

# Probing nanoscale magnetism with quantum sensors: from antiferromagnets to 2D materials

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Laboratoire Charles Coulomb  
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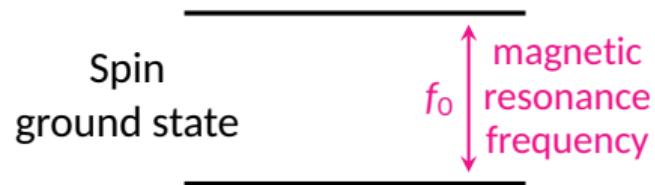
CNRS and Université de Montpellier, Montpellier, France



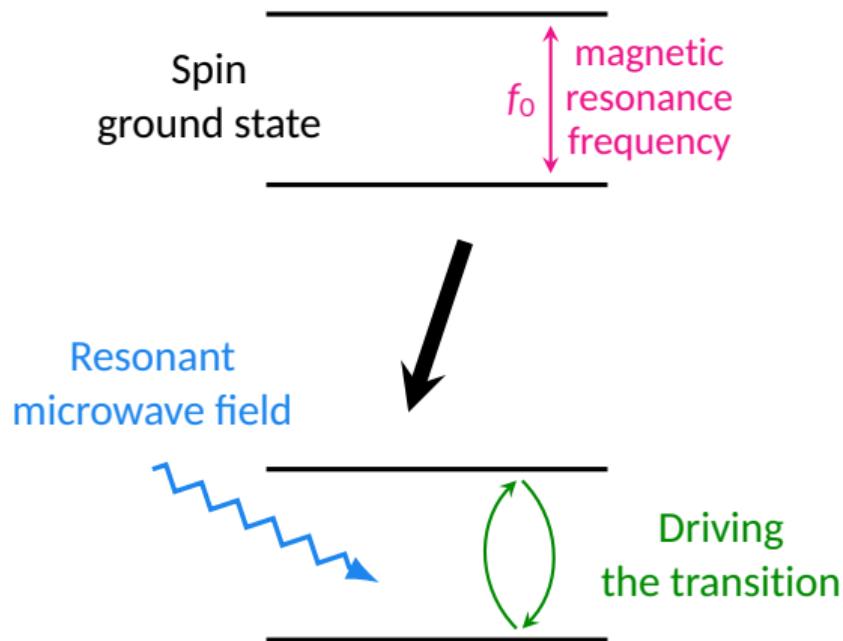
APS March Meeting, T24-005, March 7<sup>th</sup> 2024, Minneapolis

slides available at <https://magimag.eu>

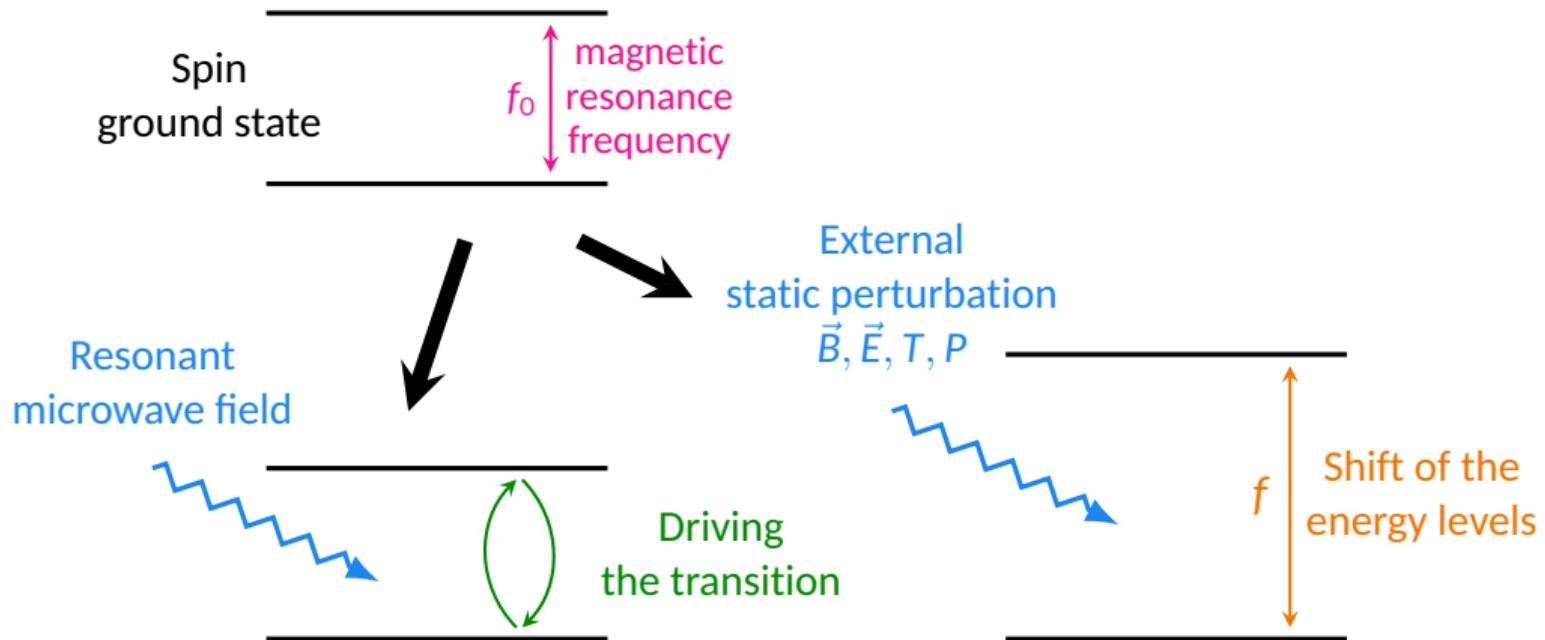
# How can we use a quantum system to probe nanomagnetism?



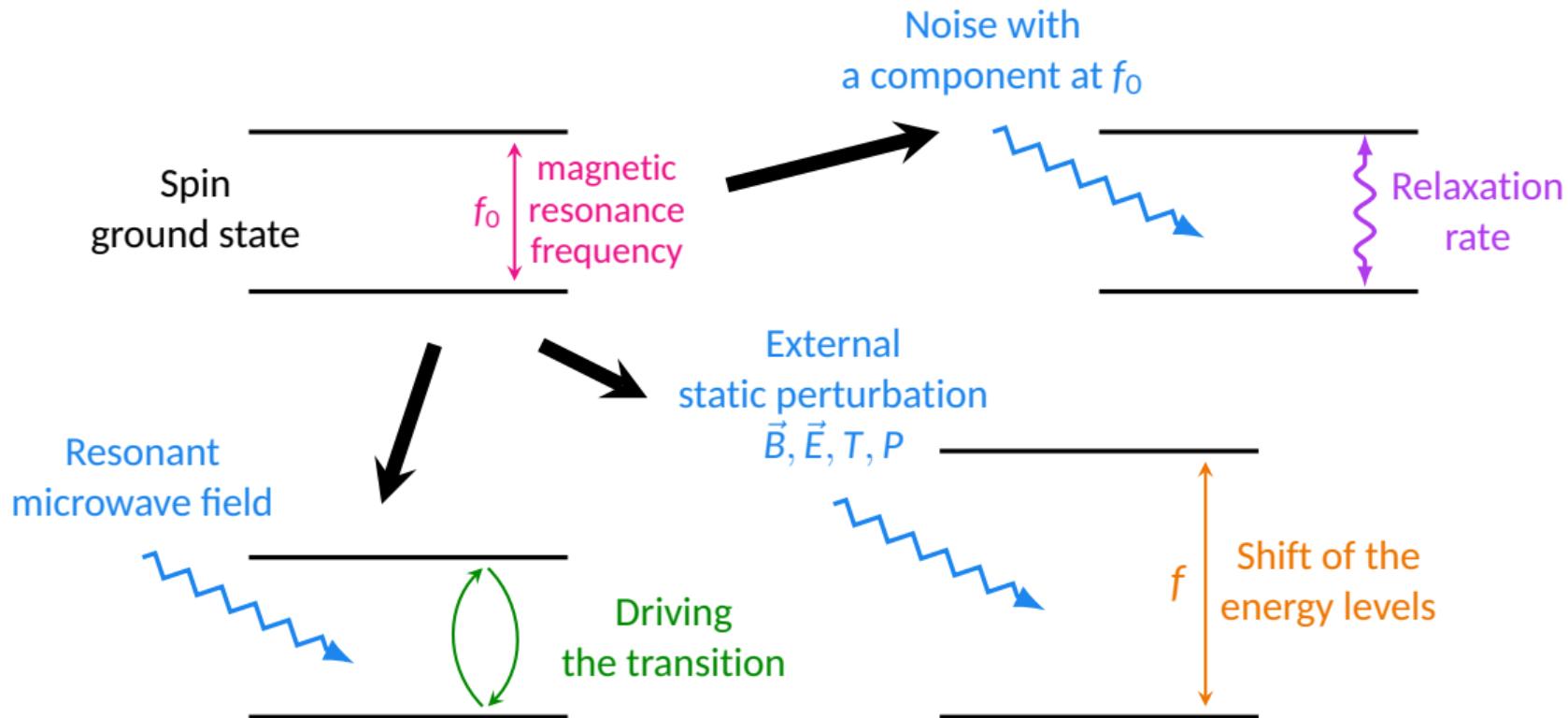
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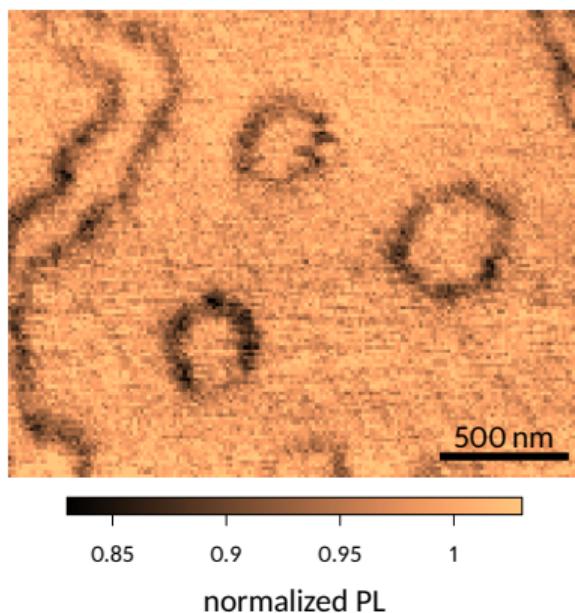
# How can we use a quantum system to probe nanomagnetism?



# Outline

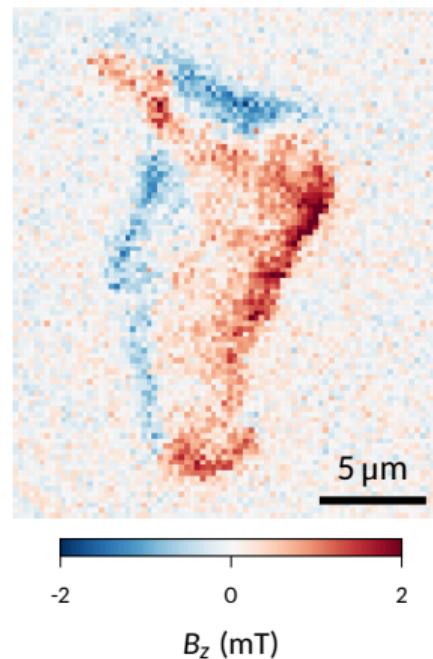
## NV centers in diamond

Probe magnetic textures using  
spin wave noise



## Boron vacancies in h-BN

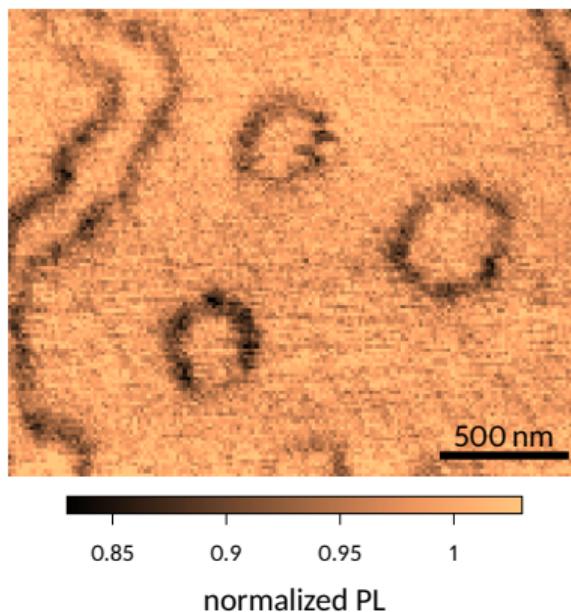
Integrate the sensor in a van der  
Waals heterostructure



# Outline

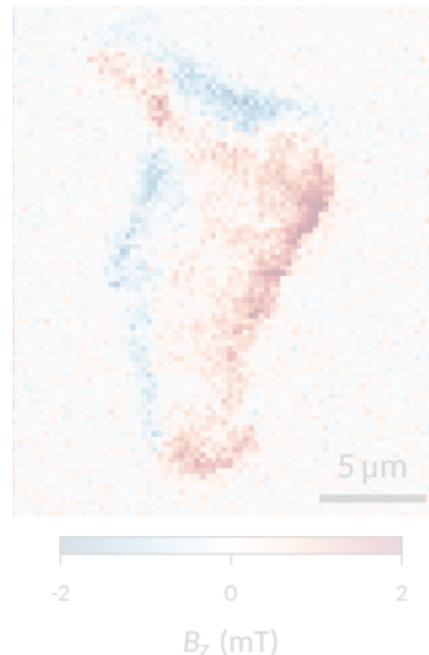
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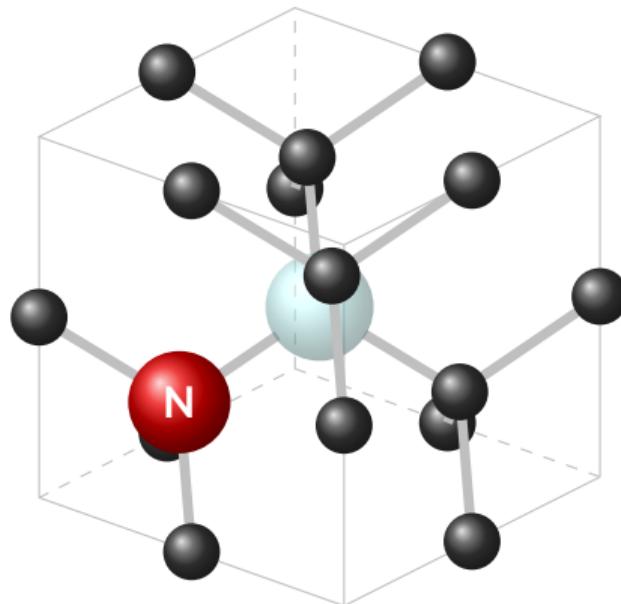


## Boron vacancies in h-BN

Integrate the sensor in a van der  
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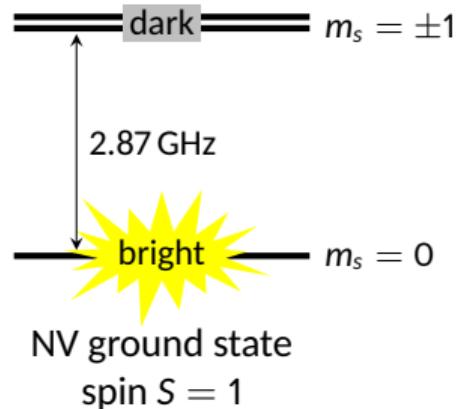
# The NV center in diamond



- Artificial atom: energy levels in the diamond bandgap
- Photostable defect
- Spin  $S=1$
- Individual defects can be isolated/implanted
- Ambient conditions

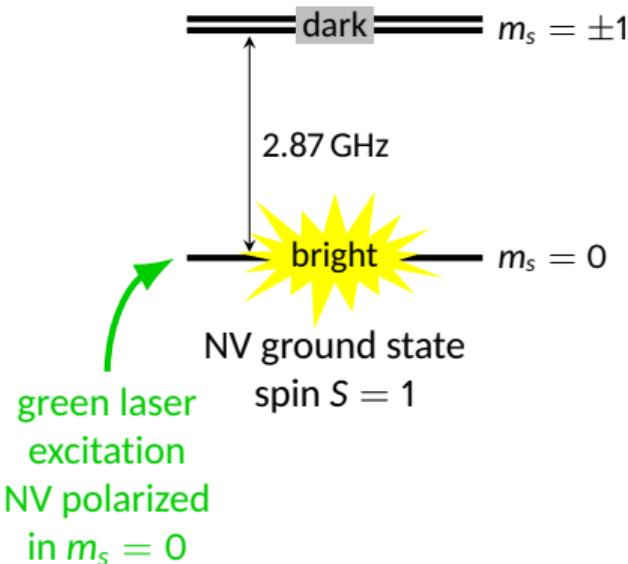
# Principle of static magnetic field measurement

Spin-dependent  
fluorescence

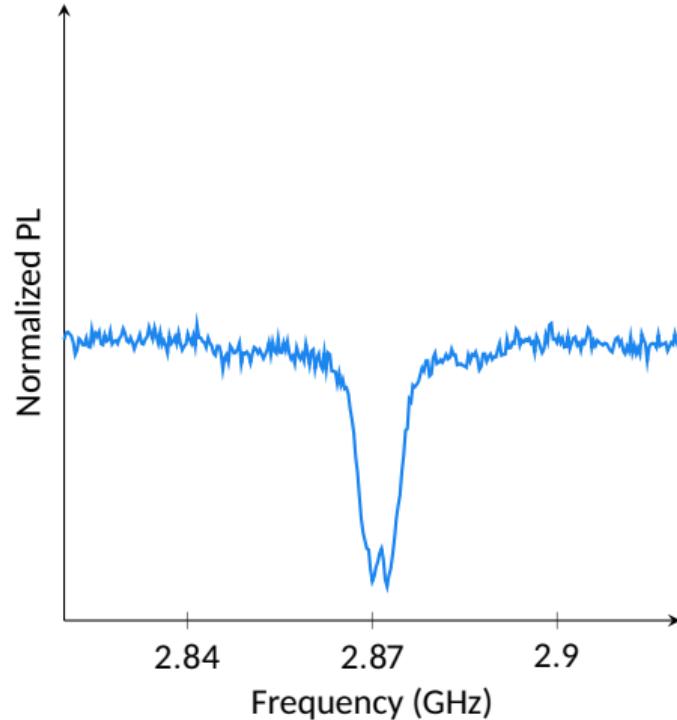
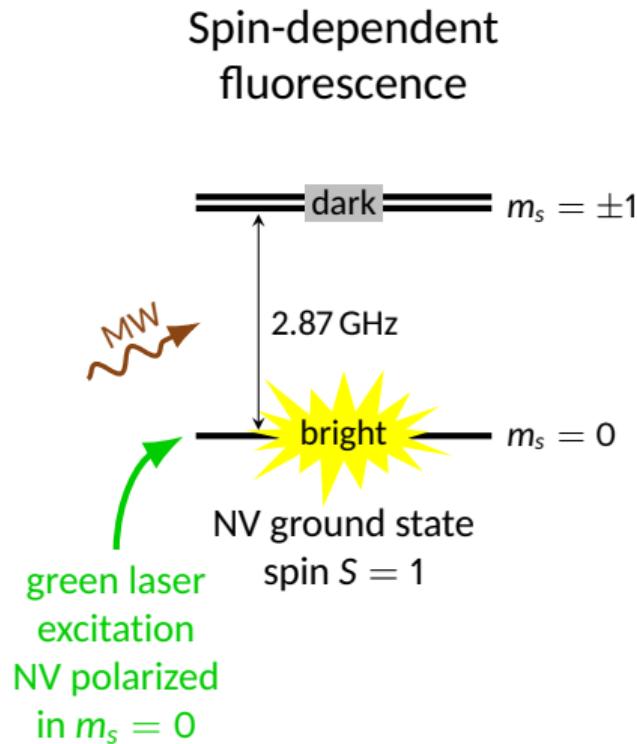


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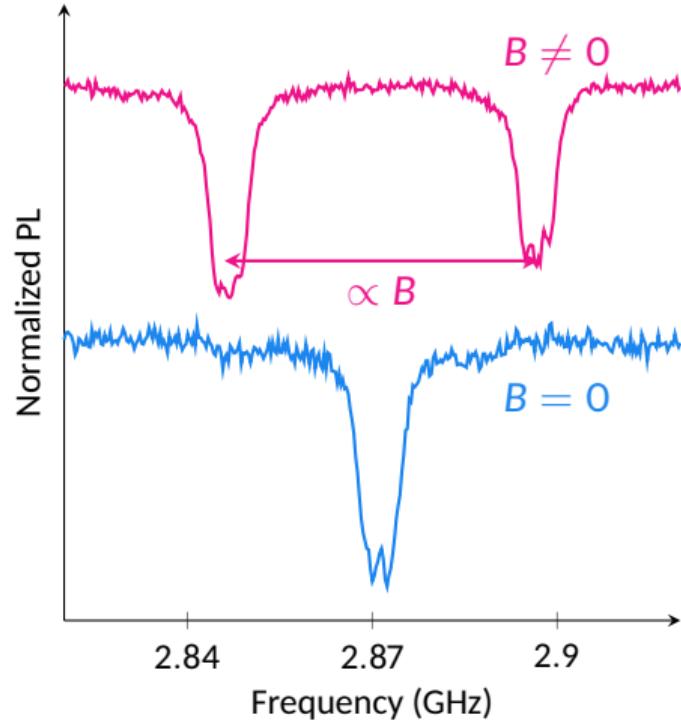
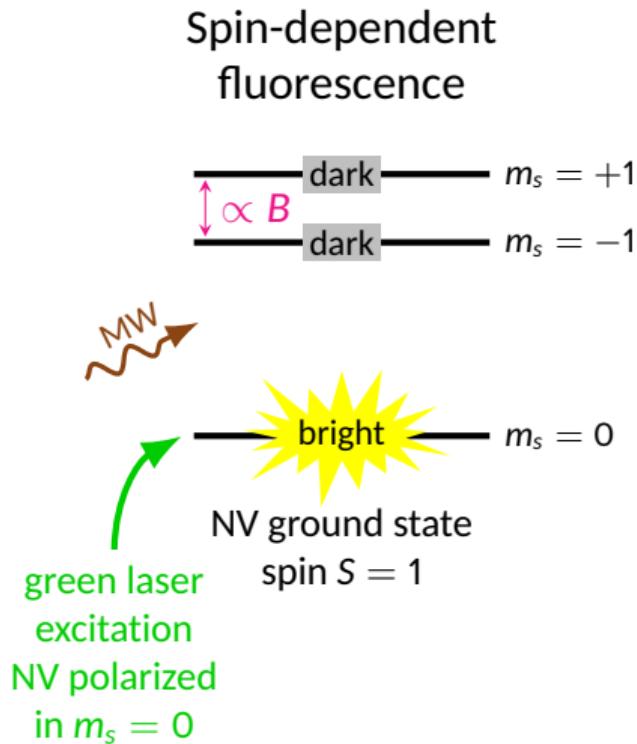
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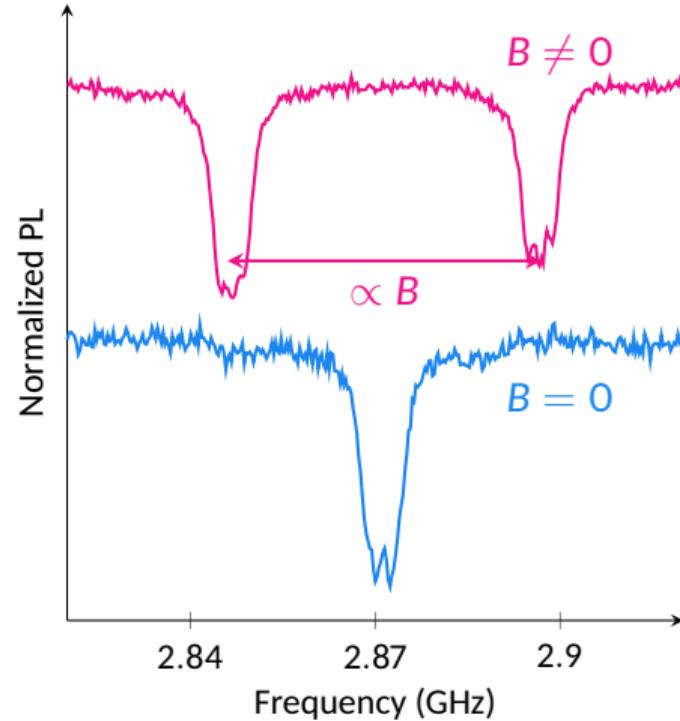
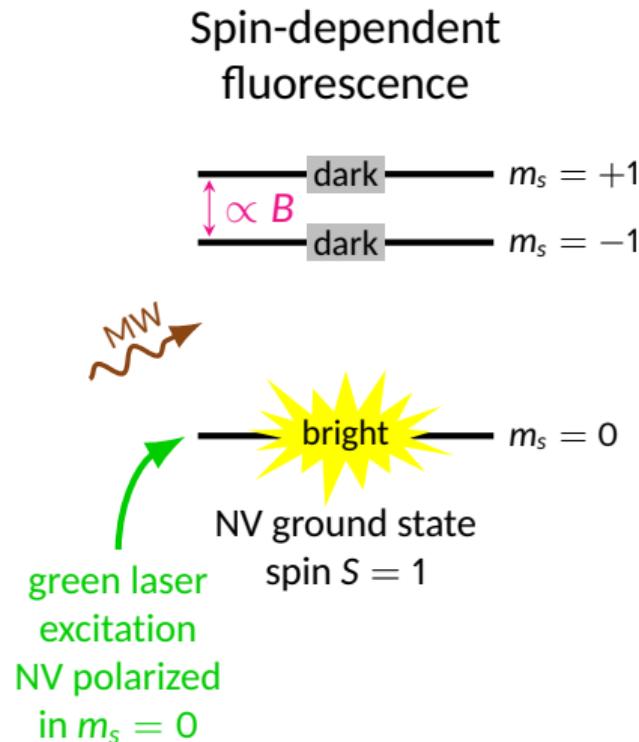
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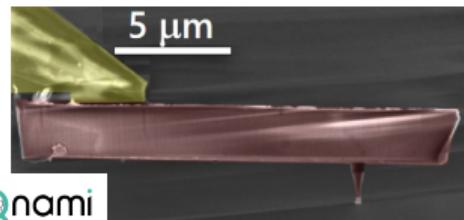
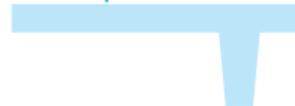
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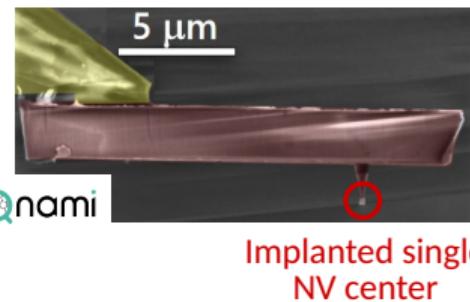
Sensitivity: a few  $\mu\text{T}/\sqrt{\text{Hz}}$

# Integration of the defect in a scanning probe microscope

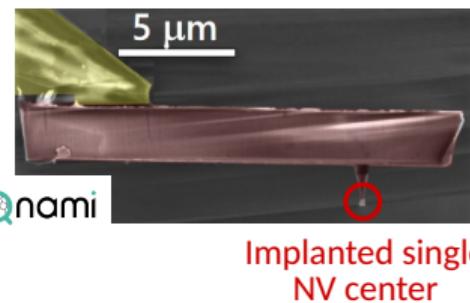
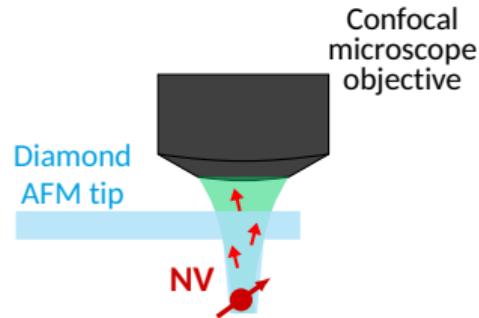
Diamond  
AFM tip



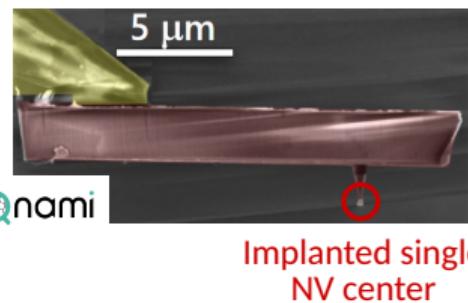
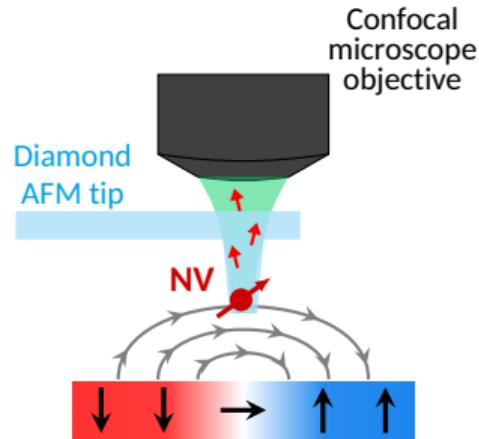
# Integration of the defect in a scanning probe microscope



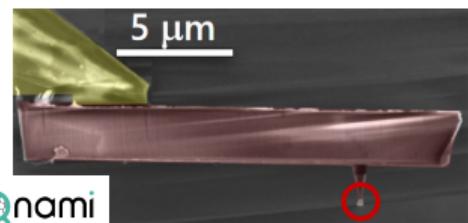
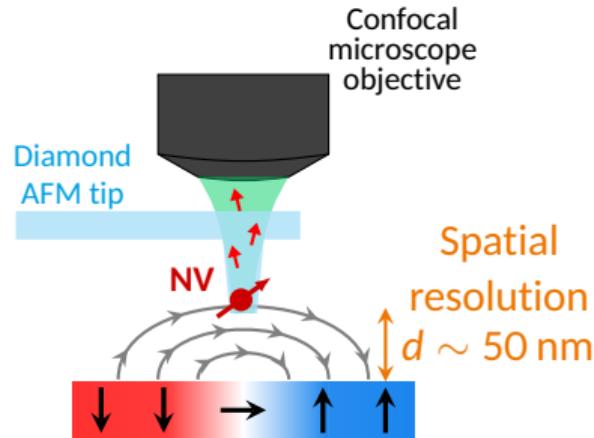
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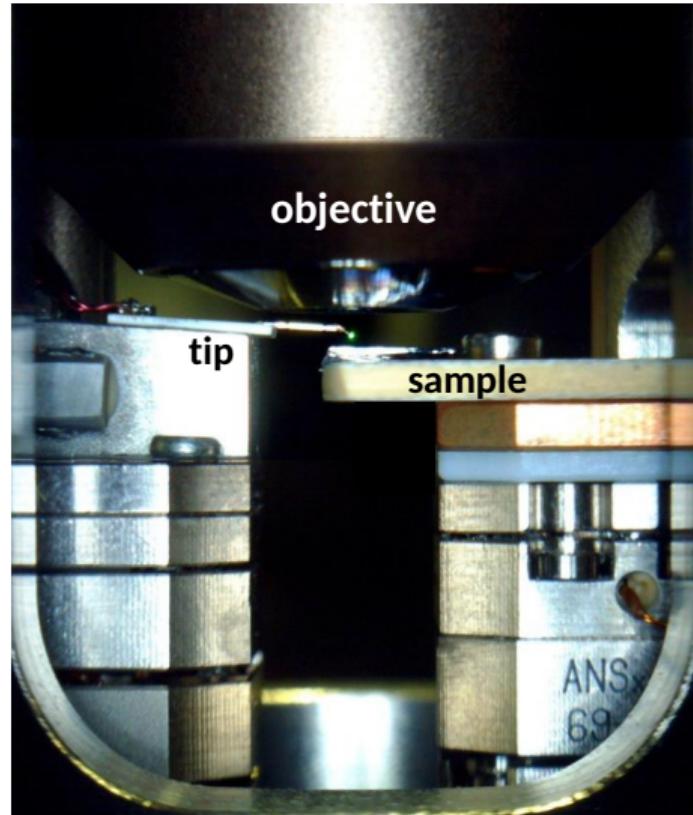
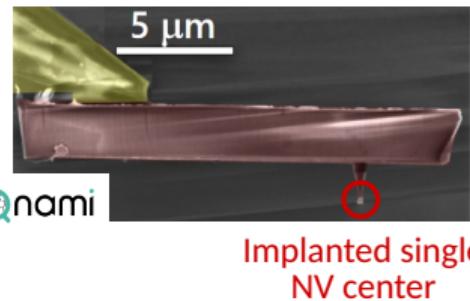
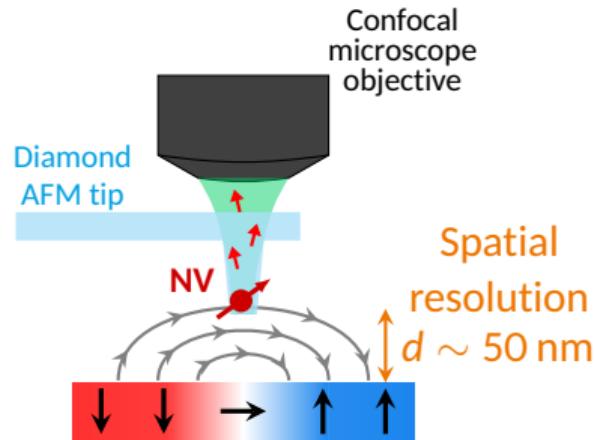


# Integration of the defect in a scanning probe microscope



Implanted single  
NV center

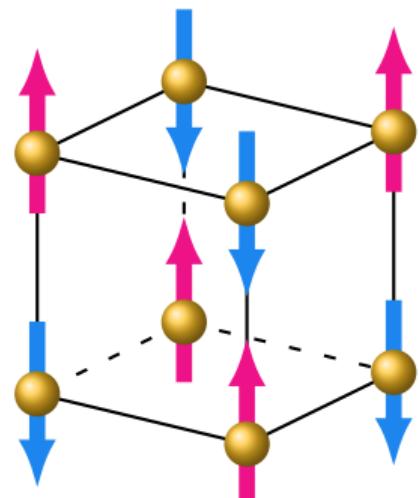
# Integration of the defect in a scanning probe microscope



# A powerful tool to image antiferromagnets

## Example: Bismuth ferrite

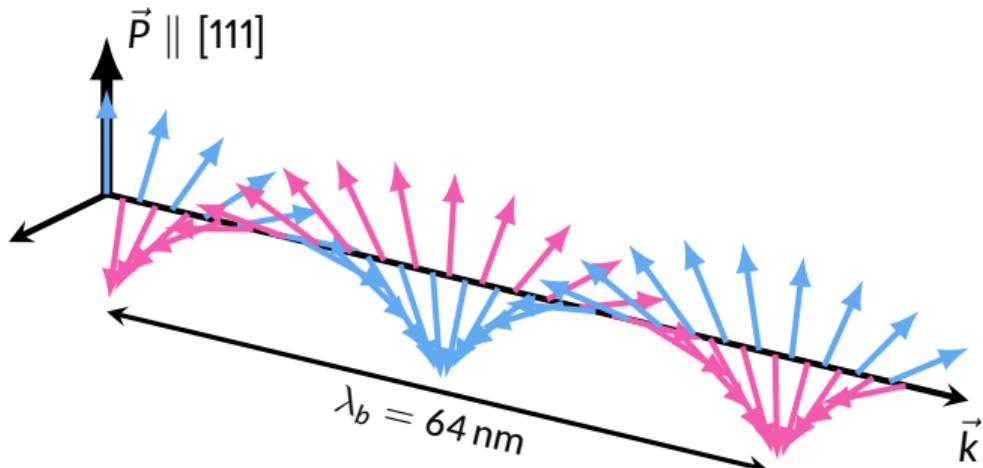
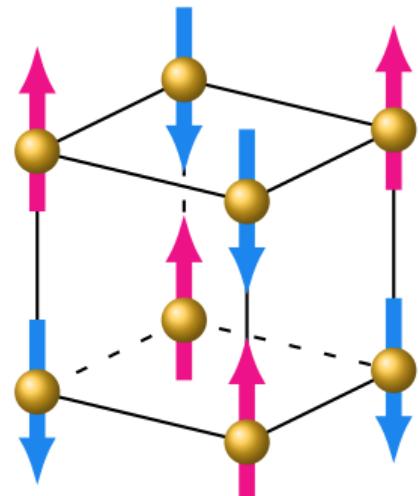
G-type antiferromagnet



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G-type antiferromagnet

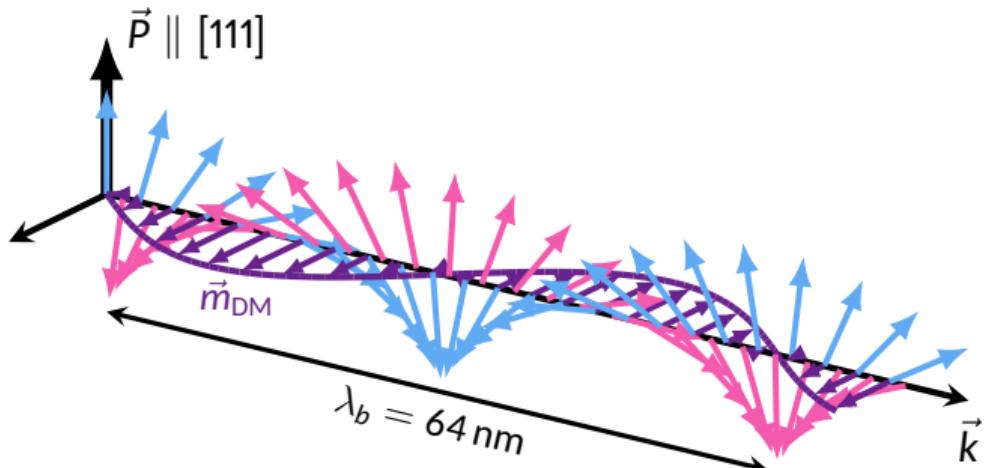
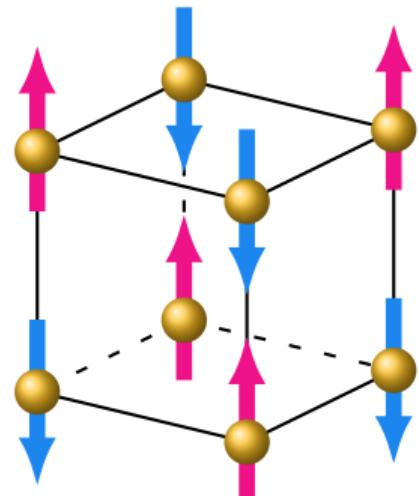


Fully compensated cycloid  
→ No stray field!

# A powerful tool to image antiferromagnets

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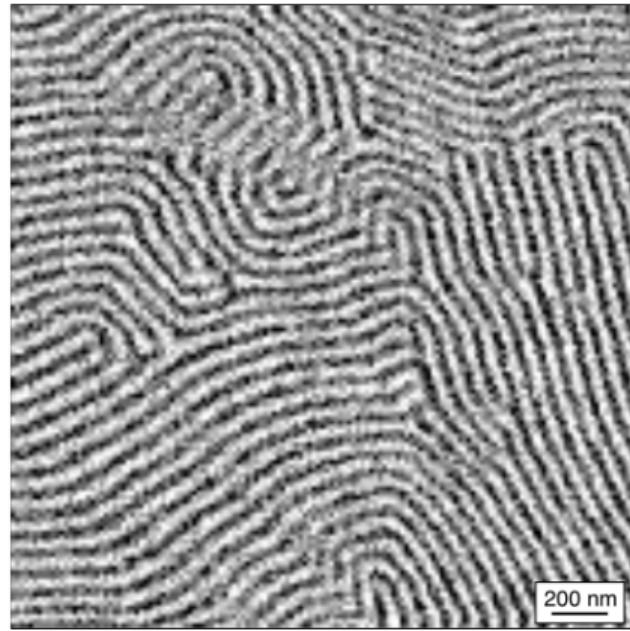
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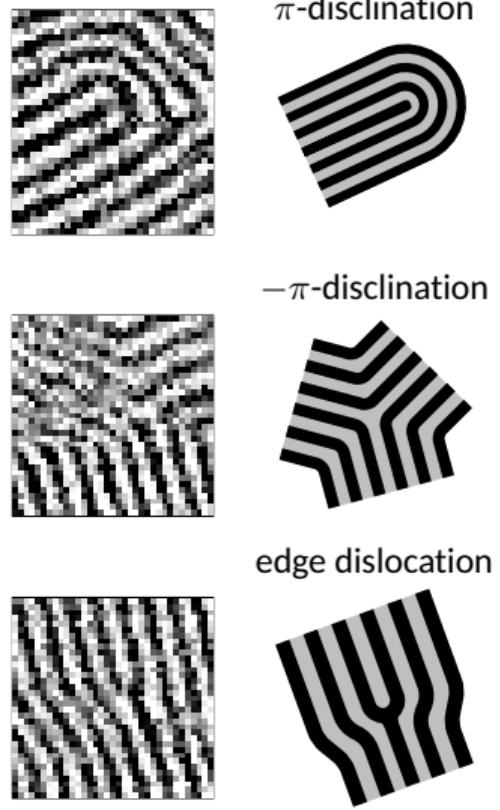
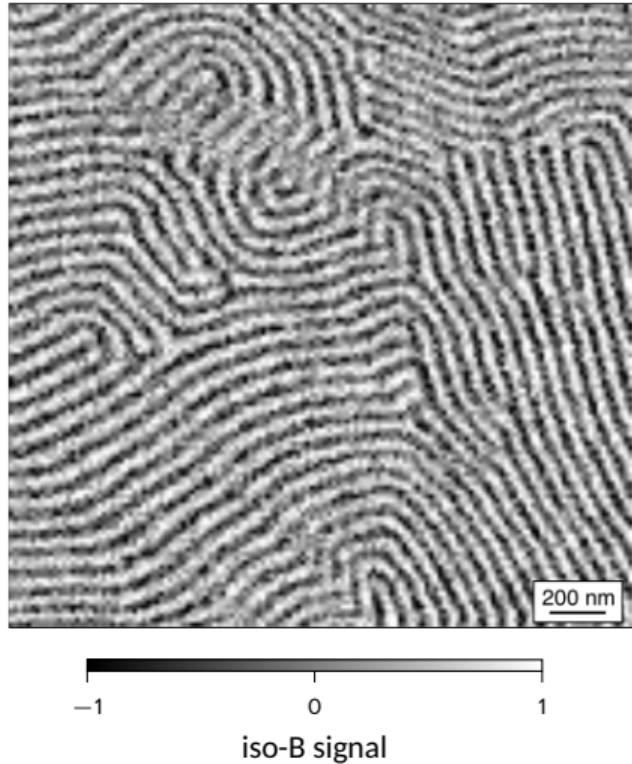
Spin density wave  
Weak uncompensated moment  
→ Small stray field

M. Ramazanoglu et al. *Phys. Rev. Lett.* 107 (2011), 207206

# Topological defects at the surface of bulk BiFeO<sub>3</sub> crystals



# Topological defects at the surface of bulk BiFeO<sub>3</sub> crystals



# Detection of magnetic noise rather than stray field

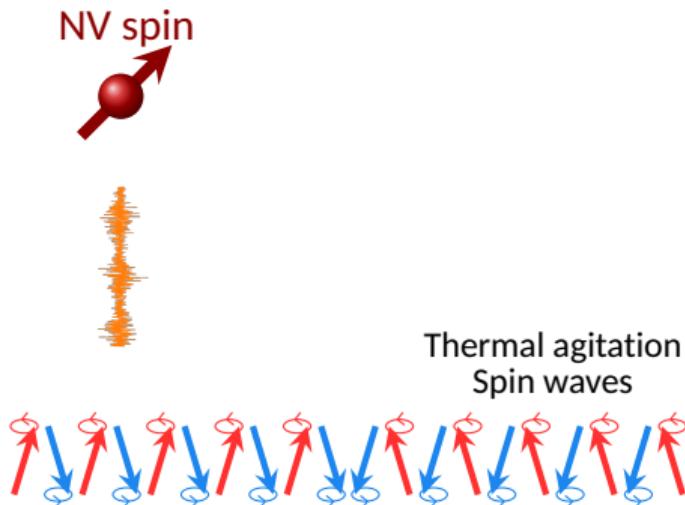
 B. Flebus *et al.* *Phys. Rev. B* 98 (2018), 180409

- Completely compensated antiferromagnets = **no static stray field** to probe
- But NV centers are also sensitive to **magnetic noise!**
- Use the different noise properties above domains and domain walls for imaging

# Detection of magnetic noise rather than stray field

 B. Flebus et al. *Phys. Rev. B* 98 (2018), 180409

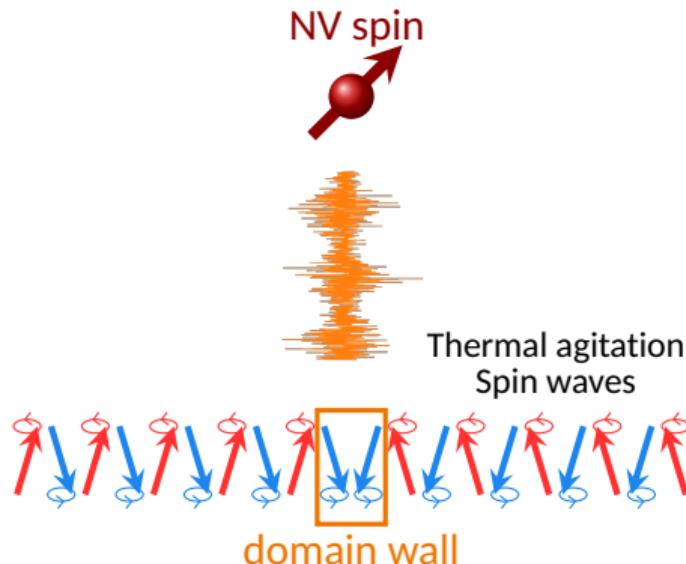
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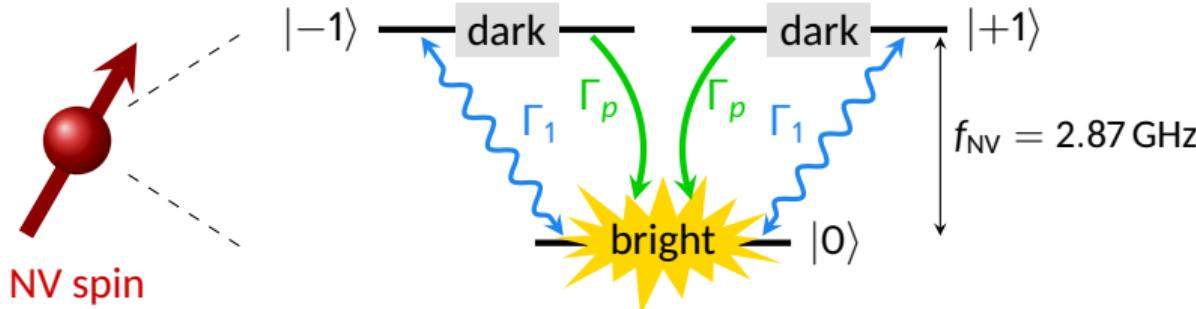
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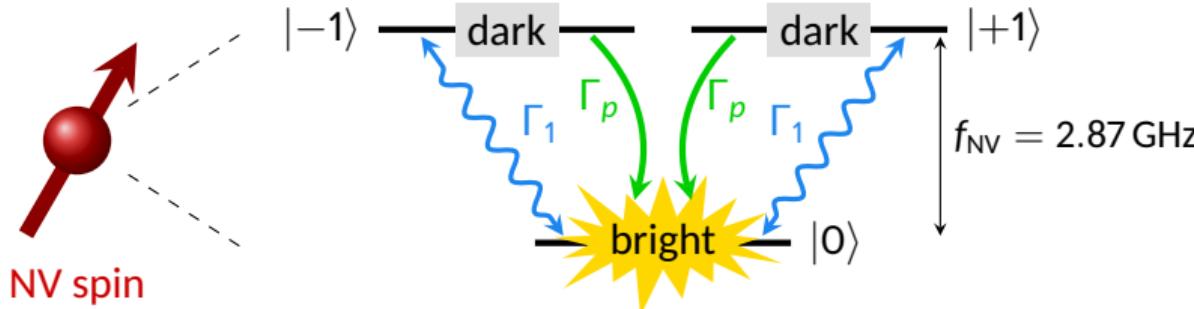


# Effect of magnetic noise on the emitted photoluminescence

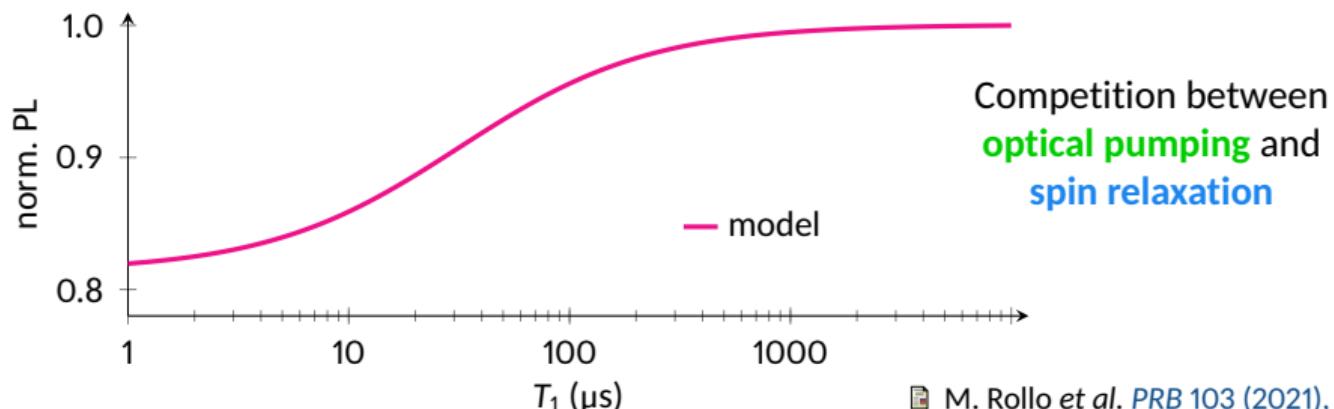


Relaxation rate  $\Gamma_1 \propto S_{B_\perp}(f_{\text{NV}})$  magnetic field spectral density at the resonance frequency  $f_{\text{NV}}$

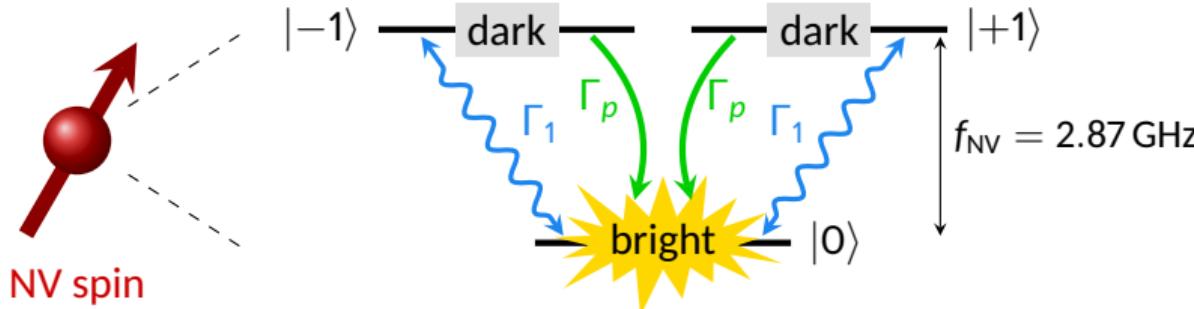
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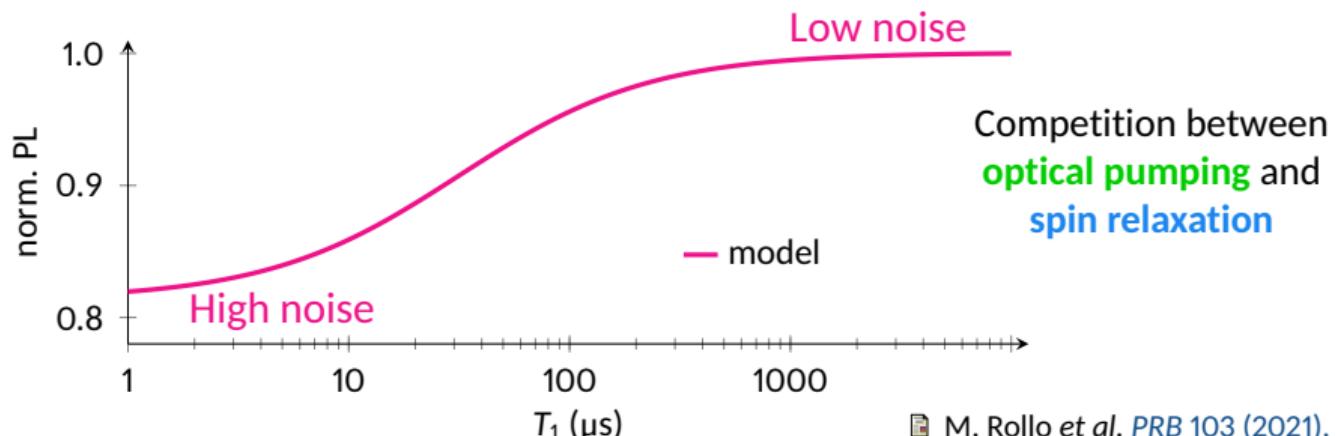
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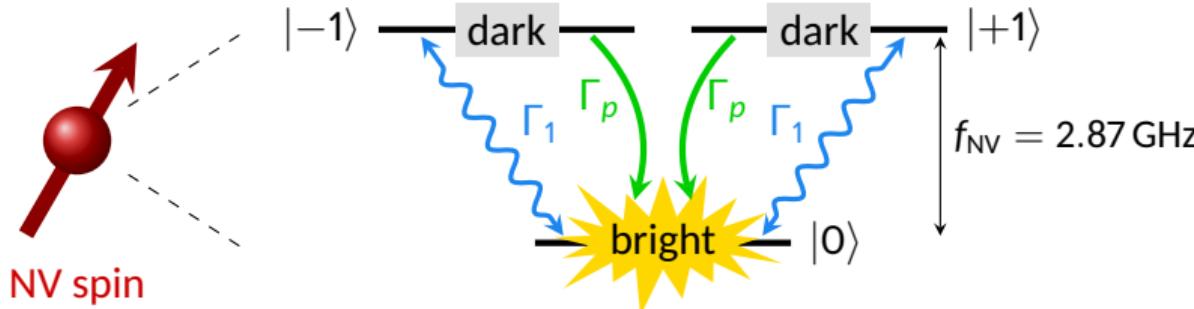
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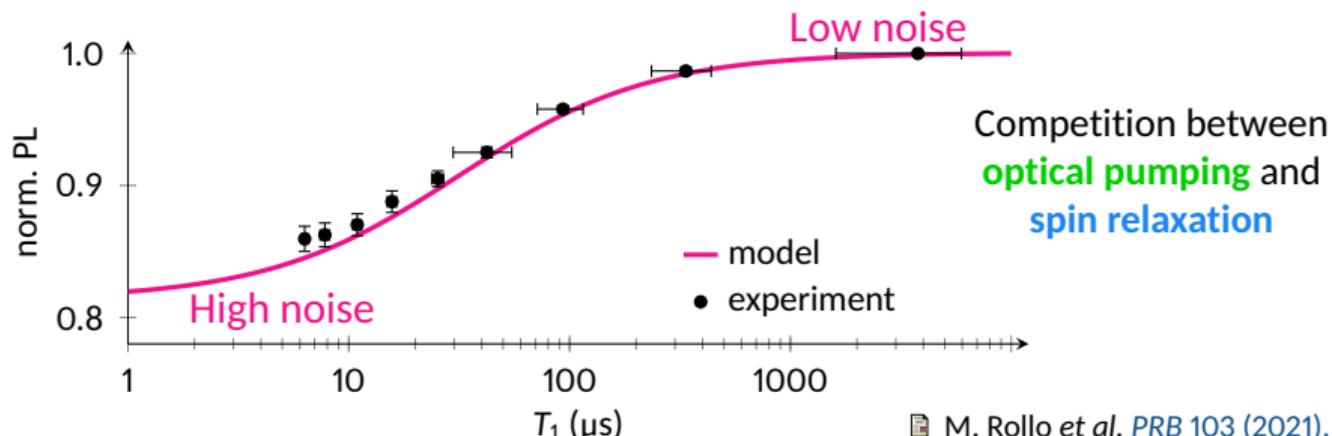
Relaxation rate  $\Gamma_1 \propto S_{B_\perp}(f_{\text{NV}})$  magnetic field spectral density at the resonance frequency  $f_{\text{NV}}$



# Effect of magnetic noise on the emitted photoluminescence



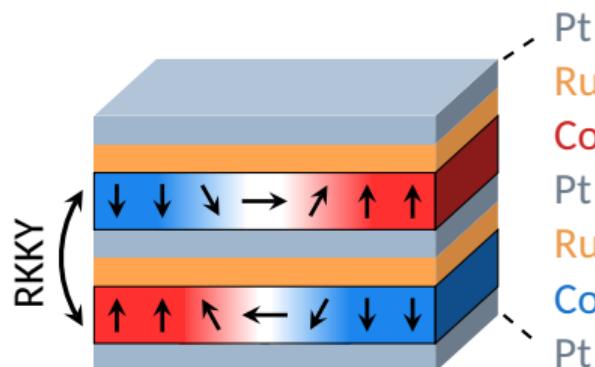
Relaxation rate  $\Gamma_1 \propto S_{B_\perp}(f_{\text{NV}})$  magnetic field spectral density at the resonance frequency  $f_{\text{NV}}$



# Synthetic antiferromagnets

Samples: LAF, Palaiseau (W. Legrand, K. Bouzehouane, N. Reyren, V. Cros)  
Spintec, Grenoble (V.-T. Pham, J. Urrestarazu, O. Boulle)

Two **ferromagnetic** layers coupled **antiferromagnetically**

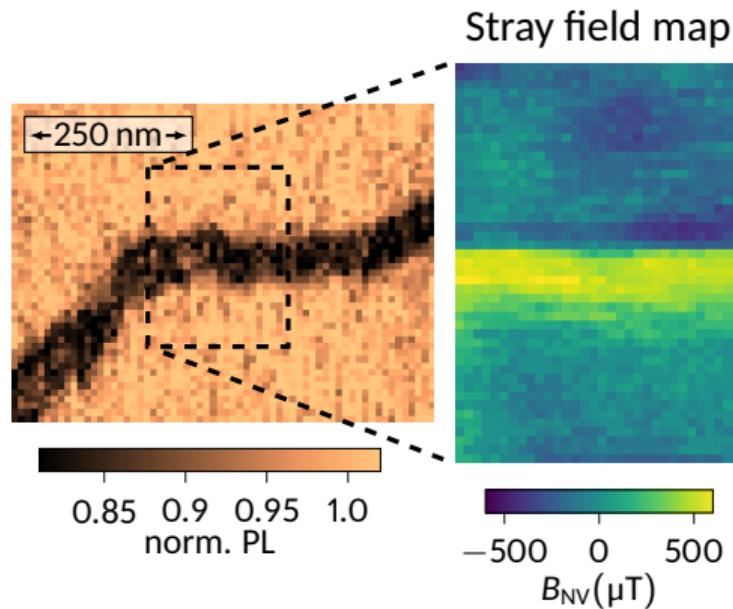
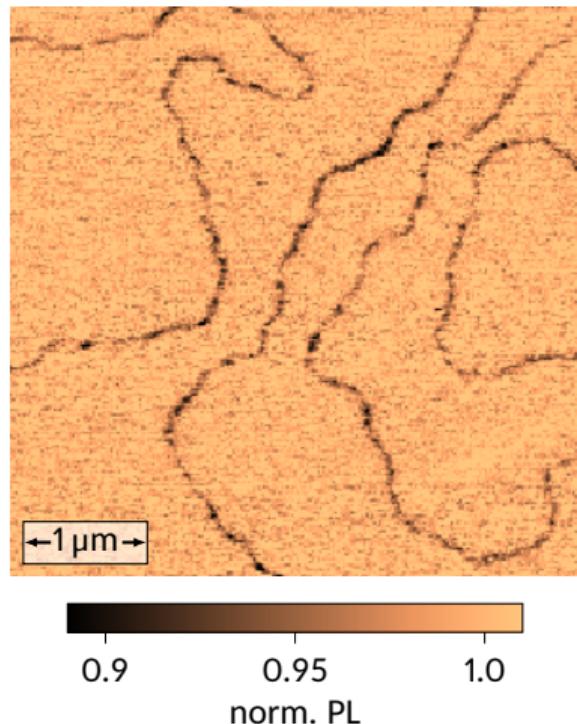


- No net magnetic moment
- Small stray field (vertical shift)
- Highly tunable properties
- Spin wave frequencies in the few GHz range

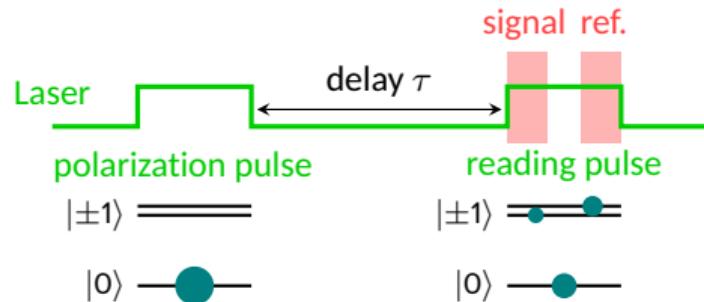
→ Perfect **test system**  
for noise imaging!

■ W. Legrand et al. *Nat. Mat.* 19 (2020), 34

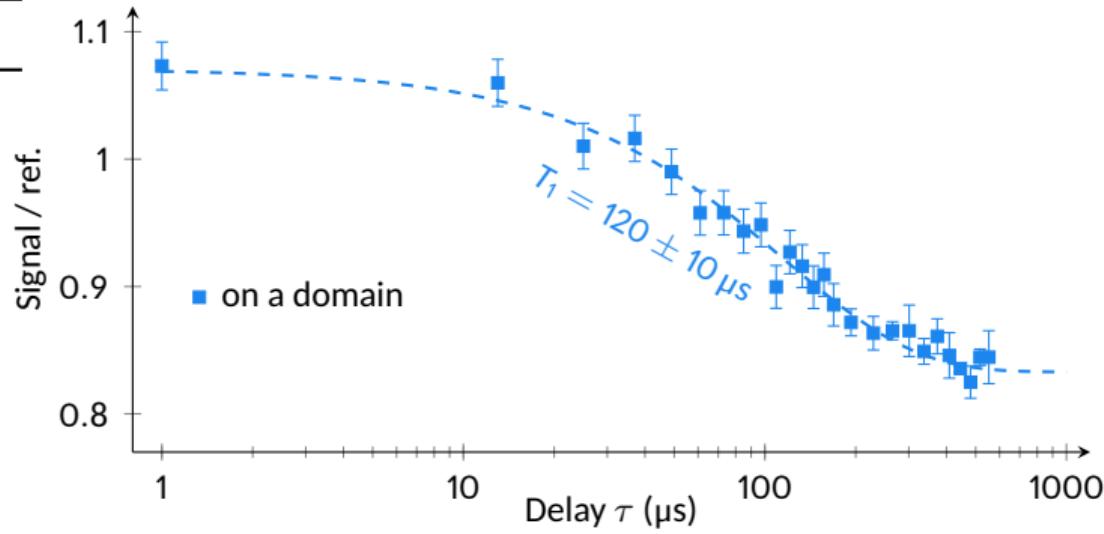
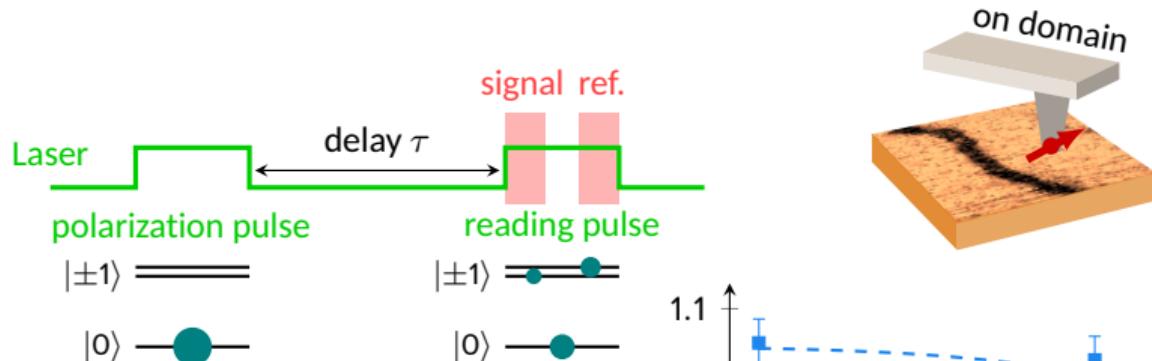
# Detection of domain walls by relaxometry



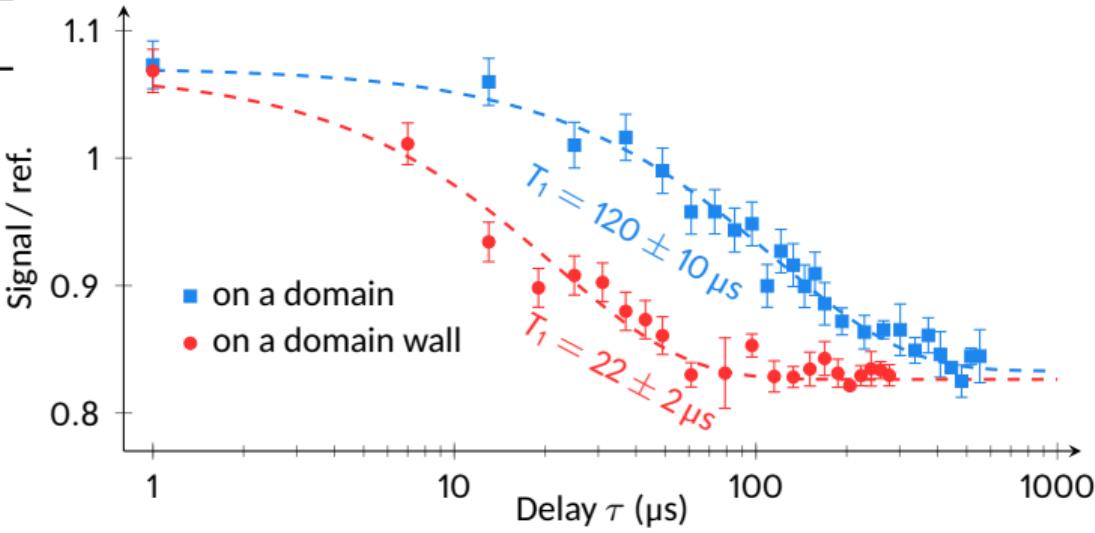
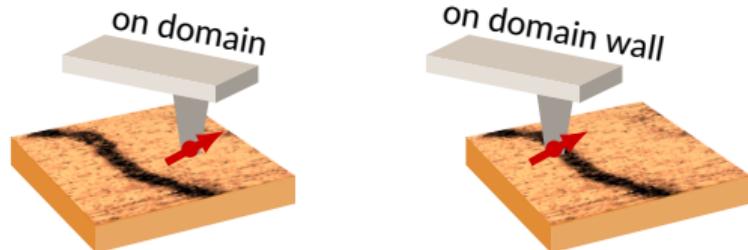
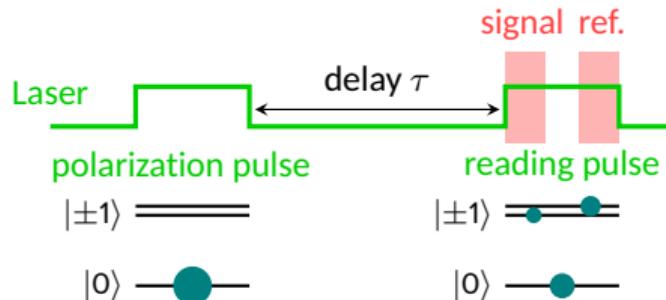
# Local variation of the relaxation time



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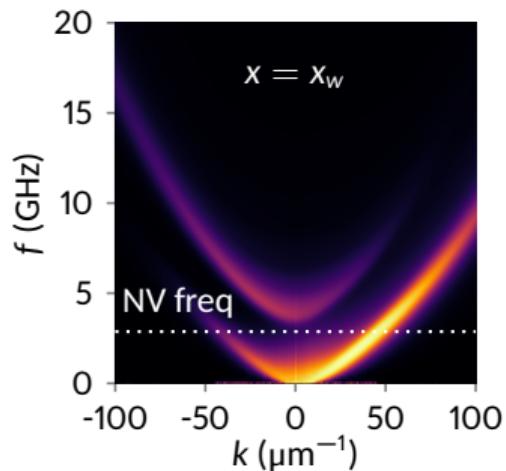
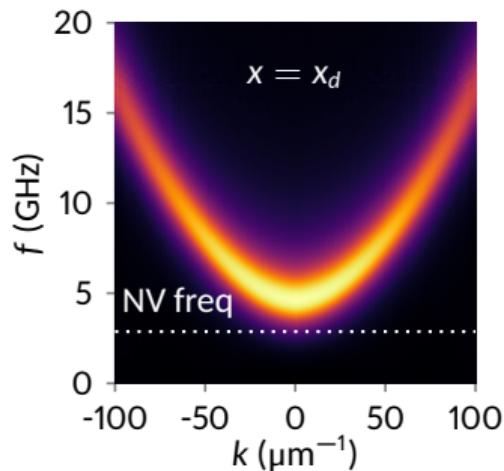
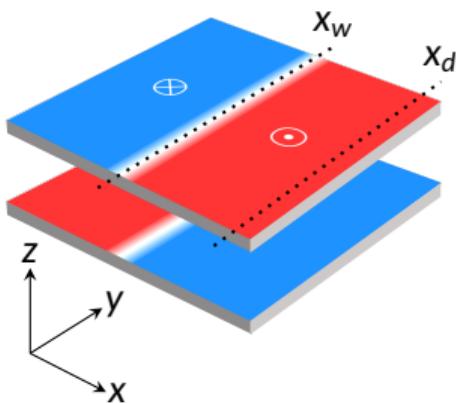


Clear diminution of  $T_1$

→ Enhancement of the spin relaxation

# Origin of the noise: spin waves

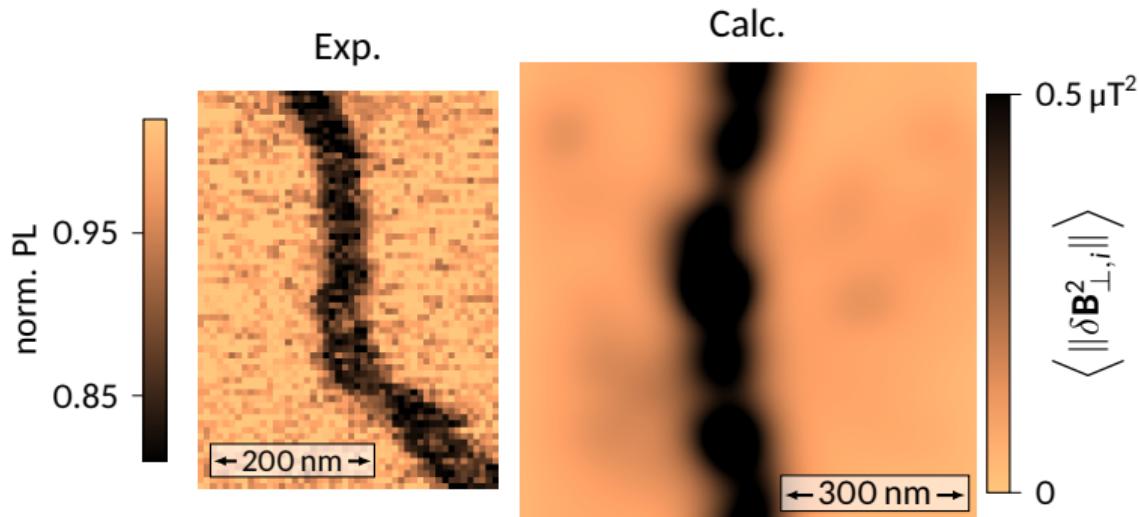
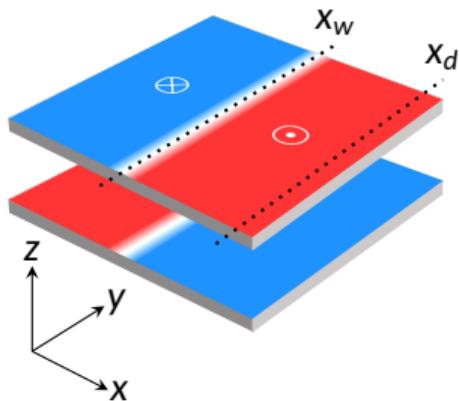
Collaboration: C2N, Palaiseau (J.-P. Adam, J.-V. Kim)



No gap in the domain walls, presence of modes at the NV frequency: **the NV center is more sensitive to the noise from the walls!**

# Origin of the noise: spin waves

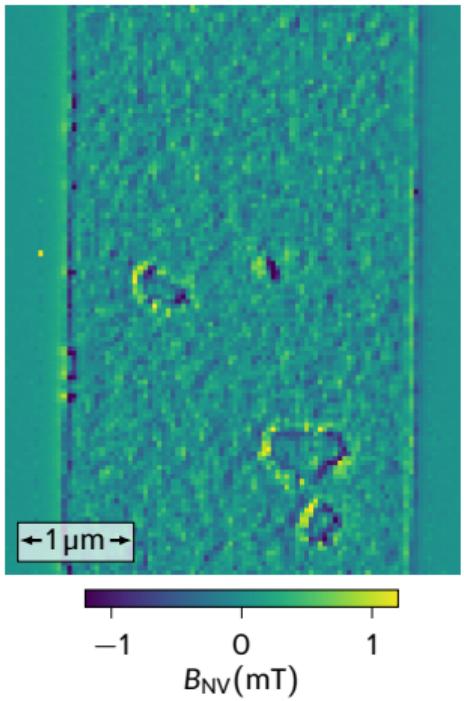
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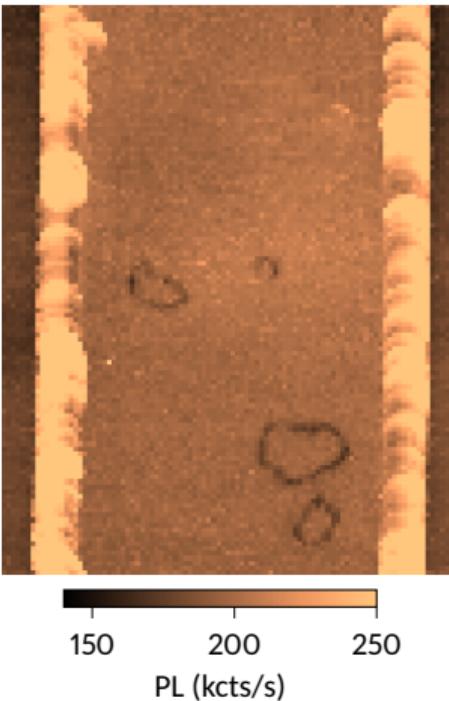
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# After applying magnetic field

NV stray field map

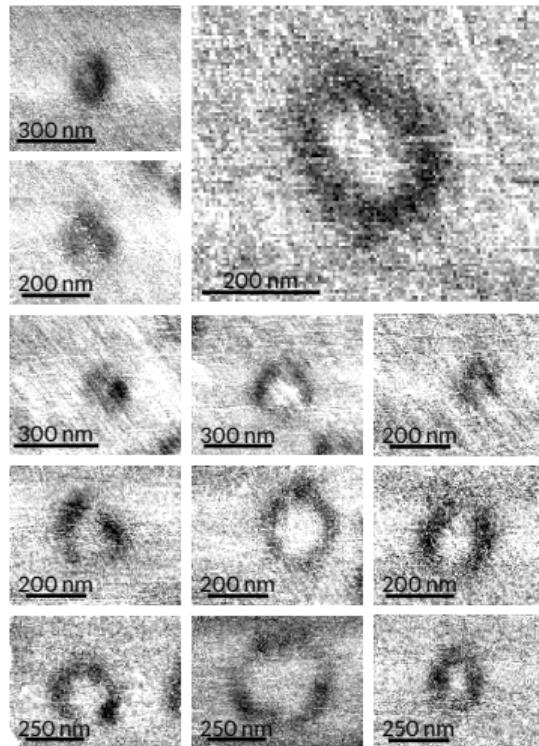


Noise (PL) map

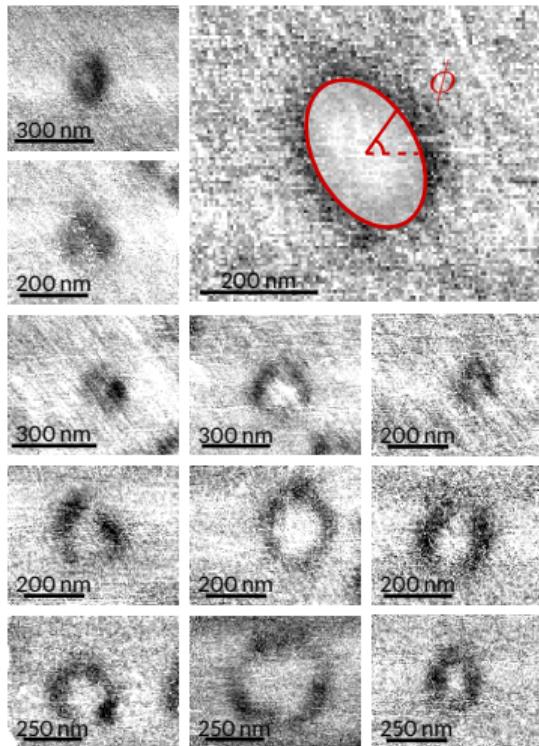


- Opp field of about 150 mT applied for nucleation
- Skyrmions and big bubbles pinned

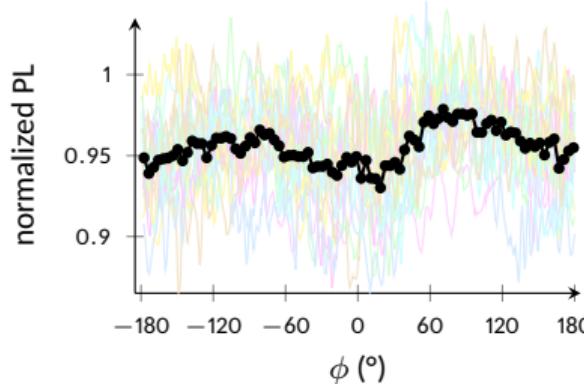
# Statistics on Néel left (CCW) skyrmions



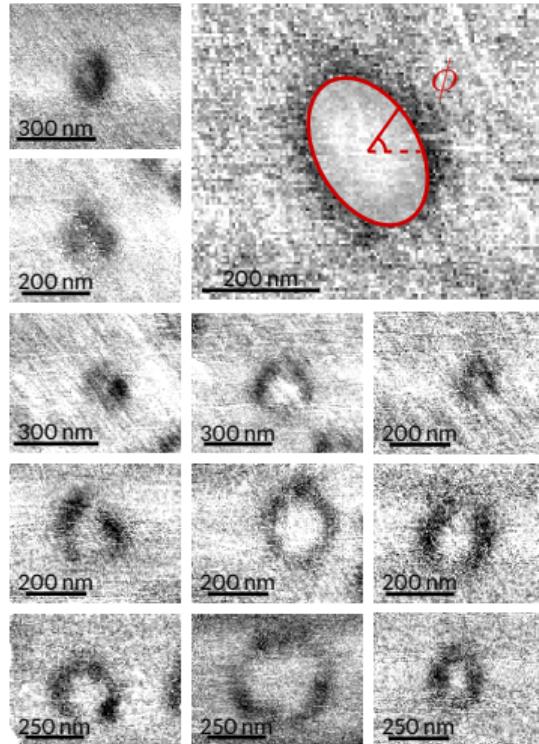
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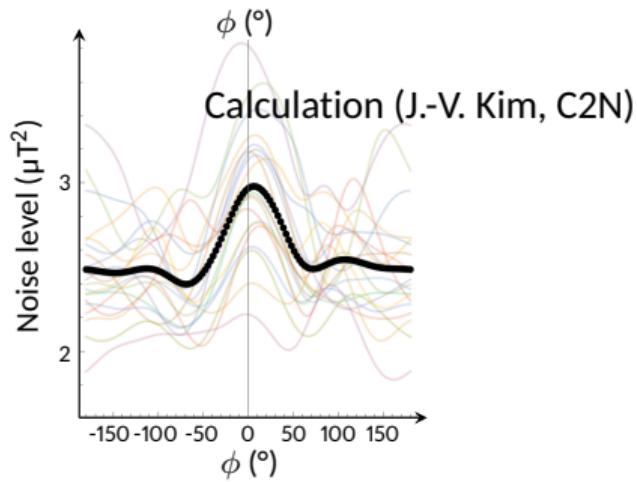
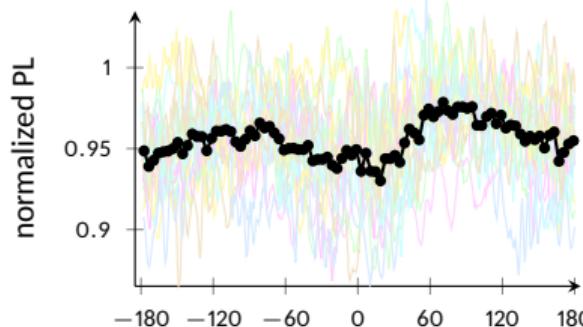
Angular variation of PL



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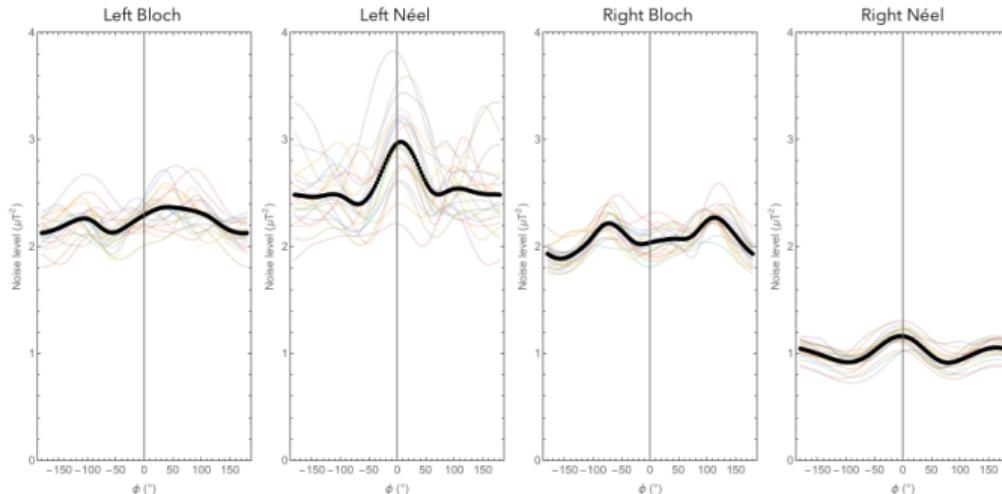


Angular variation of PL

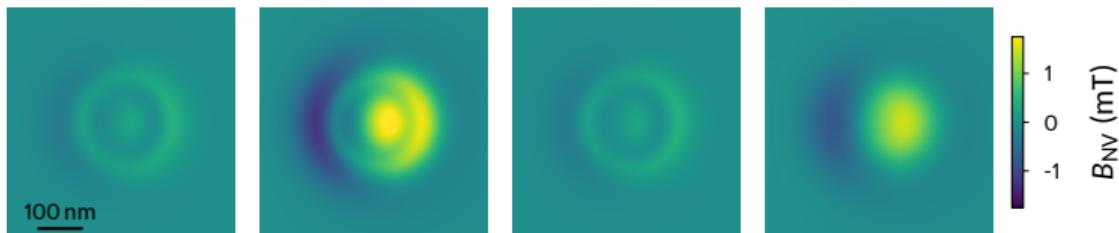


# Expected pattern on other skyrmion types

Simulated noise distribution along the contour



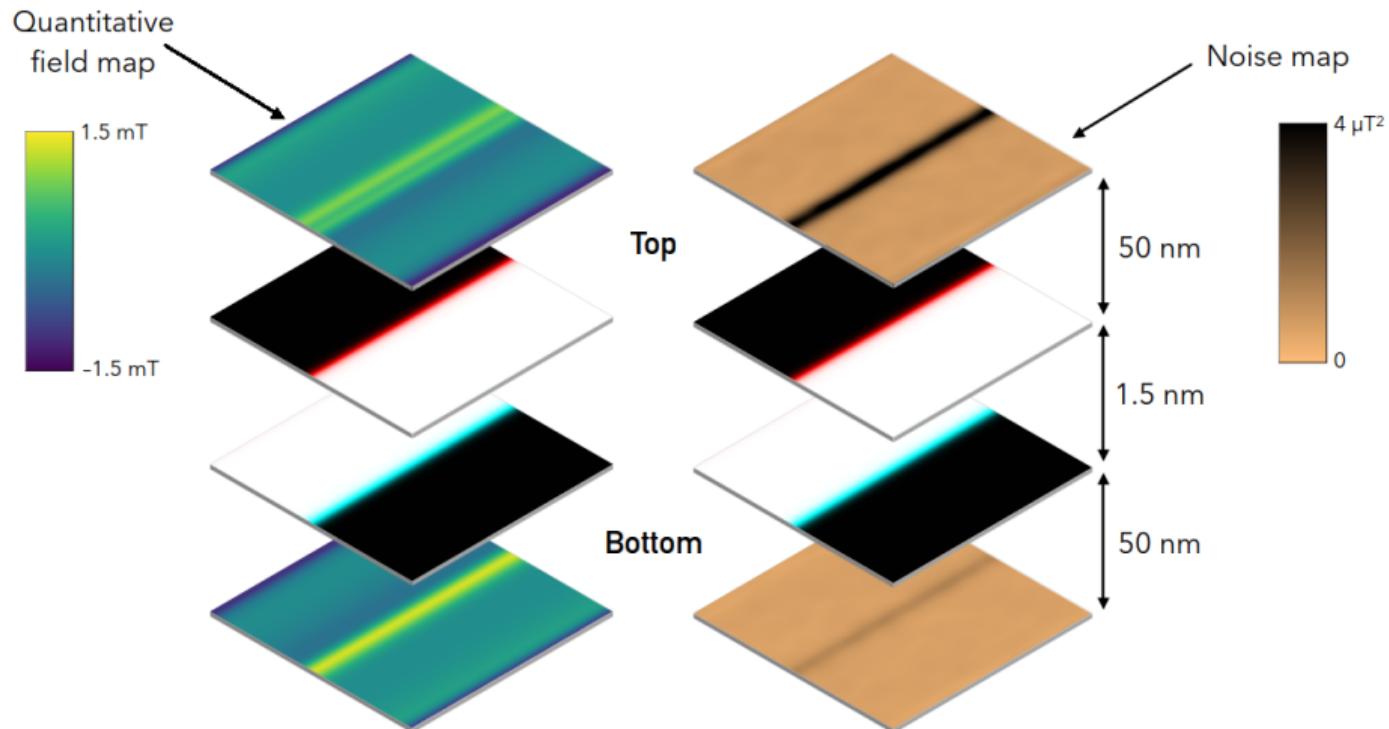
Simulated stray field maps



- The pattern allows us to identify Néel skyrmions
- Strong difference in noise amplitude expected between Néel left and Néel right skyrmions...
- ... while the stray field maps are very similar!

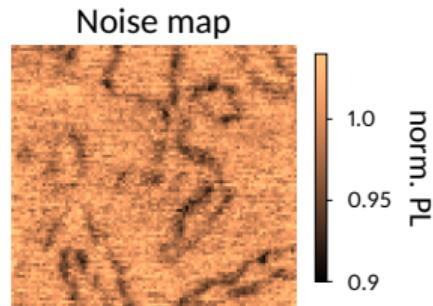
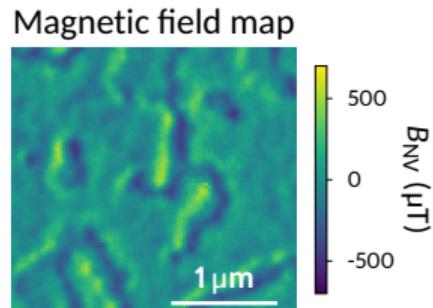
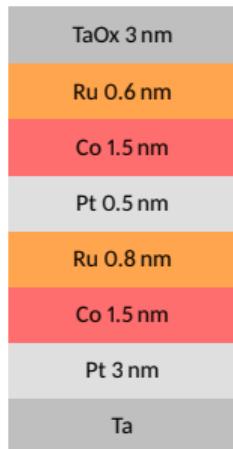
# Do we also expect this for domain walls? Yes!

Calculation: C2N, Palaiseau (J.-V. Kim)



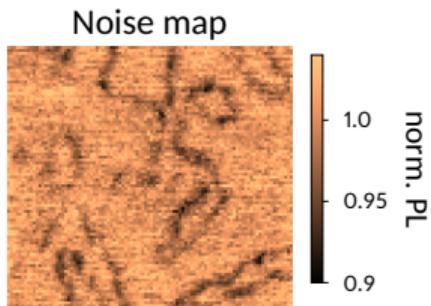
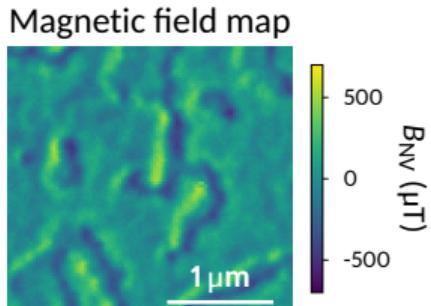
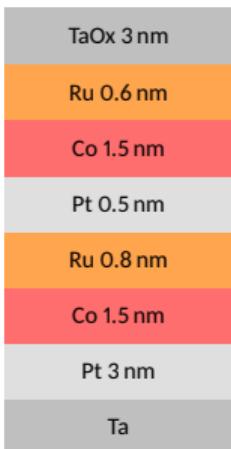
# Experiment: looking at both sides of the film

Initial stack: Néel left

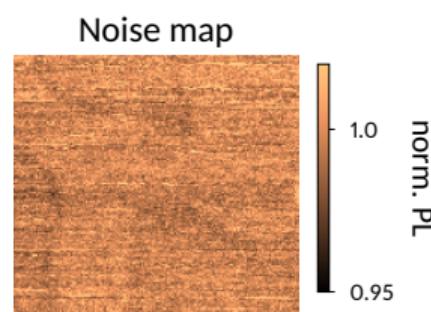
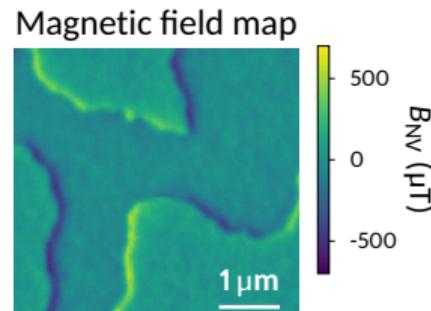
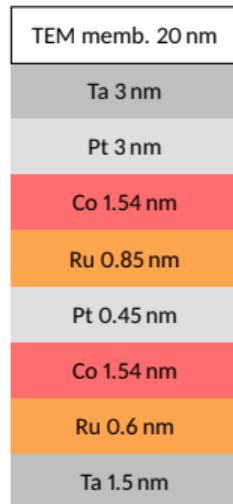


# Experiment: looking at both sides of the film

Initial stack: Néel left

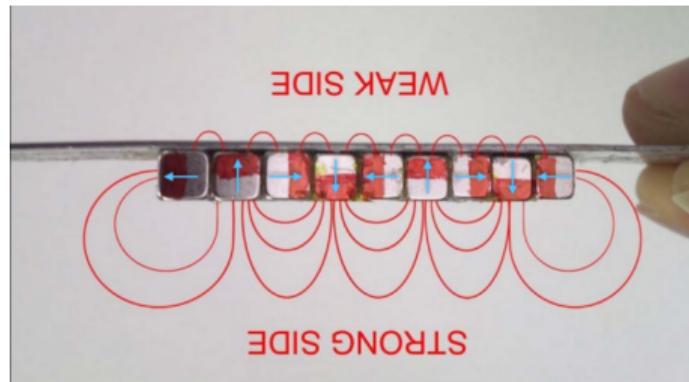
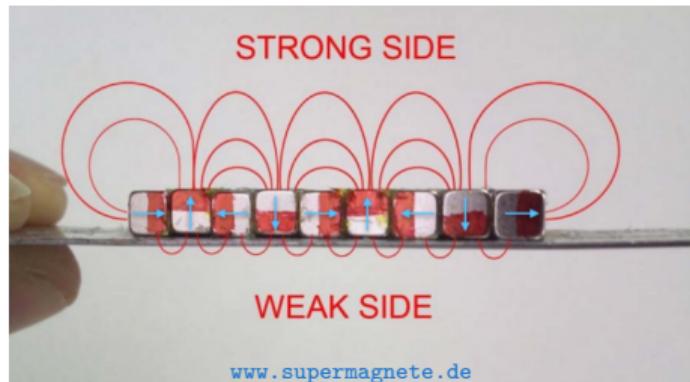


Inverted stack: Néel right



# Origin of this effect, 1st ingredient : Spin waves = fridge magnets

## Halbach arrays



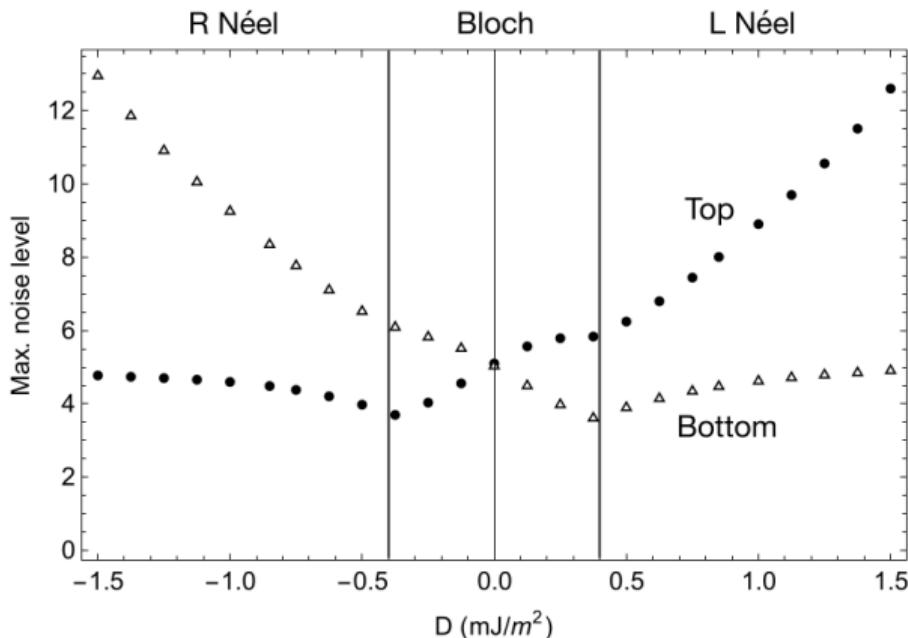
$$\vec{m}_0 \quad \bullet \quad \bullet \quad \bullet \quad \bullet \quad \bullet$$
$$\delta\vec{m} \quad \rightarrow \quad \uparrow \quad \leftarrow \quad \downarrow \quad \rightarrow \quad +\vec{k}$$

$$\vec{m}_0 \quad \bullet \quad \bullet \quad \bullet \quad \bullet \quad \bullet$$
$$\delta\vec{m} \quad \leftarrow \quad \uparrow \quad \rightarrow \quad \downarrow \quad \leftarrow \quad -\vec{k}$$

# Origin of this effect, 2nd ingredient: DMI

Calculation: C2N, Palaiseau (J.-V. Kim)

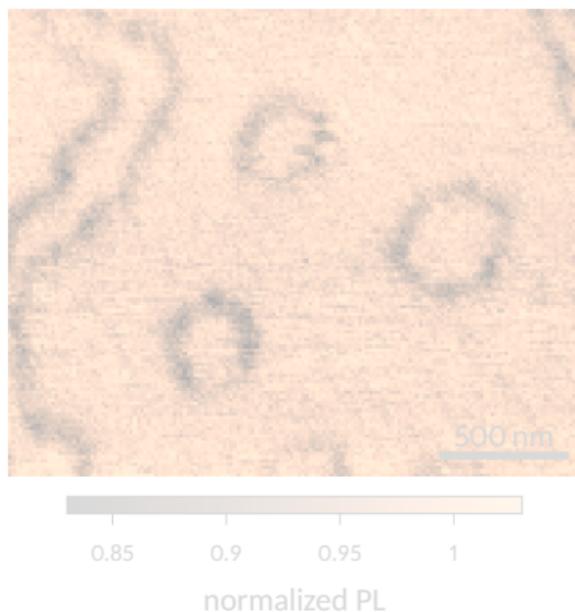
Calculation made for a **single** ferromagnetic layer



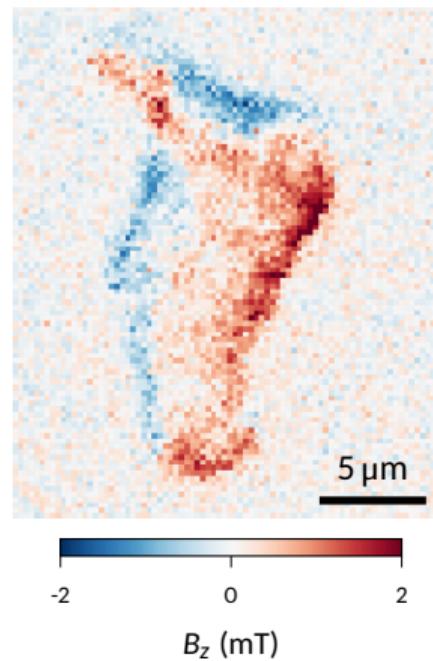
- DMI induces non-reciprocity in the spin wave dispersion
- This results in the selection of a propagation direction for the modes producing the detected noise
- **These modes create noise only on one side of the film!**

# Outline

NV centers in diamond  
Probe magnetic textures using  
spin wave noise



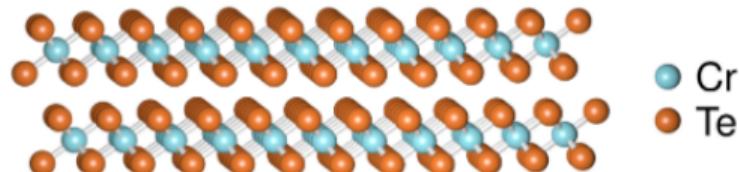
Boron vacancies in h-BN  
Integrate the sensor in a van der  
Waals heterostructure



# Imaging magnetic van der Waals materials

Collaboration: Institut Néel, Grenoble (A. Purbawati, J. Coraux, N. Rougemaille)

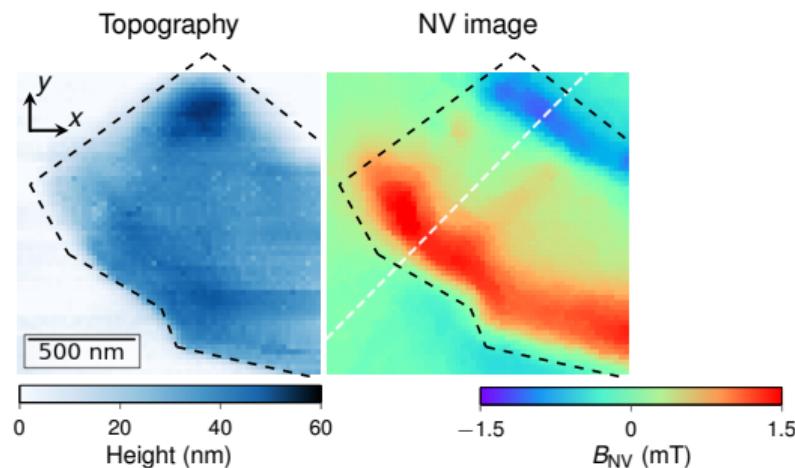
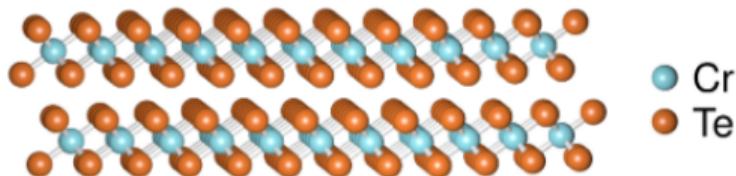
Scanning NV center magnetometry on  
 $\text{CrTe}_2$   
2D ferromagnet at room temperature  
with in-plane magnetization



# Imaging magnetic van der Waals materials

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F. Fabre et al. *Phys. Rev. Mater.* 5 (2021), 034008

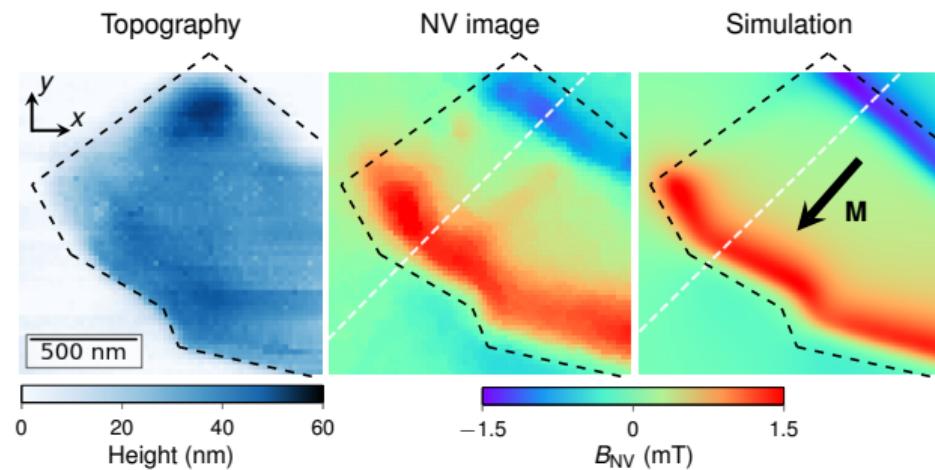
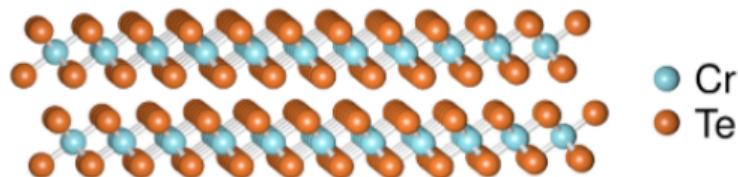
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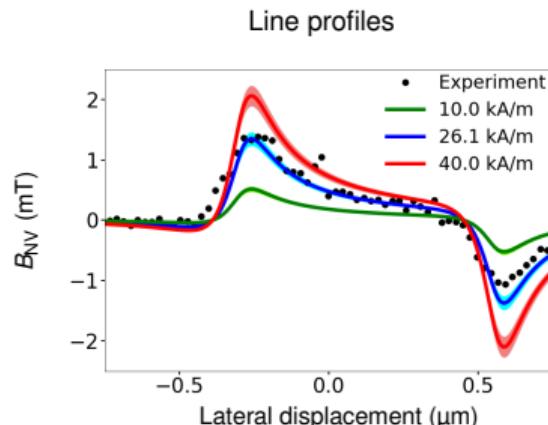
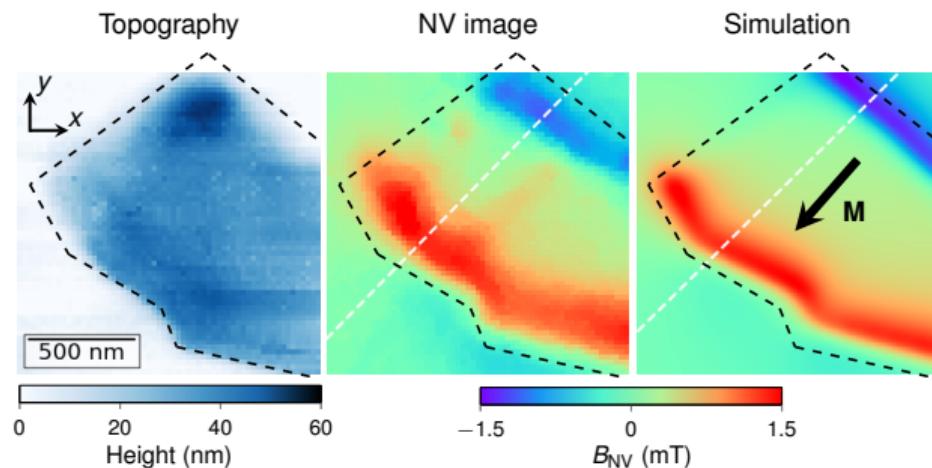
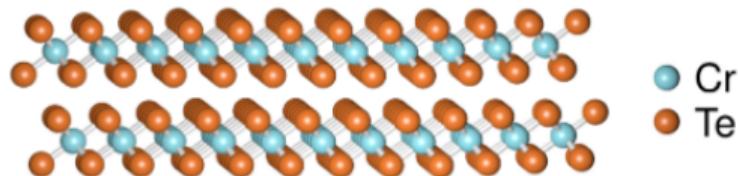


F. Fabre et al. *Phys. Rev. Mater.* 5 (2021), 034008

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Scanning NV center magnetometry on  
**CrTe<sub>2</sub>**  
2D ferromagnet at room temperature  
with in-plane magnetization



CrTe<sub>2</sub> is not stable in air → encapsulation with h-BN

F. Fabre et al. *Phys. Rev. Mater.* 5 (2021), 034008

## Defects in h-BN

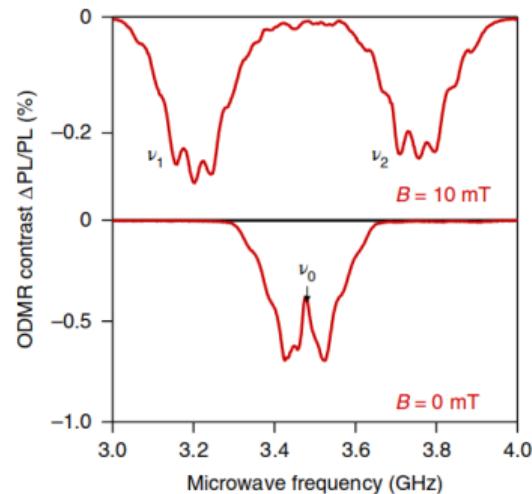
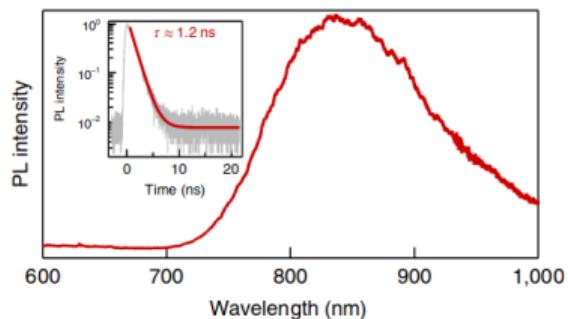
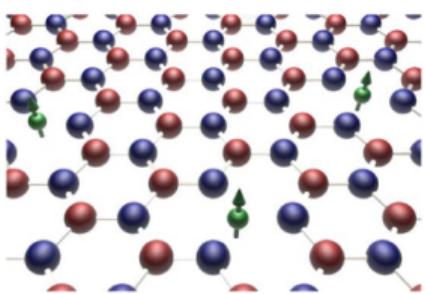
- h-BN is a wide bandgap material (about 6 eV)
- Single photon emitters were known in h-BN

 T. T. Tran et al. *Nature Nanotechnology* 11 (2016), 37

# Defects in h-BN

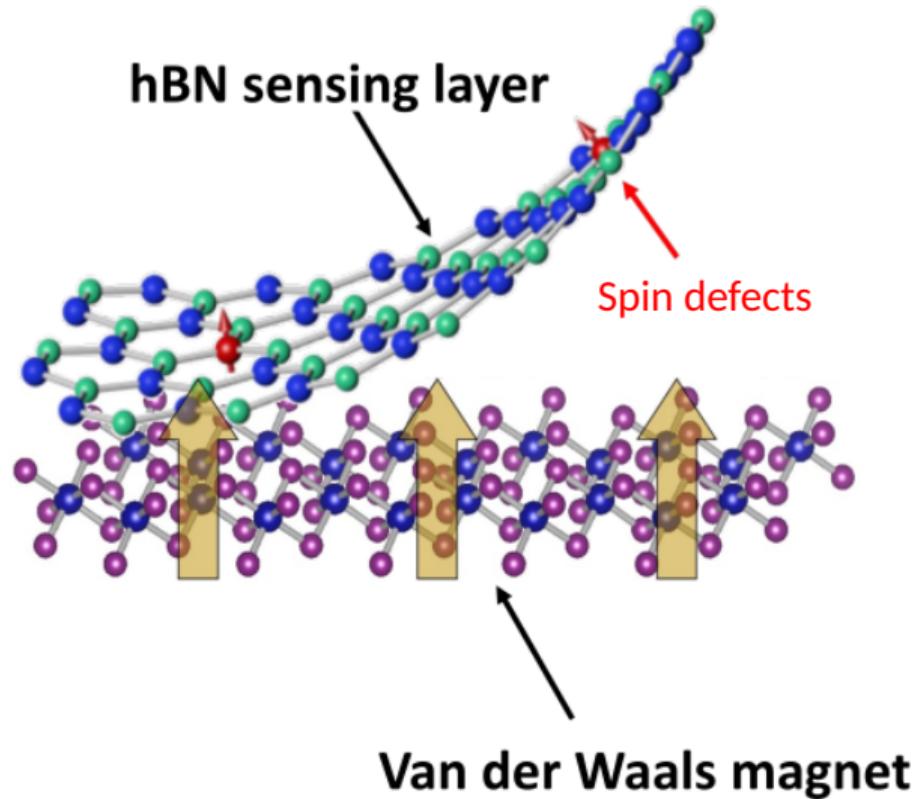
- h-BN is a wide bandgap material (about 6 eV)
- Single photon emitters were known in h-BN
- A **spin defect** was identified in 2020

■ T. T. Tran et al. *Nature Nanotechnology* 11 (2016), 37



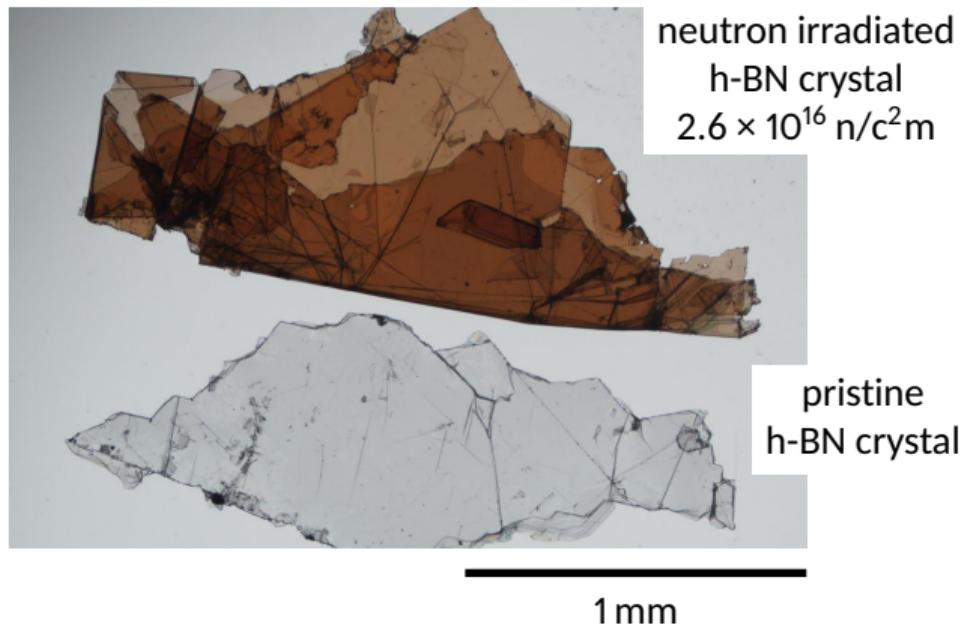
■ A. Gottscholl et al. *Nat. Mater.* 19 (2020), 540

Objective: a quantum sensing foil integrated in the van der Waals heterostructure



# Creating ensembles of boron vacancies in h-BN

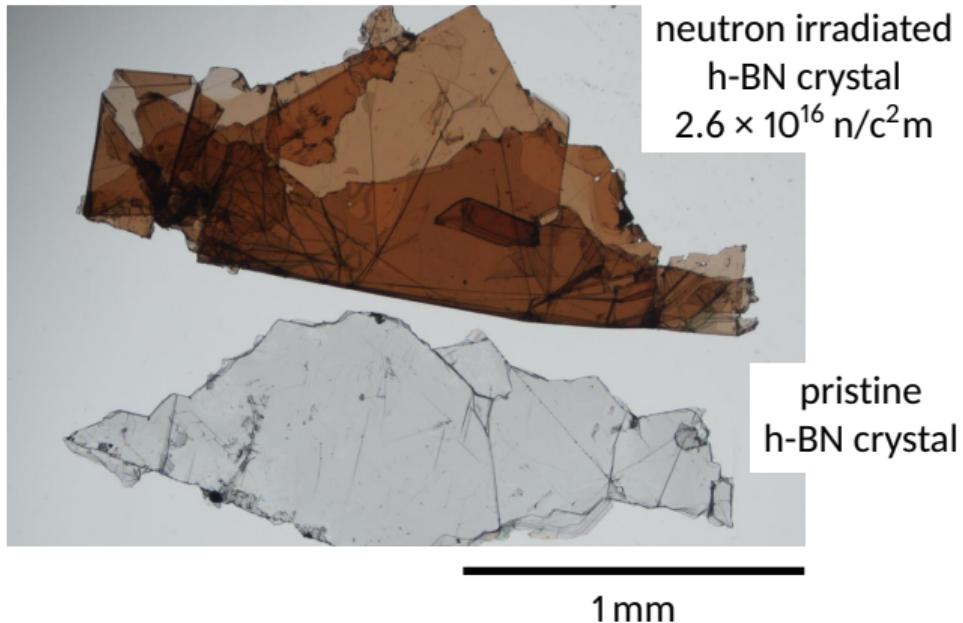
Collaboration: Kansas State University (J. Li, J. Edgar)



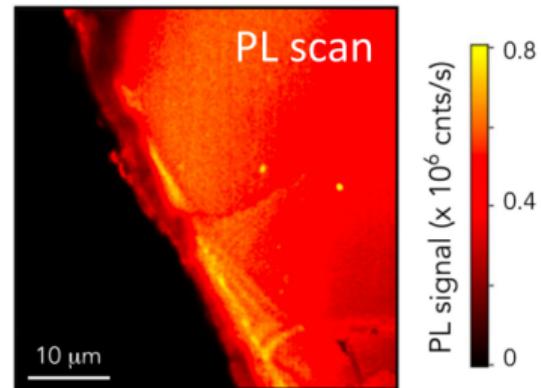
S. Liu et al. *Chem. of Mater.* 30 (2018), 6222

# Creating ensembles of boron vacancies in h-BN

Collaboration: Kansas State University (J. Li, J. Edgar)



- Excitation at 532 nm
- Ambient conditions

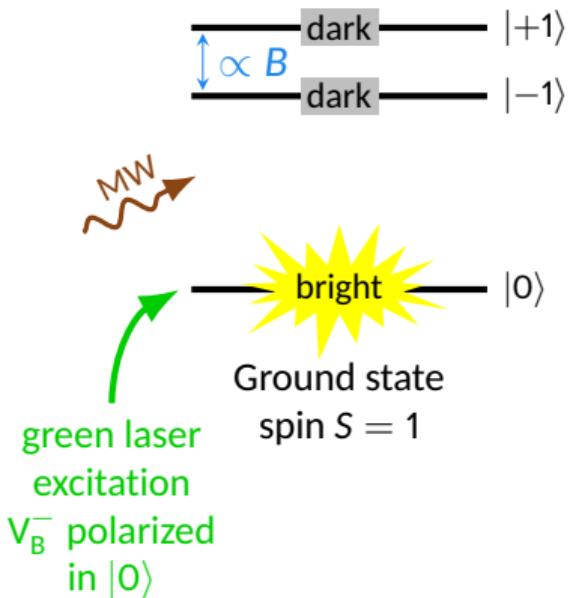


S. Liu et al. *Chem. of Mater.* 30 (2018), 6222

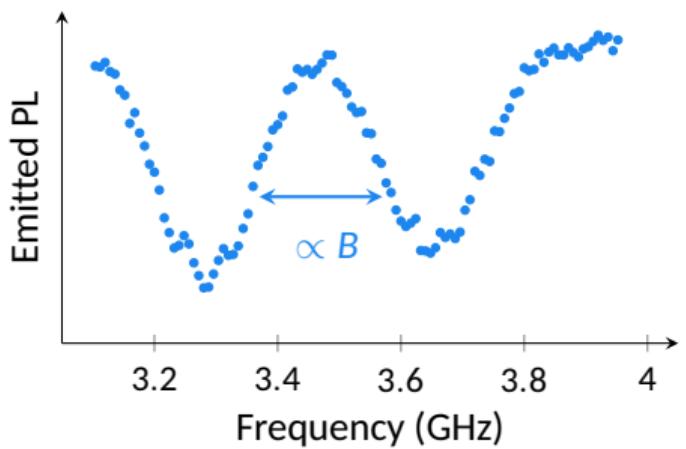
A. Haykal et al. *Nat. Commun.* 13 (2022), 4347

# Measuring magnetic fields with $V_B^-$

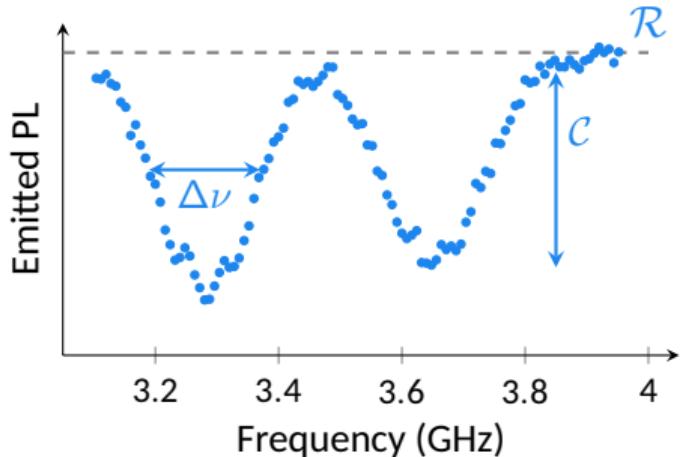
Spin-dependent  
fluorescence



Optically detected magnetic resonance



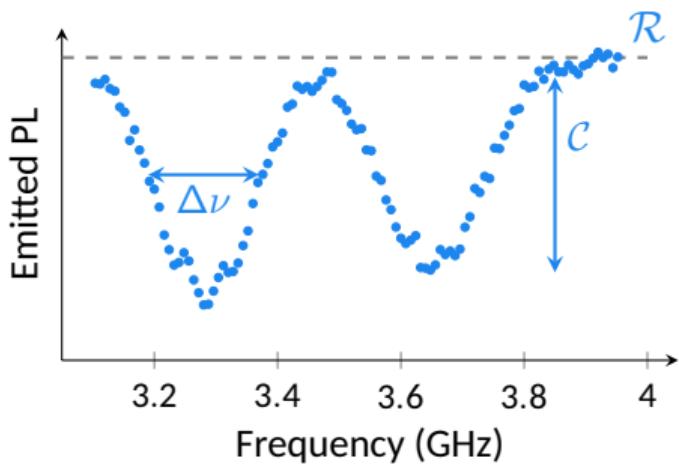
# Magnetic field sensitivity



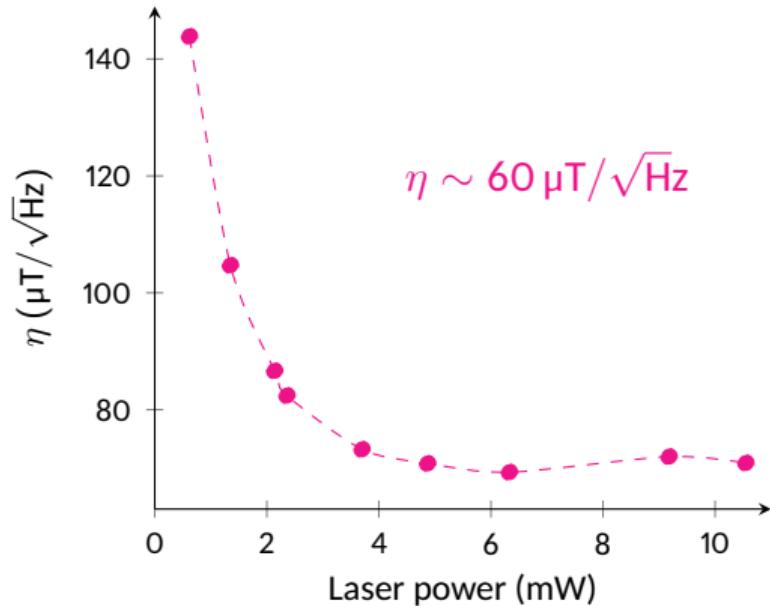
$$\eta \sim 0.7 \frac{1}{\gamma_e} \frac{\Delta\nu}{C\sqrt{R}}$$

P. Kumar et al. *Phys. Rev. Appl.* 18 (2022), L061002

# Magnetic field sensitivity



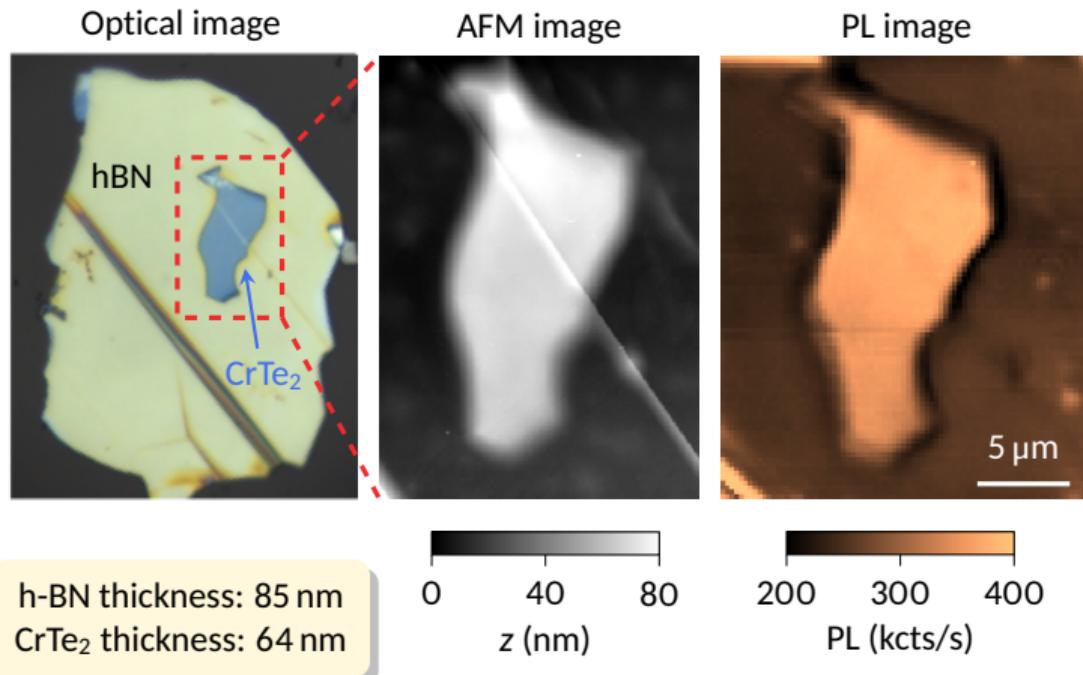
$$\eta \sim 0.7 \frac{1}{\gamma_e} \frac{\Delta\nu}{C\sqrt{R}}$$



P. Kumar et al. *Phys. Rev. Appl.* 18 (2022), L061002

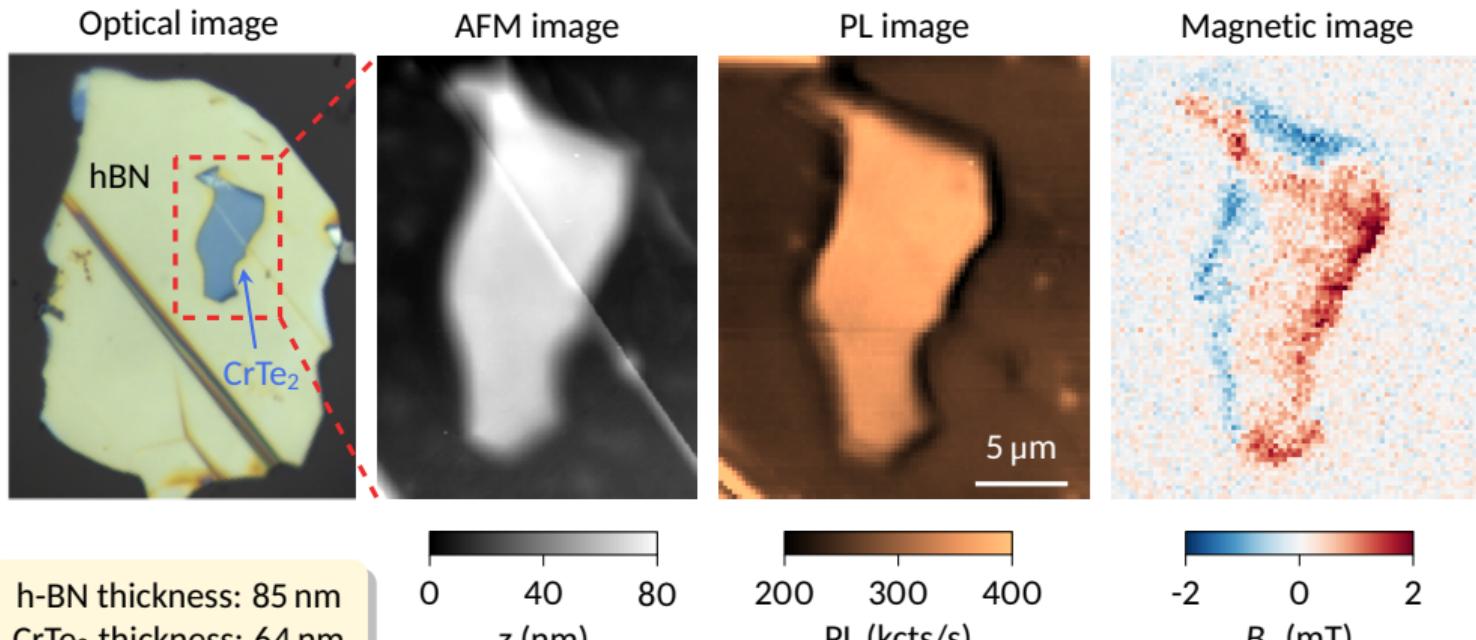
# Imaging a CrTe<sub>2</sub> flake

Collaboration: Institut Néel, Grenoble and LPCNO, Toulouse



# Imaging a CrTe<sub>2</sub> flake

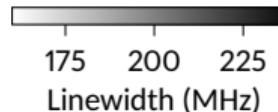
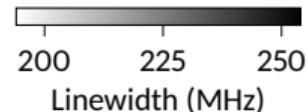
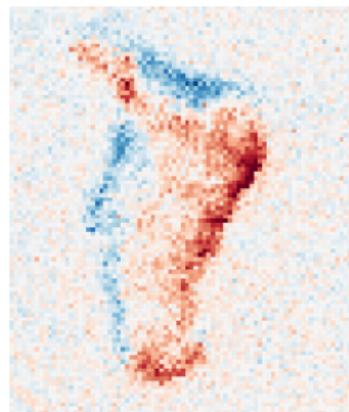
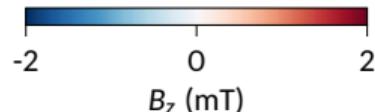
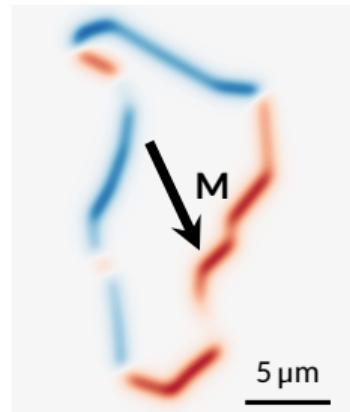
Collaboration: Institut Néel, Grenoble and LPCNO, Toulouse



# Comparison with simulations

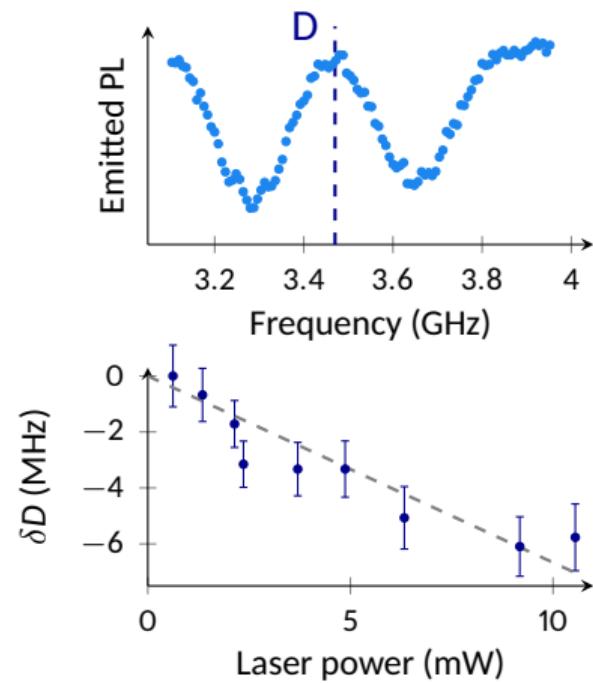
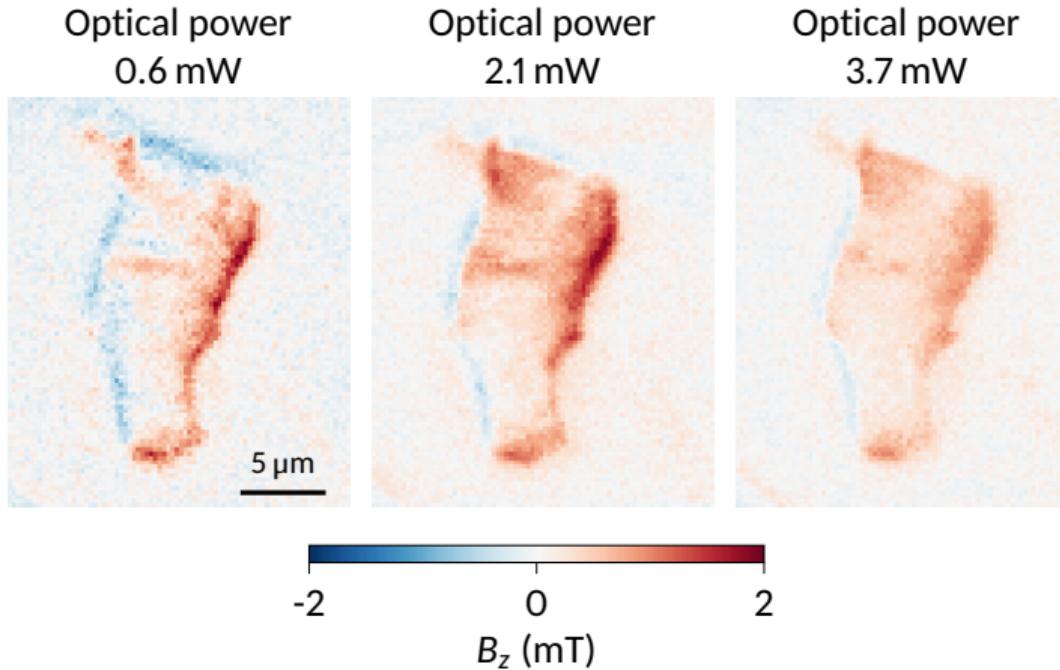
Two averaging procedures are necessary:

- Vertically, over the h-BN film thickness
- Laterally, over the gaussian profile of the laser beam



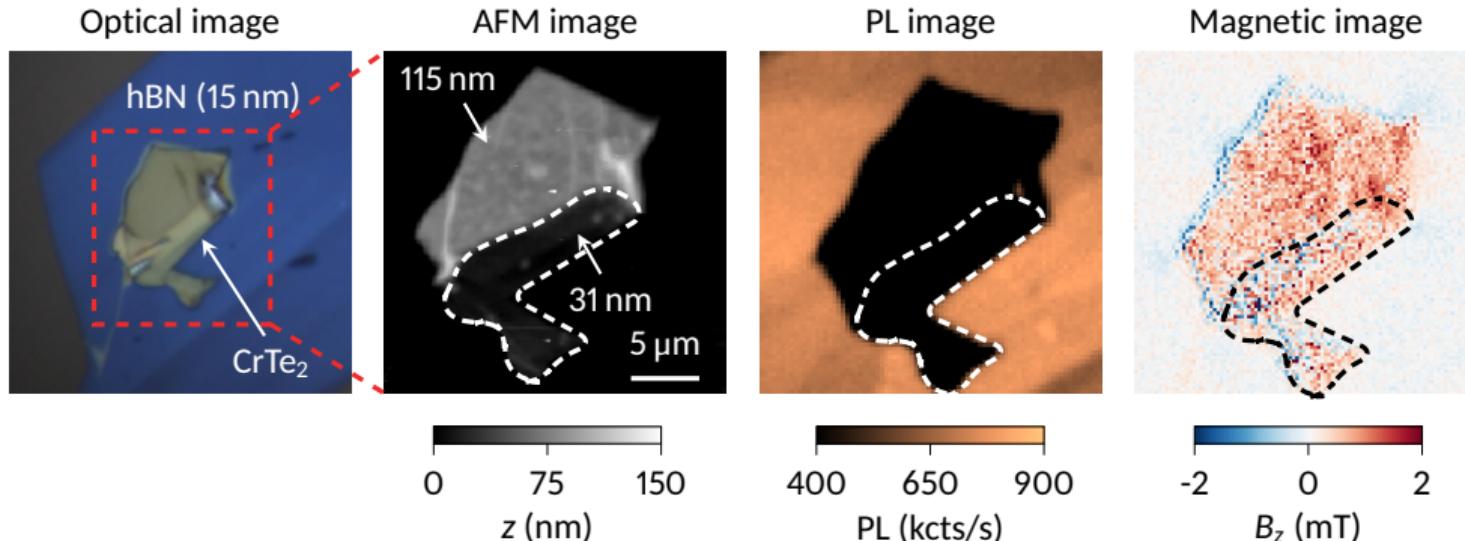
→ Being really quantitative is difficult, using thinner flakes would help!

# Effect of heating



P. Kumar et al. *Phys. Rev. Appl.* 18 (2022), L061002

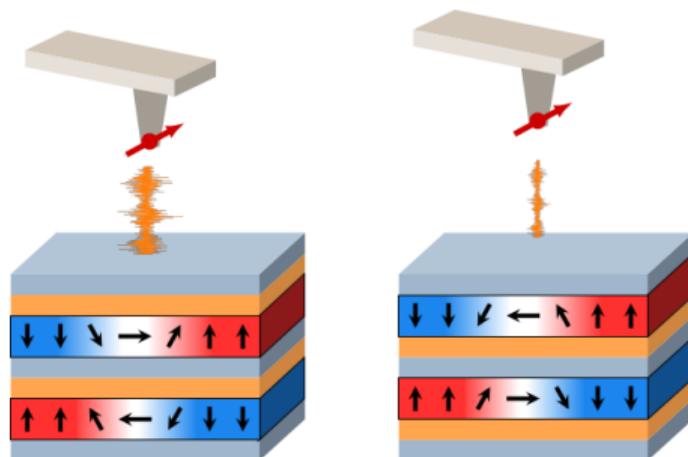
# Using thinner flakes



- PL quenching effect at the metallic surface of CrTe<sub>2</sub>
- Need for larger laser excitation power
- Heating of the magnetic material, crossing  $T_c$

# Summary

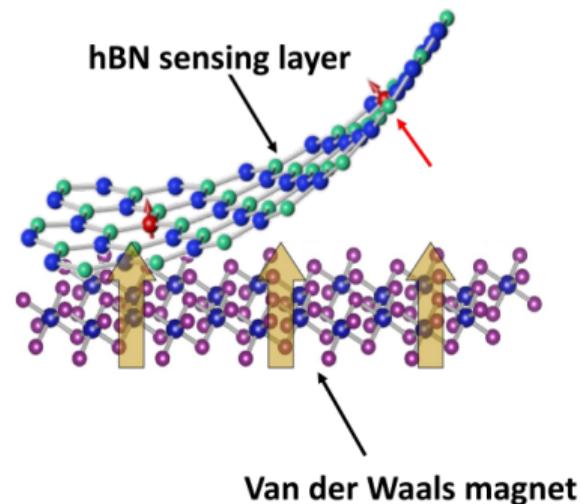
Localization and characterization of magnetic textures for thermal spin wave noise using scanning NV center microscopy



■ A. Finco et al. *Nat. Commun.* 12 (2021), 767

■ A. Finco et al. *in preparation* (2024)

Imaging van der Waals magnets with boron vacancies in hBN, integrating the sensor inside the heterostructure



■ P. Kumar et al. *Phys. Rev. Appl.* 18 (2022), L061002

■ A. J. Healey et al. *Nat. Phys.* 19 (2023), 87

■ M. Huang et al. *Nat. Commun.* 13 (2022), 5369

# Acknowledgements

## L2C, Montpellier

Pawan Kumar, Maxime Rollo, Angela Haykal, Rana Tanos,  
Florentin Fabre, Isabelle Robert-Philip, Vincent Jacques

## LAF, Palaiseau

William Legrand, Karim Bouzehouane, Fernando Ajejas,  
Nicolas Reyren, Vincent Cros

## C2N, Palaiseau

Thibaut Devolder, Jean-Paul Adam, Joo-Von Kim

## Spintec, Grenoble

Van-Tuong Pham, Joseba Urrestarazu Larranaga, Olivier Boulle

## Kansas State University

Jiahua Li, James Edgar

## Institut Néel, Grenoble

Johann Coraux, Nicolas Rougemaille

## LPCNO, Toulouse

Cédric Robert, Jules Fraunié, Pierre Renucci, Xavier Marie



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