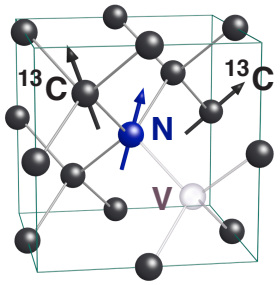


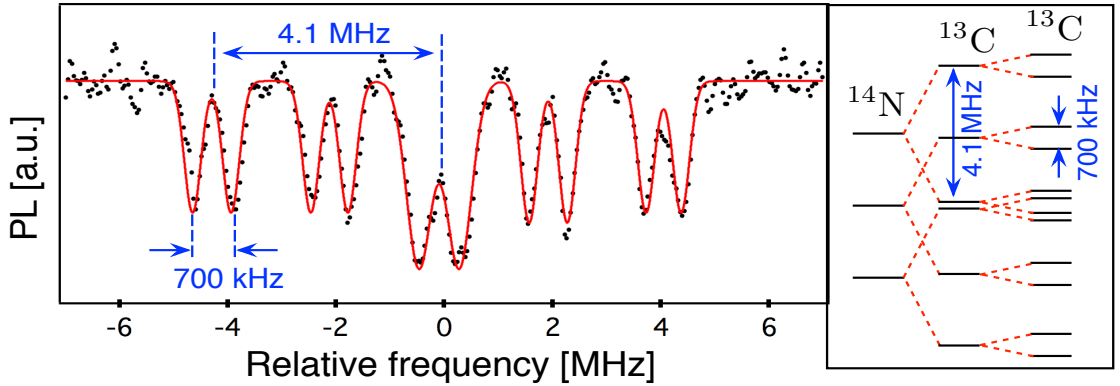
^{14}N + ^{13}C (I = 1/2)





^{14}N + ^{13}C + ^{13}C





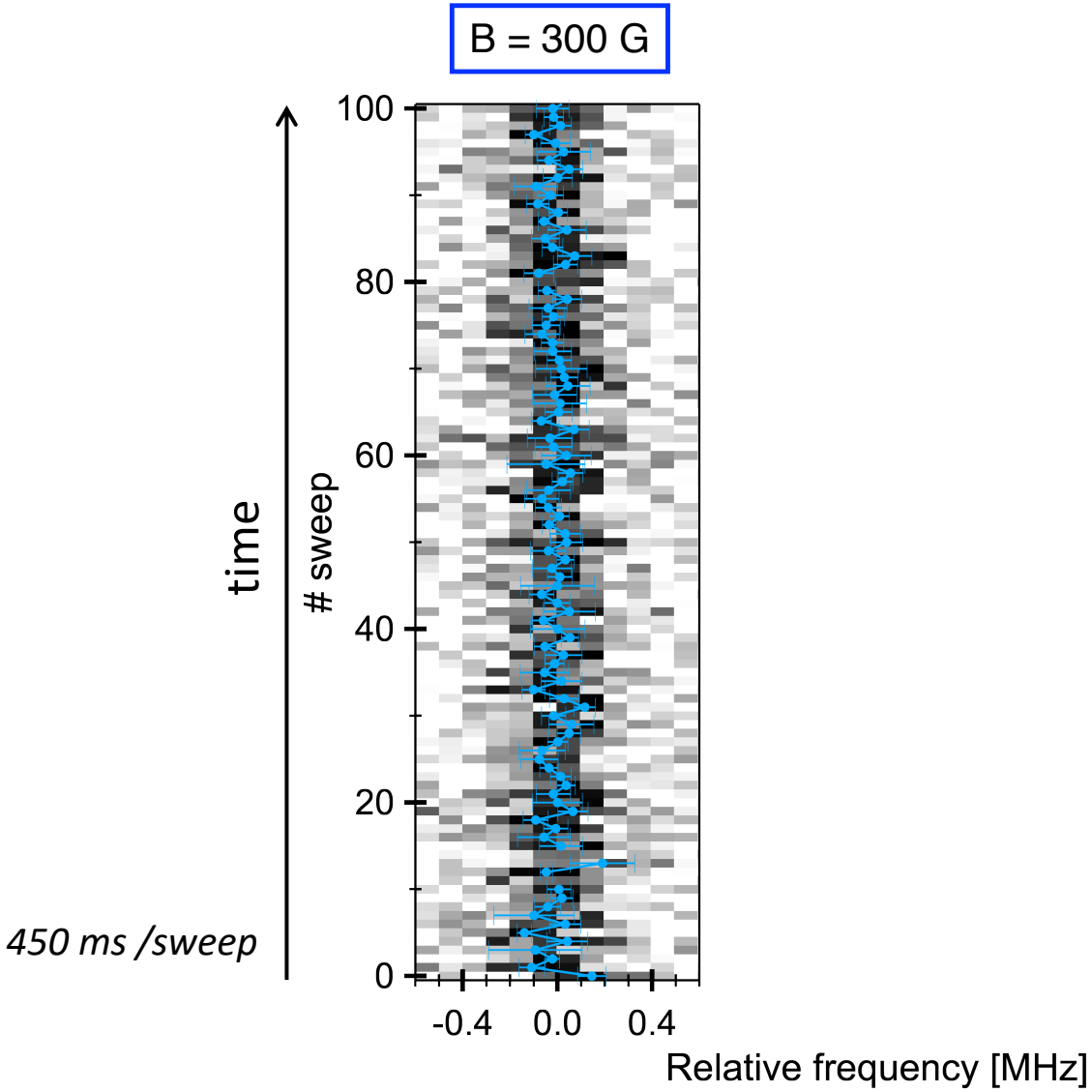
Dréau *et al.*, *PRB* **85** (2012)

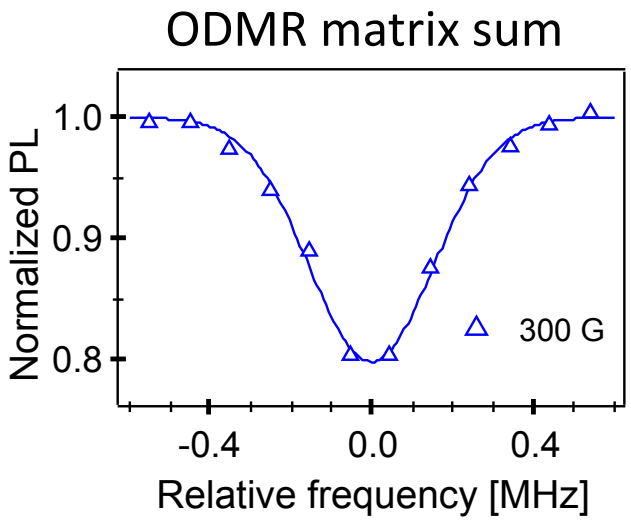
Related work:
Smeltzer *et al.*, *NJP* **13** (2011)

➢ ^{13}C lattice sites calculated by DFT:

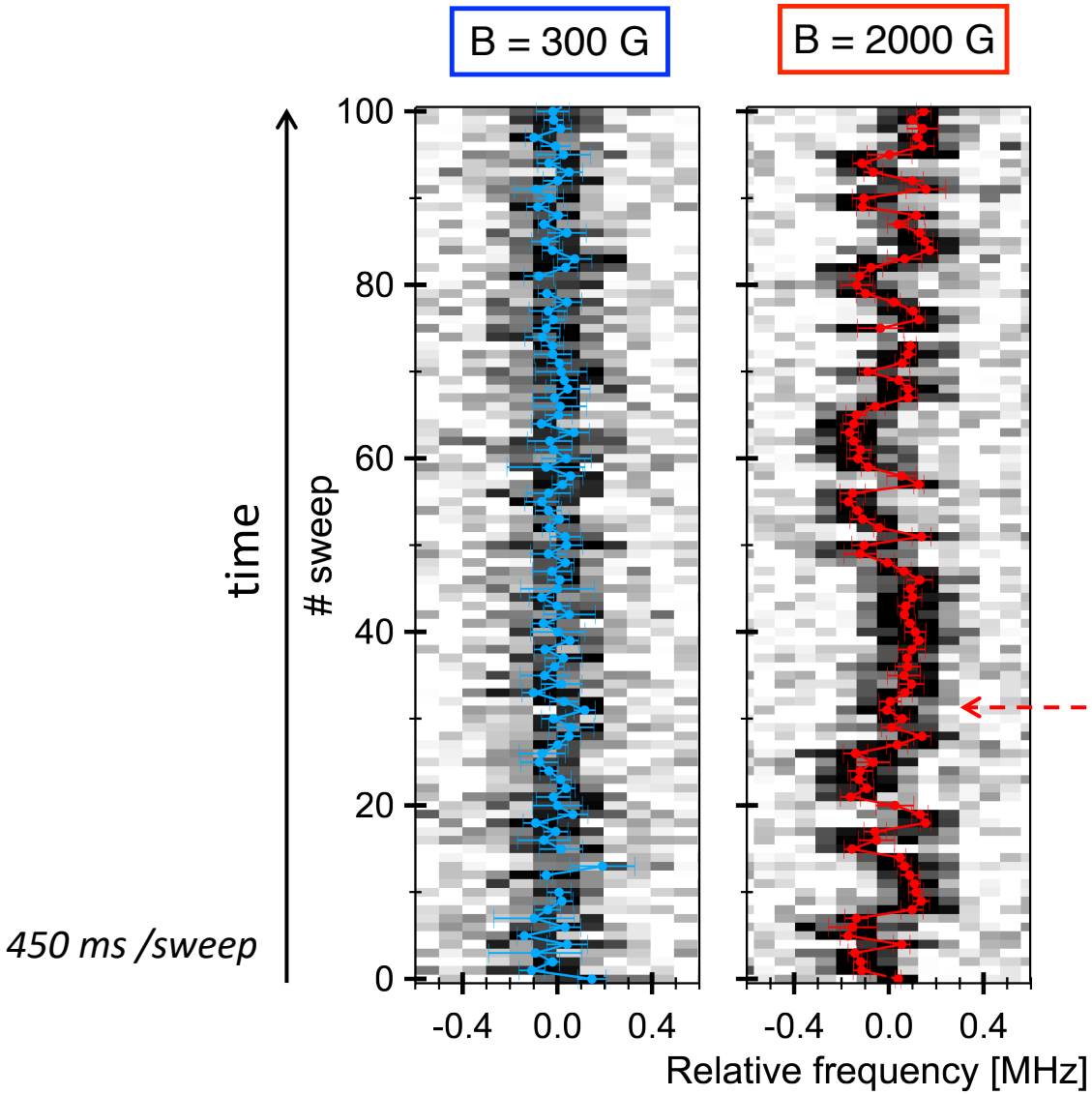
Nizovtsev *et al.*, *NJP* **16** (2014)

Real-time measurements of the hyperfine field

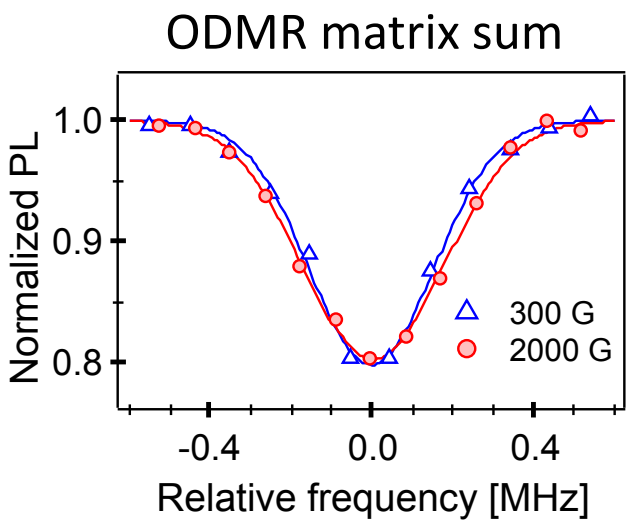




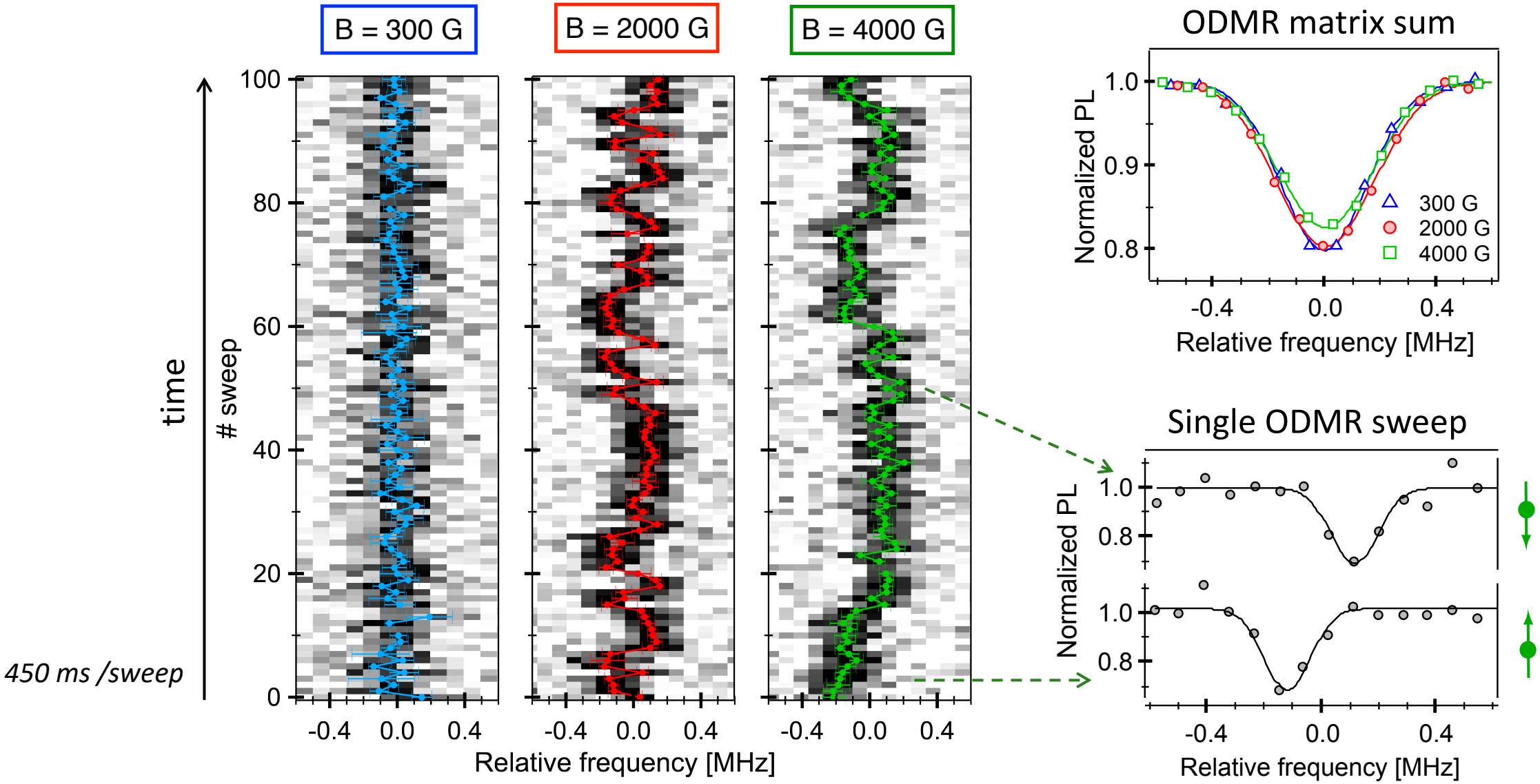
Real-time measurements of the hyperfine field



quantum jumps of a single ^{13}C nuclear spin

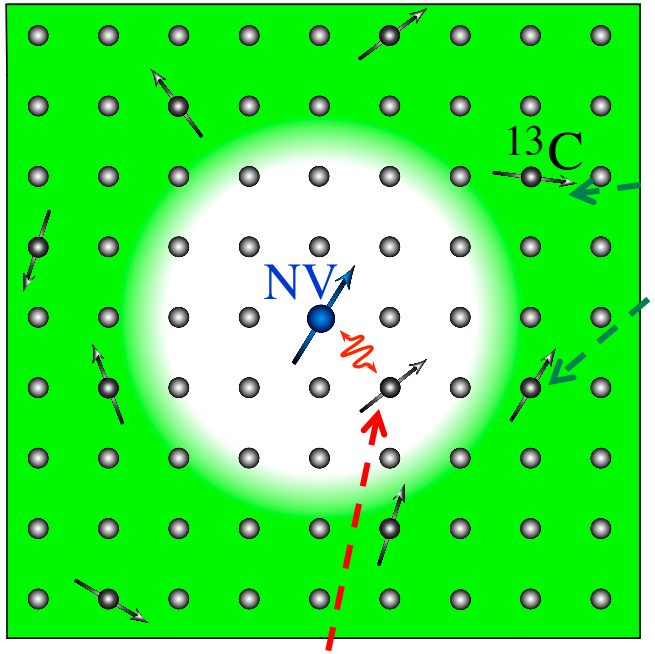


Real-time measurements of the hyperfine field



Dréau *et al.*, *PRL* **13** (2014)

Detecting weakly-coupled nuclear spins



only strongly-coupled ^{13}C nuclear spins resolvable in ODMR spectrum

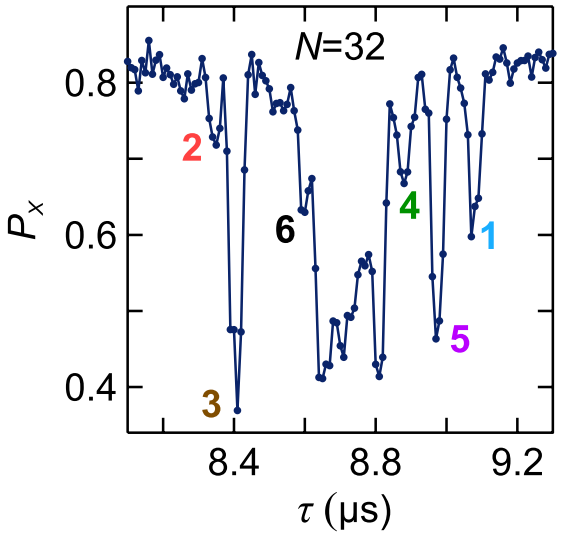
spectral resolution: **~ 200 kHz** (T_2*-limited)

weakly-coupled ^{13}C nuclear spins of the bath

coupling detectable in dynamical decoupling sequence

spectral resolution: **< 100 Hz** (T_2-limited)

Taminiau *et al.*, *PRL* **109** (2012)



Kolkowitz *et al.*, *PRL* **109** (2012)

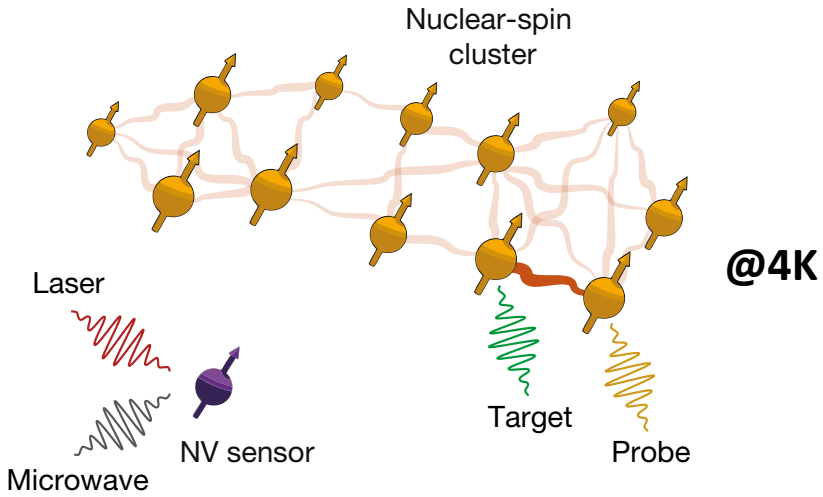
Laraoui *et al.*, *Nat. Comm.* **4** (2013)

Bradley *et al.*, PRX **9** (2019)

Imaging a 27-nuclear spin cluster

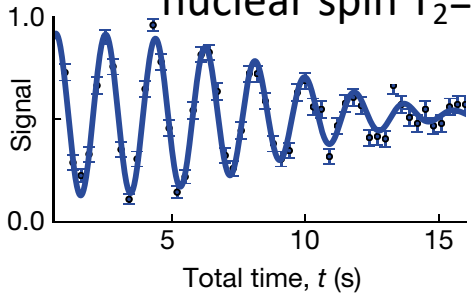
Work from T. Taminiau's group (Delft, The Netherlands)

Abobeih *et al.*, *Nature* **576** (2019)

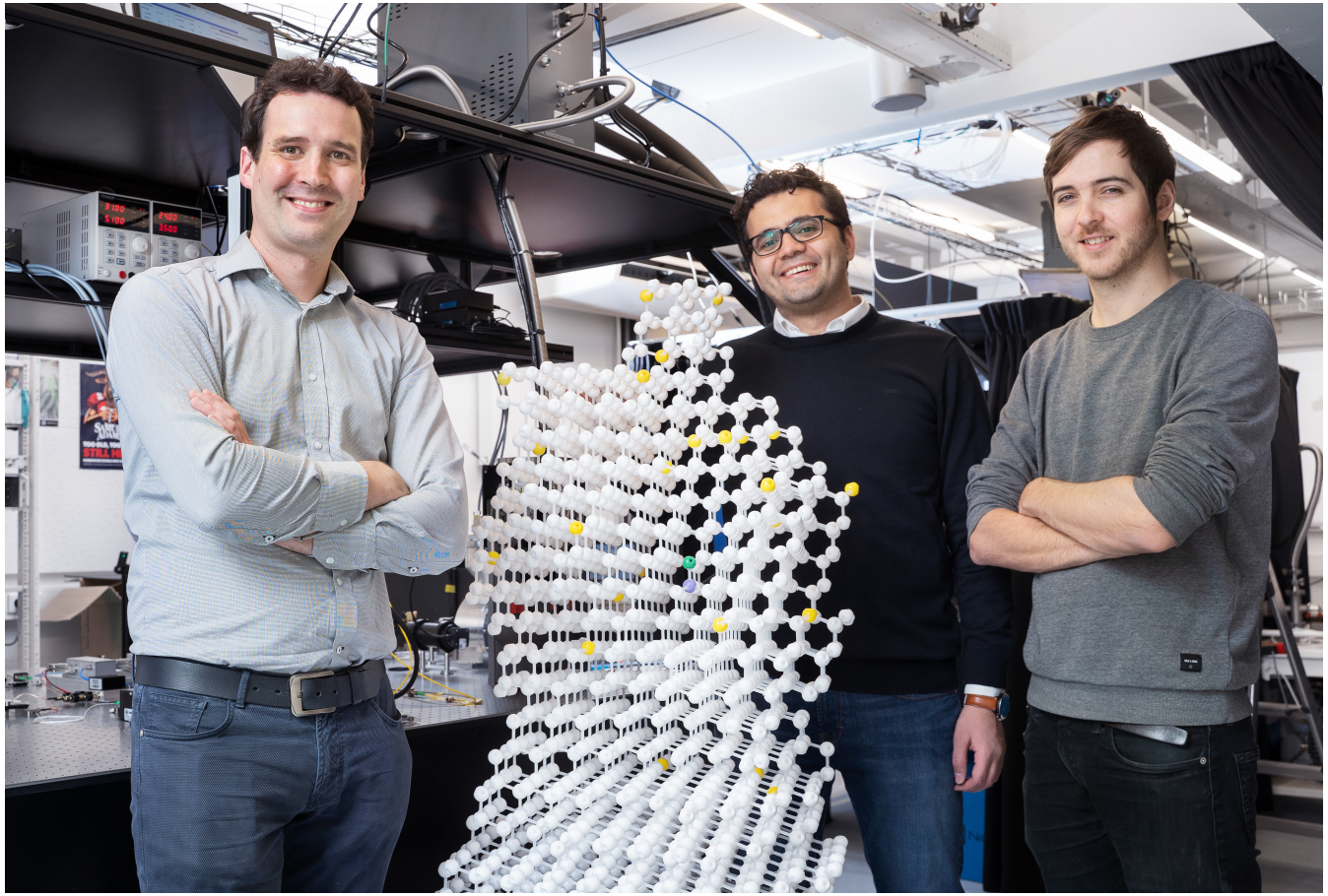


➢ NV probing nuclear-nuclear interactions with spin-echo double resonance techniques

nuclear spin T_2=11s !



spectral resolution < 80mHz !



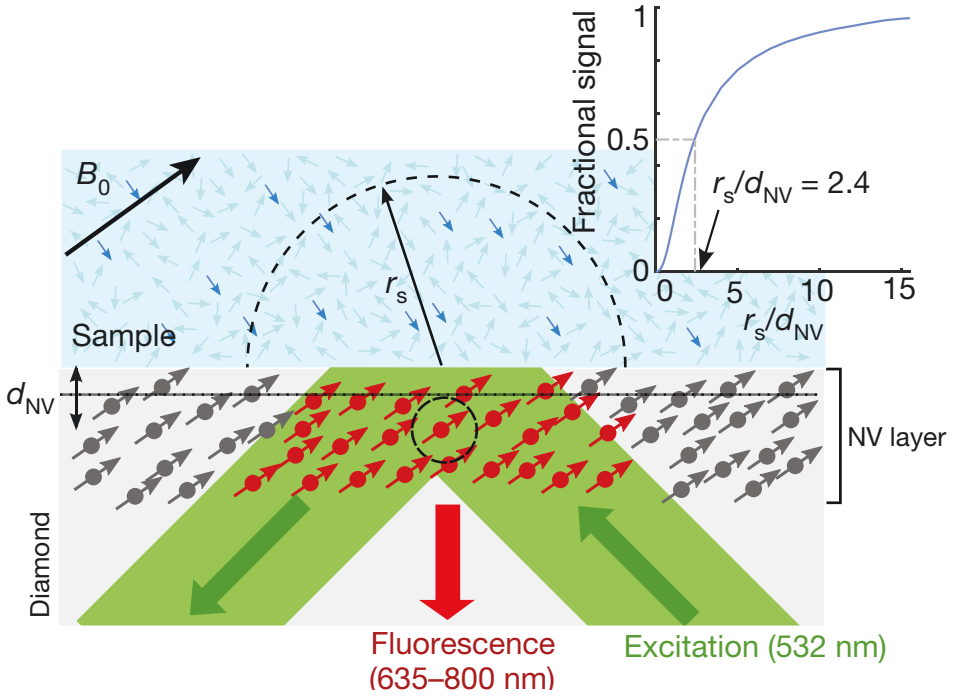
➢ ^{13}C nuclear spins in quantum registers to process Q. information

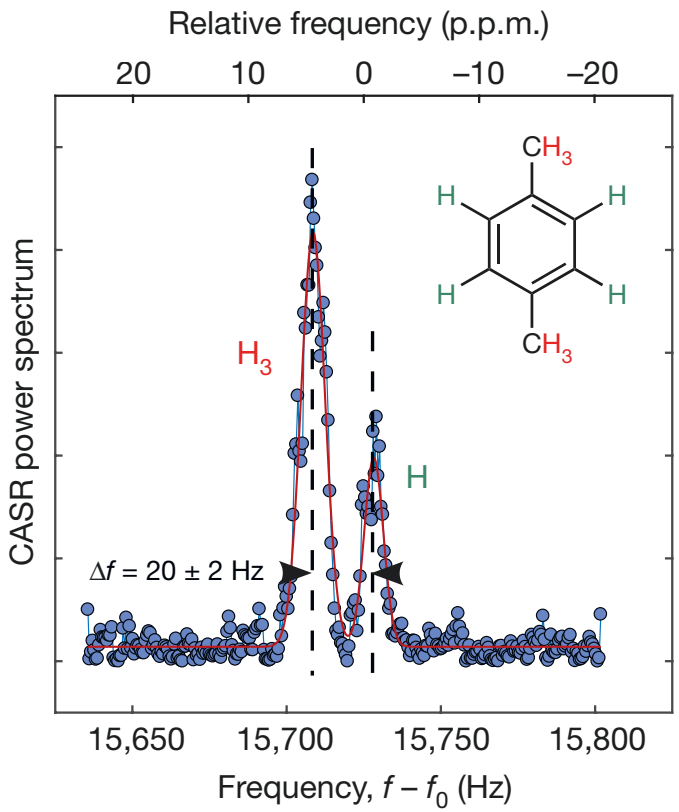
Waldherr *et al.*, *Nature* **506** (2014)
Bradley *et al.*, PRX **9** (2019)

Probing nuclear spins outside diamond

Work from R. Walsworth's group and collab. (Harvard, USA)

Glenn *et al.*, *Nature* **555** (2018)





- high-resolution NMR with a NV-ensemble
  - detection of scalar couplings and chemical shifts in molecules outside diamond

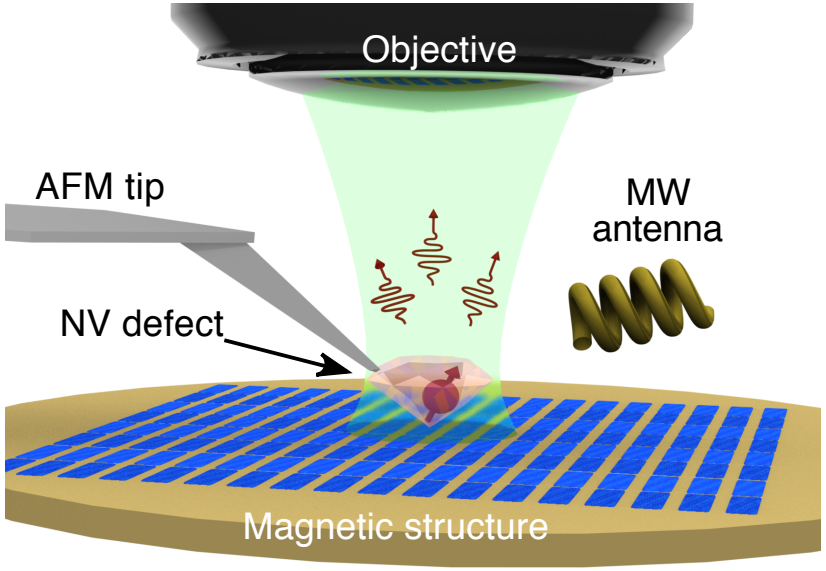
- "CARS" sequence: NV readout synchronized onto the nuclear spin precession

spectral resolution ~ 1Hz !

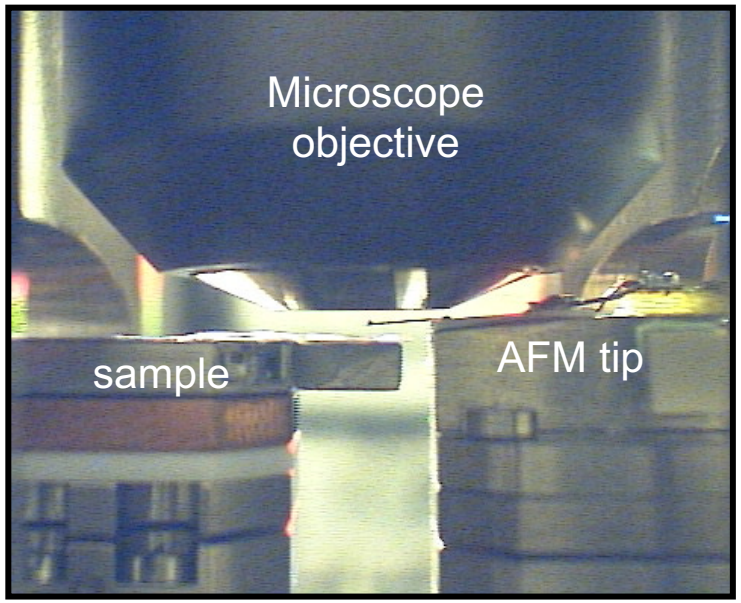
Scanning NV-magnetometry

➢ Principle

Rondin *et al.*, *APL* **100** (2012)



- ★ Quantitative and vectorial (sensitivity ~ **1 µT/Hz$^{-1/2}$**)
- ★ No magnetic back-action
- ★ Operation from 4K to 300K
- ★ Spatial resolution limited by the probe-to-sample distance ***d***

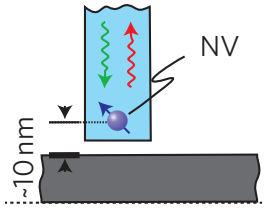


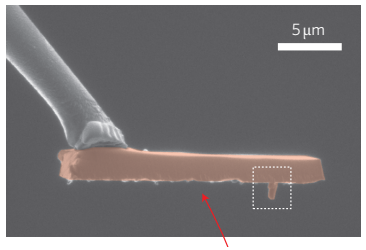
Courtesy of V. Jacques (Montpellier, France)

➢ Early experiments with nanodiamonds...

Balasubramanian et al., *Nature* **455** (2008)
Rondin *et al.*, *APL* **100** (2012), *Nat Com.* **4** (2013)

➢ ... now with all-diamond scanning probe tips





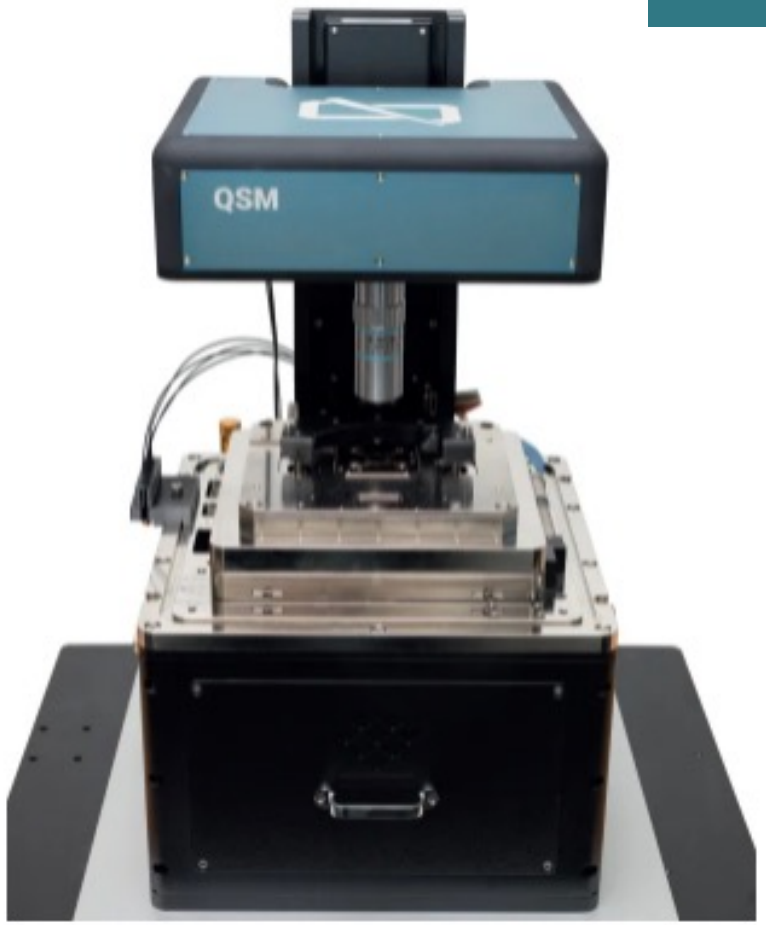
Maletinsky *et al.*, *Nat. Nano.* **7** (2012)

commercially available since 2018

2 scanning NV-magnetometers commercially available!

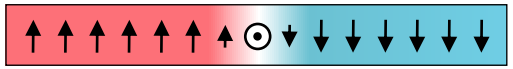
Qnami

ZABRE

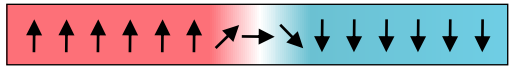


Determining the structure of domain walls - Theory

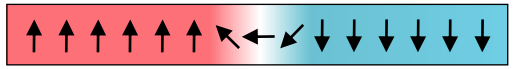
Bloch wall

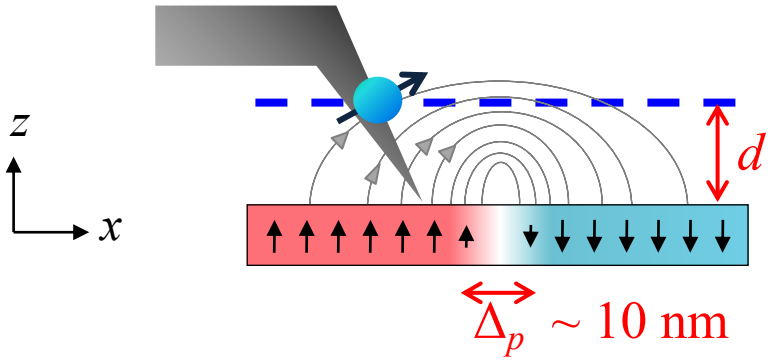


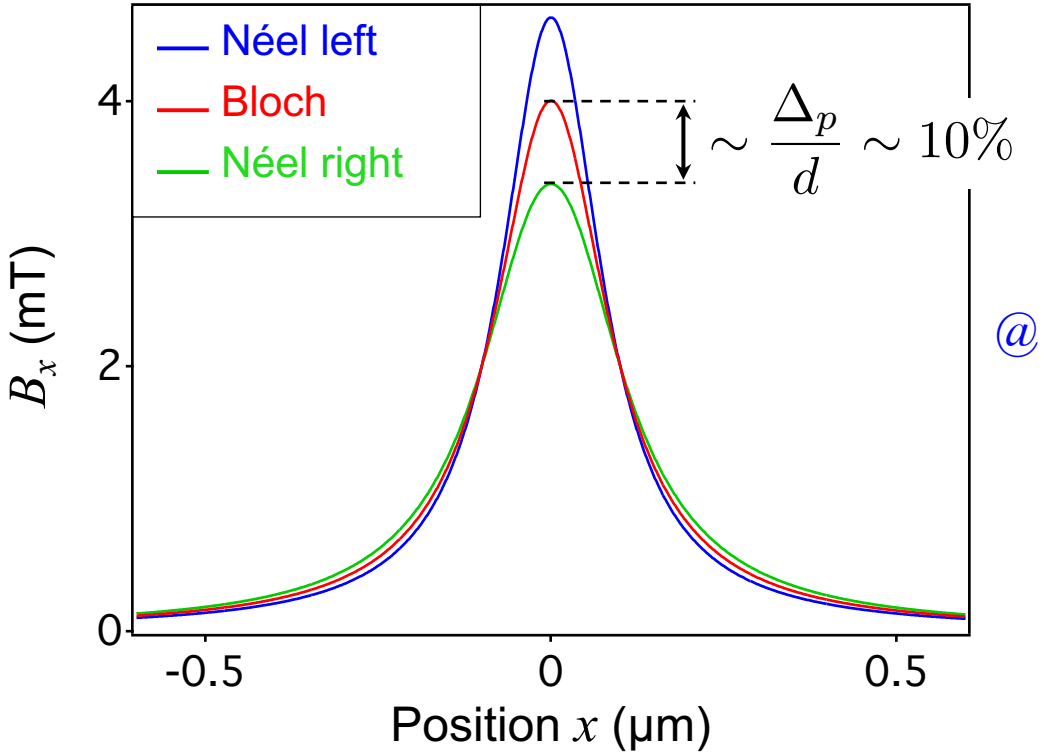
Néel wall (right)



Néel wall (left)







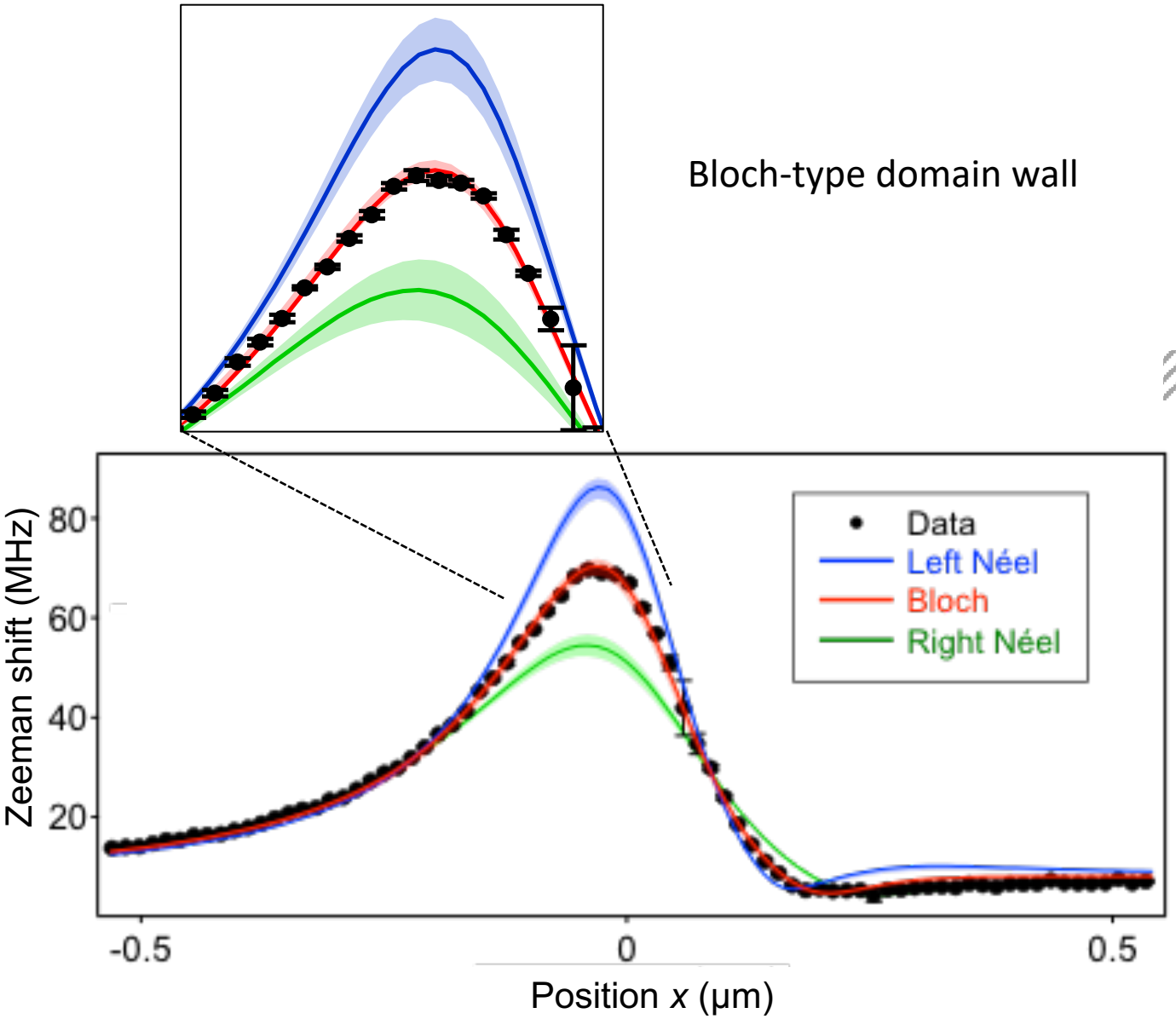
@ d = 100 nm

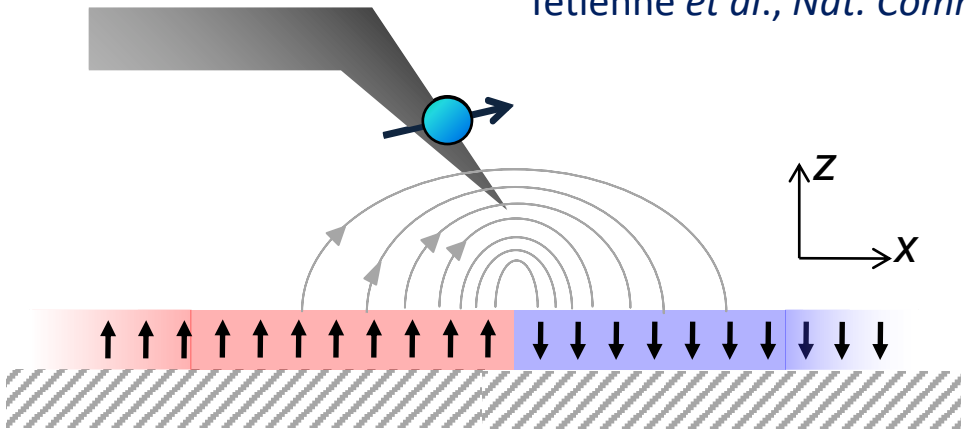
Source: Vincent Jacques (L2C)

Determining the structure of domain walls – Exp.

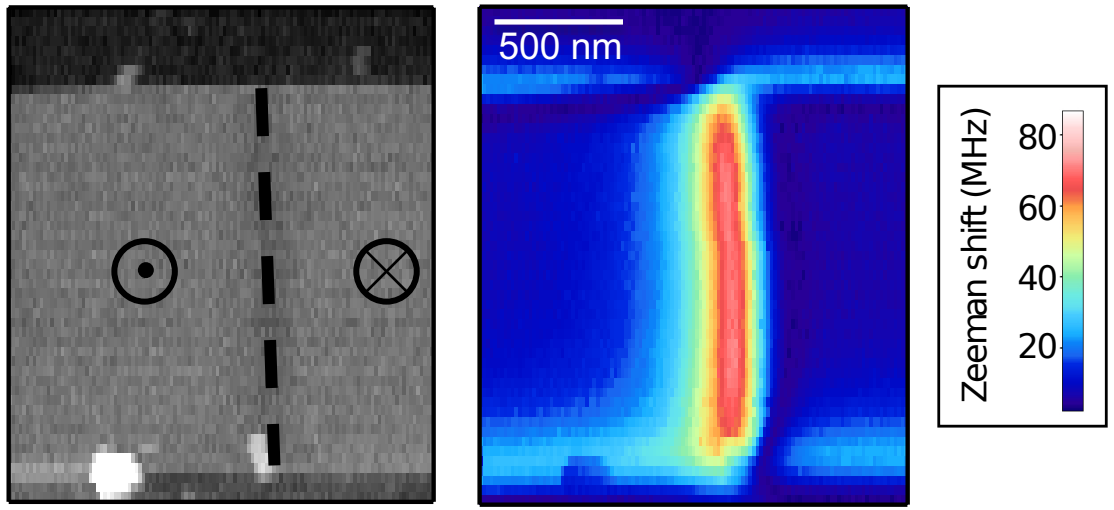
Tetienne *et al.*, *Nat. Comm.* **6** (2015)

Bloch-type domain wall



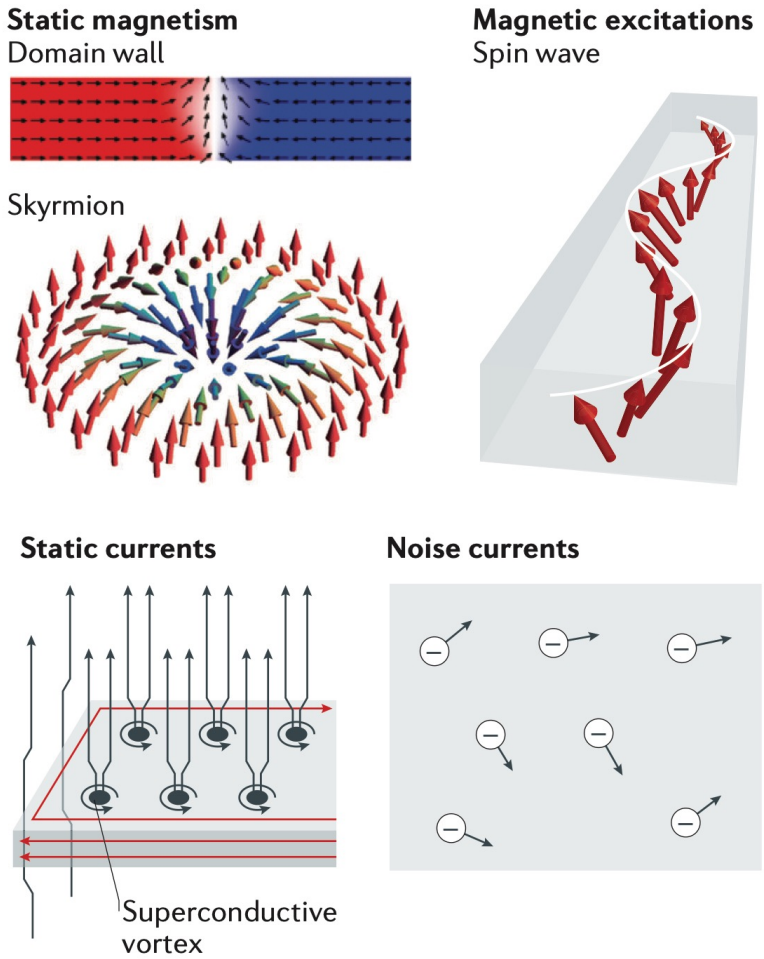


AFM



Applications of NV magnetometry

- Investigating condensed matter physics



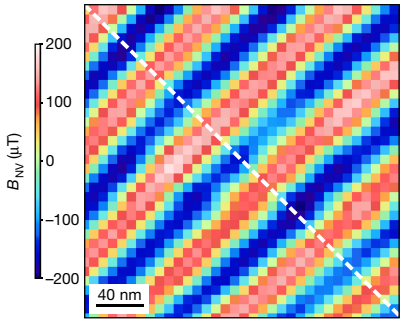
Casola *et al.*, *Nat. Rev. Mat.* **3** (2018)

- Strong developments in recent years

Reviews: Rondin *et al.*, *Rev. Prog. Phys.* **77** (2014)
Casola *et al.*, *Nat. Rev. Mat.* **3** (2018)

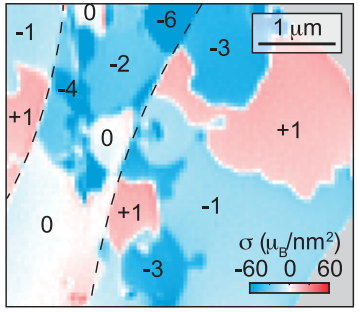
A few examples:

spin cycloid in multiferroic mat.



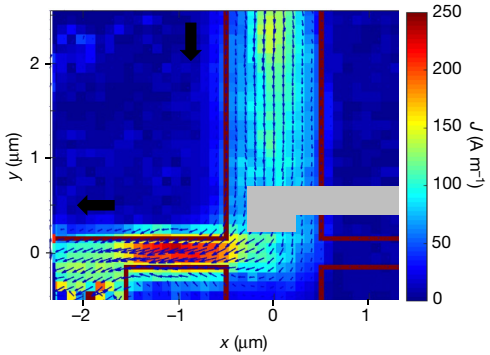
Gross *et al.*, *Nature* **549** (2017)

magnetism in 2D-materials



Thiel *et al.*, *Science* **354** (2019)

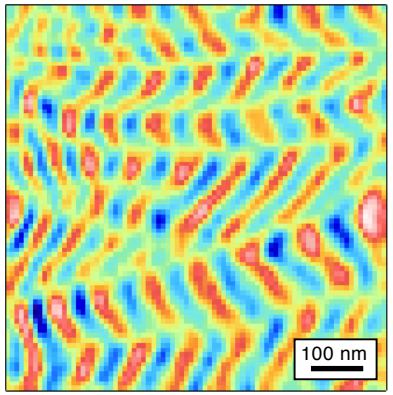
viscous electron flow in graphene



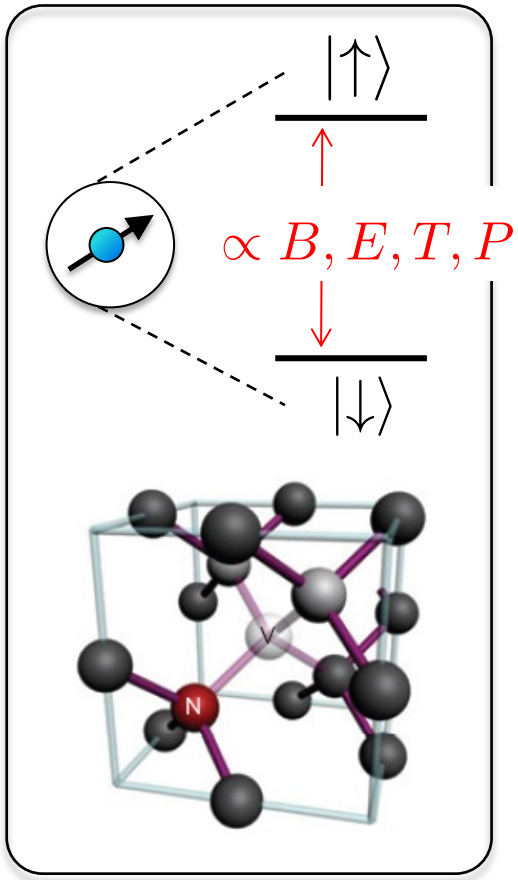
Ku *et al.*, *Nature* **583** (2020)

A multimode quantum probe

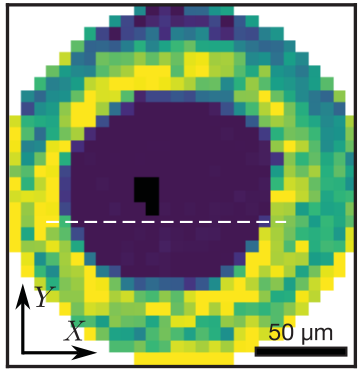
Magnetic field



Gross *et al.*, *Nature* **549** (2017)

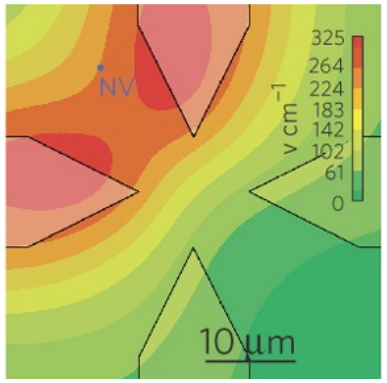


Pressure



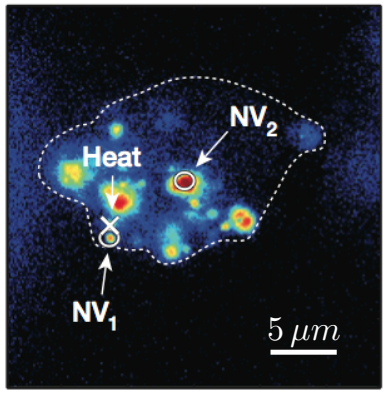
Hseih et al., *Science* ***366*** (2019)

Electric field



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

Temperature



Kucsko et al., *Nature* **500** (2013)

spin defects = highly versatile for quantum sensing

What about other defects and materials?

Many single defects already detected at single scale

Zhang et al., *Appl. Phys. Rev.* **7** (2020)