

Imaging antiferromagnetic states with scanning NV-center magnetometry

Aurore Finco, Angela Haykal, Florentin Fabre, Saddem Chouaieb,
Rana Tanos, Waseem Akhtar, Vincent Jacques

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



Hamburg, 24/09/2019

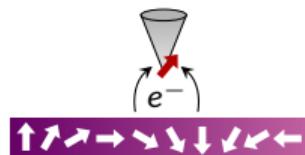
slides available at <https://magimag.eu>

How to probe non-collinear magnetism locally?



Probe directly the
sample **magnetization**

SP-STM



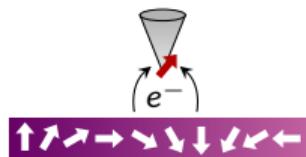
 R. Wiesendanger. *Rev. Mod. Phys.* 81 (2009), 1495–1550

How to probe non-collinear magnetism locally?



Probe directly the sample **magnetization**

SP-STM

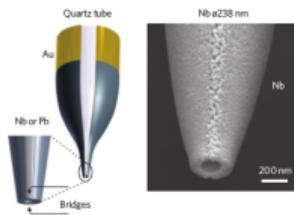


R. Wiesendanger. *Rev. Mod. Phys.* 81 (2009), 1495–1550



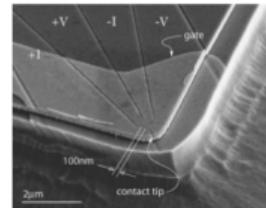
Probe the generated **stray field**

nano SQUIDs



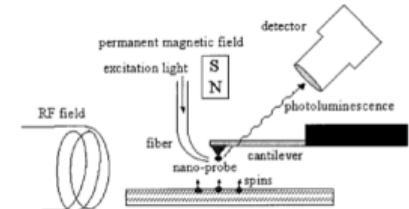
D. Vasyukov et al. *Nat. Nano.* 8 (2013), 639–644

Hall bars



C. W. Hicks et al. *Appl. Phys. Lett.* 90 (2007), 133512

Single spin



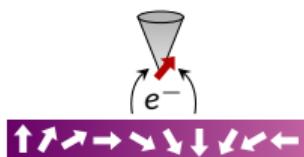
B. M. Chernobrod et al. *J. Appl. Phys.* 97 (2004), 014903

How to probe non-collinear magnetism locally?



Probe directly the sample **magnetization**

SP-STM

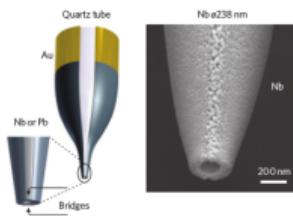


R. Wiesendanger. *Rev. Mod. Phys.* 81 (2009), 1495–1550



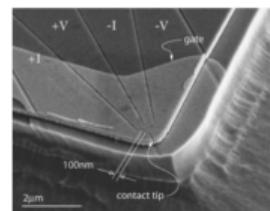
Probe the generated **stray field**

nano SQUIDs

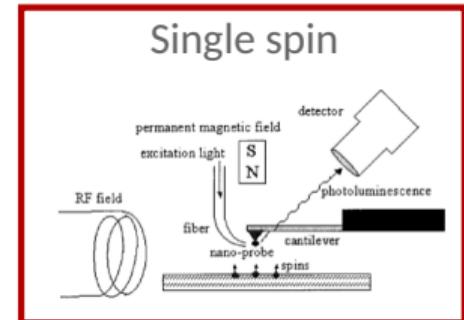


D. Vasyukov et al. *Nat. Nano.* 8 (2013), 639–644

Hall bars

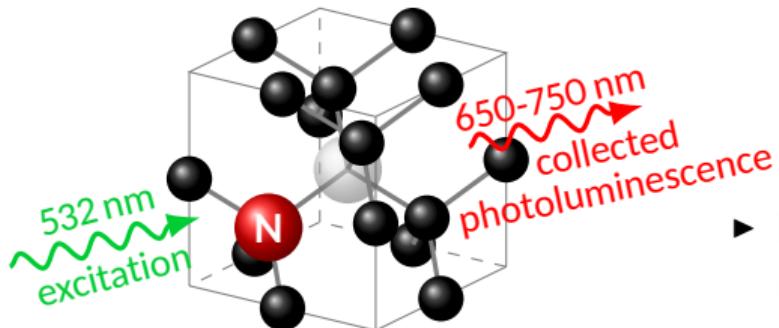


C. W. Hicks et al. *Appl. Phys. Lett.* 90 (2007), 133512



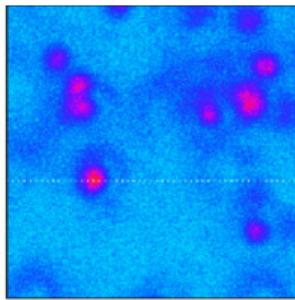
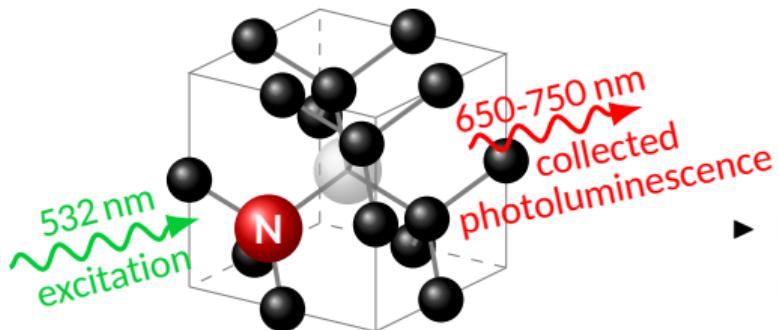
B. M. Chernobrod et al. *J. Appl. Phys.* 97 (2004), 014903

The Nitrogen Vacancy center in diamond



- ▶ Defect in the diamond lattice, a nitrogen atom next to a vacancy
- ▶ Artificial atom: discrete energy levels inside the diamond gap

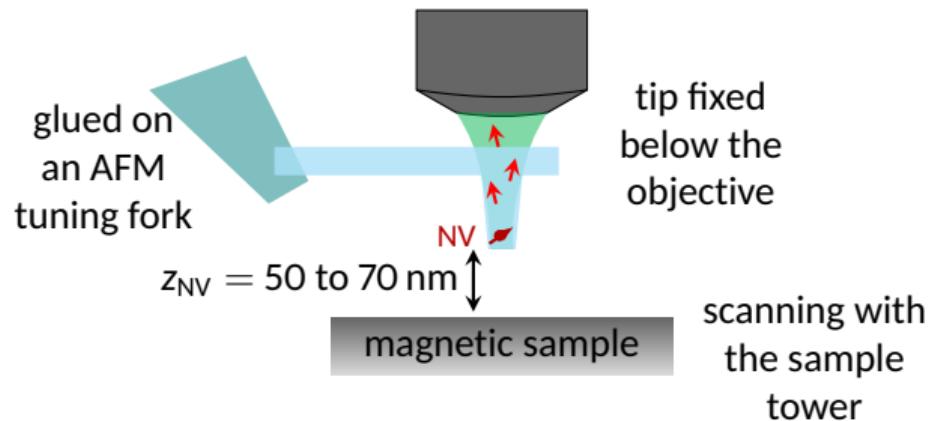
The Nitrogen Vacancy center in diamond



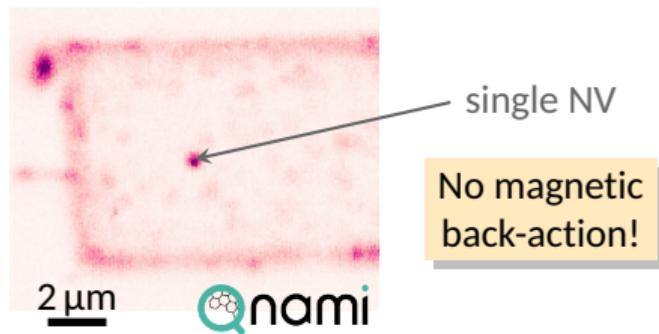
- ▶ Defect in the diamond lattice, a nitrogen atom next to a vacancy
- ▶ Artificial atom: discrete energy levels inside the diamond gap
- ▶ Stable photoluminescence at room temperature
- ▶ Single NV defects can be detected with a confocal microscope
- ▶ **Quantum magnetic field sensor**

■ A. Gruber *et al.* Science 276 (1997), 2012–2014

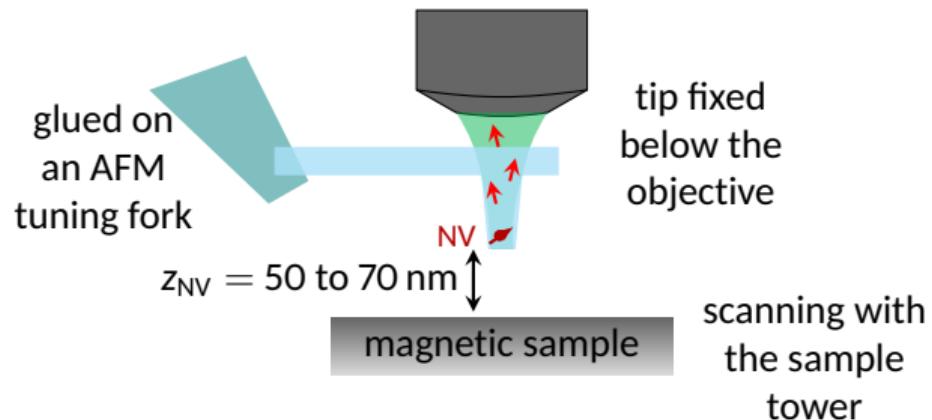
Scanning NV magnetometry



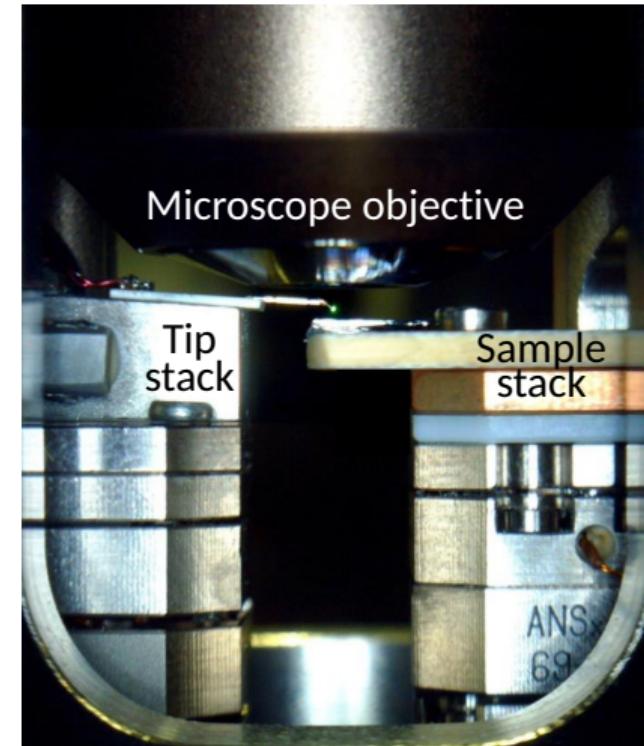
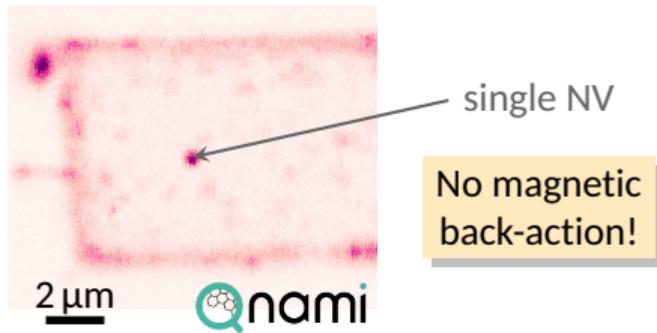
Photoluminescence scan, top view of the tip



Scanning NV magnetometry

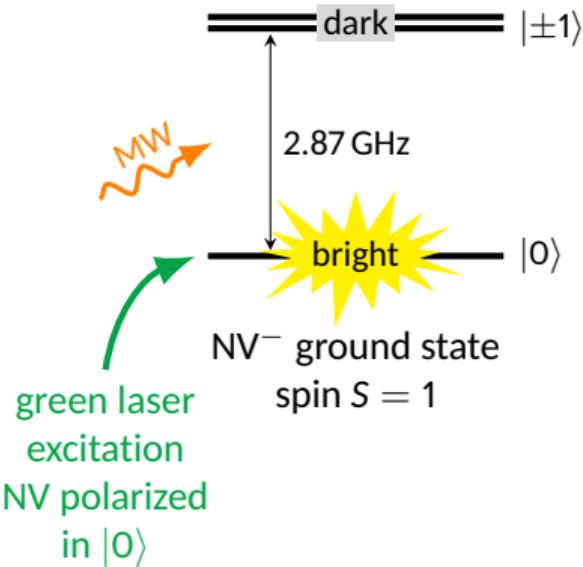


Photoluminescence scan, top view of the tip



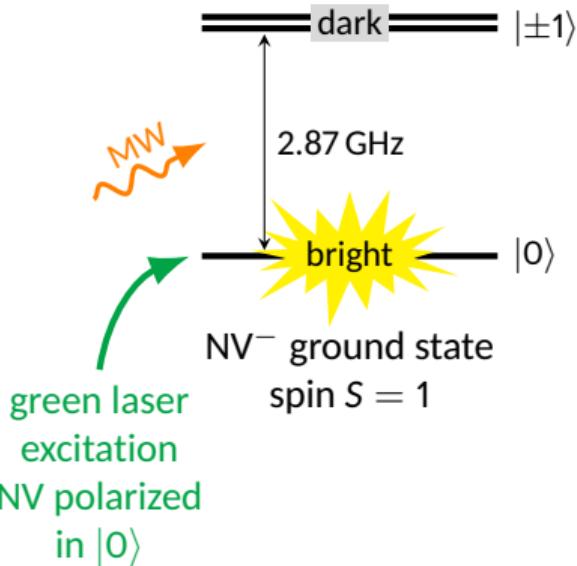
Quantitative field measurements

Spin-dependent fluorescence

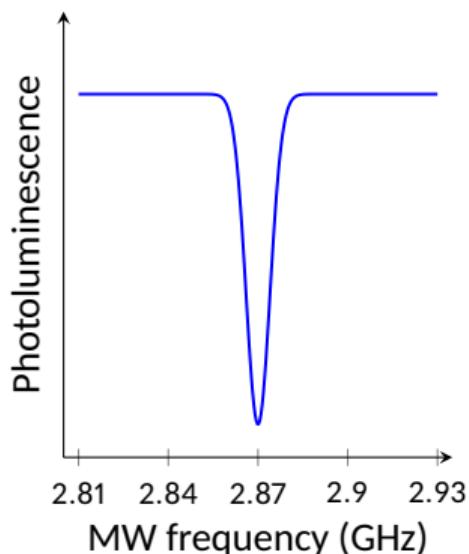


Quantitative field measurements

Spin-dependent fluorescence

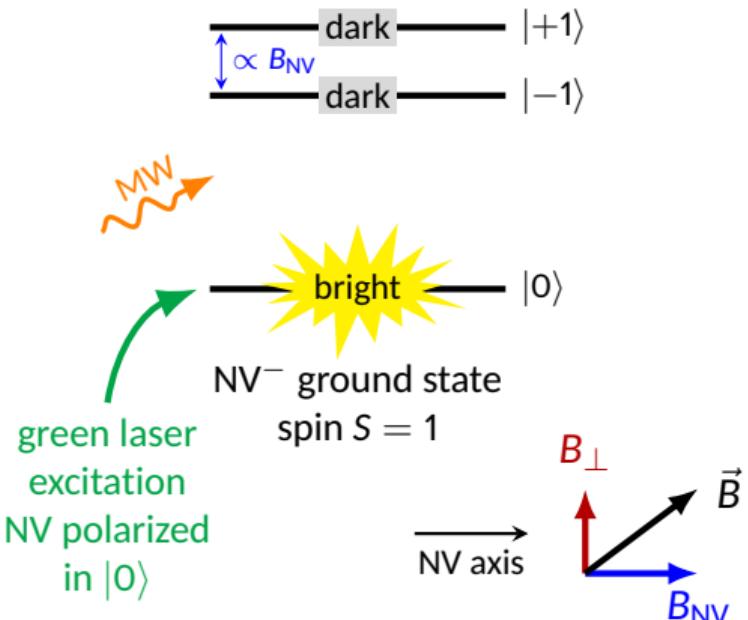


Optically Detected Magnetic Resonance

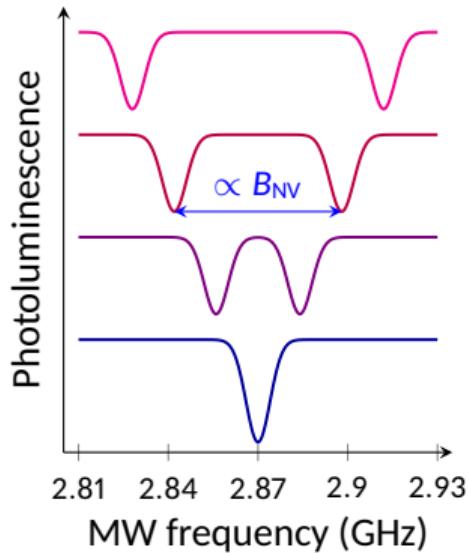


Quantitative field measurements

Spin-dependent fluorescence



Optically Detected Magnetic Resonance



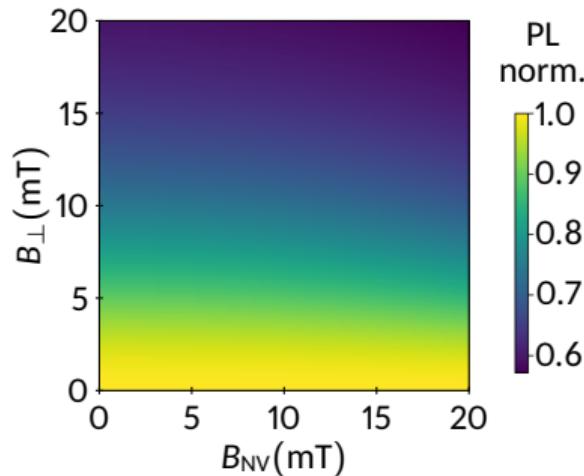
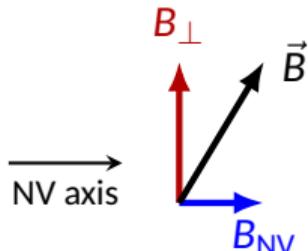
Quenching of the photoluminescence at high field

Mixing of the spin states

dark $|\psi_2\rangle$

dark $|\psi_1\rangle$

dark $|\psi_0\rangle$



J.-P. Tetienne *et al.* New J. Phys. 14 (2012), 103033

Two measurement modes

Quantitative mode

Low field regime ($B_{\perp} < 5 \text{ mT}$)

- ▶ Gives access to the precise value of the stray field along the NV axis,
sensitivity $1 \mu\text{T Hz}^{-1/2}$
- ▶ Need to measure a spectrum at each pixel to localize the resonance
- ▶ Requires a microwave excitation
- ▶ Slow, sensitive to drift

Investigation of antiferromagnets

Two measurement modes

Quantitative mode

Low field regime ($B_{\perp} < 5 \text{ mT}$)

- ▶ Gives access to the precise value of the stray field along the NV axis, sensitivity $1 \mu\text{T Hz}^{-1/2}$
- ▶ Need to measure a spectrum at each pixel to localize the resonance
- ▶ Requires a microwave excitation
- ▶ Slow, sensitive to drift

Investigation of antiferromagnets

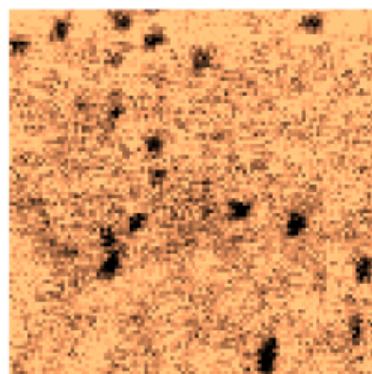
Qualitative mode

High field regime ($B_{\perp} > 5 \text{ mT}$)

- ▶ Localize the areas producing a large stray field
- ▶ Only need to record the photoluminescence at each pixel
- ▶ No microwave excitation required
- ▶ Strength of the measured field unknown

Study of ferromagnets

Zero-field skyrmions in exchange-biased magnetic layers

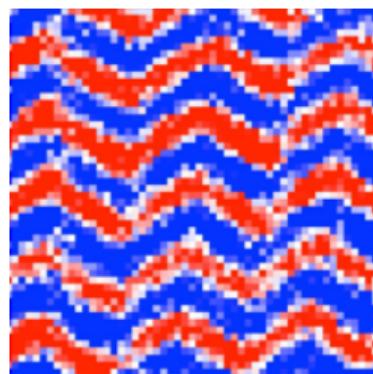


Collaboration



O. Boulle group, Grenoble

Influence of epitaxial strain on the cycloid in the multiferroic BiFeO₃

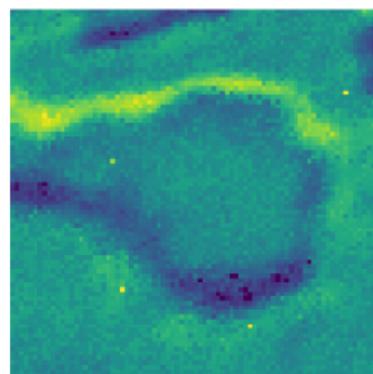


Collaboration



S. Fusil and V. Garcia group
M. Viret group, Palaiseau

Detection of domain wall magnons in a synthetic antiferromagnet

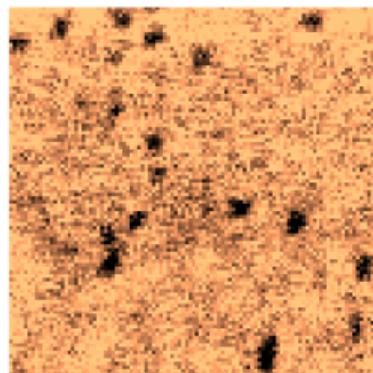


Collaboration



V. Cros group
J.-V. Kim, Palaiseau

Zero-field skyrmions in exchange-biased magnetic layers

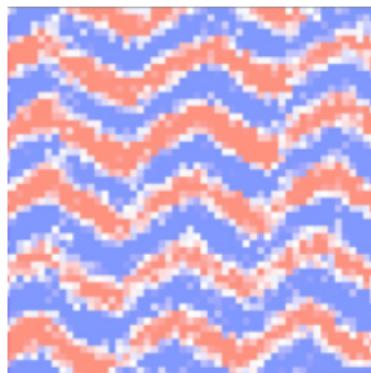


Collaboration



O. Boulle group, Grenoble

Influence of epitaxial strain on the cycloid in the multiferroic BiFeO₃

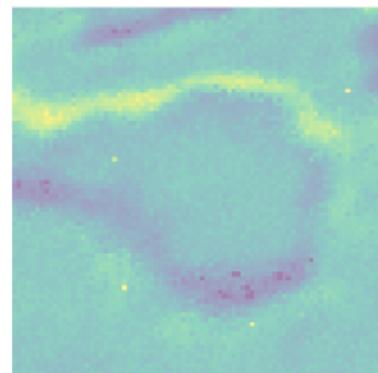


Collaboration



S. Fusil and V. Garcia group
M. Viret group, Palaiseau

Detection of domain wall magnons in a synthetic antiferromagnet



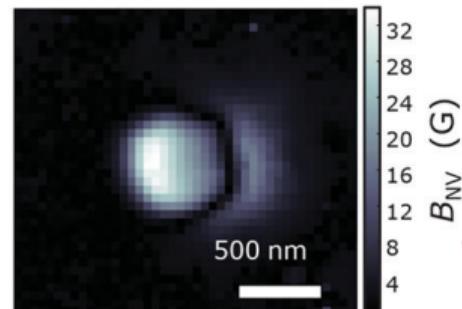
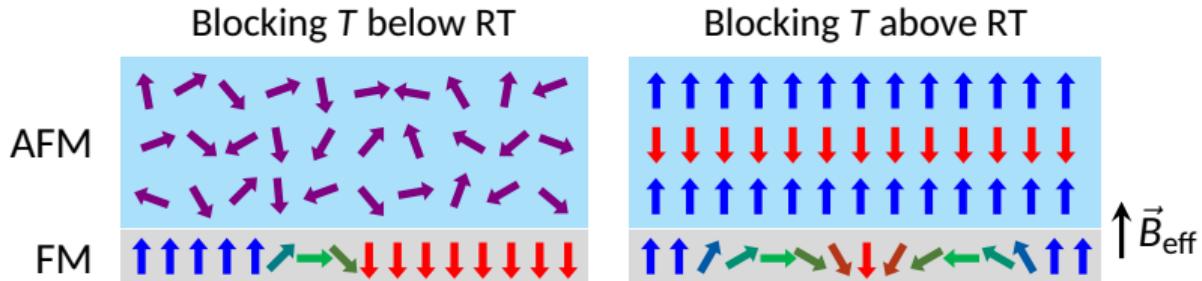
Collaboration



V. Cros group
J.-V. Kim, Palaiseau

Use exchange bias as an effective field

Goal: stable zero-field skyrmions at room temperature without confinement

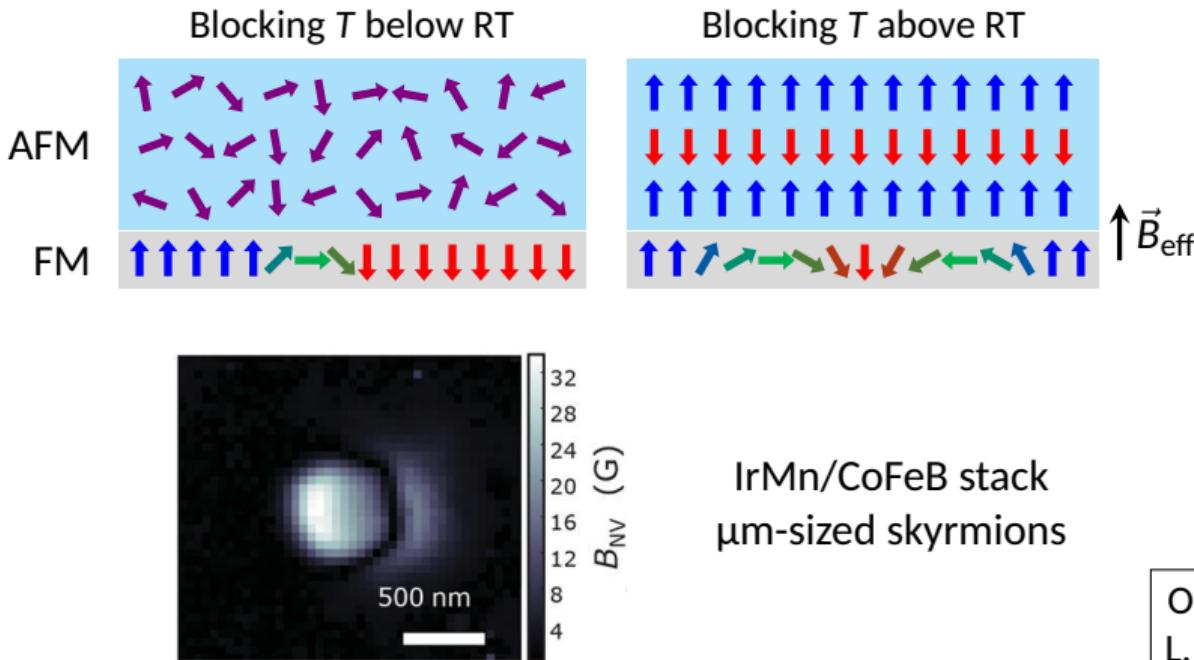


IrMn/CoFeB stack
 μm -sized skyrmions

■ G. Yu *et al.* *Nano Lett.* 18 (2018), 980–986

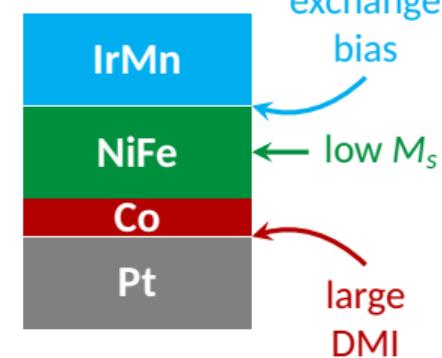
Use exchange bias as an effective field

Goal: stable zero-field skyrmions at room temperature without confinement



G. Yu et al. Nano Lett. 18 (2018), 980–986

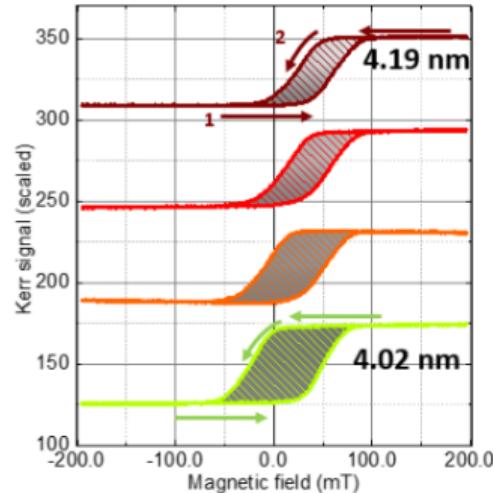
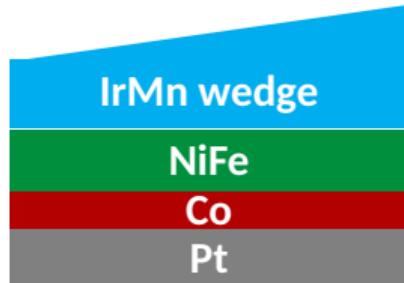
Strategy to reduce
the skyrmion size



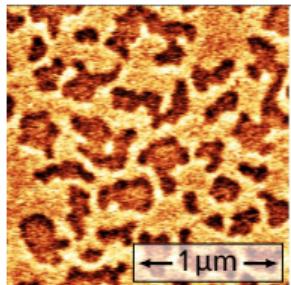
O. Boulle, G. Rana
L. Buda-Prejbeanu



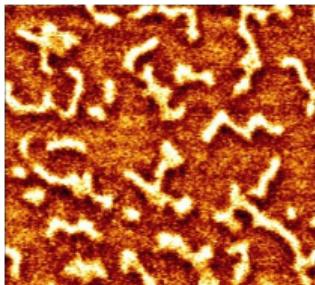
Optimization of the sample parameters



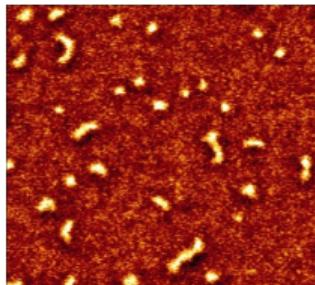
$$t_{\text{IrMn}} = 4.02 \text{ nm}$$



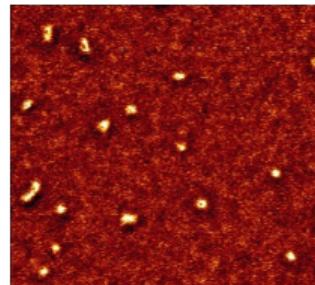
$$t_{\text{IrMn}} = 4.11 \text{ nm}$$



$$t_{\text{IrMn}} = 4.15 \text{ nm}$$

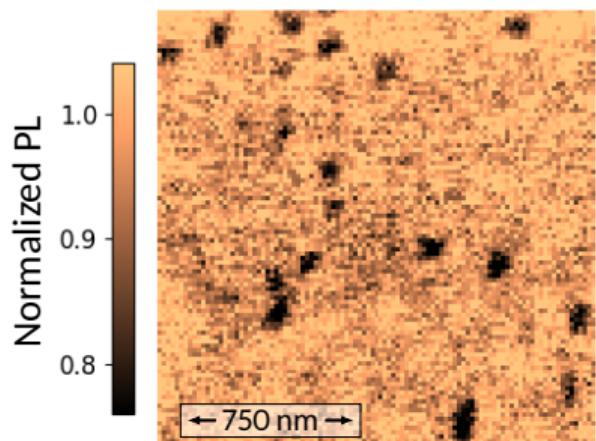


$$t_{\text{IrMn}} = 4.19 \text{ nm}$$

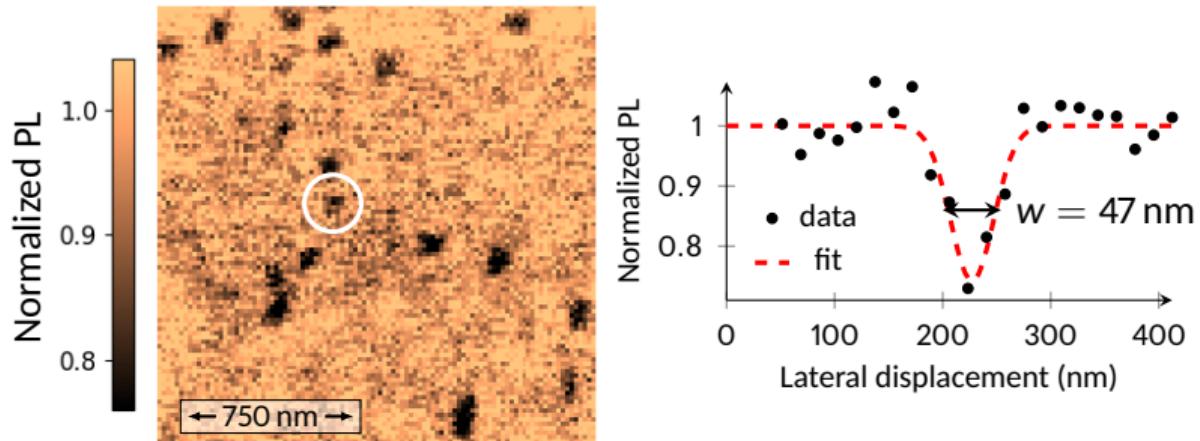


MFM
images

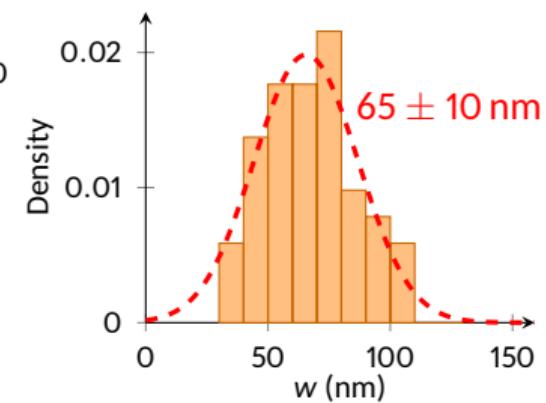
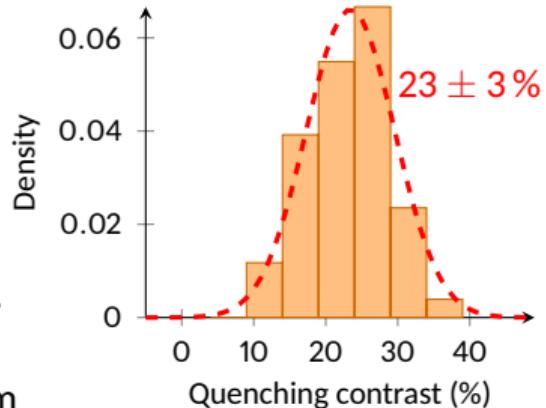
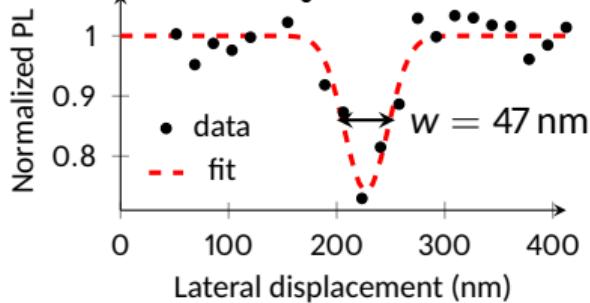
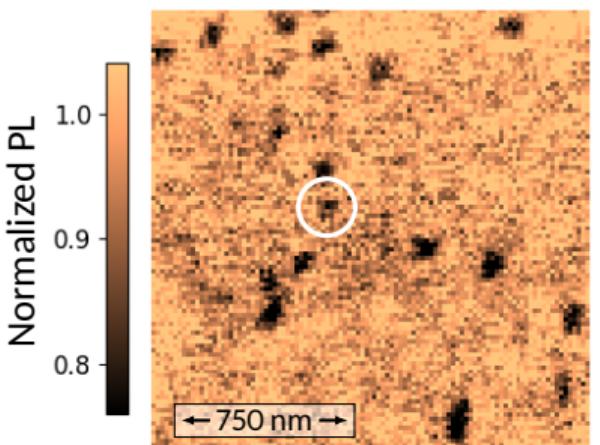
Magnetic skyrmions in qualitative “quenching” mode



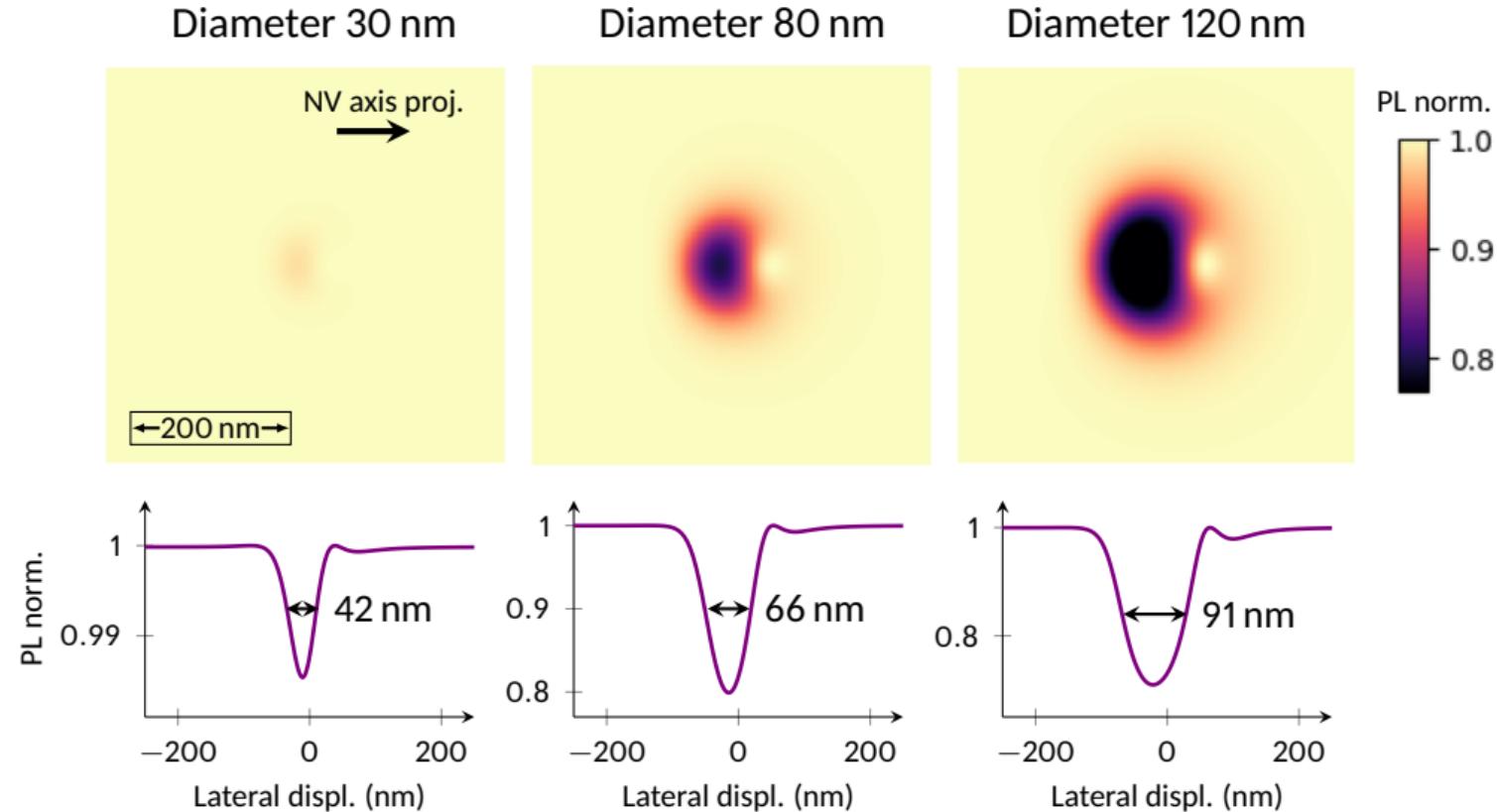
Magnetic skyrmions in qualitative “quenching” mode



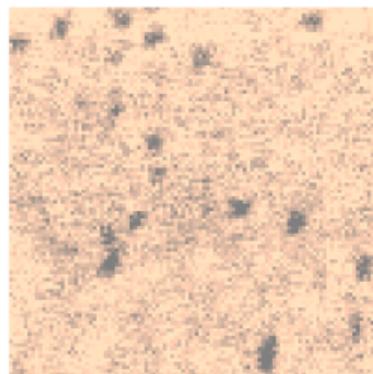
Magnetic skyrmions in qualitative “quenching” mode



Comparison with simulations



Zero-field skyrmions in exchange-biased magnetic layers

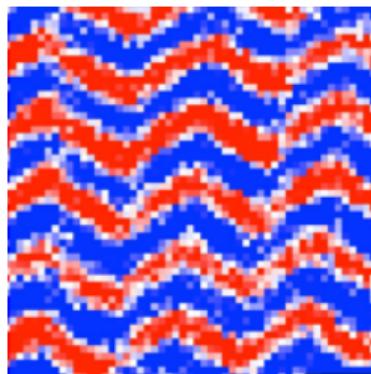


Collaboration



O. Boulle group, Grenoble

Influence of epitaxial strain on the cycloid in the multiferroic BiFeO₃

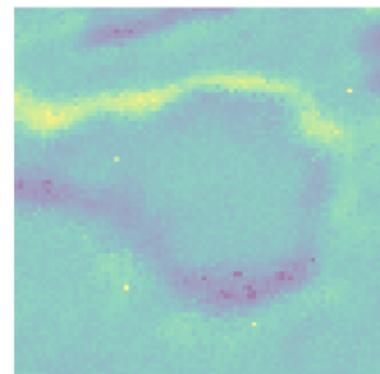


Collaboration



S. Fusil and V. Garcia group
M. Viret group, Palaiseau

Detection of domain wall magnons in a synthetic antiferromagnet



Collaboration



V. Cros group
J.-V. Kim, Palaiseau

How to control the spin state of antiferromagnets?

With magnetic field? No, antiferromagnets are insensitive to external fields!



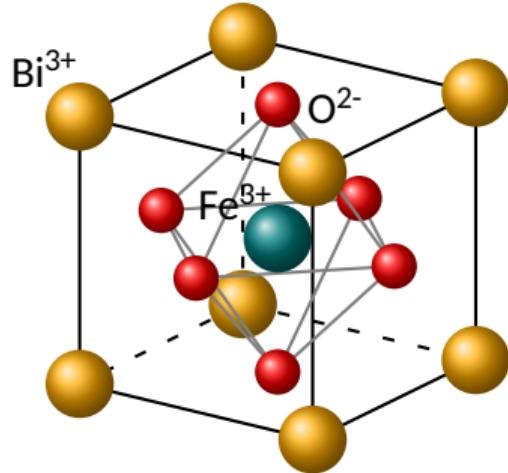
P. Wadley et al. *Science* 351 (2016), 587–590

H. Yan et al. *Nat. Nano.* 14 (2019), 131

Here: apply strain on a multiferroic to combine strain and magnetoelectric effects

BiFeO_3 , a room temperature multiferroic

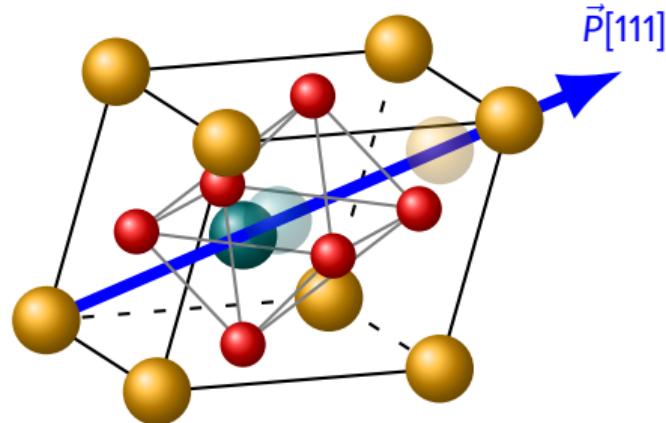
Electric polarization



Paraelectric phase ($T > 1100 \text{ K}$)

BiFeO_3 , a room temperature multiferroic

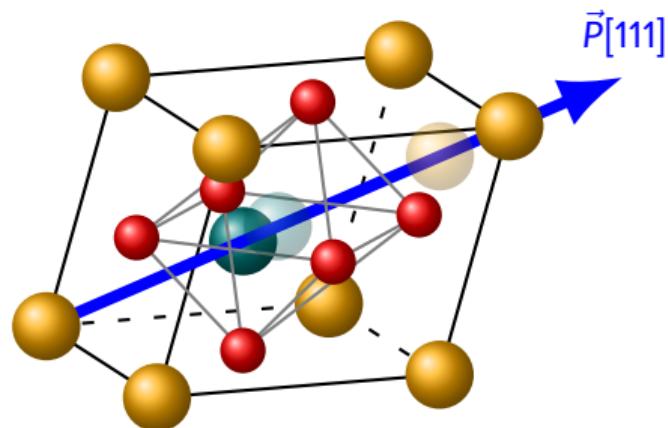
Electric polarization



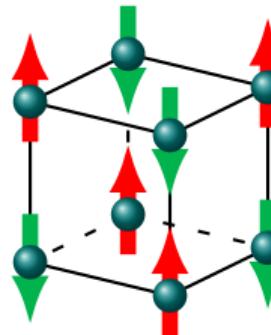
Ferroelectric phase ($T < 1100 \text{ K}$)

BiFeO_3 , a room temperature multiferroic

Electric polarization



Magnetism

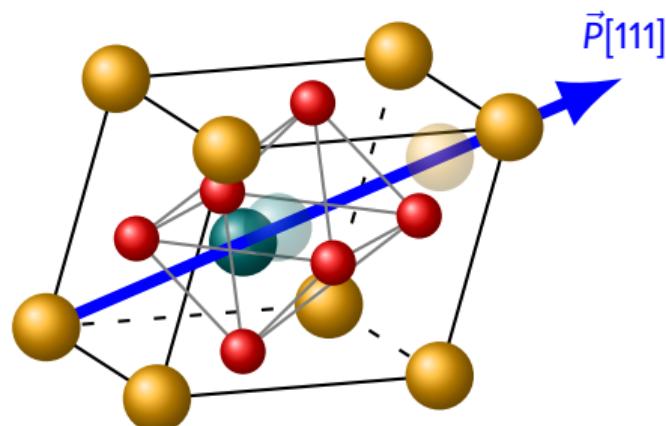


G-type
antiferromagnet

Ferroelectric phase ($T < 1100 \text{ K}$)

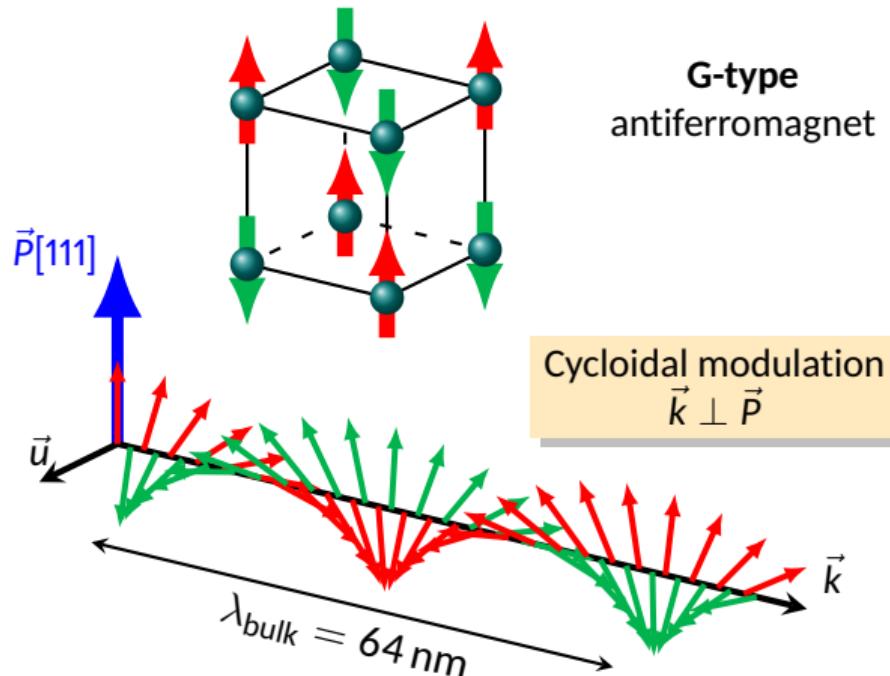
BiFeO_3 , a room temperature multiferroic

Electric polarization

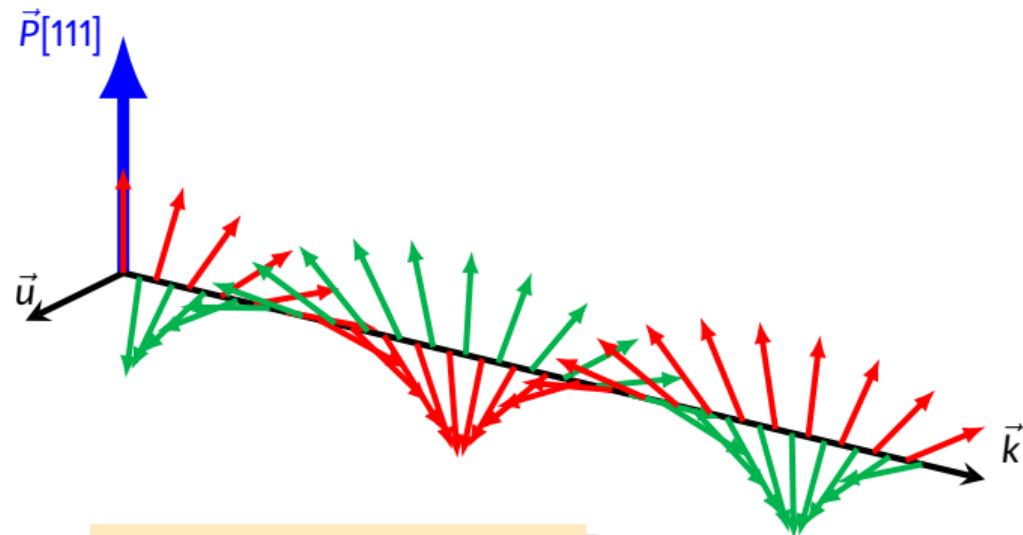


Ferroelectric phase ($T < 1100 \text{ K}$)

Magnetism

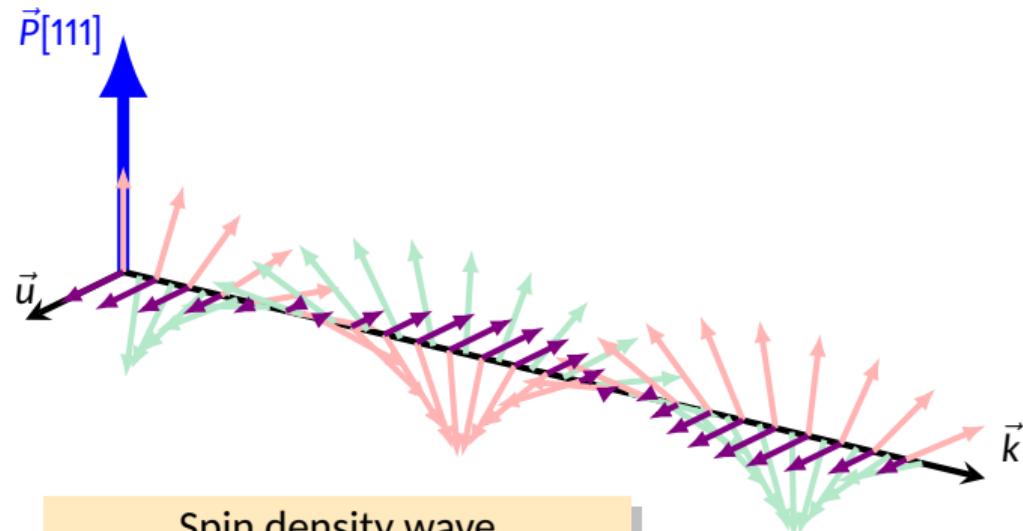


Origin of the stray field: spin density wave



Fully compensated cycloid
→ No stray field!

Origin of the stray field: spin density wave

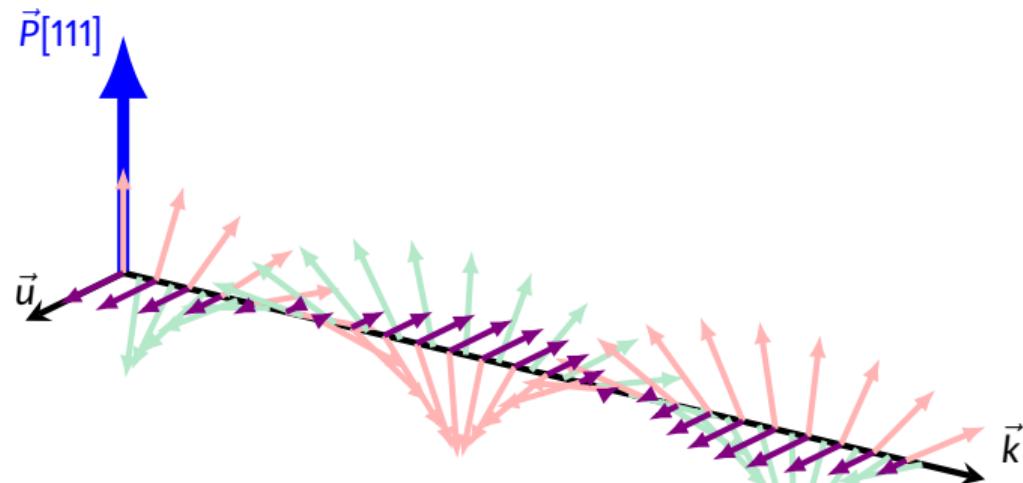


Spin density wave
Weak uncompensated moment
→ **Small stray field**

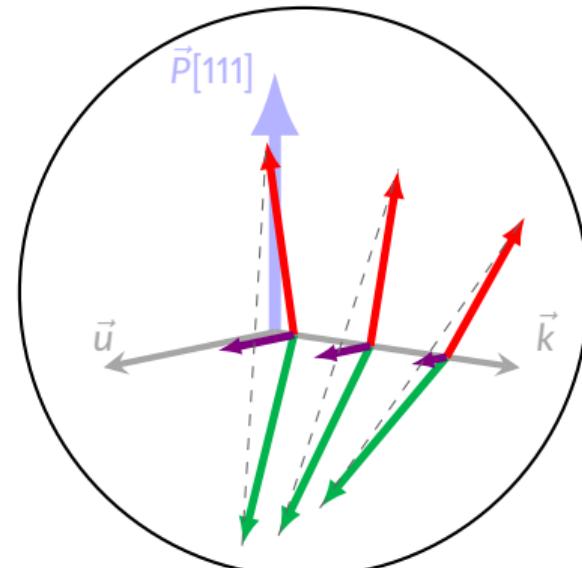


M. Ramazanoglu et al. Phys. Rev. Lett. 107 (2011), 207206

Origin of the stray field: spin density wave



Spin density wave
Weak uncompensated moment
→ **Small stray field**



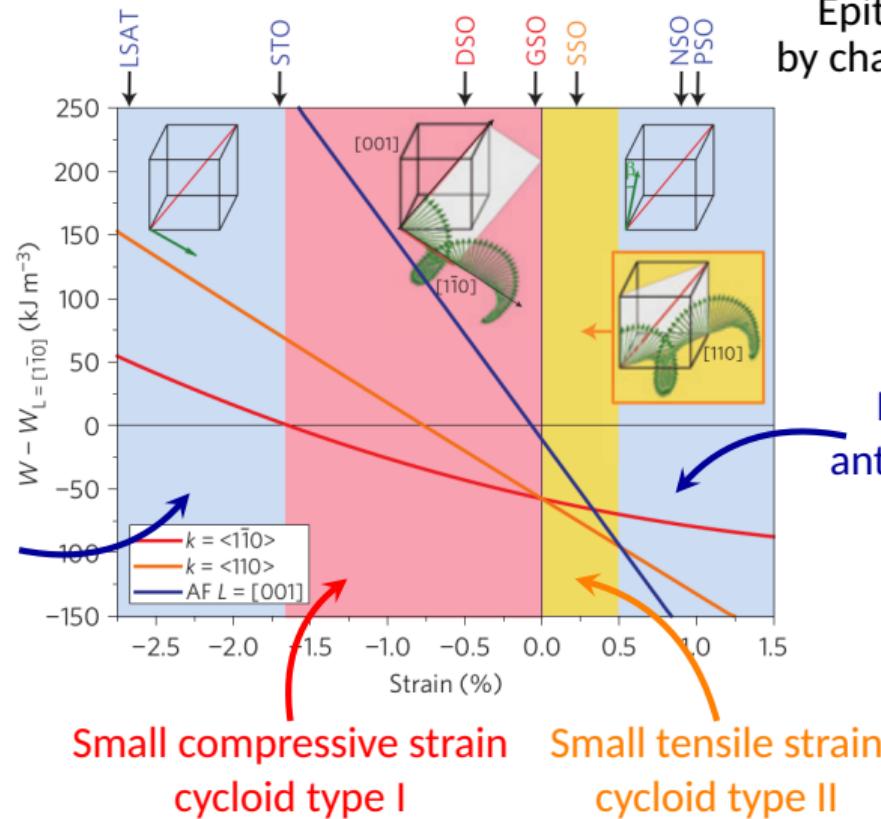
■ M. Ramazanoglu et al. Phys. Rev. Lett. 107 (2011), 207206

Known effect of epitaxial strain on the cycloid

Phase diagram obtained from spectroscopic measurements

Epitaxial strain tuned by changing the substrate

Large compressive strain antiferromagnetic order

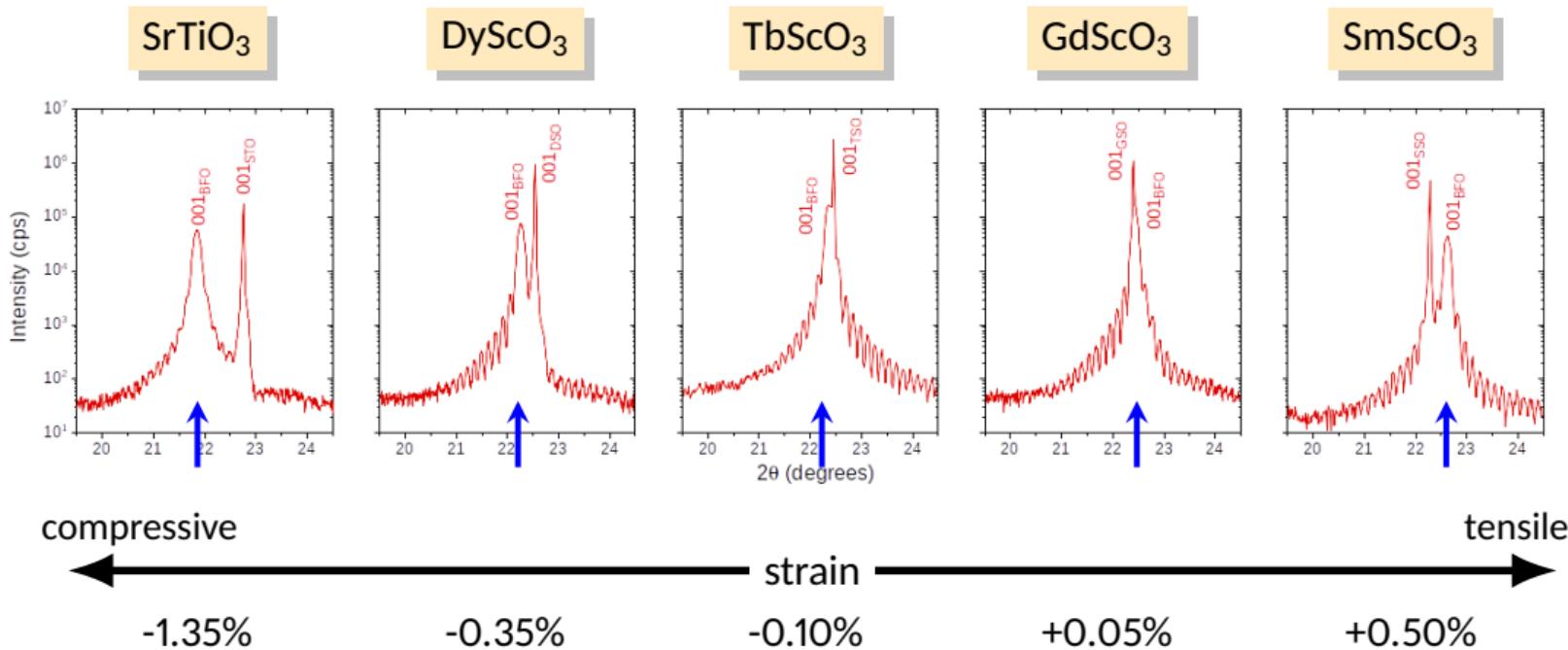


Tuning of epitaxial strain

BiFeO₃ thin film samples grown on various substrates

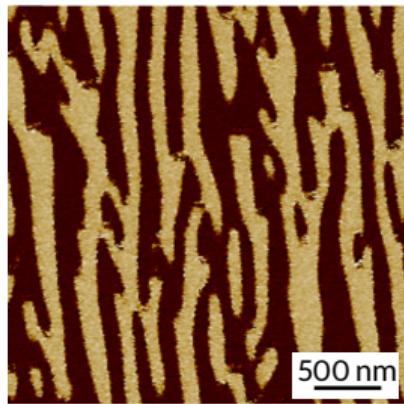


Samples from UMR CNRS/Thales
J. Fischer, V. Garcia, S. Fusil

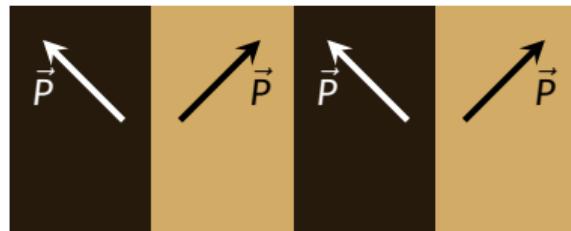


NV imaging of the cycloid, iso-B mode

DyScO₃, strain -0.35%

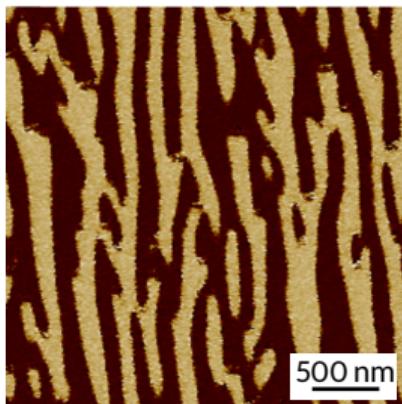


PFM image
ferroelectric domains

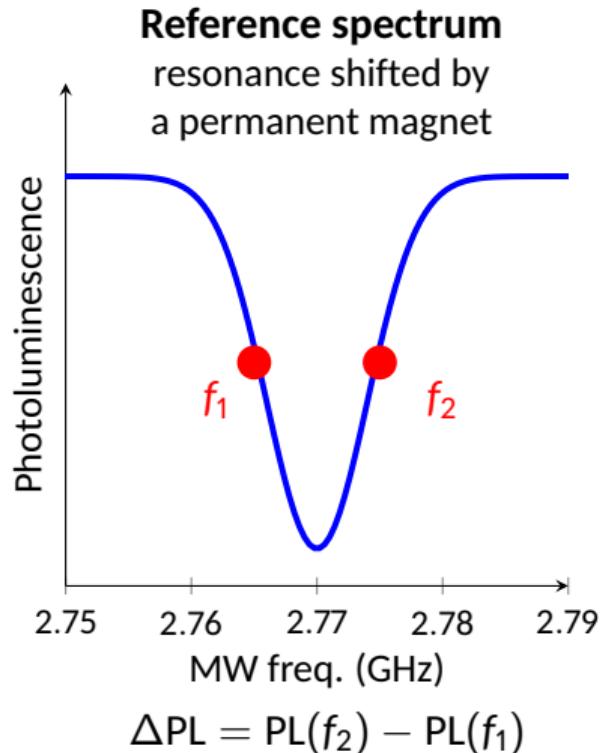
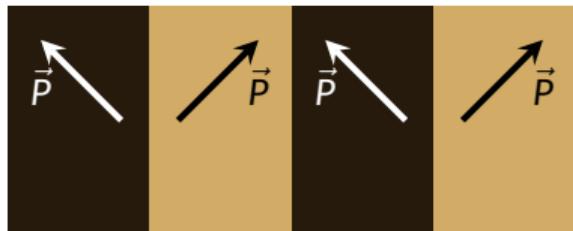


NV imaging of the cycloid, iso-B mode

DyScO₃, strain -0.35%

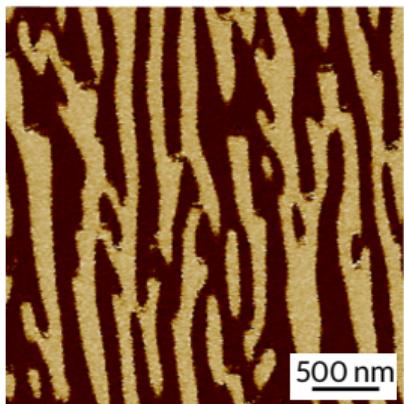


PFM image
ferroelectric domains

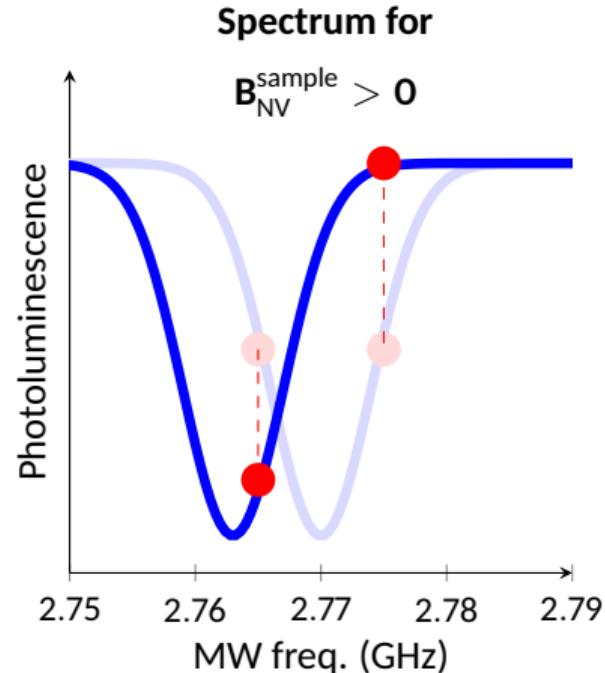
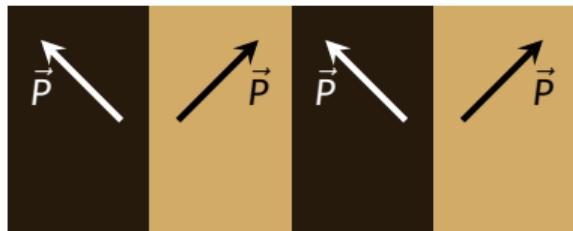


NV imaging of the cycloid, iso-B mode

DyScO₃, strain -0.35%



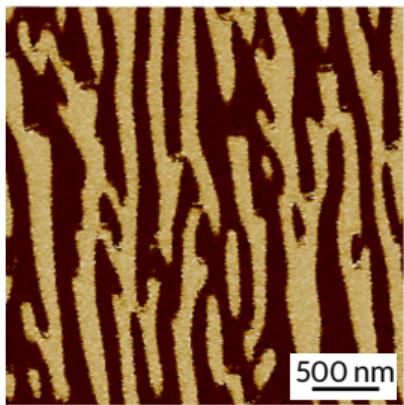
PFM image
ferroelectric domains



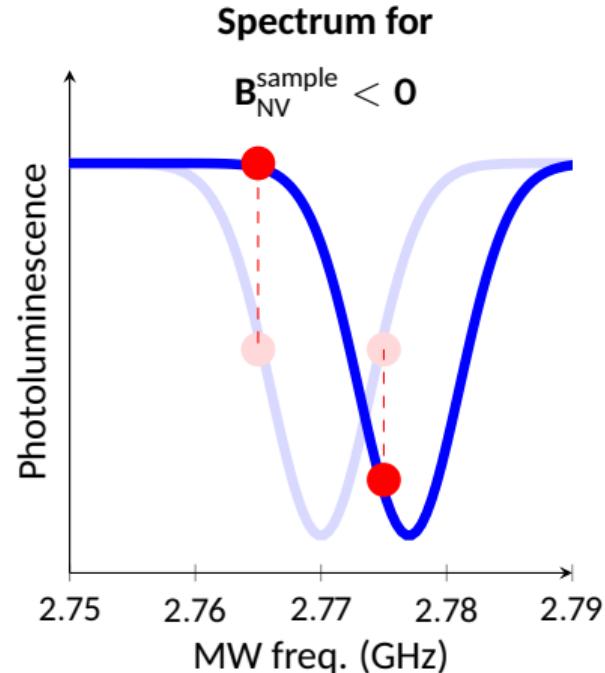
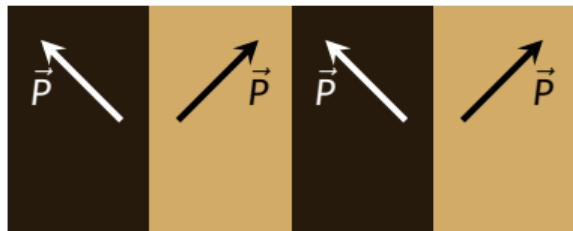
$$\Delta PL = PL(f_2) - PL(f_1)$$
$$\Delta PL > 0$$

NV imaging of the cycloid, iso-B mode

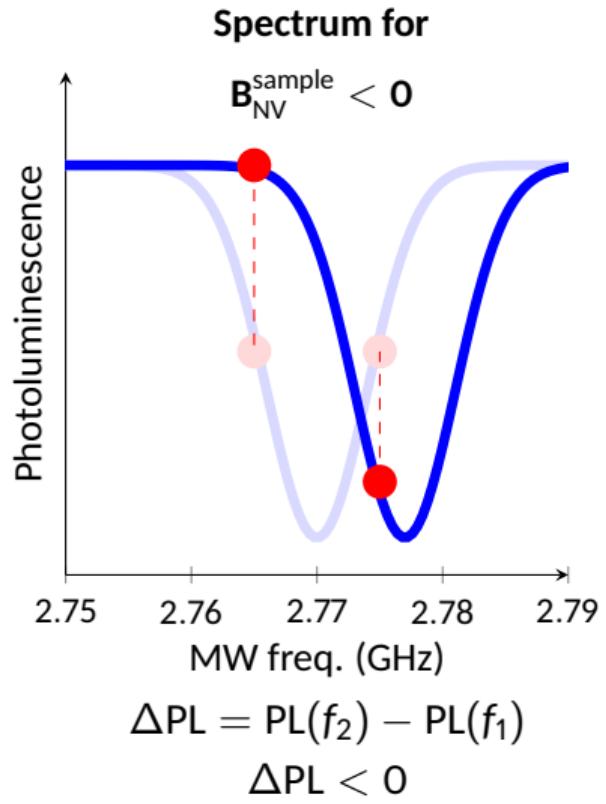
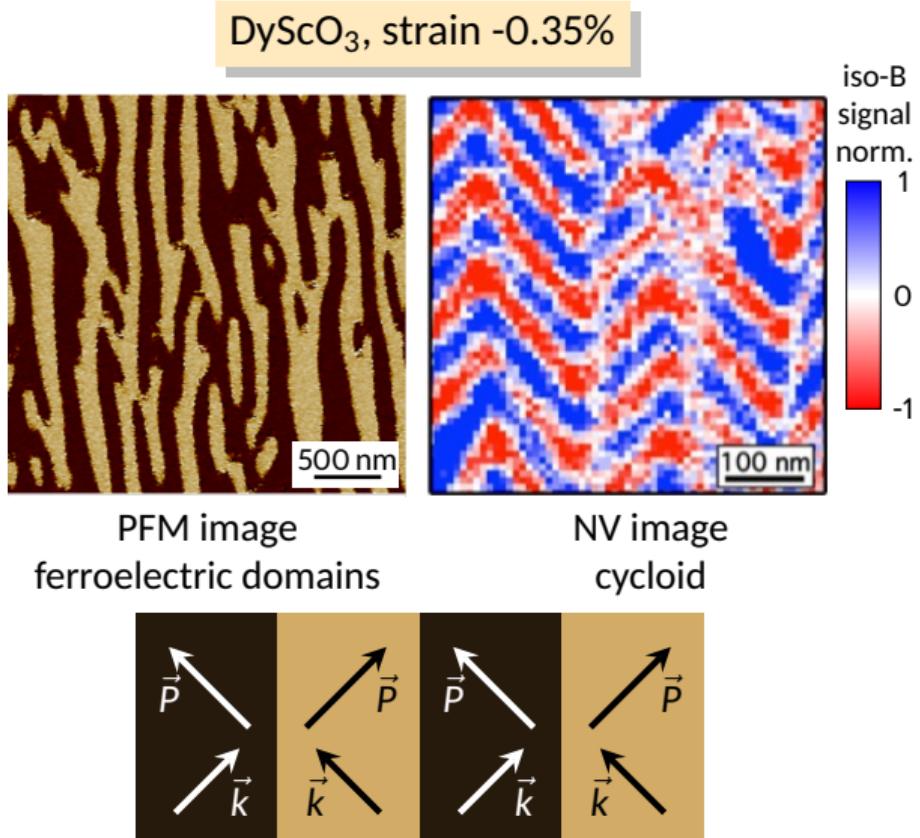
DyScO₃, strain -0.35%



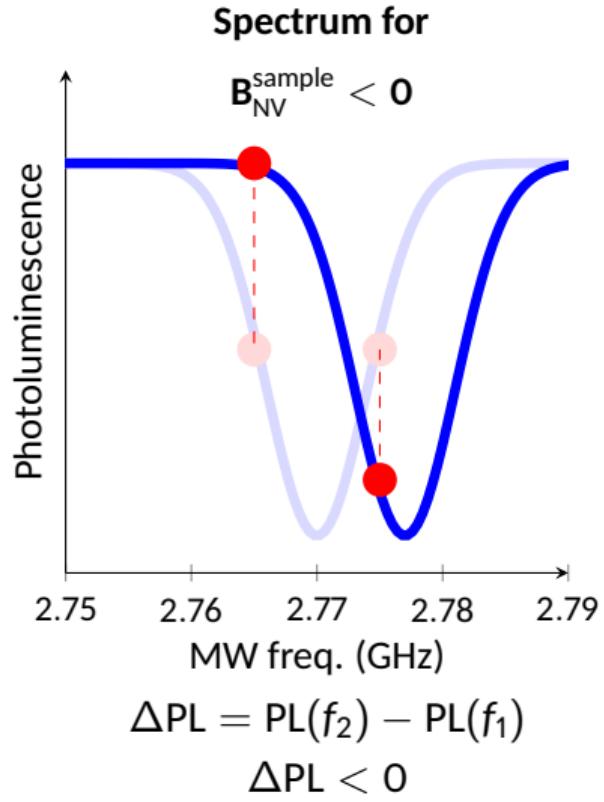
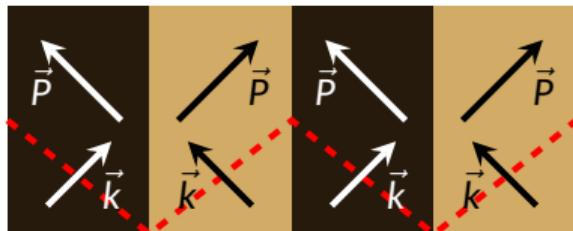
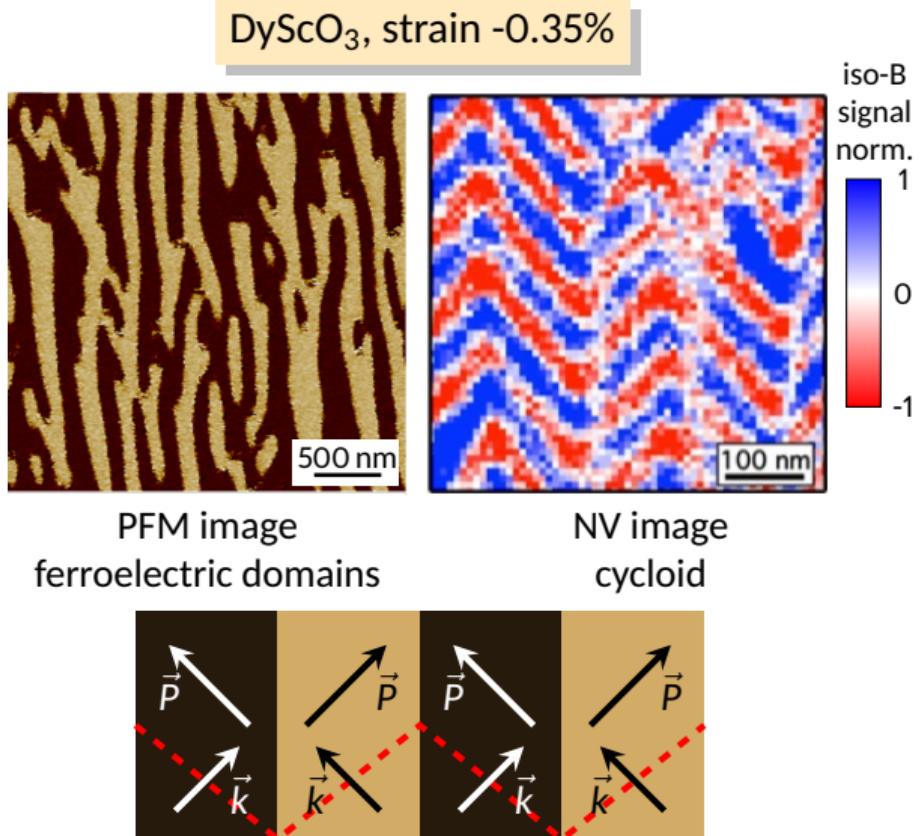
PFM image
ferroelectric domains



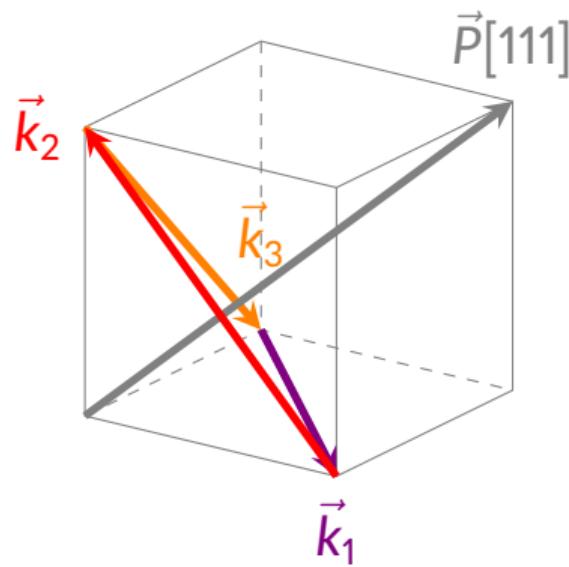
NV imaging of the cycloid, iso-B mode



NV imaging of the cycloid, iso-B mode



The type I cycloid

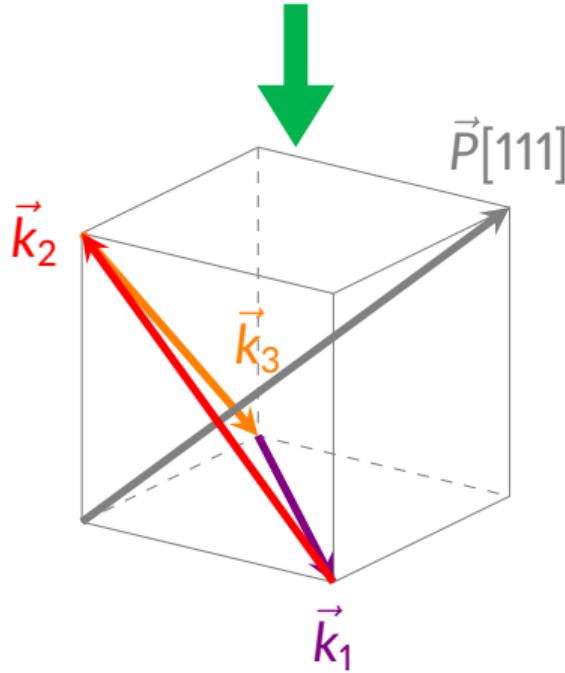


$$\vec{k}_1 \parallel [1\bar{1}0]$$

$$\vec{k}_2 \parallel [\bar{1}01]$$

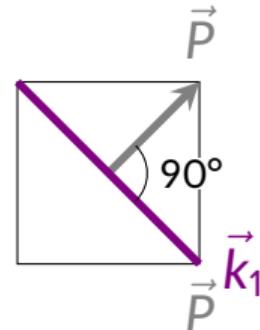
$$\vec{k}_3 \parallel [01\bar{1}]$$

The type I cycloid

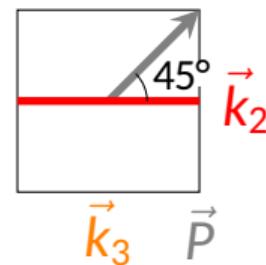


$$\begin{aligned}\vec{k}_1 &\parallel [1\bar{1}0] \\ \vec{k}_2 &\parallel [\bar{1}01] \\ \vec{k}_3 &\parallel [01\bar{1}]\end{aligned}$$

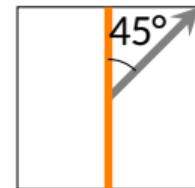
Top view



$$\begin{aligned}\lambda_{IP} &= \lambda_{bulk} \\ 64 \text{ nm}\end{aligned}$$



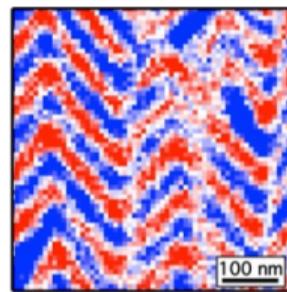
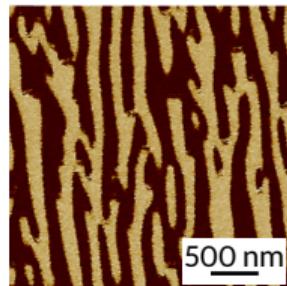
$$\begin{aligned}\lambda_{IP} &= \sqrt{2}\lambda_{bulk} \\ 90 \text{ nm}\end{aligned}$$



$$\begin{aligned}\lambda_{IP} &= \sqrt{2}\lambda_{bulk} \\ 90 \text{ nm}\end{aligned}$$

Stripy ferroelectric domains

DyScO₃



-0.35%

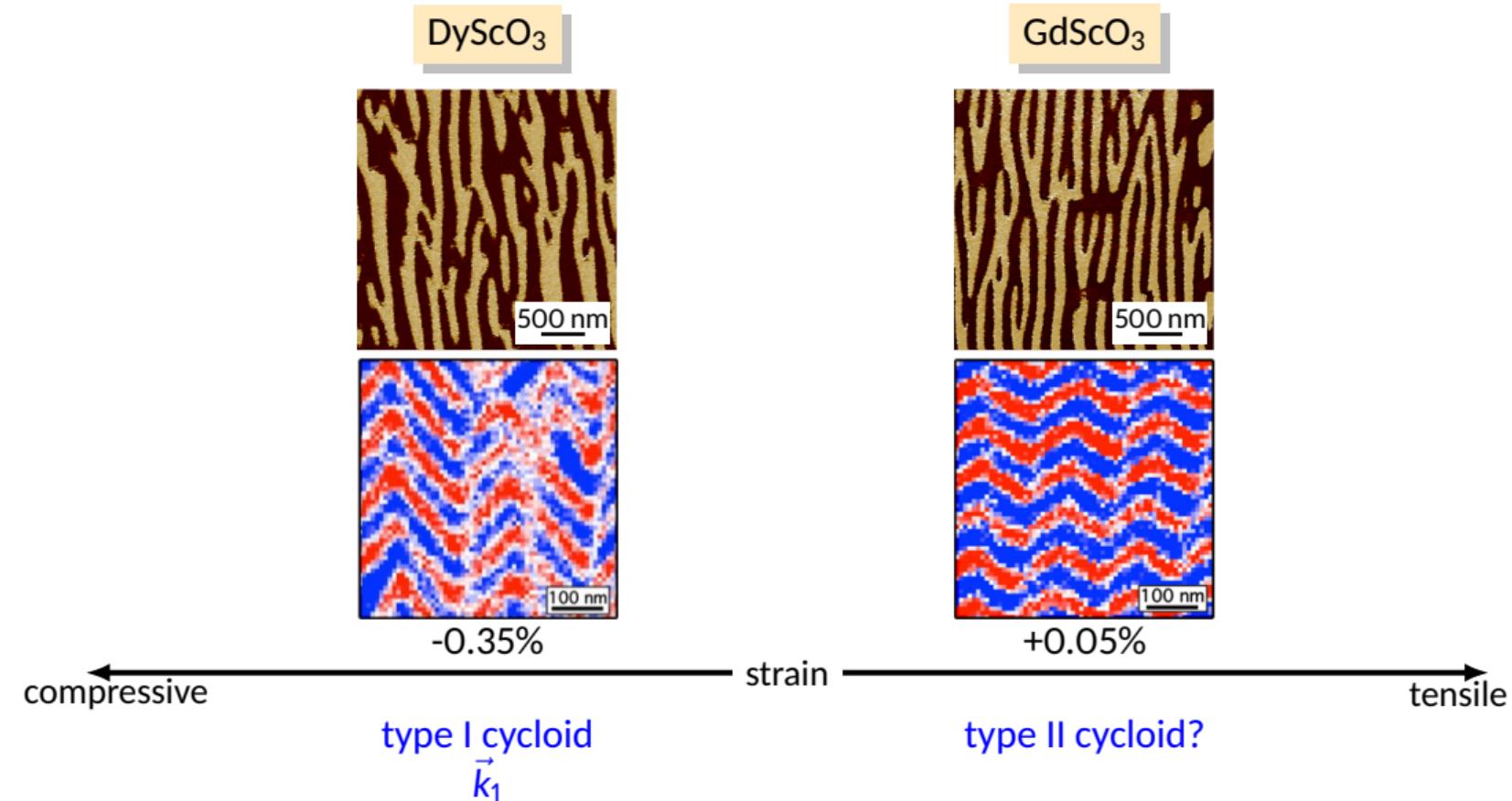
compressive

strain

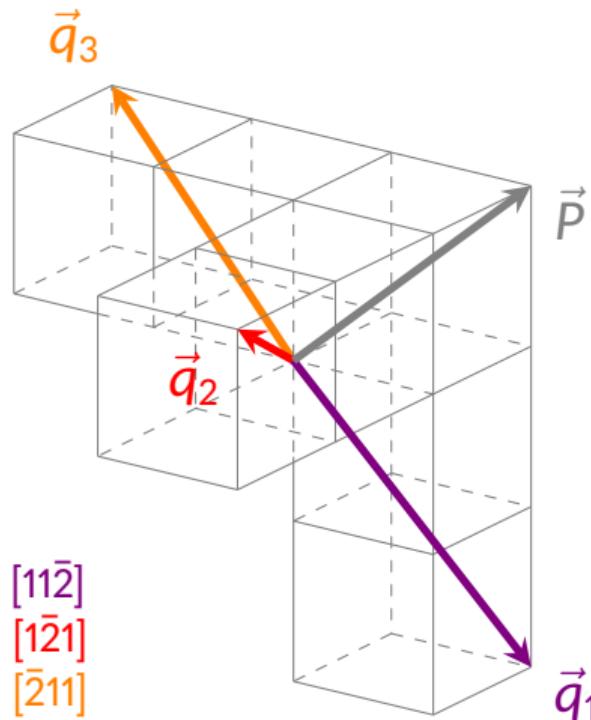
tensile

type I cycloid
 \vec{k}_1

Stripy ferroelectric domains



The type II cycloid

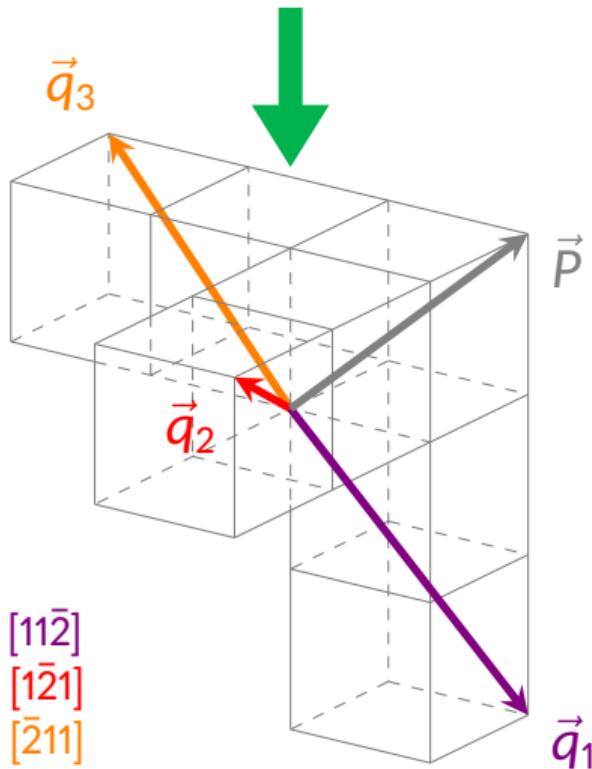


$$\vec{q}_1 \parallel [11\bar{2}]$$

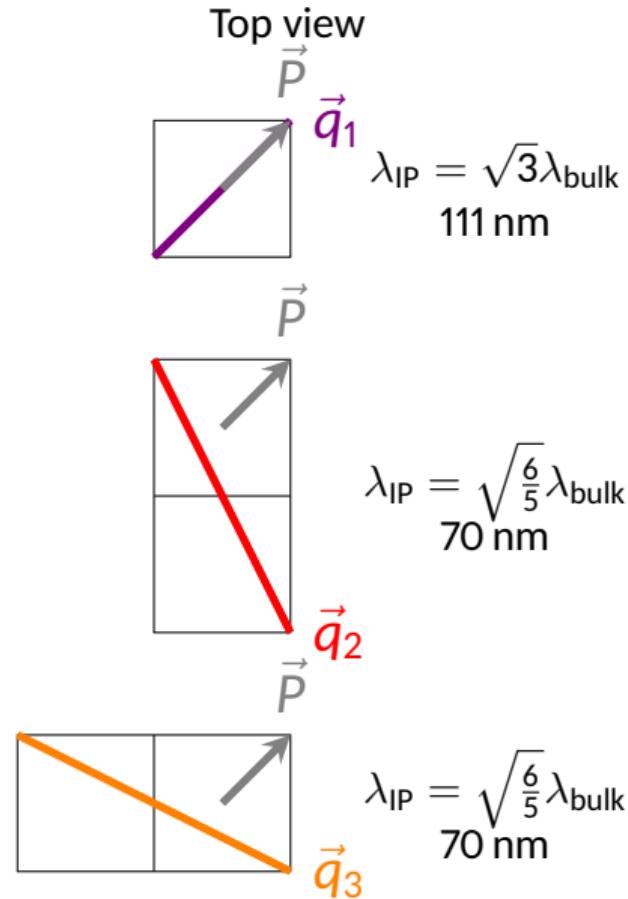
$$\vec{q}_2 \parallel [\bar{1}\bar{2}1]$$

$$\vec{q}_3 \parallel [\bar{2}11]$$

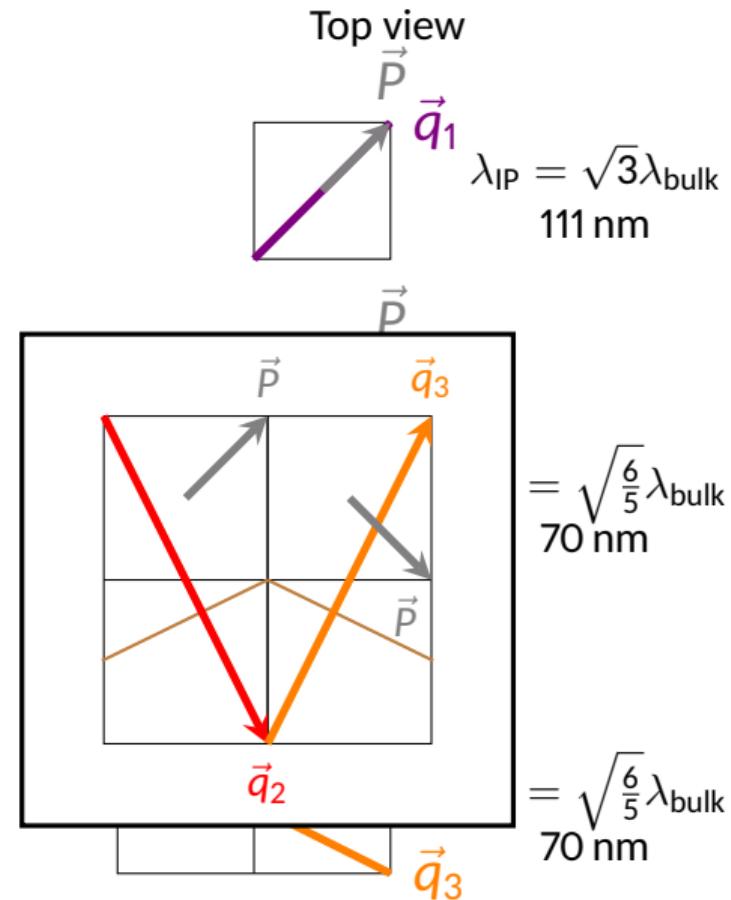
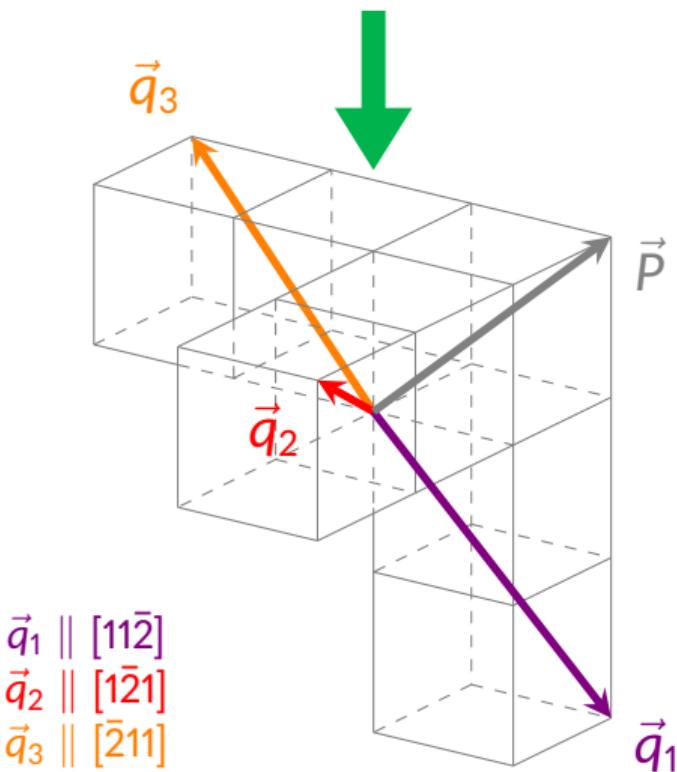
The type II cycloid



D. Sando et al. Nat. Mater. 12 (2013), 641–646



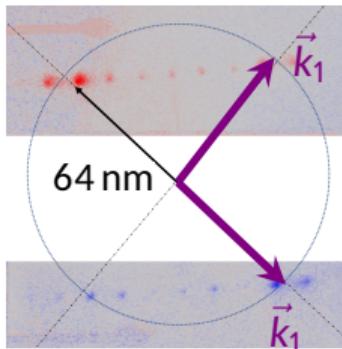
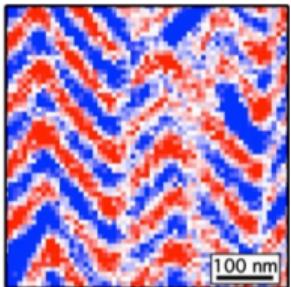
The type II cycloid



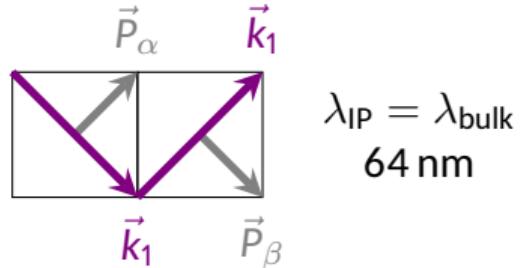
X-ray diffraction

DyScO₃

type I cycloid
 \vec{k}_1



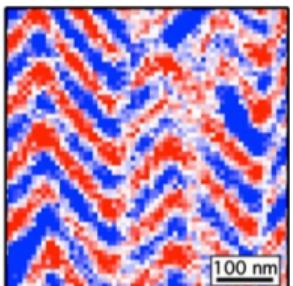
N. Jaouen, J.-Y. Chauleau, M. Viret



X-ray diffraction

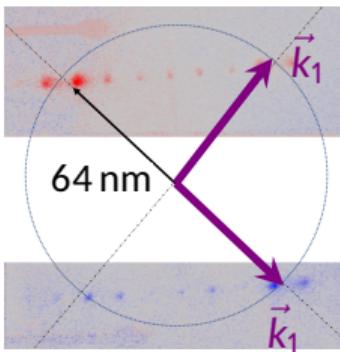
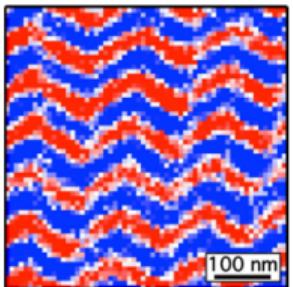
DyScO₃

type I cycloid
 \vec{k}_1

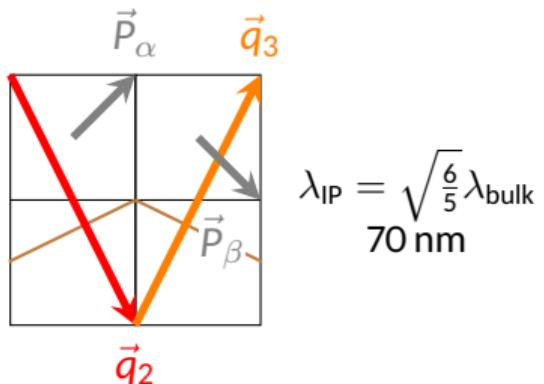
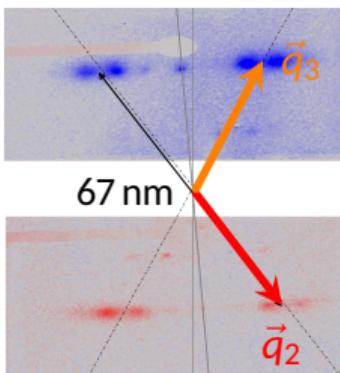
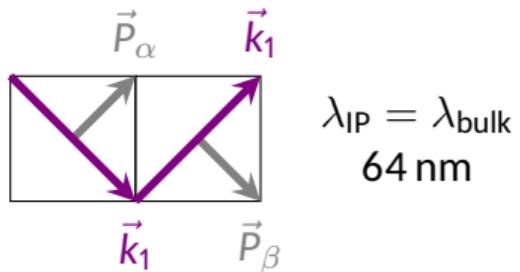


GdScO₃

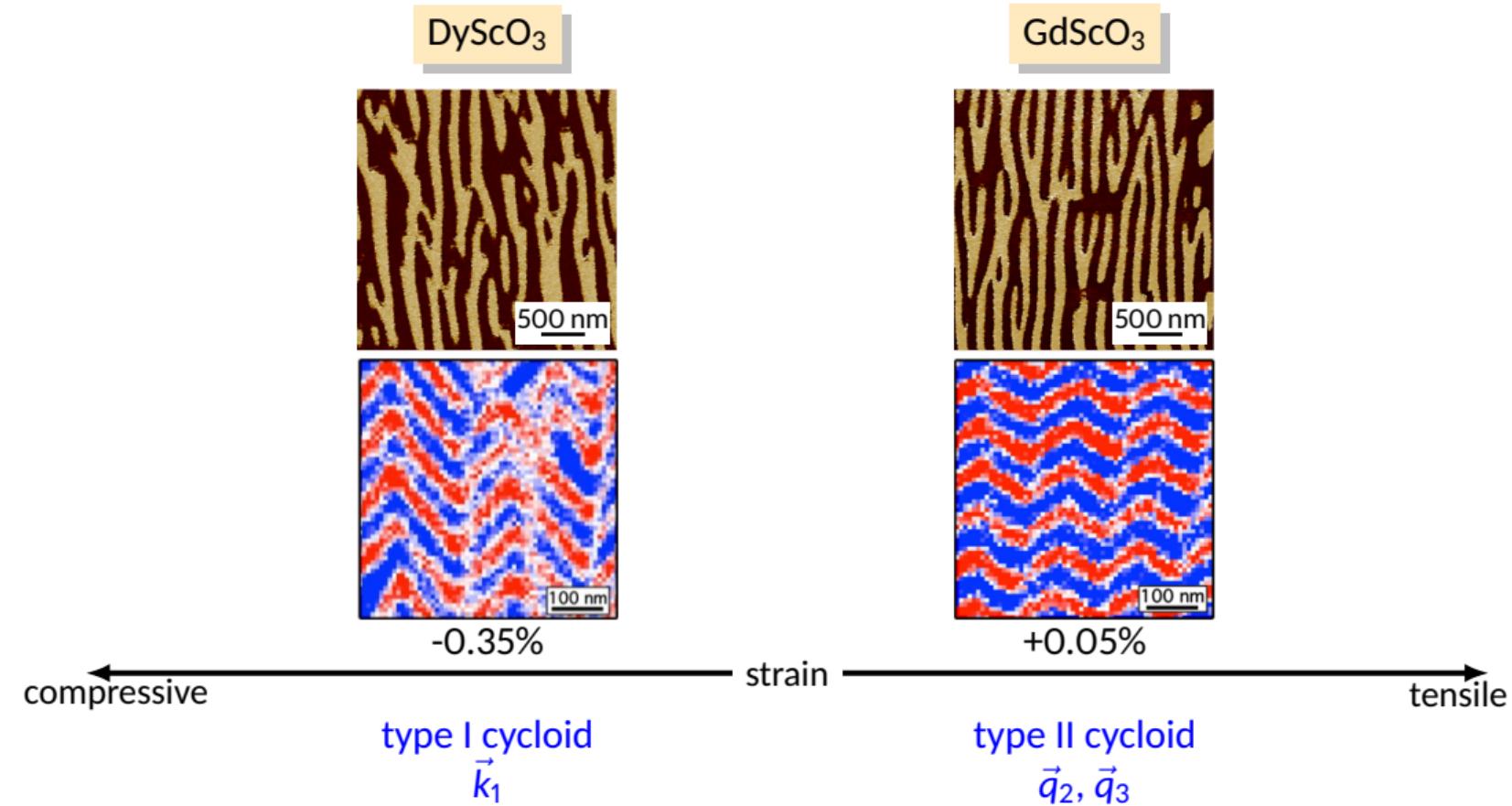
type II cycloid
 \vec{q}_2, \vec{q}_3



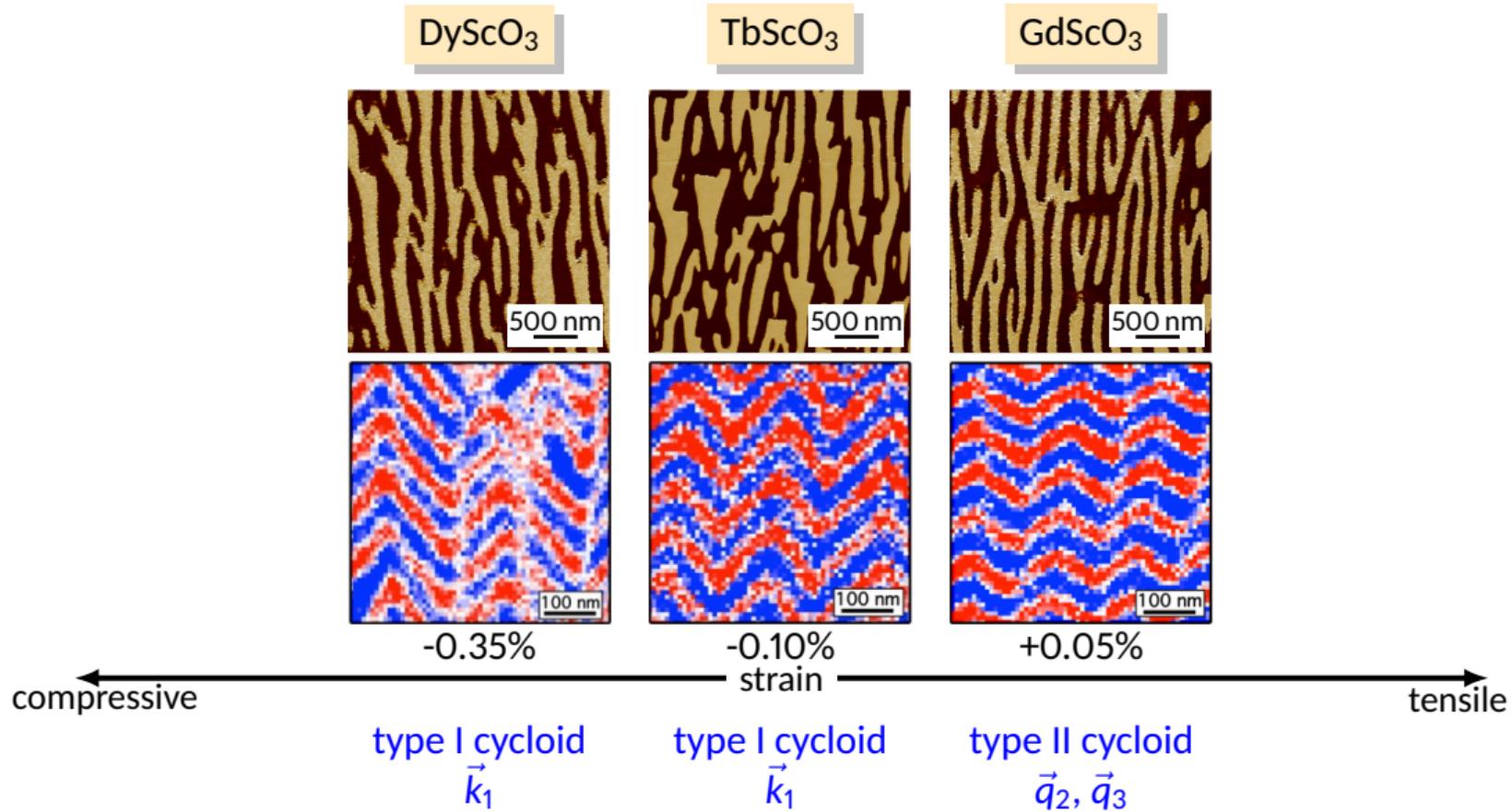
N. Jaouen, J.-Y. Chauleau, M. Viret



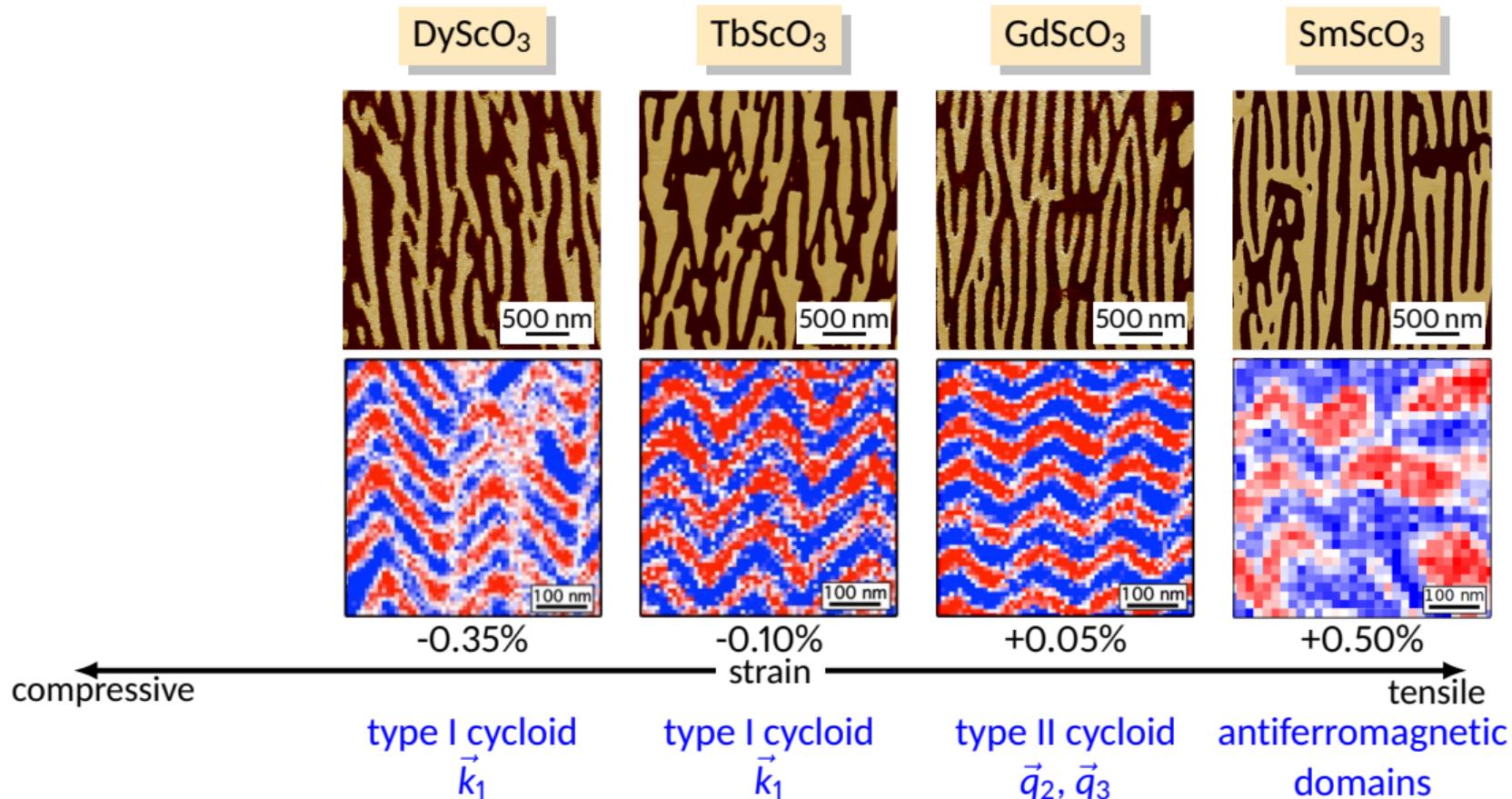
Stripy ferroelectric domains



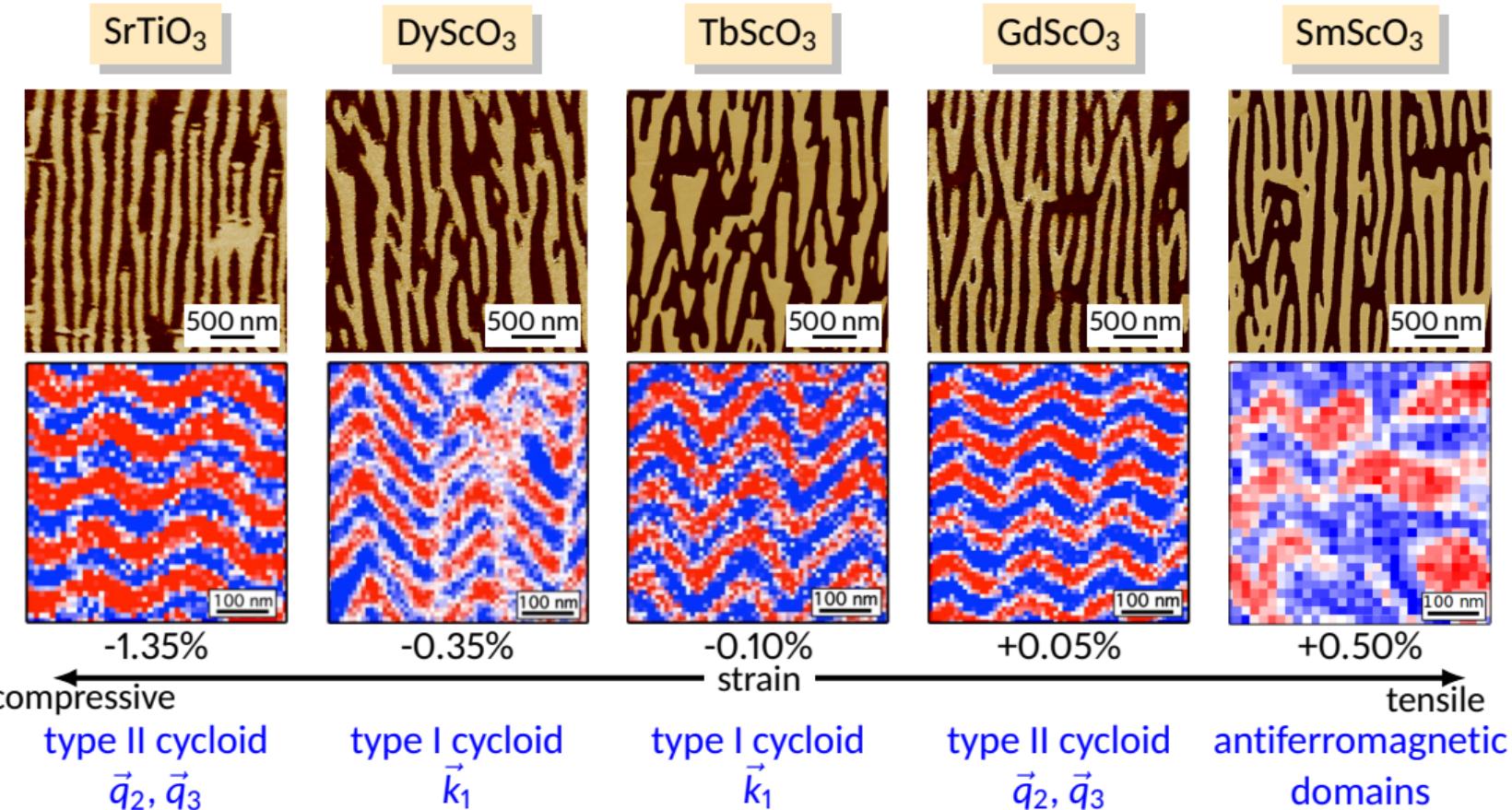
Stripy ferroelectric domains



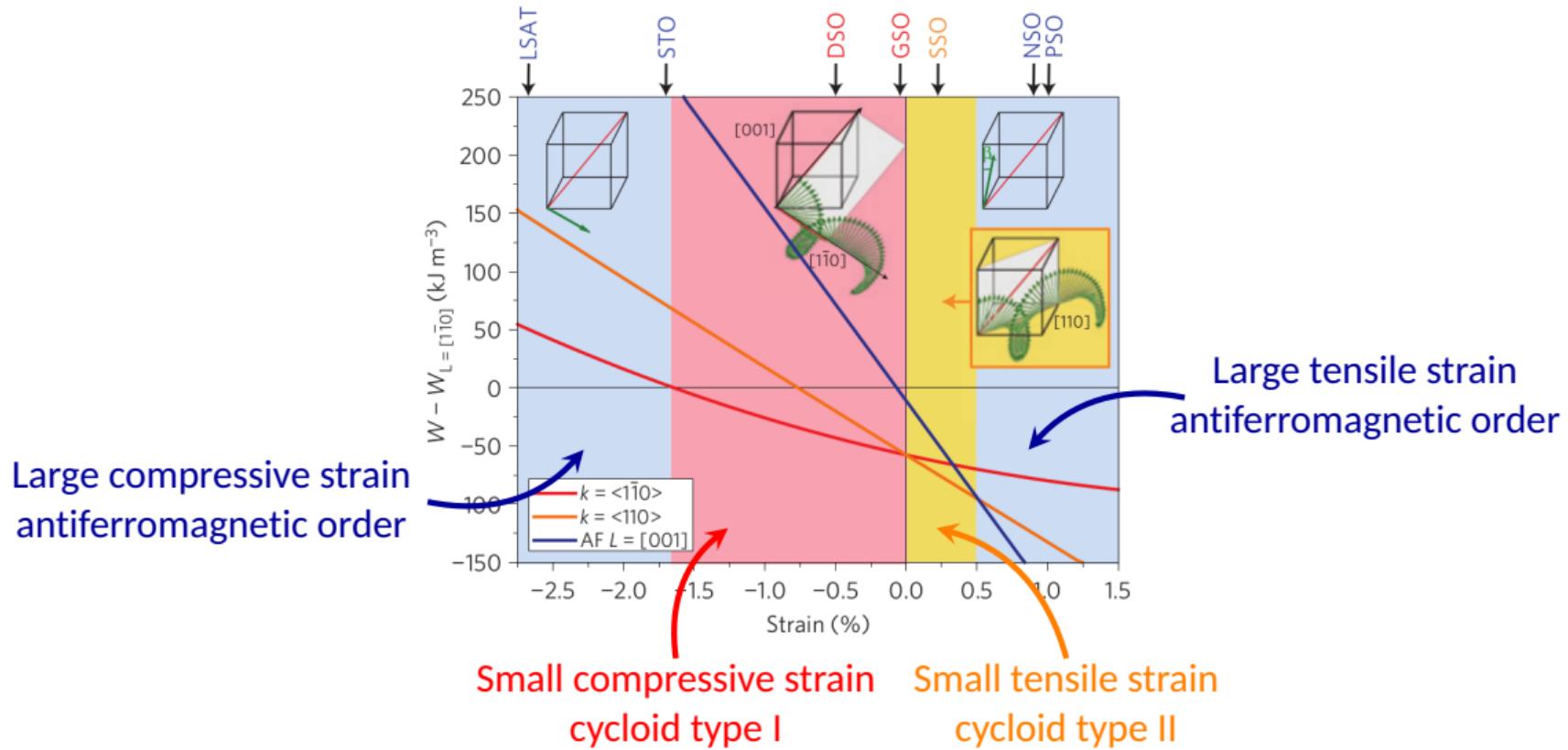
Stripy ferroelectric domains



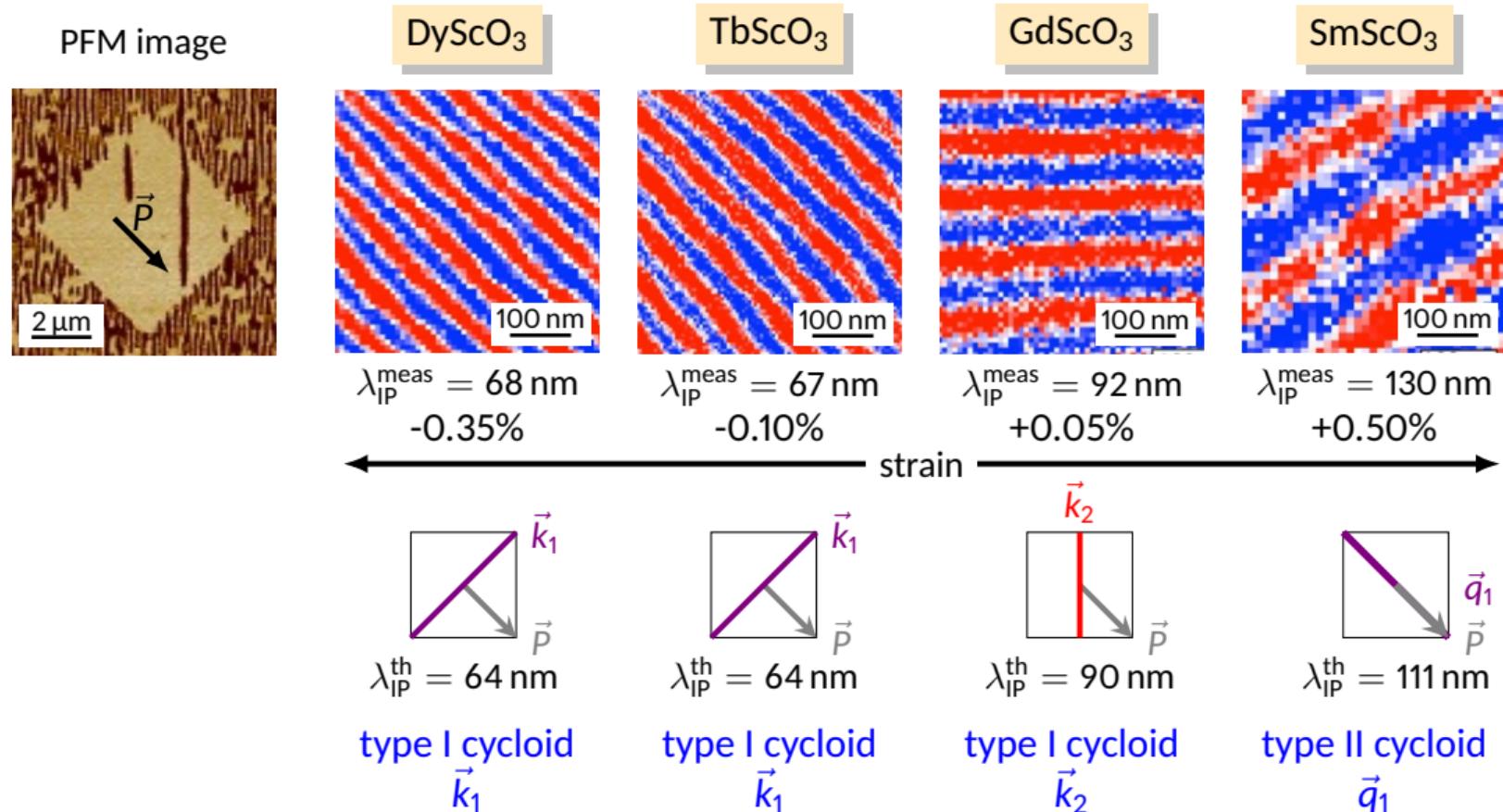
Stripy ferroelectric domains



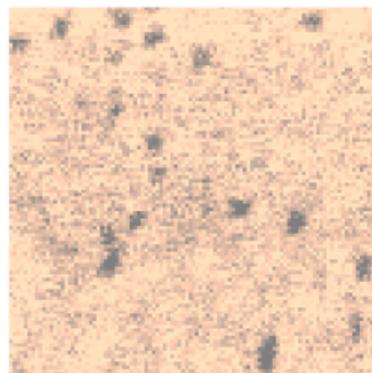
Known effect of epitaxial strain on the cycloid



Manipulation via magnetoelectric coupling



Zero-field skyrmions in exchange-biased magnetic layers

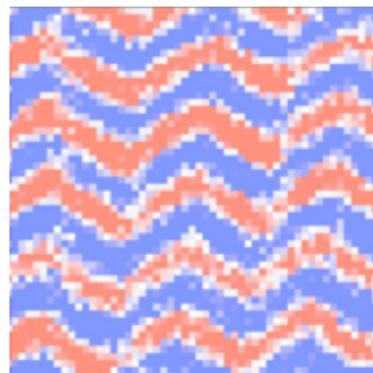


Collaboration



O. Boulle group, Grenoble

Influence of epitaxial strain on the cycloid in the multiferroic BiFeO₃

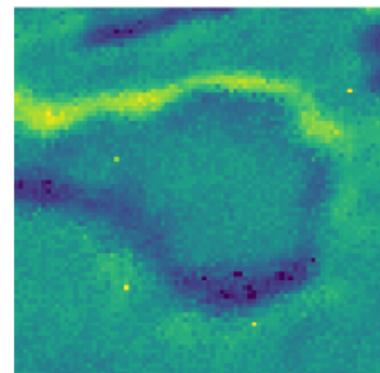


Collaboration



S. Fusil and V. Garcia group
M. Viret group, Palaiseau

Detection of domain wall magnons in a synthetic antiferromagnet



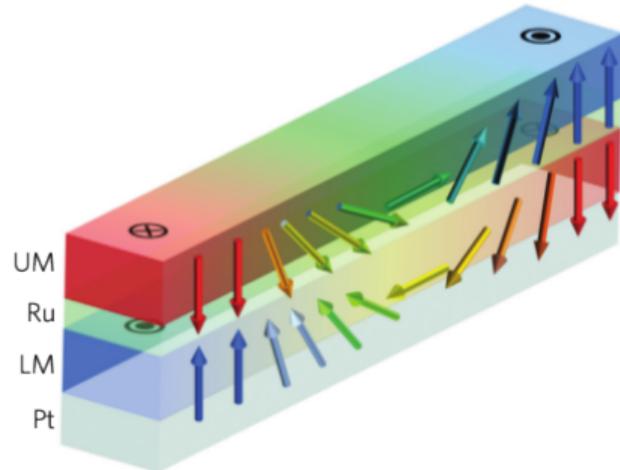
Collaboration



V. Cros group
J.-V. Kim, Palaiseau

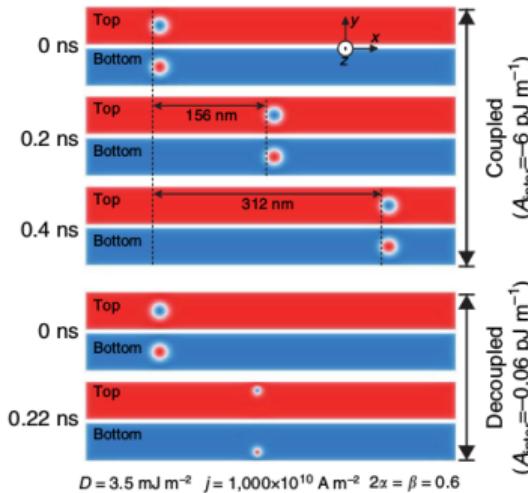
Synthetic antiferromagnets

Two ferromagnetic layers antiferromagnetically coupled through RKKY interaction



Fast current-induced domain wall motion up to 750 m s^{-1}

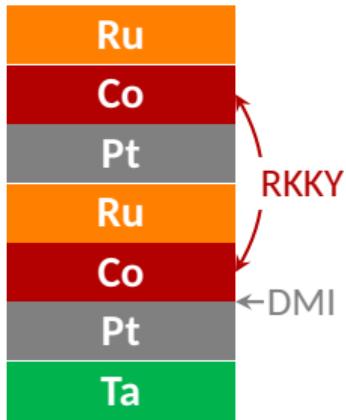
S.-H. Yang *et al.* *Nat. Nano.* 10 (2015), 221–226



Compensation of the dipolar field:
→ smaller skyrmions
→ no skyrmion Hall effect

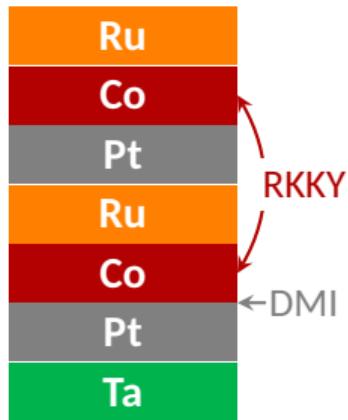
X. Zhang *et al.* *Nat. Commun.* 7 (2016), 10293

Sample optimization

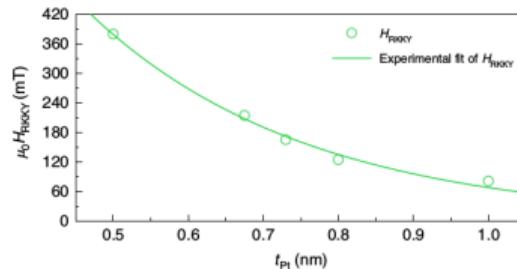
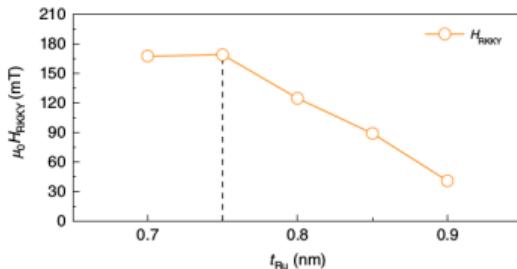


Samples from UMR
CNRS/Thales
W. Legrand, F. Ajejas,
Y. Sassi, V. Cros

Sample optimization



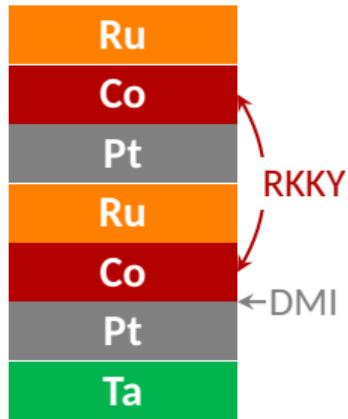
Tuning the RKKY coupling with the Ru and Pt thicknesses



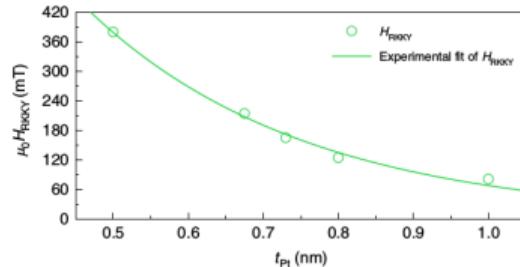
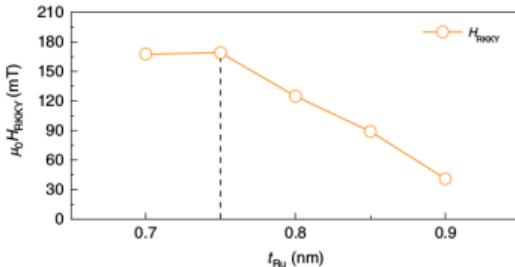
Samples from UMR
CNRS/Thales
W. Legrand, F. Ajedas,
Y. Sassi, V. Cros



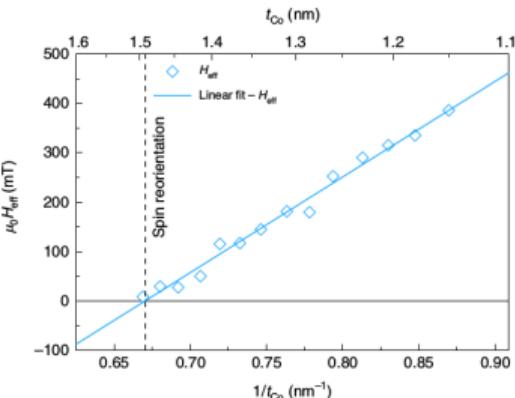
Sample optimization



Tuning the RKKY coupling with the Ru and Pt thicknesses



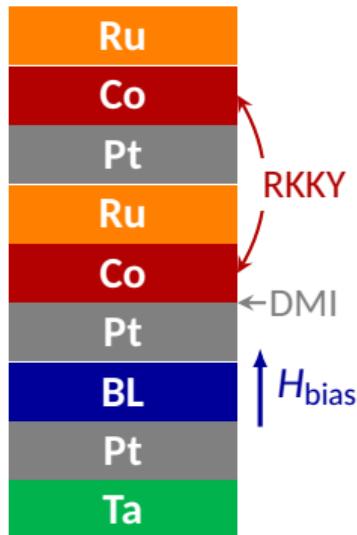
Anisotropy fixed by the Co thickness



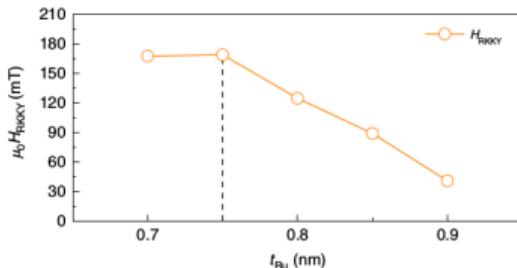
Samples from UMR
CNRS/Thales
W. Legrand, F. Ajedas,
Y. Sassi, V. Cros



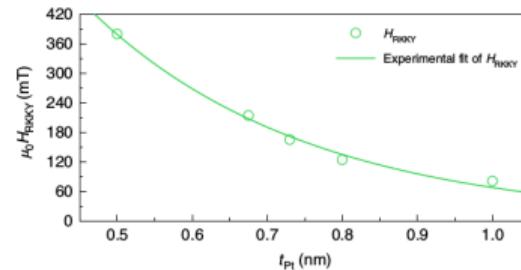
Sample optimization



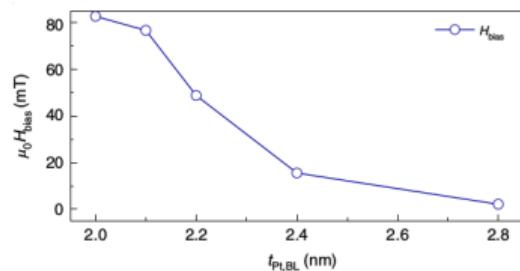
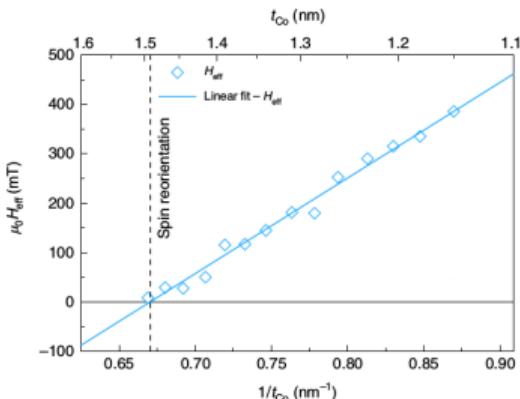
Tuning the RKKY coupling with the Ru and Pt thicknesses



Anisotropy fixed by the Co thickness



Bias field adjusted by the Pt spacer



Samples from UMR
CNRS/Thales
W. Legrand, F. Ajedas,
Y. Sassi, V. Cros

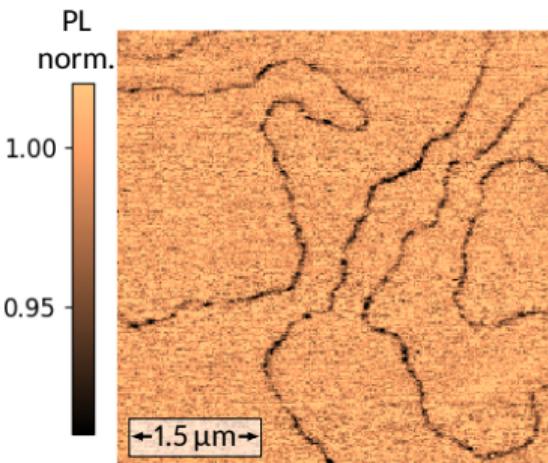


NV images of the non-collinear magnetic structures

Fully compensated SAF

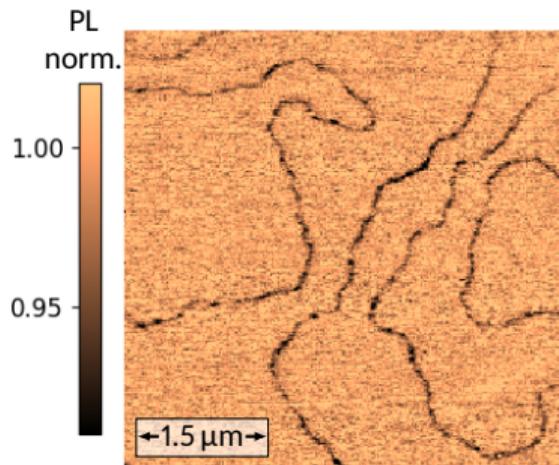
Perpendicular anisotropy

Large out-of-plane domains

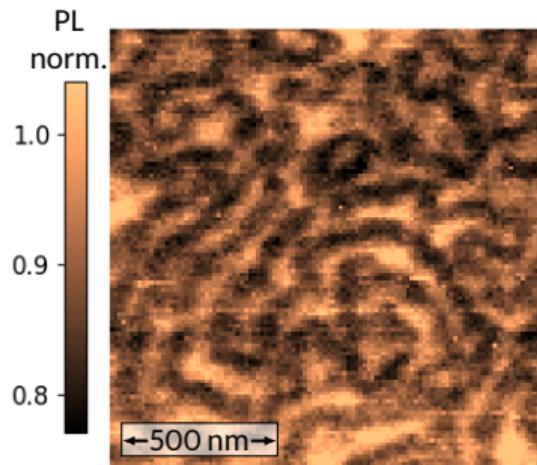


NV images of the non-collinear magnetic structures

Fully compensated SAF
Perpendicular anisotropy
Large out-of-plane domains

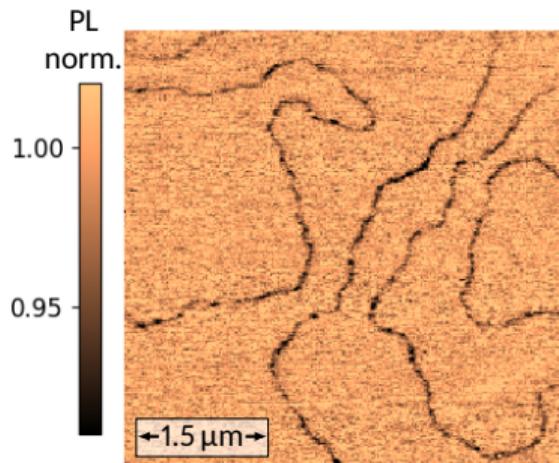


Fully compensated SAF
Vanishing anisotropy
Spin spiral

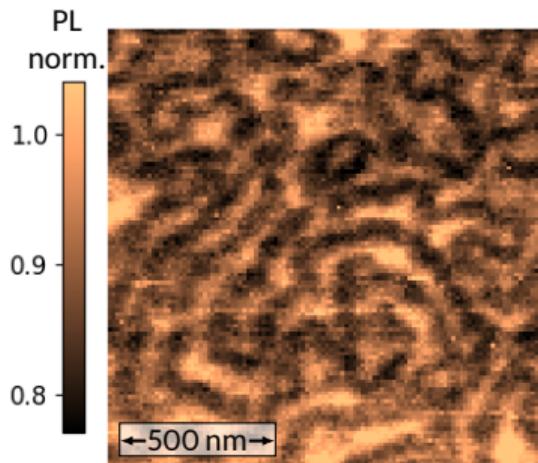


NV images of the non-collinear magnetic structures

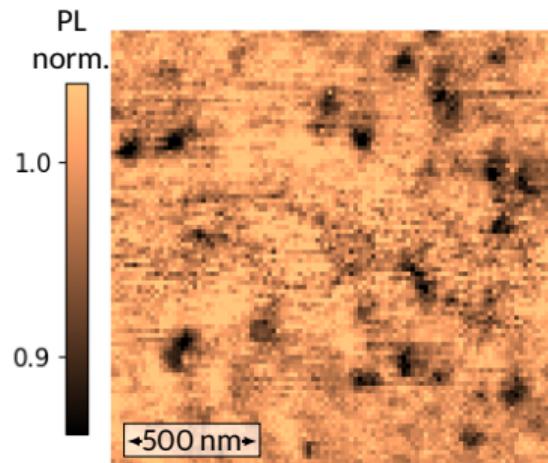
Fully compensated SAF
Perpendicular anisotropy
Large out-of-plane domains



Fully compensated SAF
Vanishing anisotropy
Spin spiral

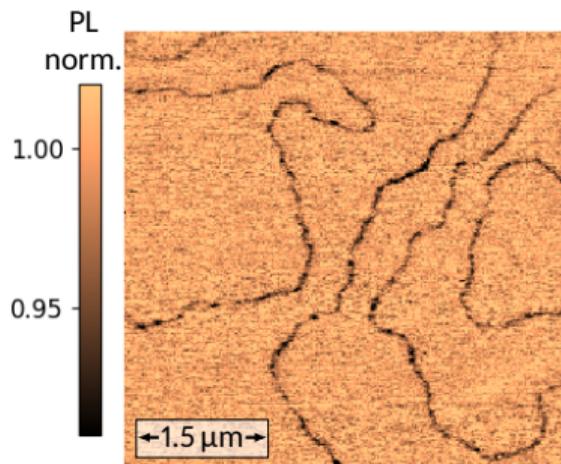


Fully compensated SAF
Biased by a FM layer
Antiferromagnetic skyrmions

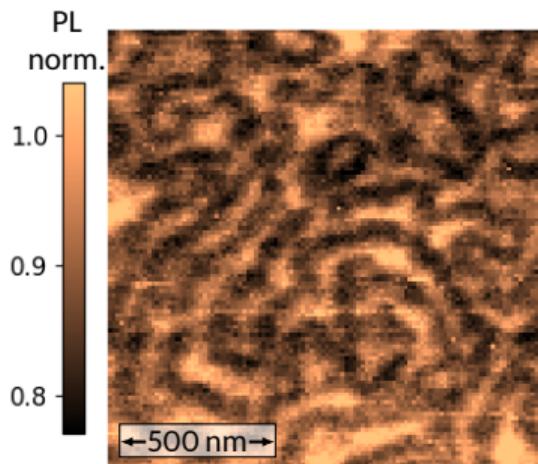


NV images of the non-collinear magnetic structures

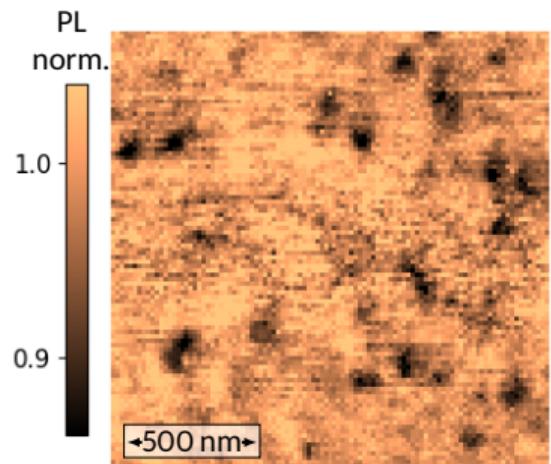
Fully compensated SAF
Perpendicular anisotropy
Large out-of-plane domains



Fully compensated SAF
Vanishing anisotropy
Spin spiral



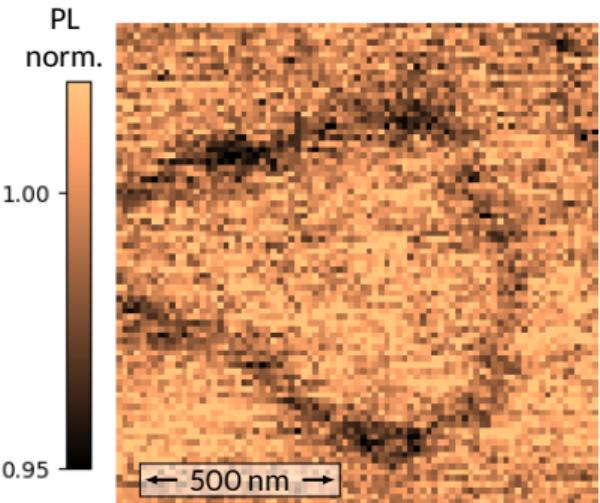
Fully compensated SAF
Biased by a FM layer
Antiferromagnetic skyrmions



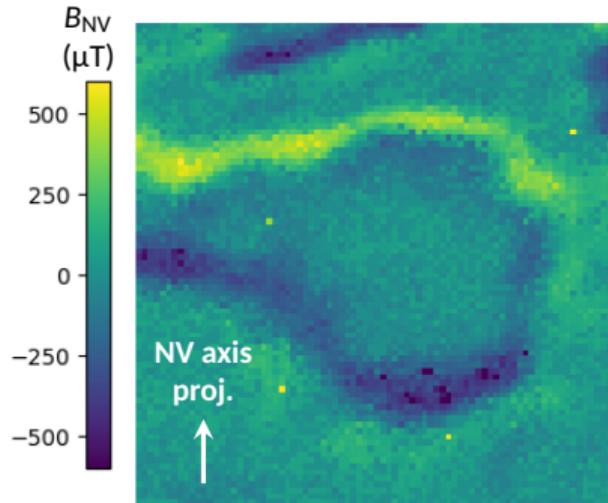
→ Are we really in high-field mode ?

Quantitative imaging of the domain walls

Photoluminescence map

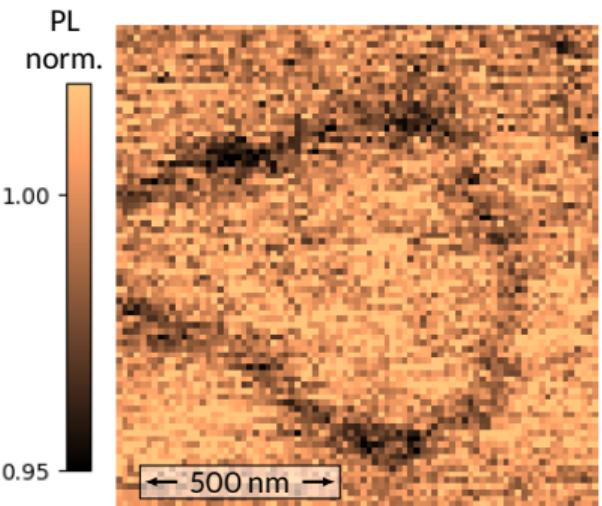


Stray field map

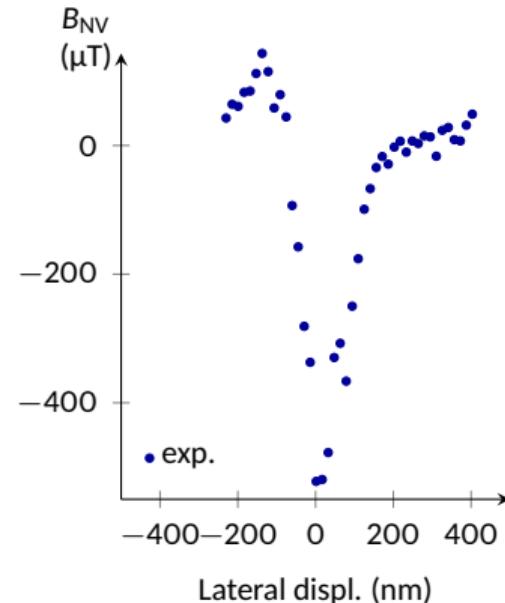
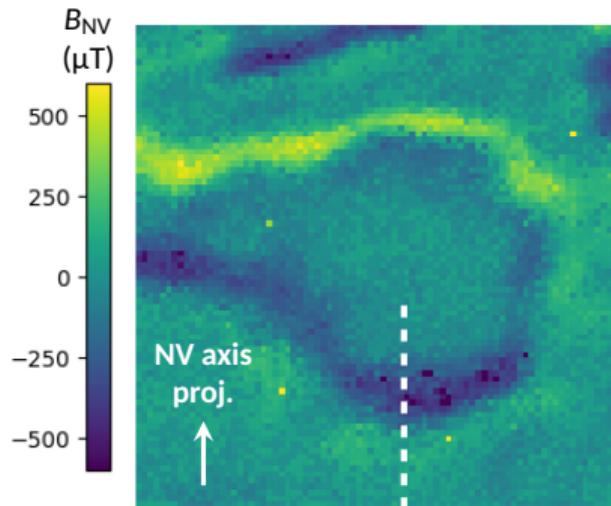


Quantitative imaging of the domain walls

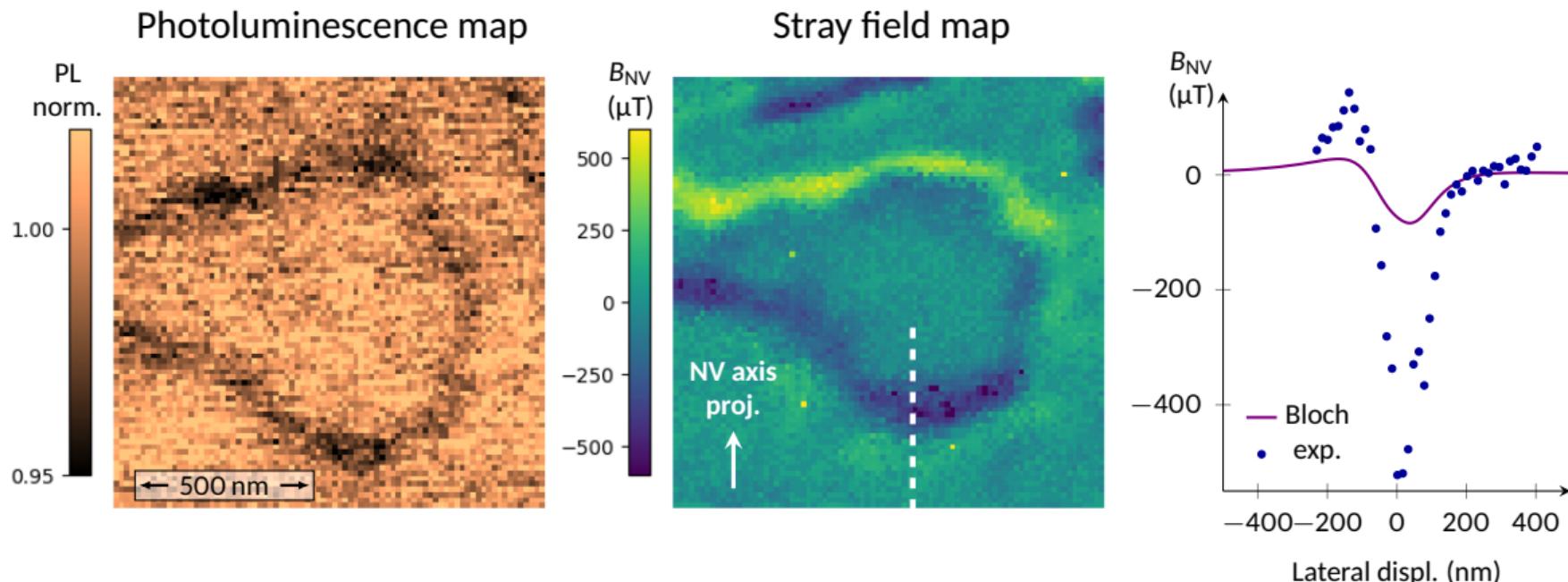
Photoluminescence map



Stray field map

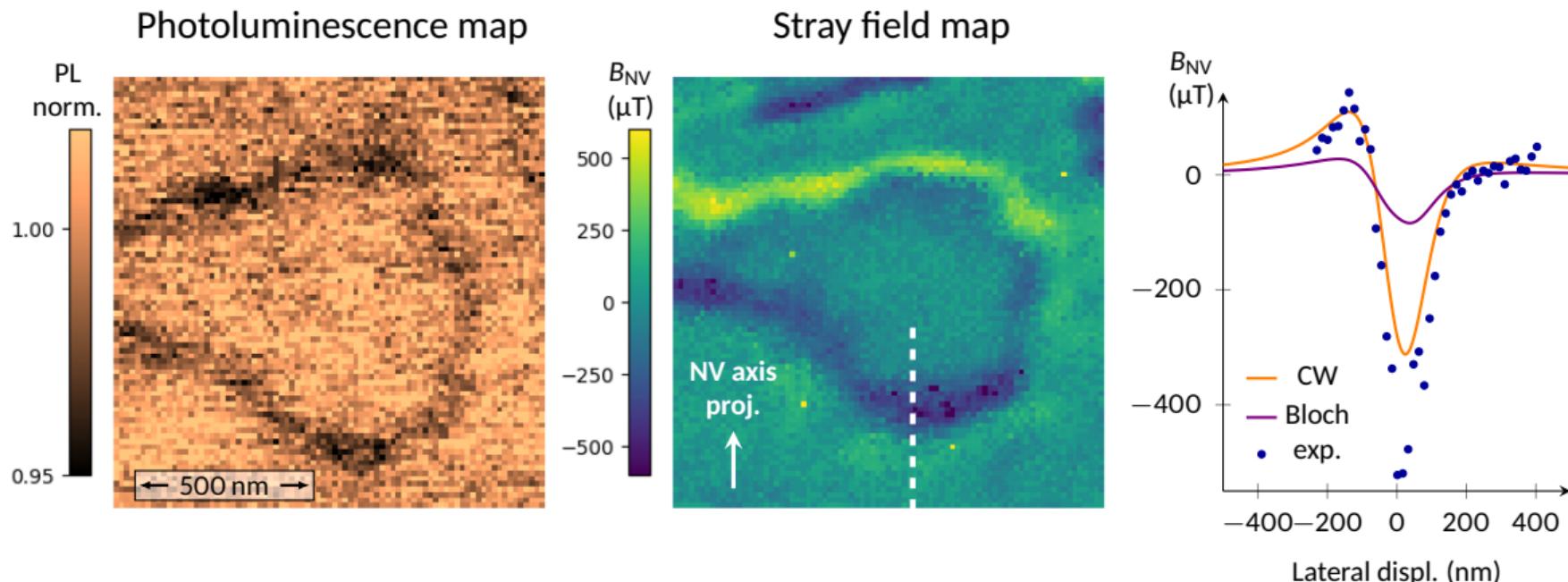


Quantitative imaging of the domain walls



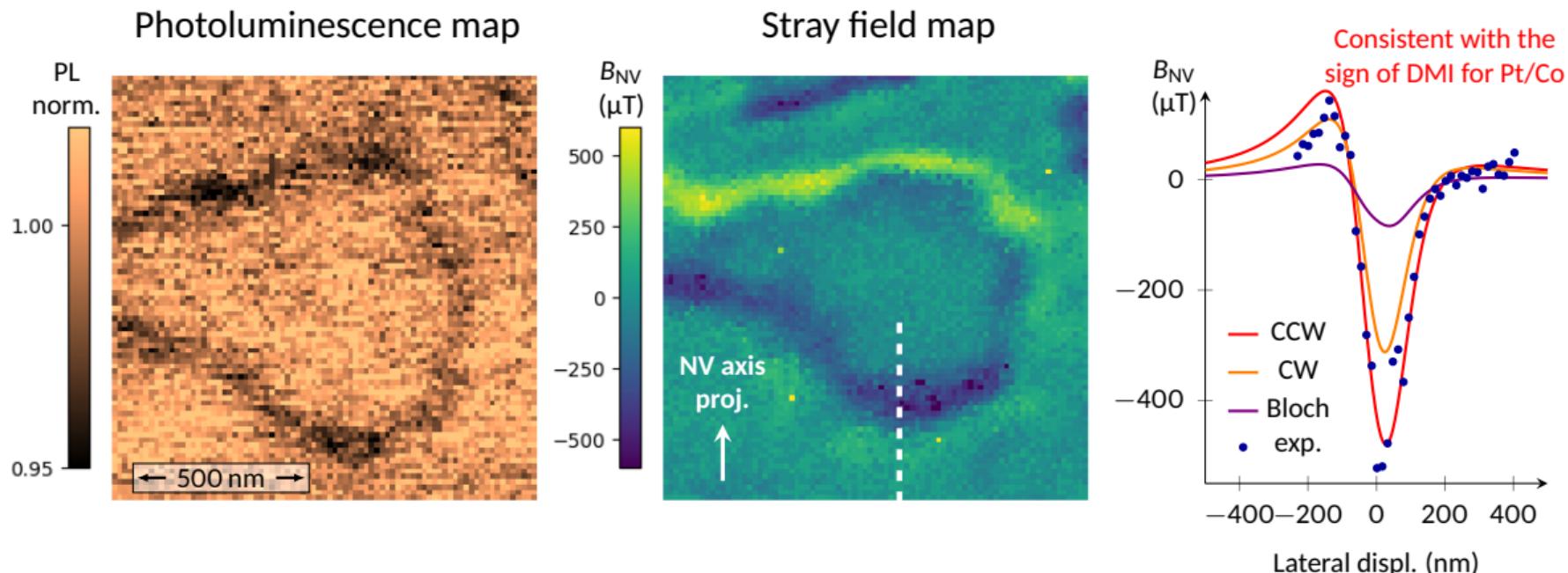
From micromagnetic calculations including the dipolar couplings:
→ Domain wall width top layer: 37 nm
→ Domain wall width bottom layer: 26 nm

Quantitative imaging of the domain walls



From micromagnetic calculations including the dipolar couplings:
→ Domain wall width top layer: 37 nm
→ Domain wall width bottom layer: 26 nm

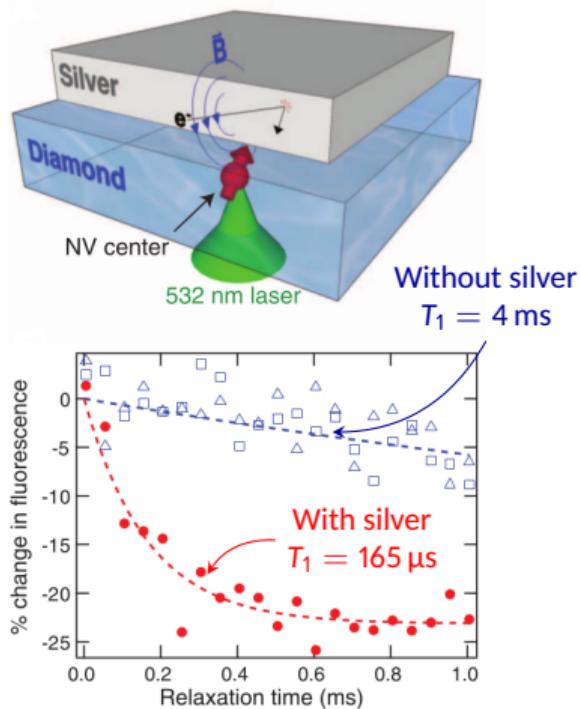
Quantitative imaging of the domain walls



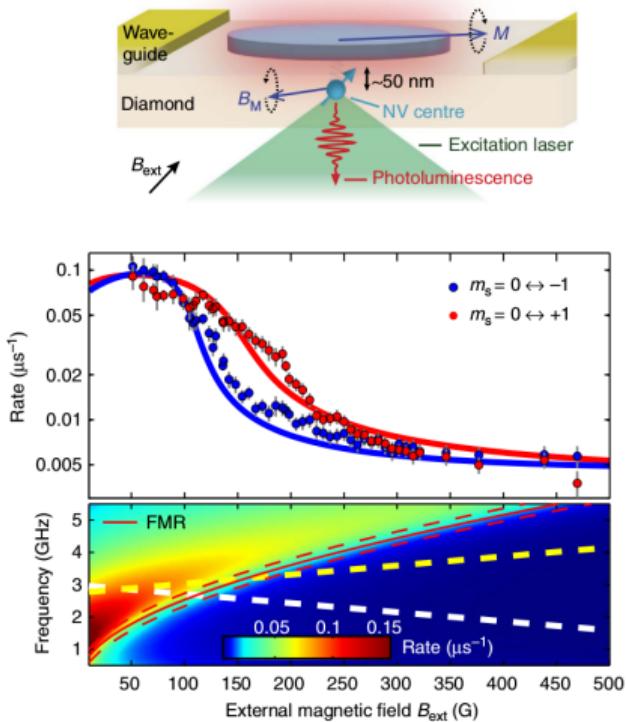
From micromagnetic calculations including the dipolar couplings:
→ Domain wall width top layer: 37 nm
→ Domain wall width bottom layer: 26 nm

Probing magnetic noise with the NV center

Detection of Johnson noise



Detection of ferromagnetic resonance

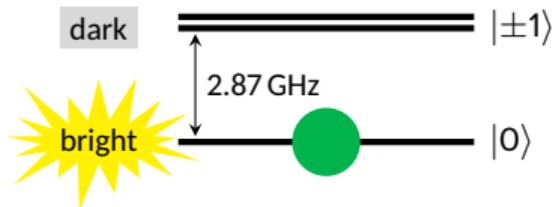


S. Kolkowitz *et al.* *Science* 347 (2015), 1129–1132

T. van der Sar *et al.* *Nat. Commun.* 6 (2015), 7886

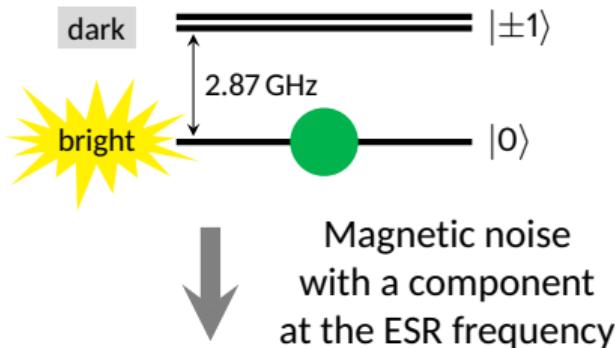
Enhancement of the spin relaxation

Optical pumping
NV spin polarized
in $m_s = 0$

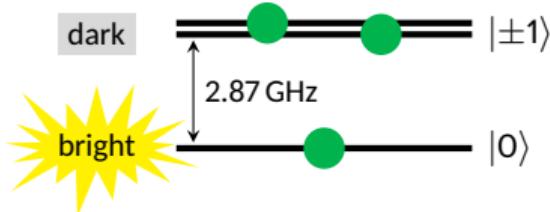


Enhancement of the spin relaxation

Optical pumping
NV spin polarized
in $m_s = 0$

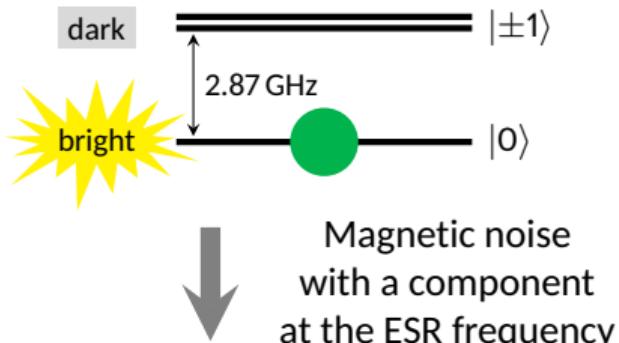


Faster spin
relaxation
(smaller T_1)



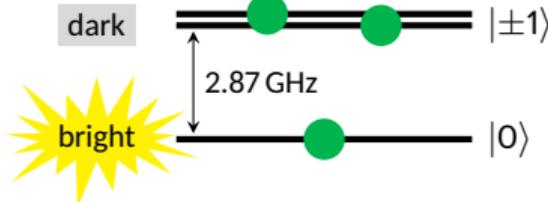
Enhancement of the spin relaxation

Optical pumping
NV spin polarized
in $m_s = 0$

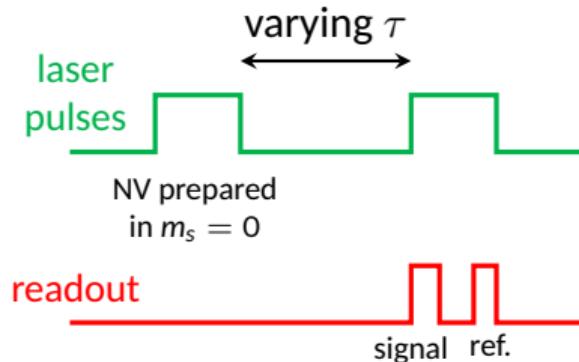


Overall decrease of
the photoluminescence
not related to the DC B_{\perp} field

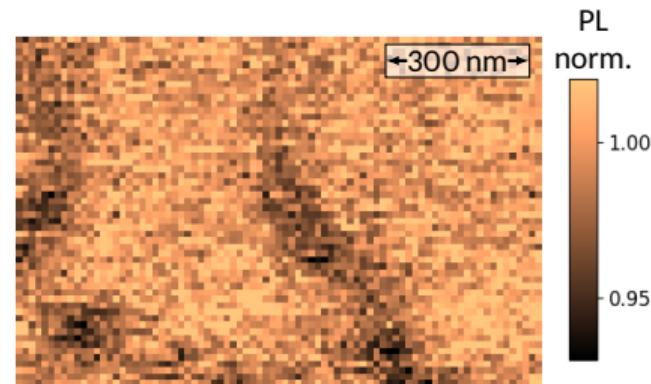
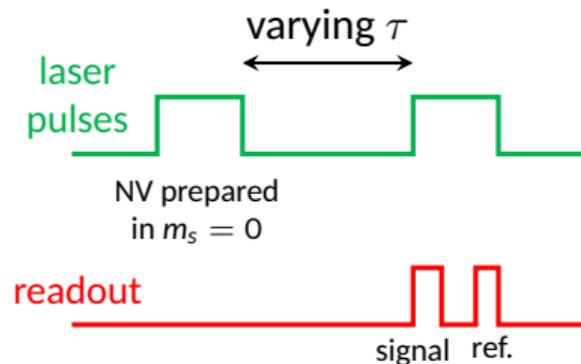
Faster spin
relaxation
(smaller T_1)



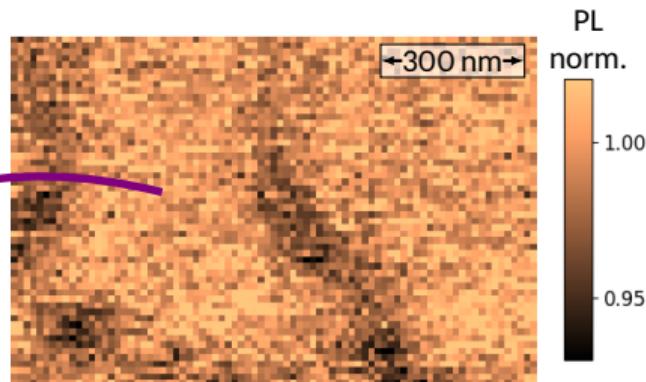
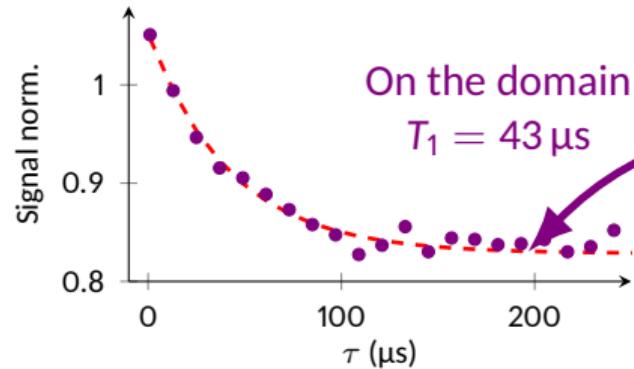
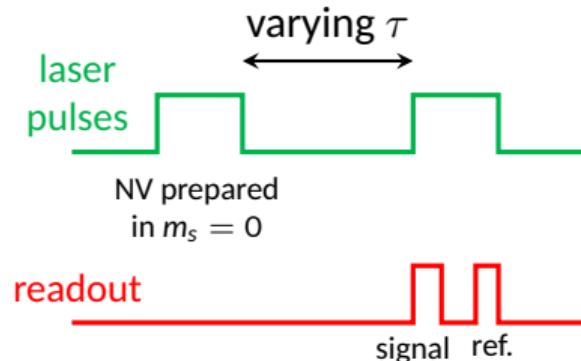
Measurements of the spin relaxation time T_1



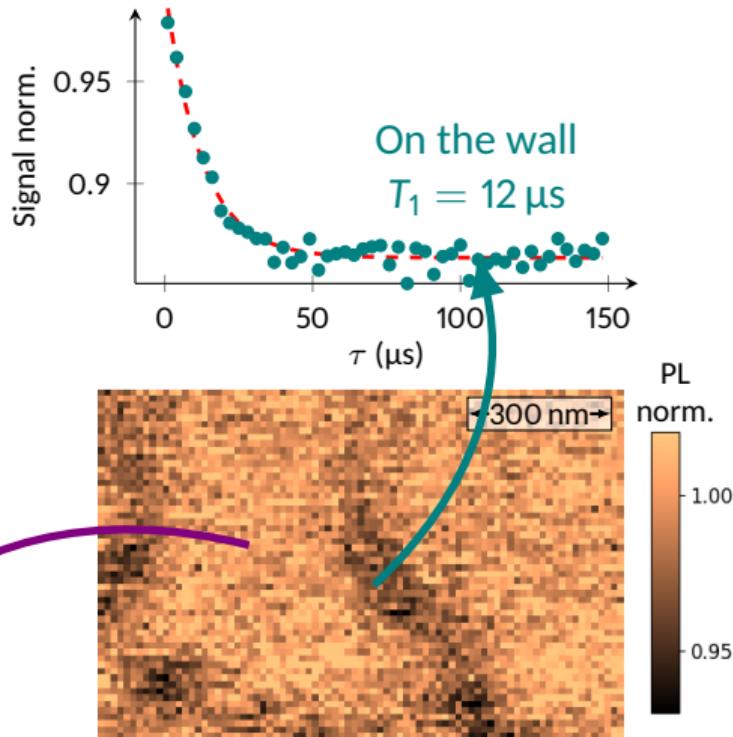
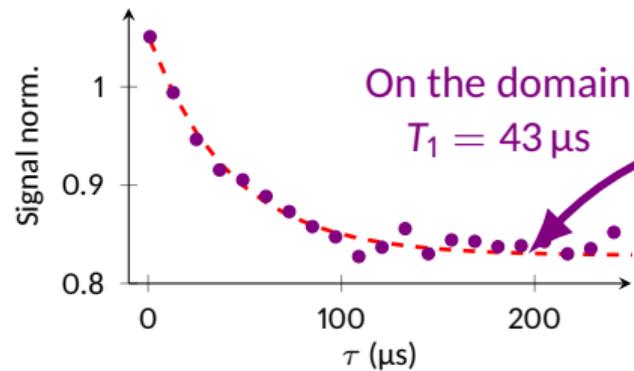
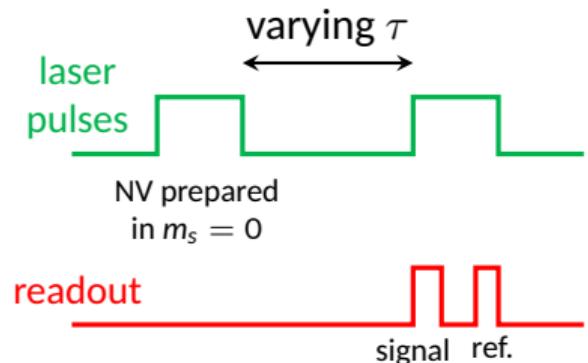
Measurements of the spin relaxation time T_1



Measurements of the spin relaxation time T_1



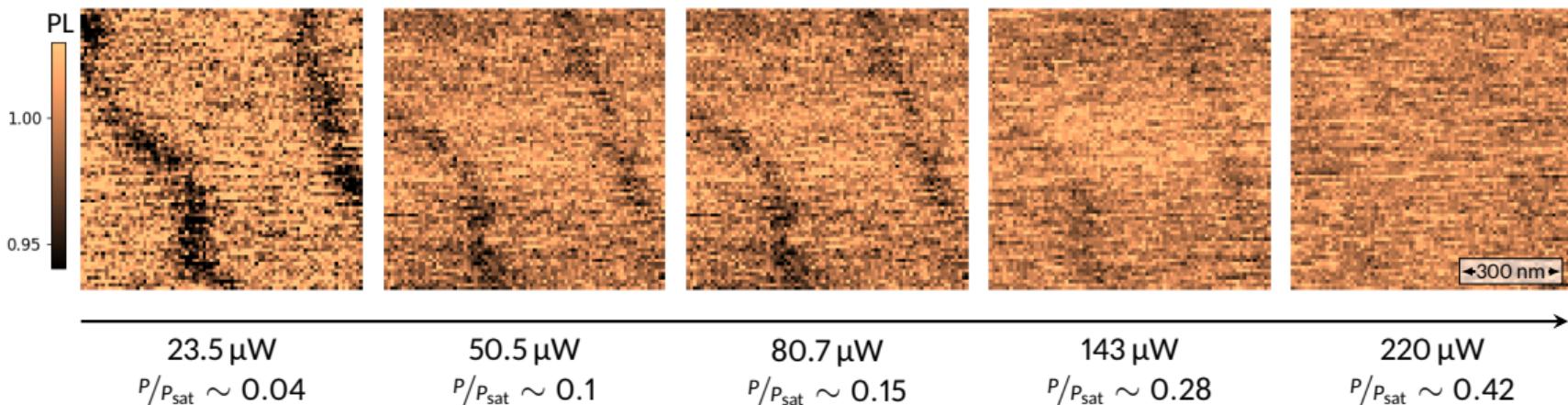
Measurements of the spin relaxation time T_1



Optical power dependence

Increase of the optical power

- More efficient polarization of the NV in the bright $m_s = 0$ state
- Effect of the spin relaxation enhancement less visible



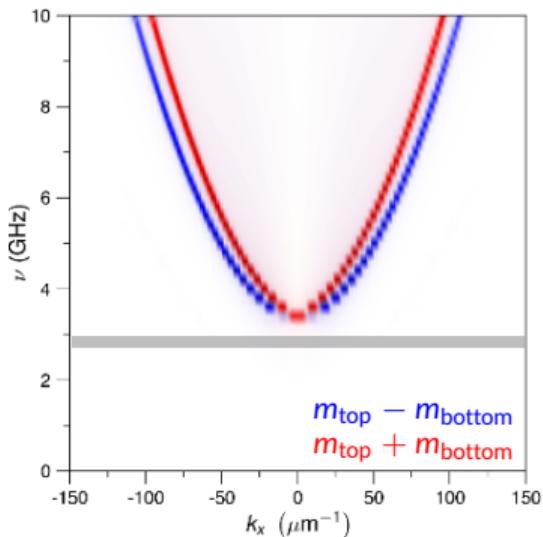
Stronger contrast for low optical power!

Magnon dispersion

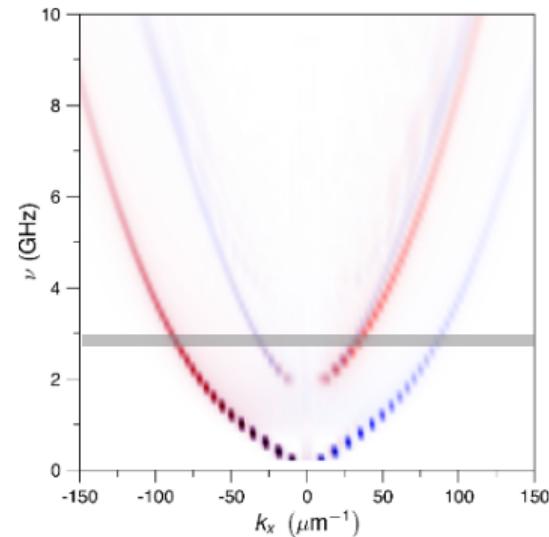
Simulation parameters:

- $M_s = 1.1 \text{ MA m}^{-1}$
- $A = 10 \text{ pJ m}^{-1}$
- $K = 0.8 \text{ MJ m}^{-3}$
- $D = 0.5 \text{ mJ m}^{-2}$
- $A_{\text{RKKY}} = 0.19 \text{ mJ m}^{-2}$

In the domains



In the domain walls



Calculations by
J.-V. Kim

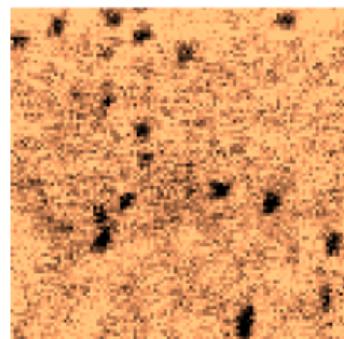
We probe inside the gap!

We can probe thermally
activated magnons!

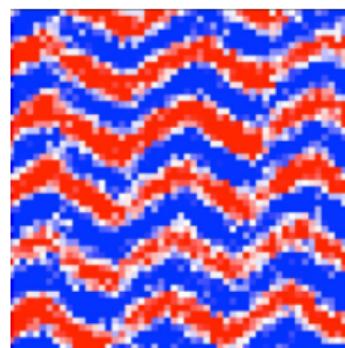
Summary

With a scanning NV-magnetometer, you can:

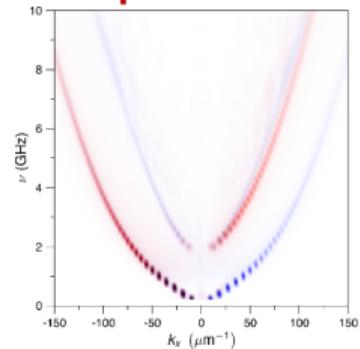
Study ferromagnets **at zero field** in “quenching” mode



Investigate **antiferromagnets** in quantitative mode



Detect magnetic structures **using their excitation spectrum**



Stabilization of zero-field skyrmions in exchanged-bias layers

Exploration of the strain-dependent phase diagram of BiFeO₃ in real-space

New imaging mode based on magnetic noise

Acknowledgments

L2C, Montpellier

Angela Haykal
Rana Tanos
Saddem Chouaieb
Florentin Fabre
Waseem Akhtar
Vincent Jacques

Spintec, Grenoble

Gaurav Rana
Olivier Boulle
Liliana
Buda-Prejbeanu

UMR CNRS/Thales, Palaiseau

Johanna Fischer
Cécile Carrétéro
Manuel Bibes
Stéphane Fusil
Vincent Garcia

CEA, Saclay

Jean-Yves Chauleau
Théophile Chirac
Michel Viret
Synchrotron Soleil
Nicolas Jaouen
C2N, Palaiseau
Joo-Von Kim

UMR CNRS/Thales, Palaiseau

William Legrand
Fernando Ajejas
Yannis Sassi
Karim
Bouzehouane
Nicolas Reyren
Vincent Cros



European Research Council
Established by the European Commission



DEFENSE ADVANCED
RESEARCH PROJECTS AGENCY

