

# Exploring Spins at Surfaces by Spin-Polarized STM

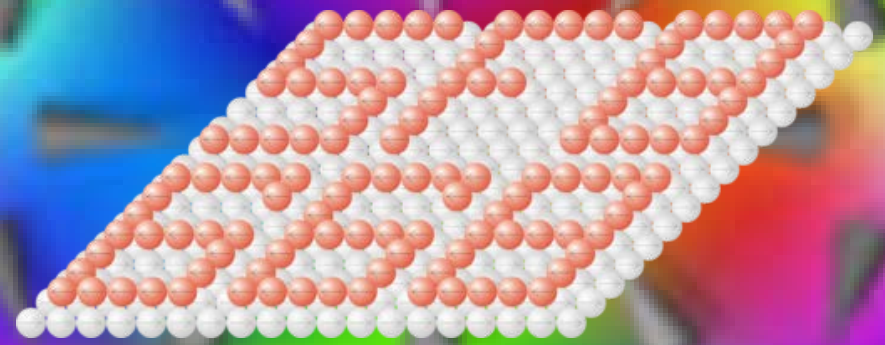
**Roland Wiesendanger**

ERC Advanced Research Group FUIRORE  
Interdisciplinary Nanoscience Center Hamburg  
University of Hamburg

Jungiusstrasse 11, D-20355 Hamburg, Germany



***www.nanoscience.de***

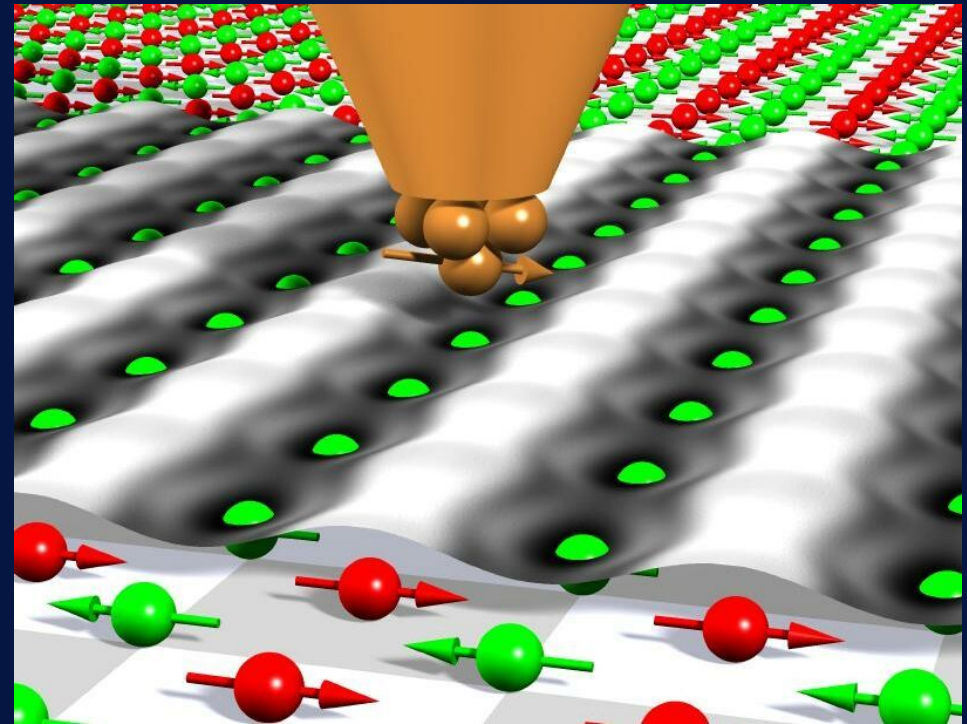


# Spin-Polarized STM for Revealing & Manipulating Complex Spin Textures on the Atomic Scale

## Correlation between

- atomic structure
- electronic structure
- spin structure

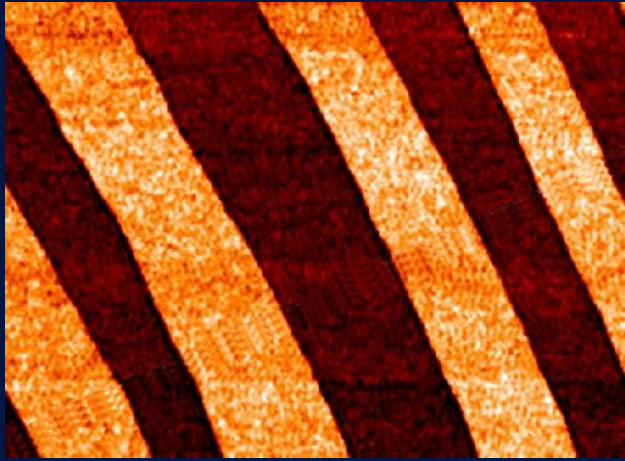
at ultimate spatial, time  
and energy resolution !



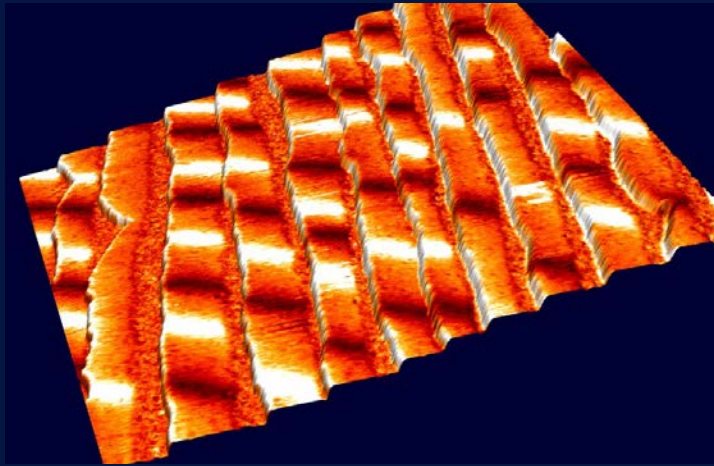
$$I_{sp} = I_0 [1 + P_S P_T \cos(\vec{M}_S, \vec{M}_T)]$$

- R. Wiesendanger *et al.*, Phys. Rev. Lett. **65**, 247 (1990)
- R. Wiesendanger *et al.*, Science **255**, 583 (1992)
- R. Wiesendanger, Rev. Mod. Phys. **81**, 1495 (2009)

# Milestones in the Development & Application of SP-STM



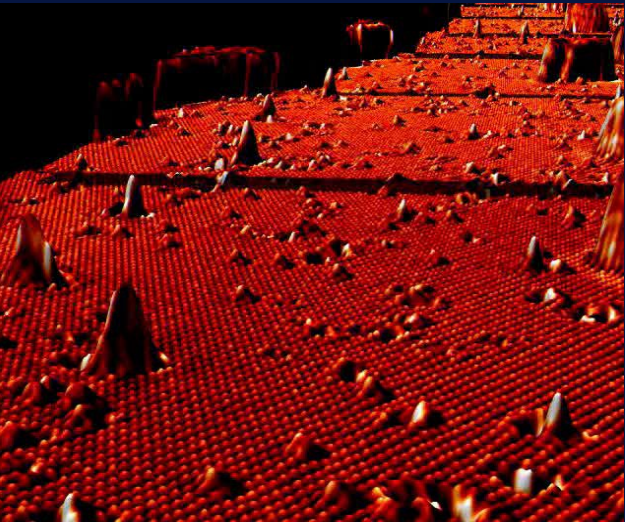
Phys. Rev. Lett. **65**, 247 (1990)  
Phys. Rev. Lett. **85**, 4606 (2000)



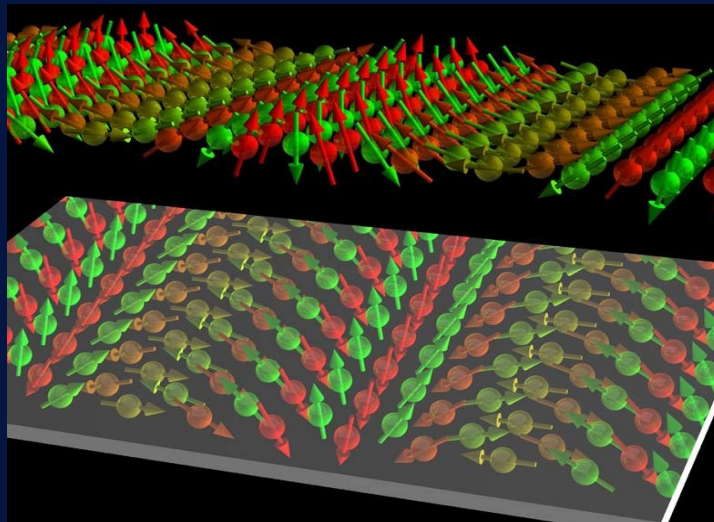
Science **292**, 2053 (2001)  
Phys. Rev. Lett. **88**, 057201 (2002)



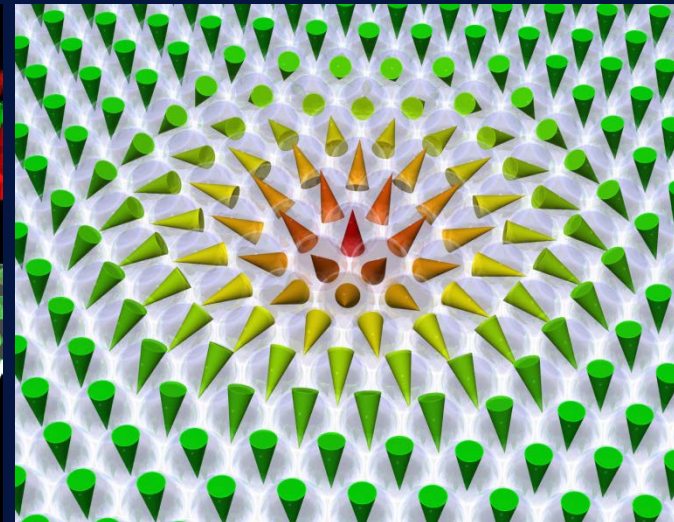
Science **298**, 577 (2002)



Science **255**, 583 (1992)  
Science **288**, 1805 (2000)



Nature **447**, 190 (2007)  
Phys. Rev. Lett. **101**, 027201 (2008)

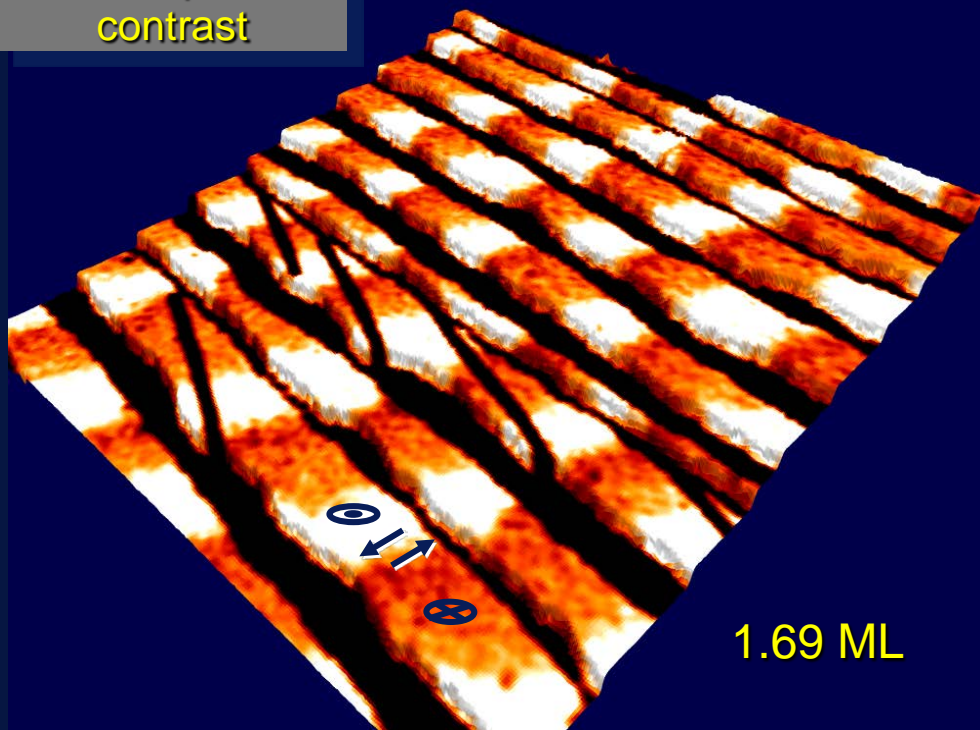


Nature Phys. **7**, 713 (2011)  
Science **341**, 6146 (2013)

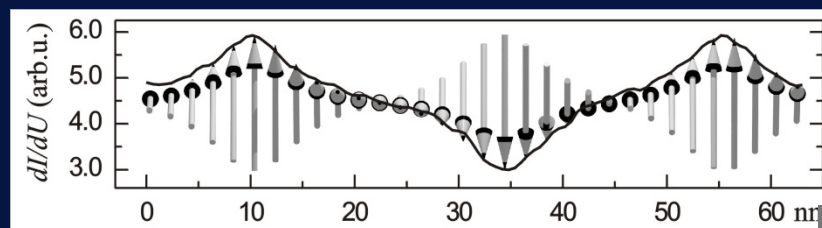
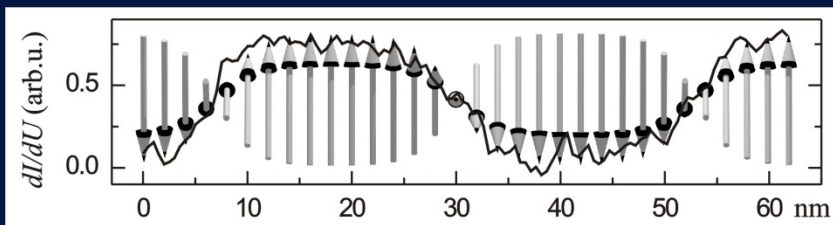
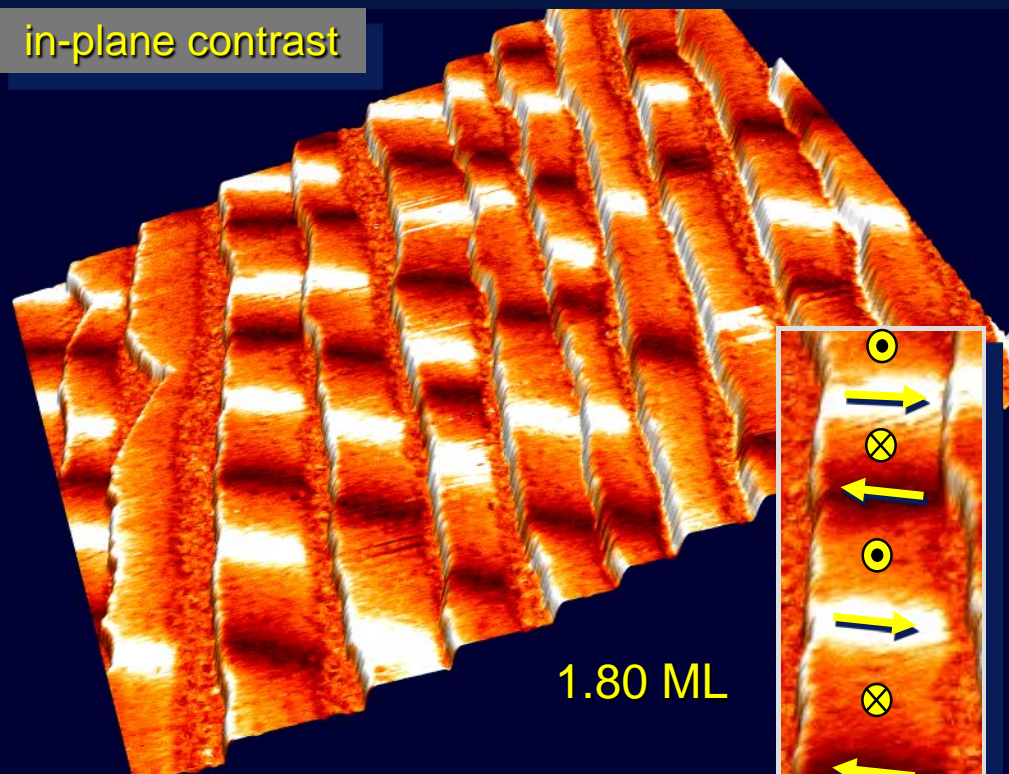
# Domain Structure of Double-Layer Fe Nanowires

3D-Composites of 200 nm × 200 nm  
topography (height) and magn. signal (colour)

out-of-plane  
contrast



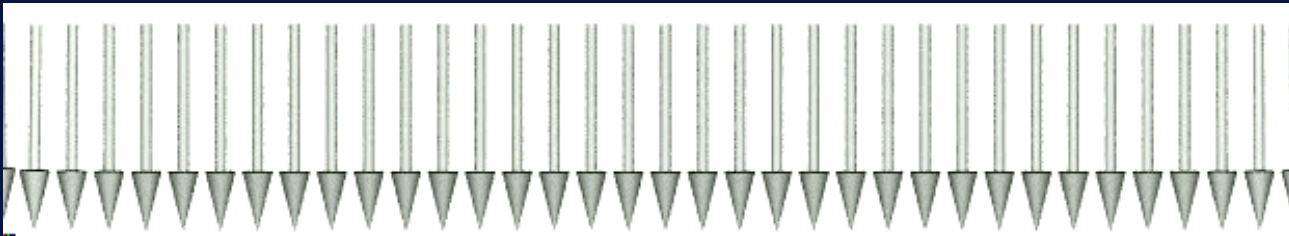
in-plane contrast



[110]

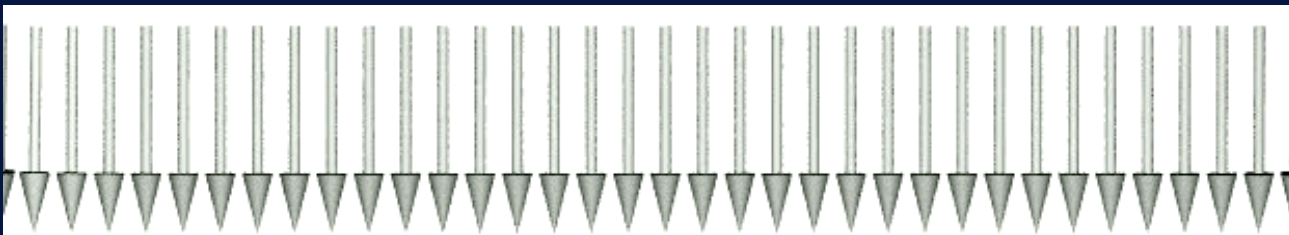
# Pairs of Winding and Unwinding Walls

## unwinding walls



- opposite sense of rotation
- domains can be annihilated

## winding walls

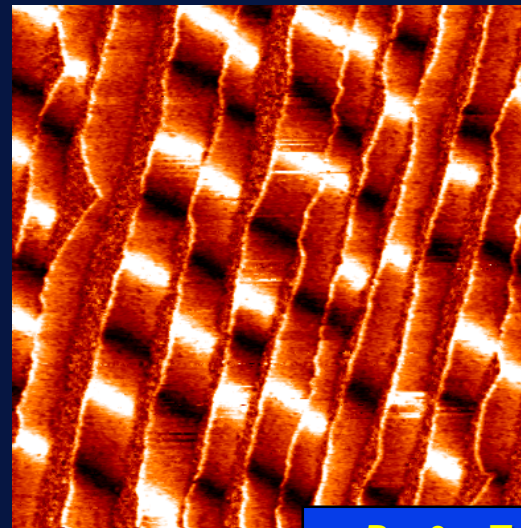


- equal sense of rotation
- stable in saturation fields

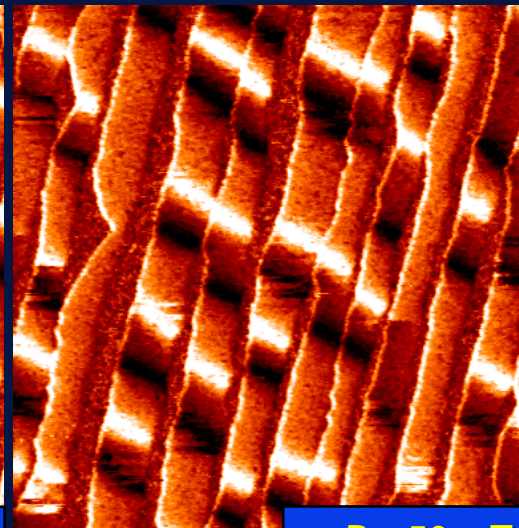
→ indistinguishable in their *perpendicular* component,  
but **distinguishable** in their *in-plane* component.

# Chiral Domain Walls in a perpendicular Field

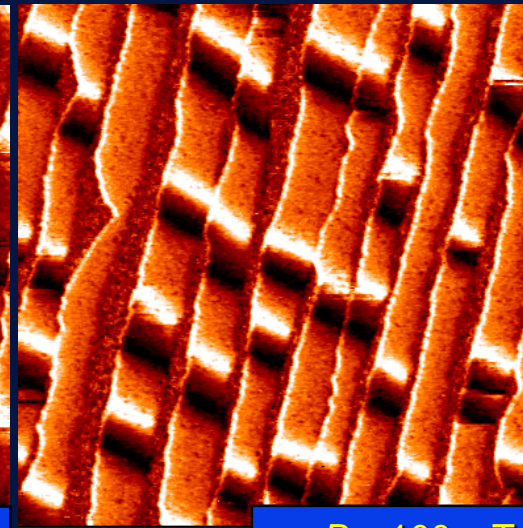
(unique rotational sense is a consequence of DM-interaction)



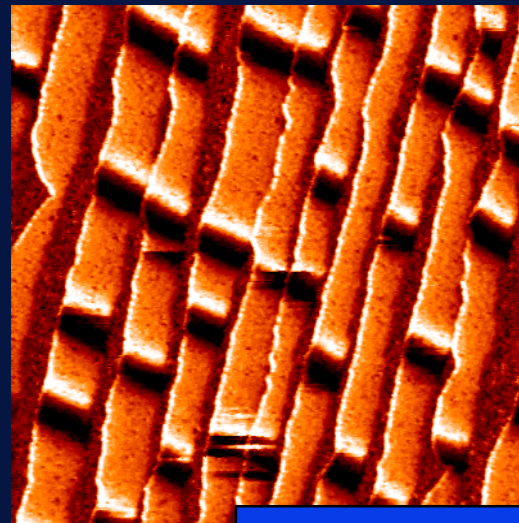
$B = 0 \text{ mT}$



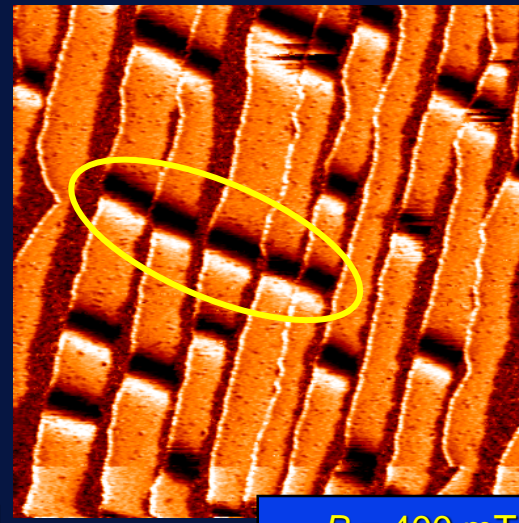
$B = 50 \text{ mT}$



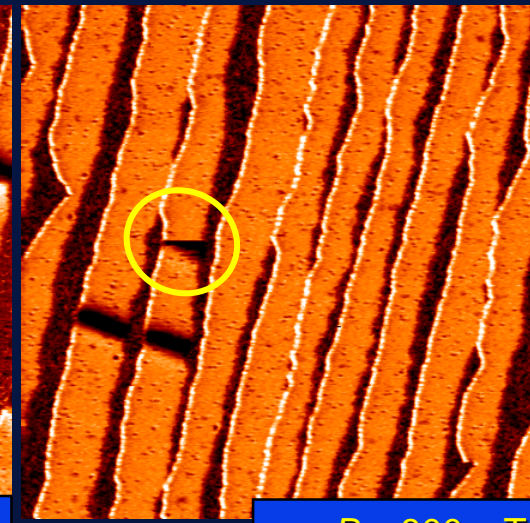
$B = 100 \text{ mT}$



$B = 200 \text{ mT}$



$B = 400 \text{ mT}$



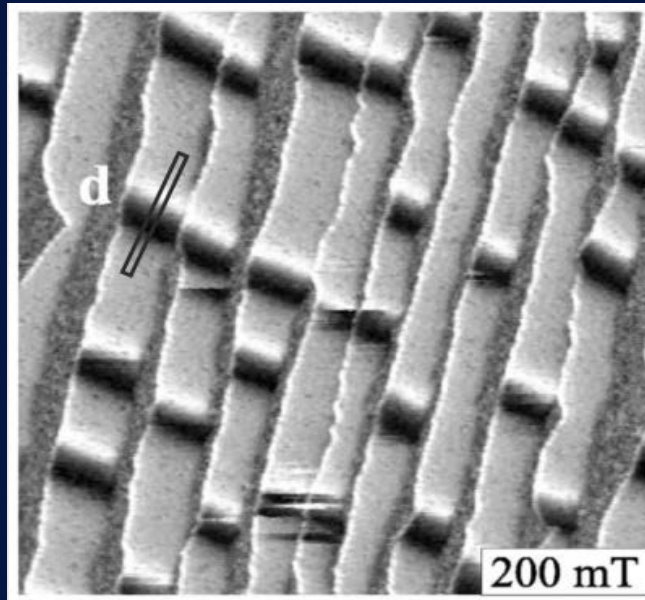
$B = 800 \text{ mT}$

1.8 ML Fe/W(110)

200 x 200 nm<sup>2</sup>

# Atomic-Scale Profile of 360° Domain Walls: Comparison of Experiment with Theory

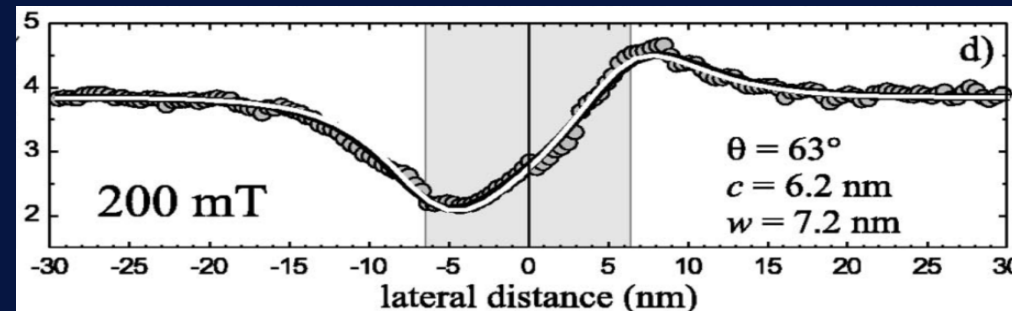
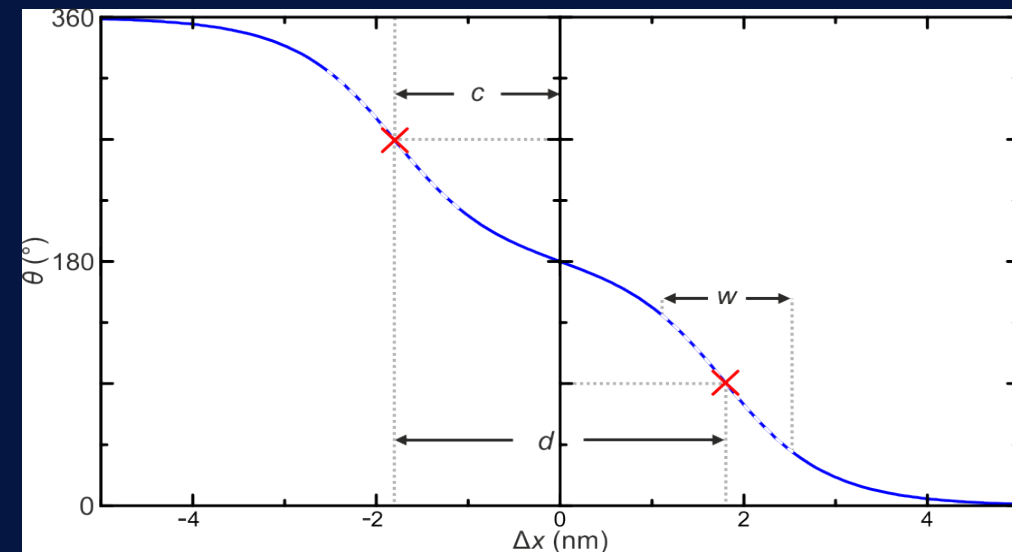
Chiral 360°-domain walls in DL Fe/W(110):



O. Pietzsch *et al.*:  
 Science **292**, 2053 (2001)  
 A. Kubetzka *et al.*:  
 PRL **88**, 057201 (2002)  
 A. Kubetzka *et al.*:  
 Phys. Rev. B **67**, 020401 (2003)  
 E. Y. Vedmedenko *et al.*:  
 Phys. Rev. B **75**, 104431 (2007)

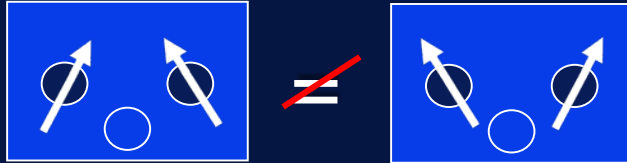
Analytical formula for two 180° domain walls:

$$\theta(\rho, c, w) = \begin{cases} \sum_{+,-} \left[ \arcsin \left( \tanh \frac{-\rho \pm c}{w/2} \right) \right] + \pi & |B_z > 0 \\ \sum_{+,-} \left[ \arcsin \left( \tanh \frac{-\rho \pm c}{w/2} \right) \right] & |B_z < 0 \end{cases}$$



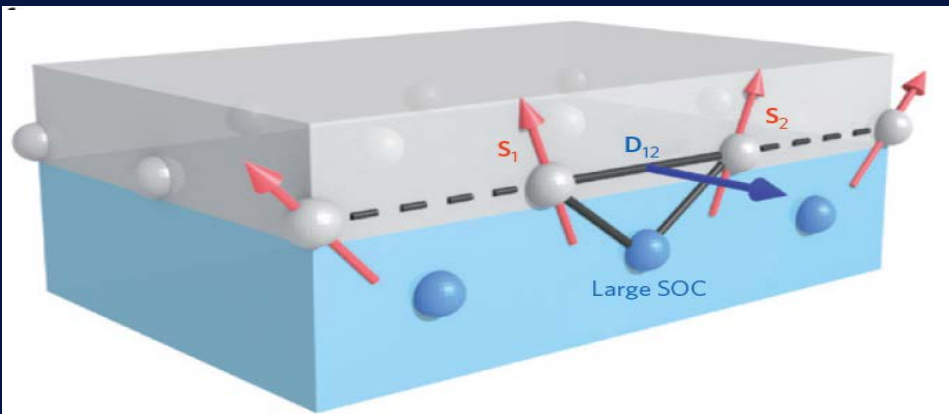
# Interface-Driven Non-collinear Spin States

Dzyaloshinskii-Moriya interaction  
due to spin-orbit coupling



when inversion symmetry is broken

**DM always to be considered  
at surfaces and interfaces**



$$E_{\text{DM}} = \sum_{i,j} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$$

I. Dzyaloshinskii,  
J. Phys. Chem. Solids **4**, 241 (1958).  
T. Moriya, Phys. Rev. **120**, 91 (1960).

## SPSTM:

- chiral domain walls (2001-2007)
- cycloidal spin spirals with unique rotational sense (since 2007)
- chiral skyrmions (since 2011)



A. Fert, in:  
Materials Science Forum, **59-60**, 439 (1990).  
A. Crépieux and C. Lacroix,  
J. Magn. Magn. Mat. **182**, 341 (1990).

M. Bode et al., Nature **447**, 190 (2007)  
P. Ferriani et al., PRL **101**, 27201 (2008)  
S. Meckler et al., PRL **103**, 157201 (2009)  
M. Menzel et al., PRL **108**, 197204 (2012)

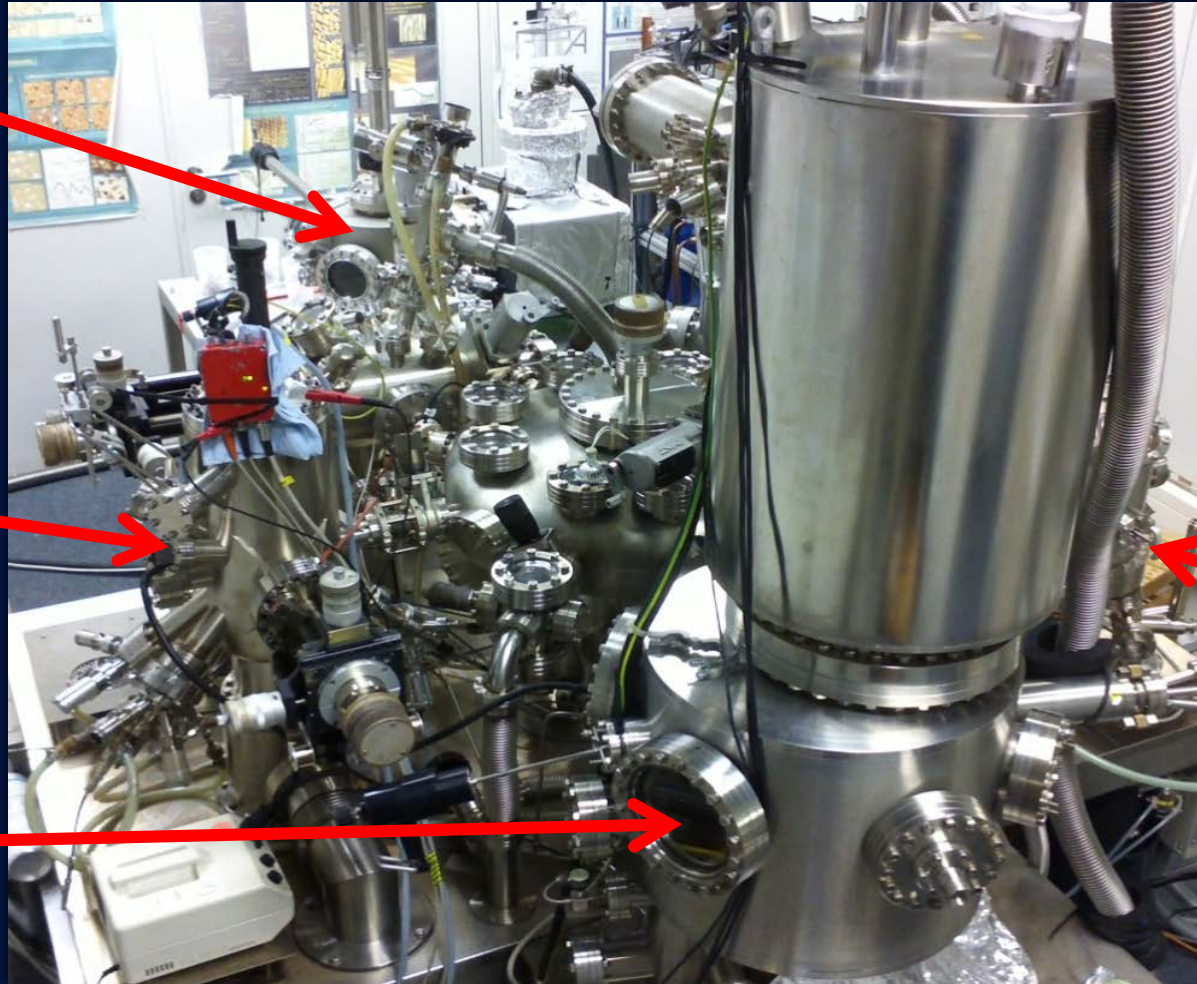


# NANOLAB in Hamburg: Metal-MBE combined with SP-STM

SP-STM  
 $T = 1.3 - 4.2 \text{ K}$   
 $B_{\perp} = 9 \text{ T}$

MBE-STM  
 $T = 300 \text{ K}$

SP-STM  
 $T = 8 - 13 \text{ K}$   
 $B_{\perp} = 2.5 \text{ T}$

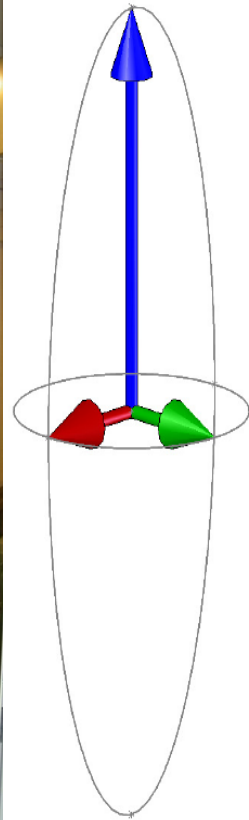
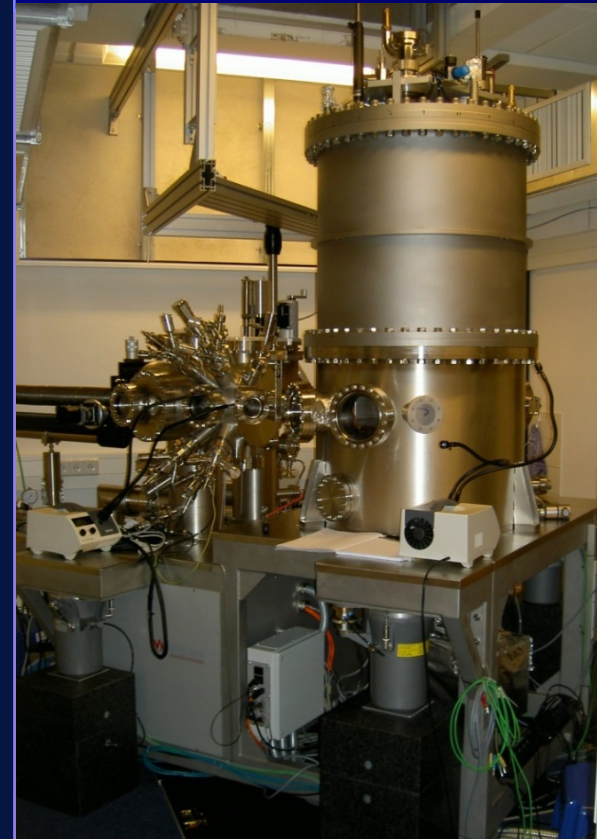


VT-STM  
 $T = 30 - 1000 \text{ K}$

→ *in situ* preparation of STM tip and sample in UHV

# 3D Control of Tip Magnetization & Magnetic Contrast

## Setup and Magnetic Field Range



Field strengths  
single direction:

$B_x, B_y: 1.3 \text{ T}$

$B_z: 5 \text{ T}$

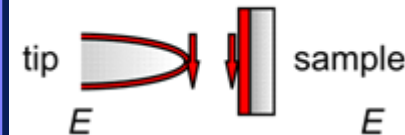
3D mode:

$B_x, B_y: 1 \text{ T}$

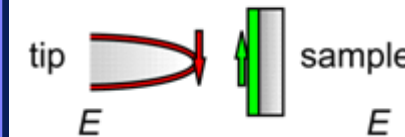
$B_z: 3.5 \text{ T}$

## Contrast Mechanism

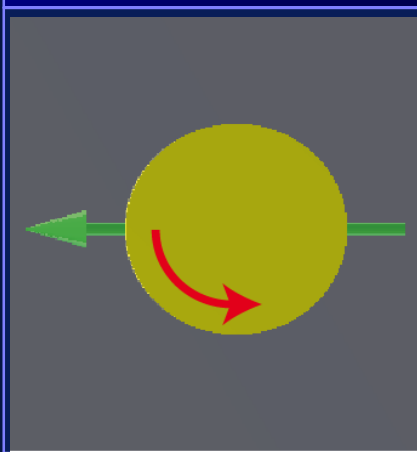
parallel configuration



antiparallel configuration



## Top View of the Tip



Alignment of the  
tip-magnetization  
in the external field



Investigation of the  
absolute magnetization  
direction in the sample is possible

# Magnetization Orientation in the Domain Walls

*In-Plane Magnetic Field Rotation*



→ Right-rotating cycloidal spin spiral !  
(S. Meckler et al., PRL 103, 157201 (2009))

# Cycloidal Spin Spiral observed for 1 ML Mn on W(110)



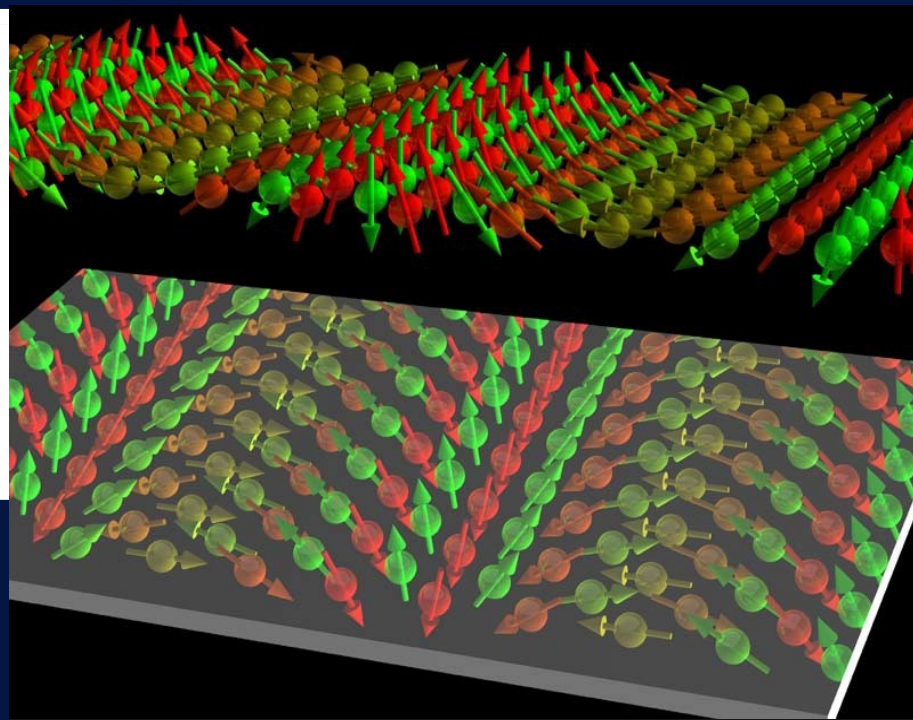
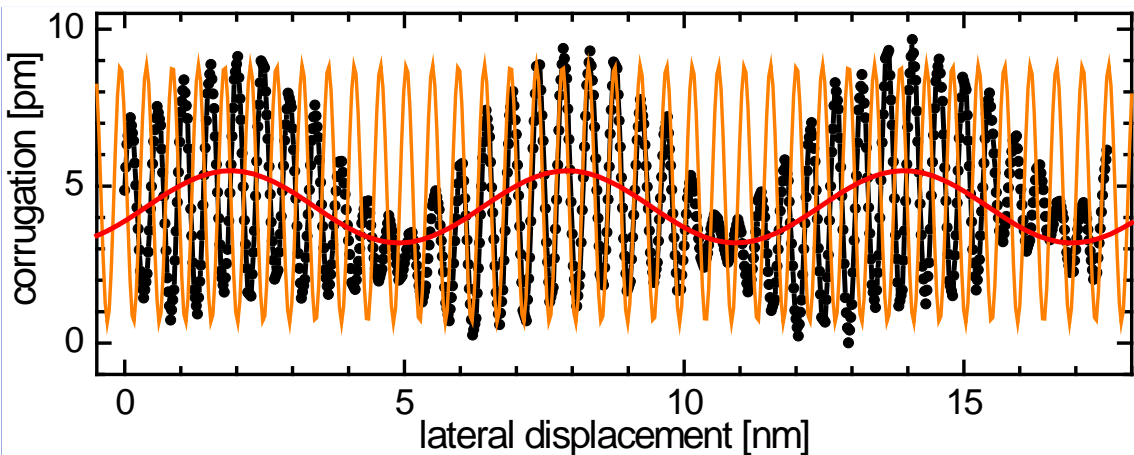
tip: Cr-coated W-tip

sample: 0.8 ML Mn/W(110)

STM:  $U_{\text{bias}} = +3 \text{ mV}$

$I_{\text{tun}} = 15 \text{ nA}$

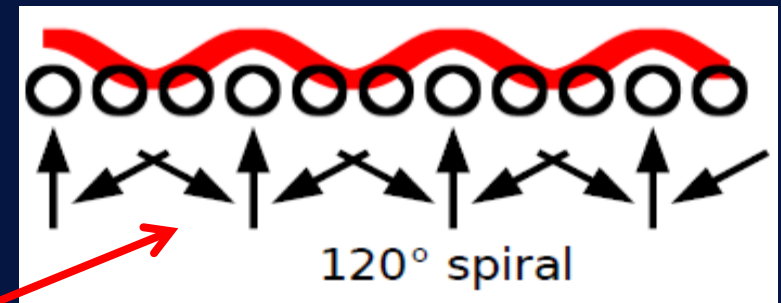
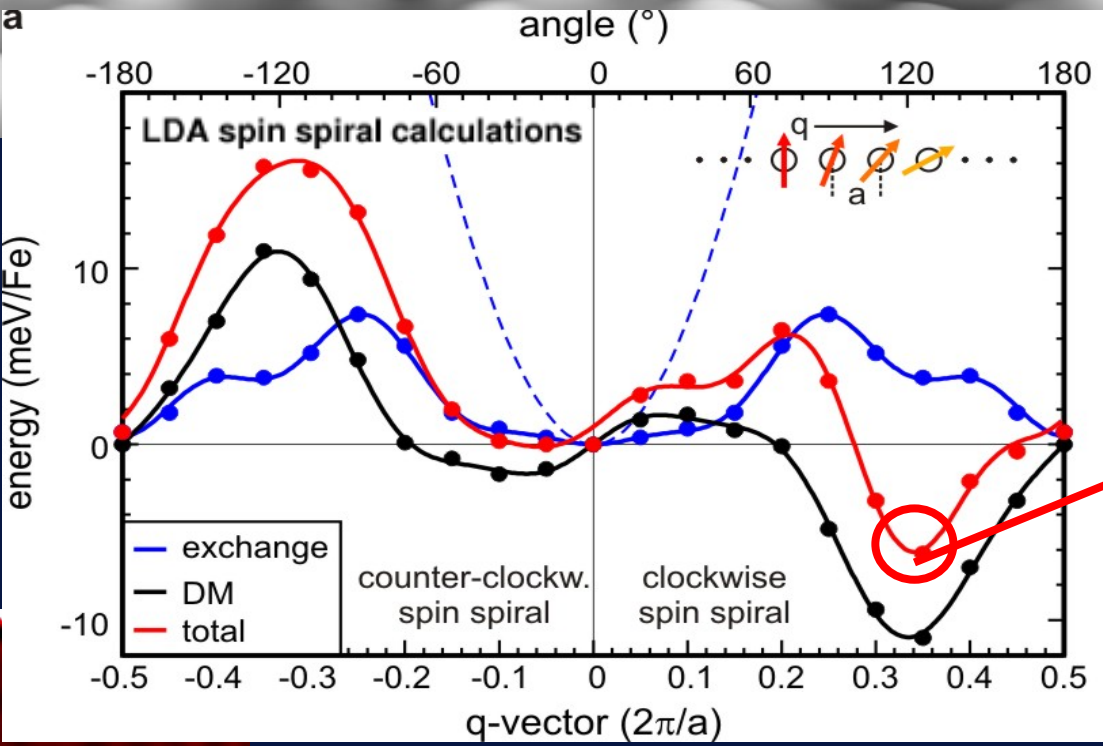
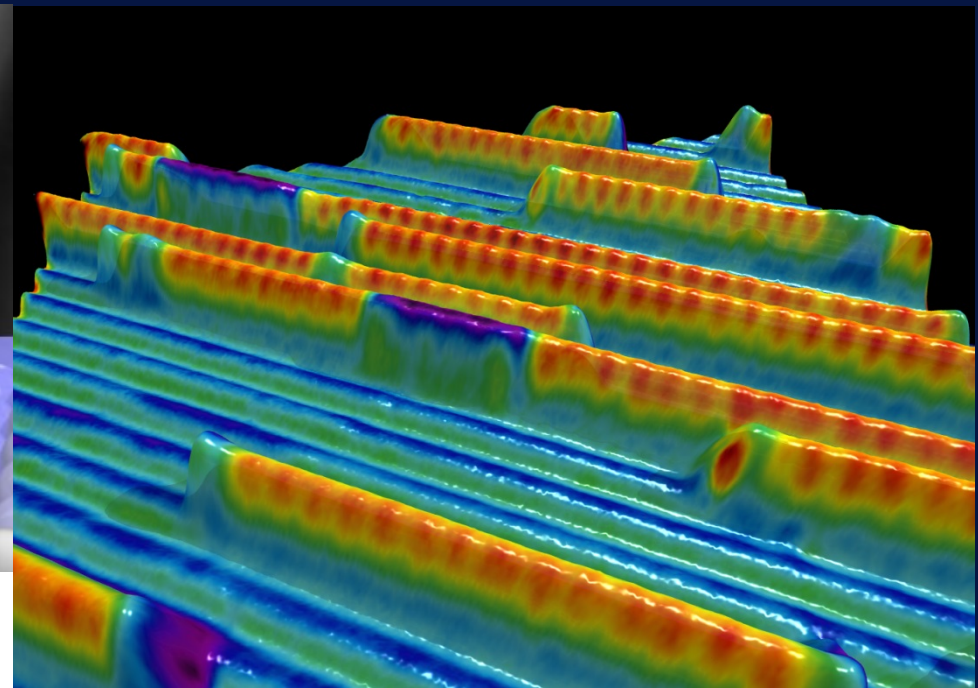
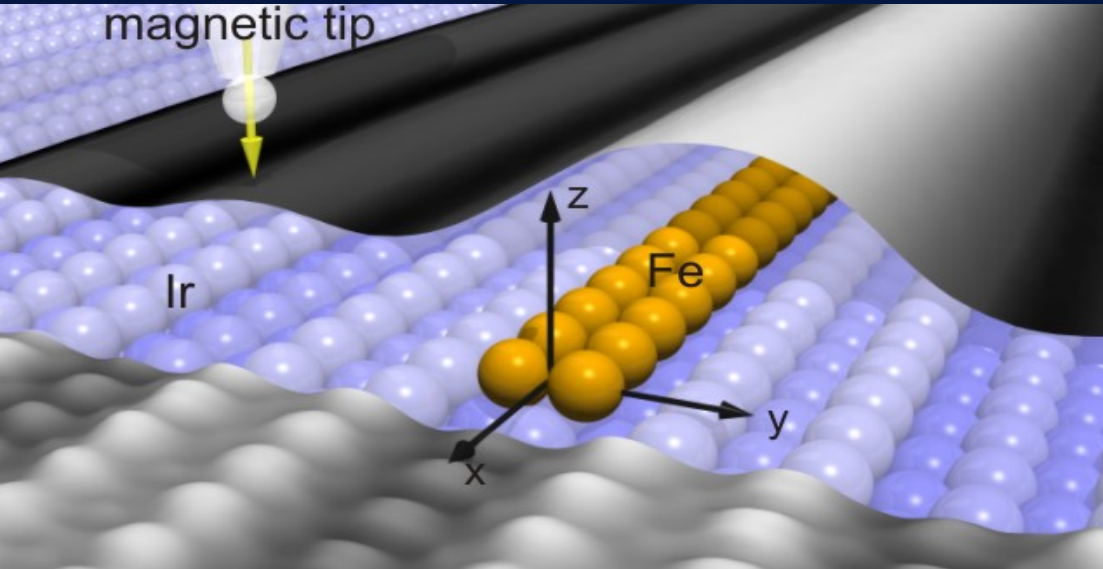
$T = 13 \text{ K}$



M. Bode et al., Nature 447, 190 (2007)



# 120° Spin Spiral in Bi-Atomic Fe Chains on Ir(001)

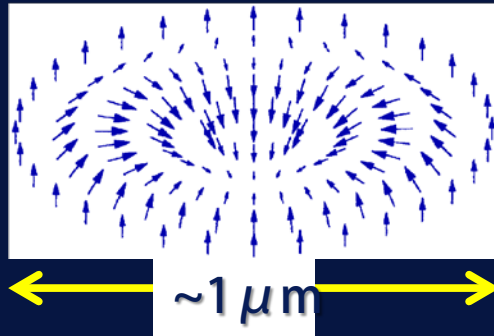


→ DM-interaction is responsible for chiral spin spiral formation !

M. Menzel et al., PRL 108, 197204 (2012)

# Chiral Magnetic Skyrmions: From Theoretical Predictions to Experimental Observations

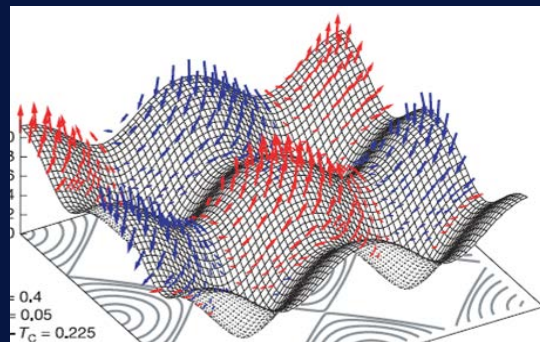
Theoretical predictions:



~1  $\mu\text{m}$

→ stable defect of FM film

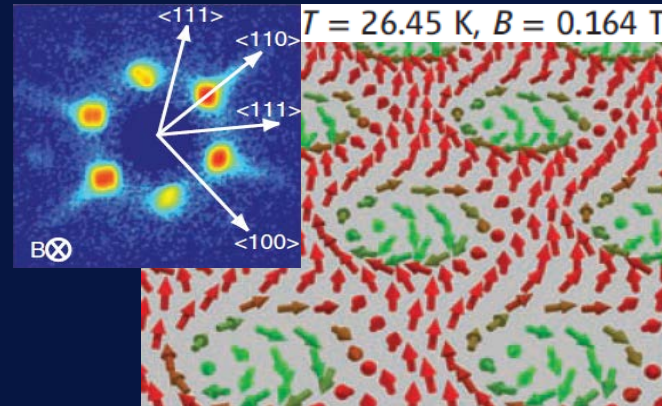
□ A. Bogdanov & D. Yablonskii, Sov. Phys. JETP 68, 101 (1989)



→ stable lattices with modulated

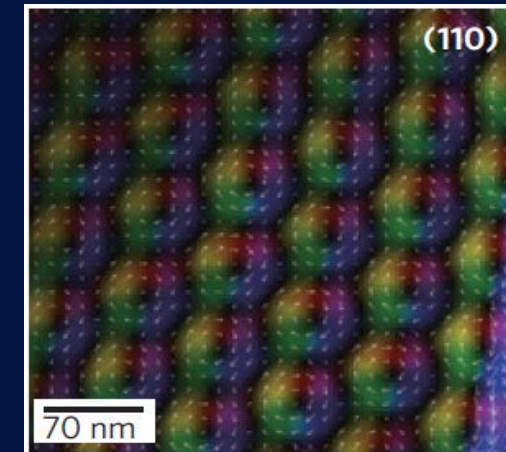
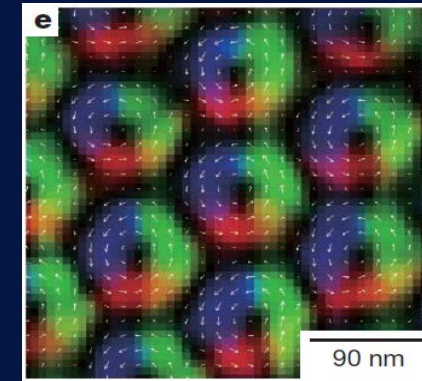
$M$  □ U.K. Rößler *et al.*, Nature 442, 797 (2006)

Experimental findings:



Skyrmion lattices in bulk crystals lacking inversion symmetry:

MnSi (bulk)  
FeCoSi (~100nm film)  
FeGe (15 — 100nm films)

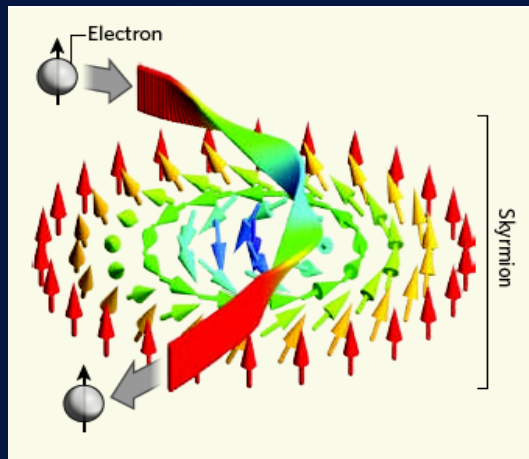


□ S. Mühlbauer *et al.*, Science 323, 915 (2009)  
□ X.Z. Yu *et al.*, Nature 465, 901 (2010)  
□ X.Z. Yu *et al.*, Nature Mater. 10, 106 (2011)

# Motivation for Studying Magnetic Skyrmions

## SPINTRONICS:

Electrons interact very efficiently with non-collinear magnetic states



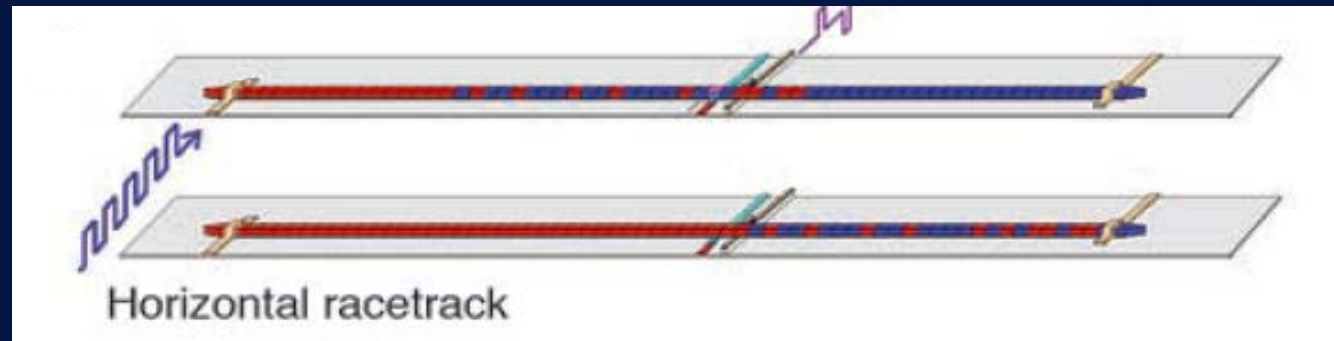
C. Pfleiderer, A. Rosch, Nature **465**, 880 (2010)

How can skyrmions be created and annihilated (or written and deleted) in a controlled fashion?

Do skyrmions exist in ultrathin magnetic films and multilayers?

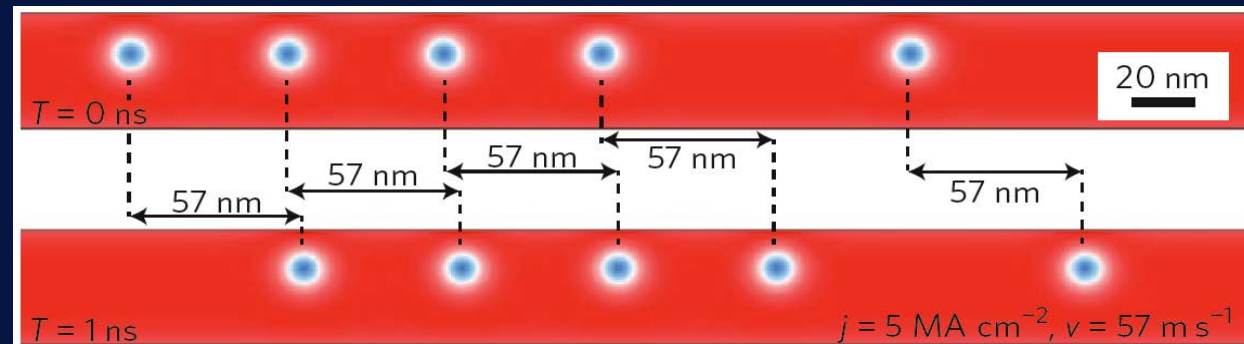
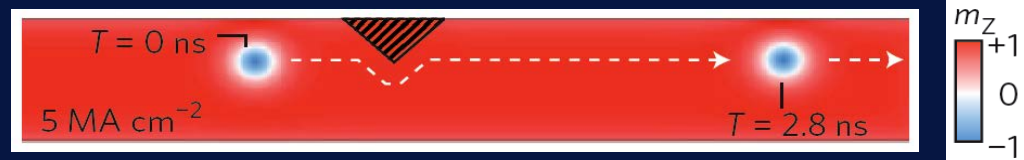
## MAGNETIC MEMORIES:

„Racetrack memory“ based on shifting domain walls



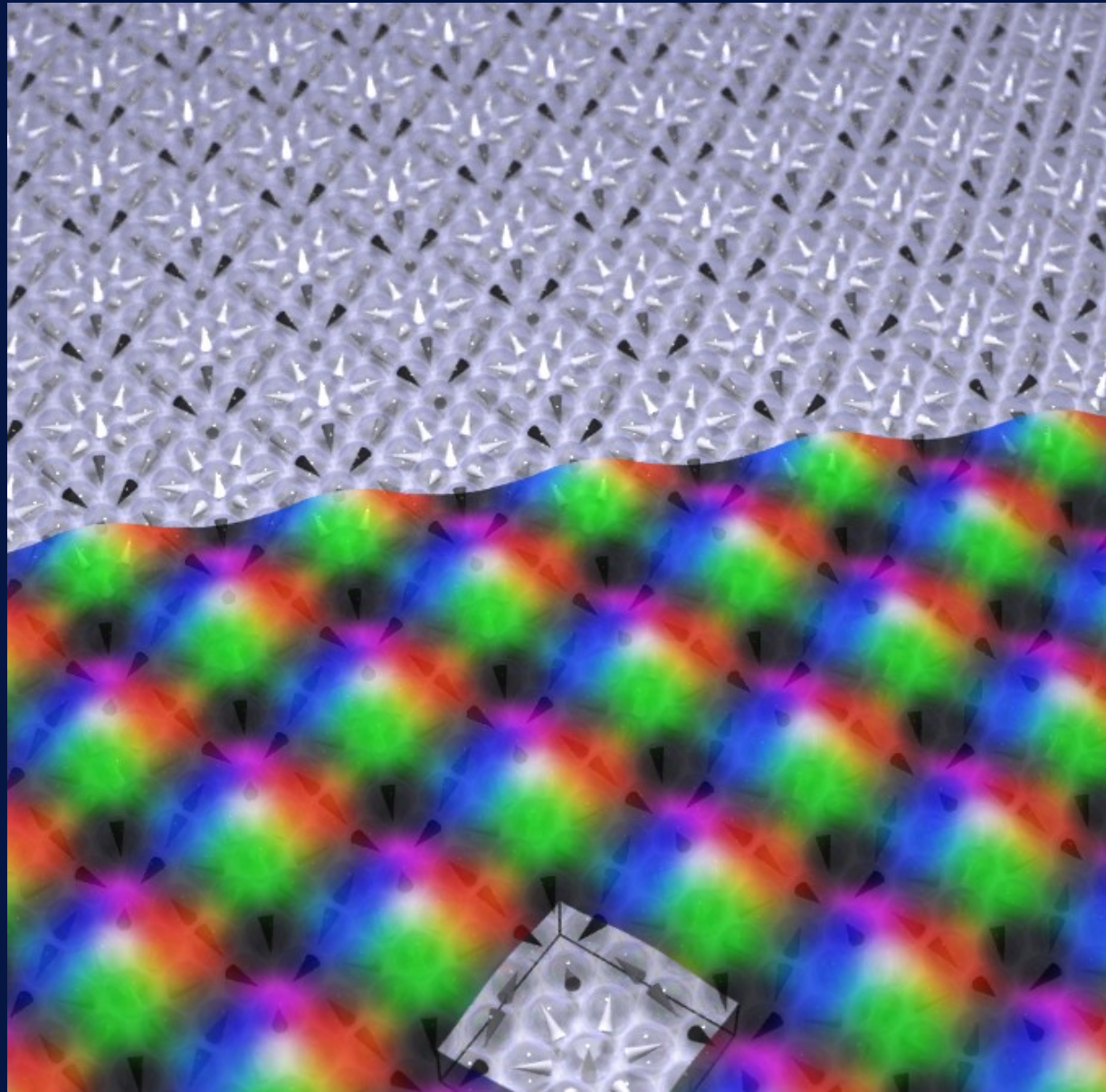
S.S.P. Parkin et al., Science **320**, 190 (2008)

Much smaller current densities needed to move skyrmions:



A. Fert et al., Nature Nanotech. **8**, 152 (2013)

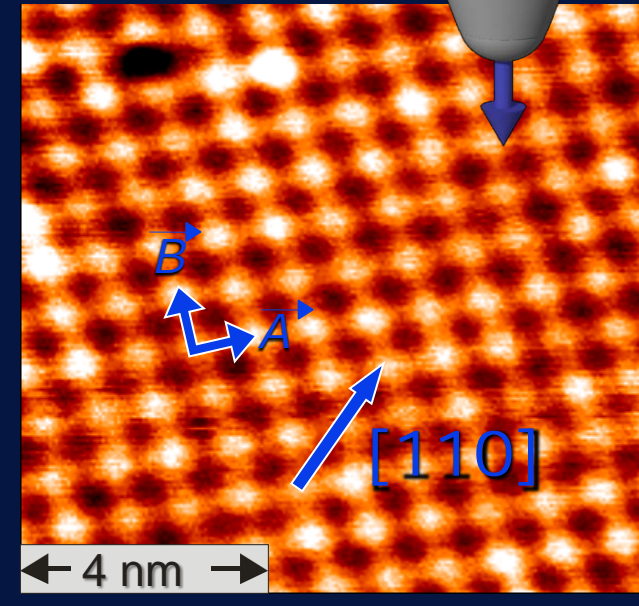
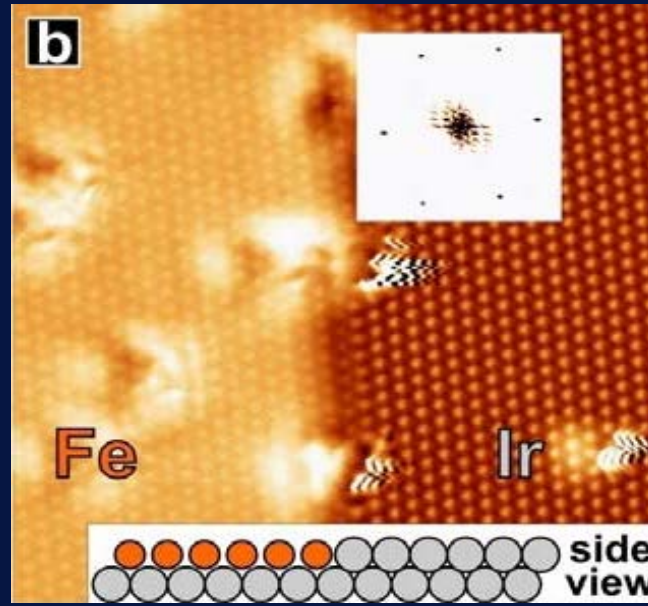
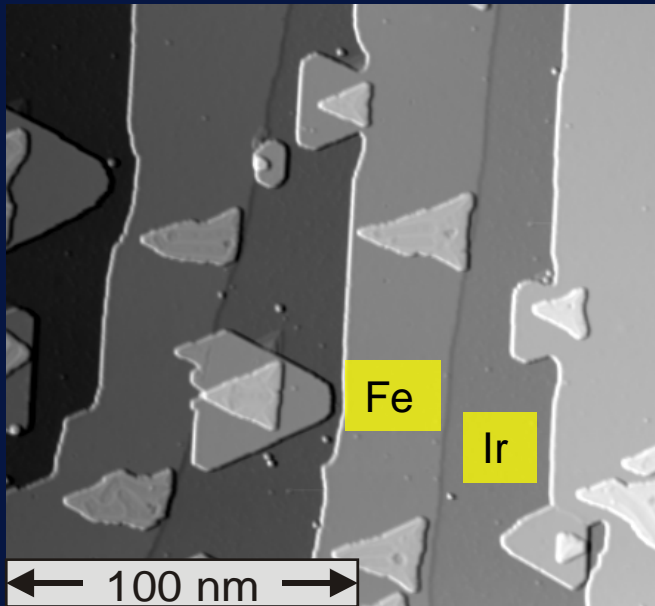
# Discovery of Interface-Driven Chiral Skyrmionic Lattices in a Monolayer of Fe on Ir(111)





# Fe on Ir(111) with out-of-plane magnetized SP-STM tip

topography



0.6 AL Fe on Ir(111)

1<sup>st</sup> AL Fe grows pseudomorphically

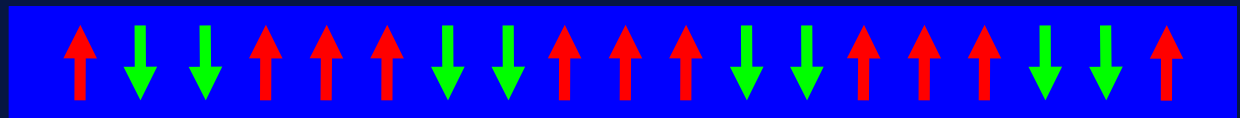
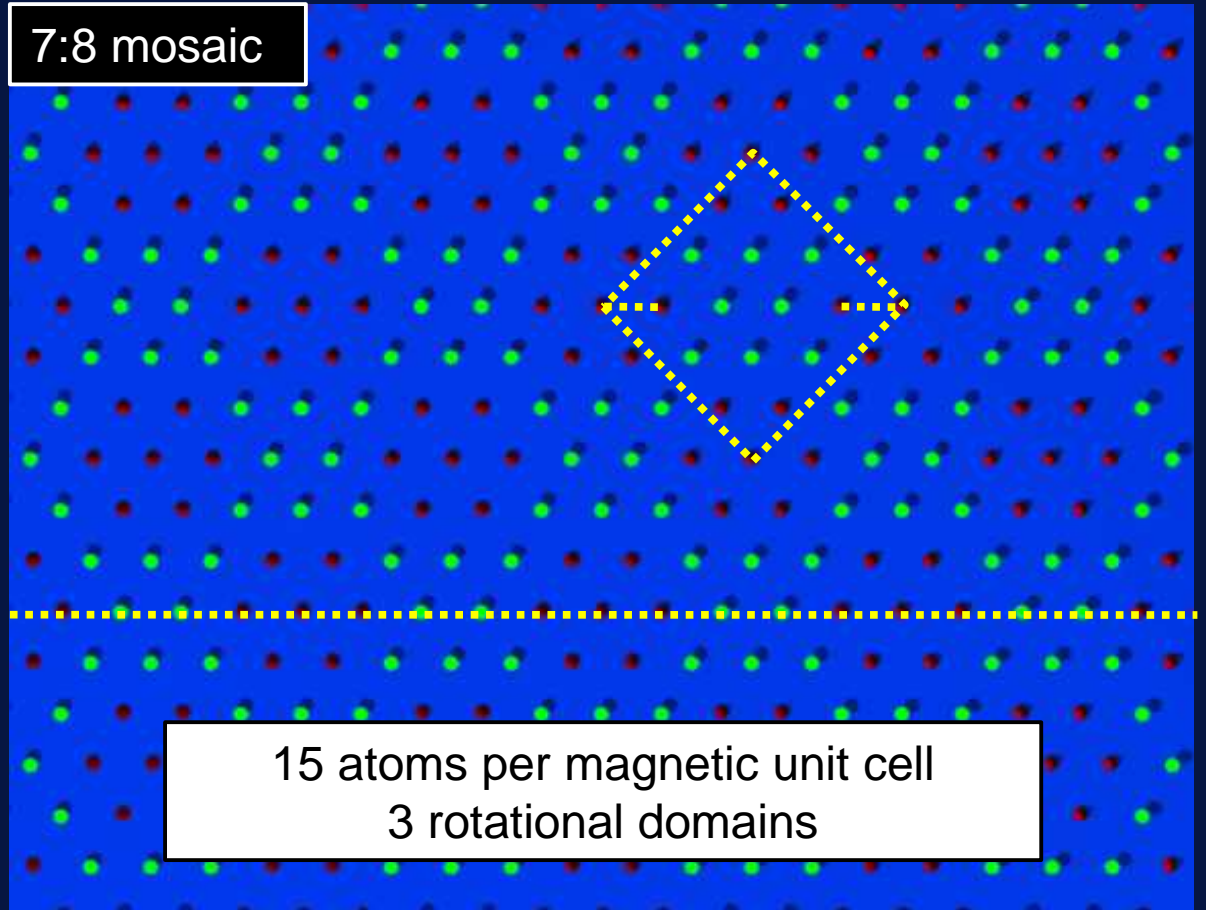
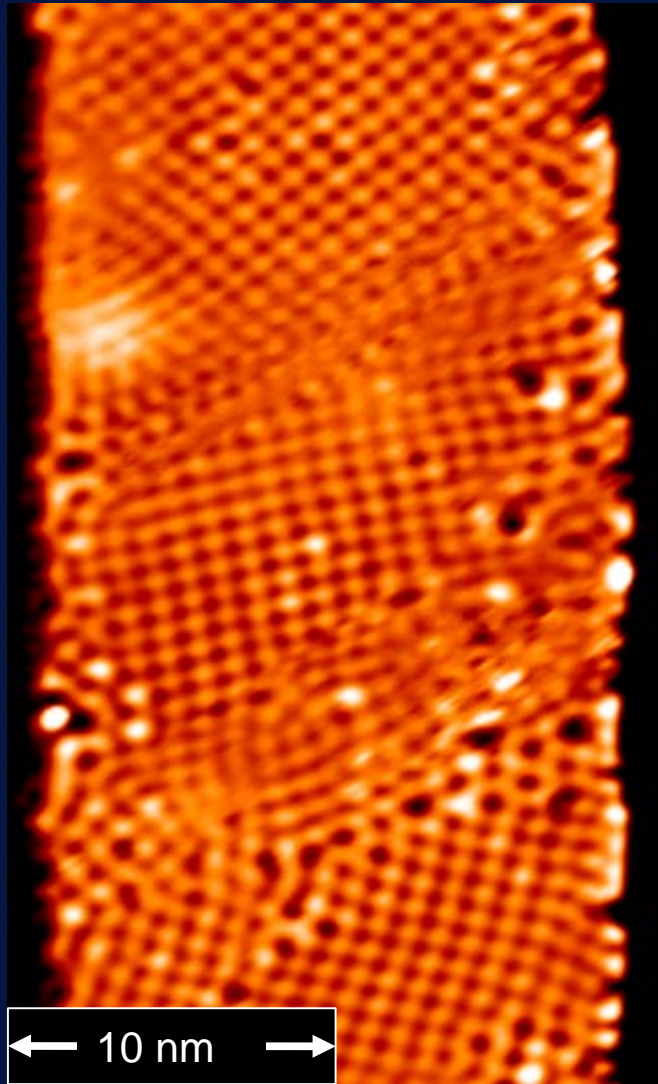
→ hexagonally arranged surface atoms

square magnetic unit cell ~15 atoms  
 $\vec{A}, \vec{B} \approx 1 \text{ nm}, \pm 45^\circ$  to close packed row  
out-of-plane component

K. von Bergmann *et al.*, Phys. Rev. Lett. **96**, 167203 (2006)  
K. von Bergmann *et al.*, New J. Phys. **9**, 396 (2007)



# Fe/Ir(111) with out-of-plane Magnetized SP-STM Tip

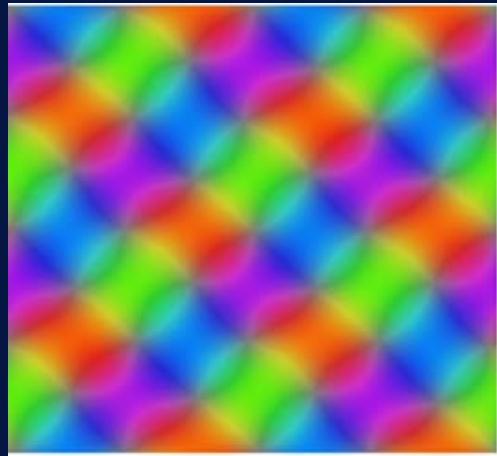


side view

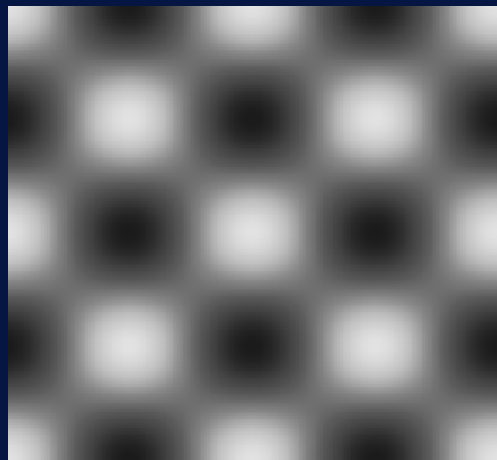
K. von Bergmann *et al.*, Phys. Rev. Lett. **96**, 167203 (2006)  
K. von Bergmann *et al.*, New J. Phys. **9**, 396 (2007)



# 3D Vectorial Spin Map of 1 ML Fe on Ir(111)



in-plane magnetization



out-of-plane magnetization



# Microscopic Origin of the Skyrmion Lattice

$$H = - \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i,j} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) + \sum_i A_i (S_i^z)^2$$

exchange

Dzyaloshinskii-  
Moriya

anisotropy

$$- \sum_{ij} B_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j)^2 - \sum_{ijkl} K_{ijkl} [(\mathbf{S}_i \mathbf{S}_j)(\mathbf{S}_k \mathbf{S}_l) + (\mathbf{S}_j \mathbf{S}_k)(\mathbf{S}_l \mathbf{S}_i) - (\mathbf{S}_i \mathbf{S}_k)(\mathbf{S}_j \mathbf{S}_l)]$$

biquadratic

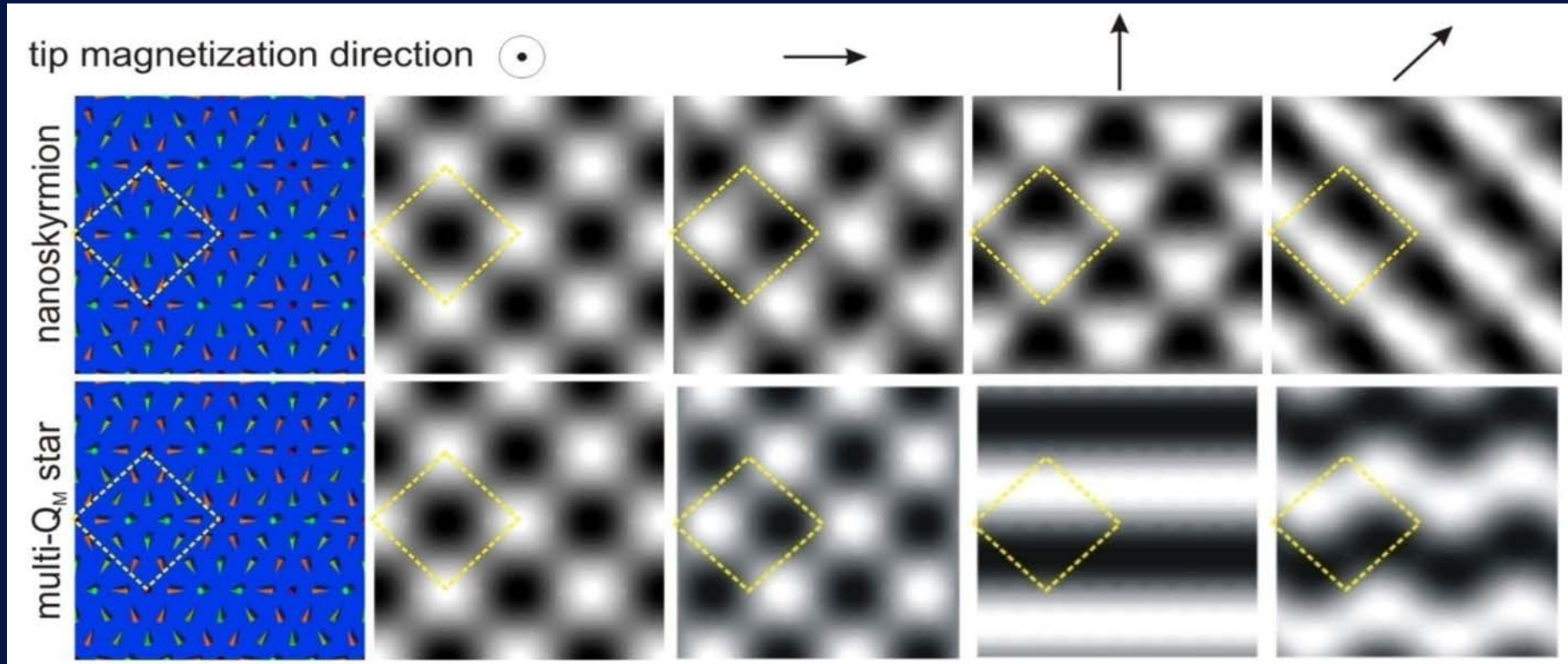
4-spin

- Dzyaloshinskii-Moriya interaction chooses skyrmion lattice out of several possible 2D spin textures for Fe on Ir(111)
- due to 4-spin interaction 2D spin textures are favored over ferromagnetic and 1D spin spiral states

→ Nanoskyrmion lattice is energetically favorable even in zero field !

# Nanoskyrmion Lattice vs. Superposition of Spin Spirals

Simulation of SP-STM images for nanoskyrmion lattice and multi-Q state



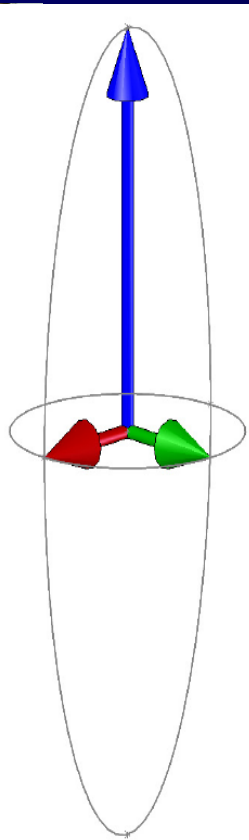
- both states show stripes for certain magnetization directions of the tip
- **but:** distance of stripes in multi-Q state is  $\sqrt{2}$  larger than in experiment and observed stripes are in a different crystallographic direction

→ **multi-Q state can be excluded by comparison of experimental data with simulation**



# 3D Control of Tip Magnetization & Vector-Resolved Spin Contrast

## Setup and Magnetic Field Range



Field strengths  
single direction:

$B_x, B_y: 1.3 \text{ T}$

$B_z: 5 \text{ T}$

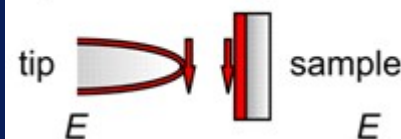
3D mode:

$B_x, B_y: 1 \text{ T}$

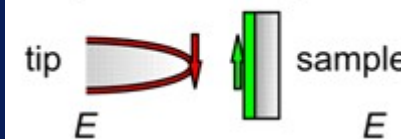
$B_z: 3.5 \text{ T}$

## Contrast Mechanism

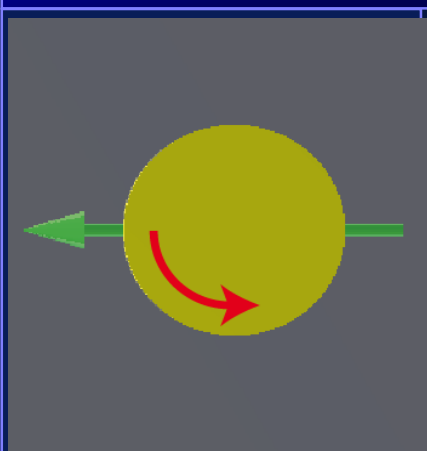
parallel configuration



antiparallel configuration



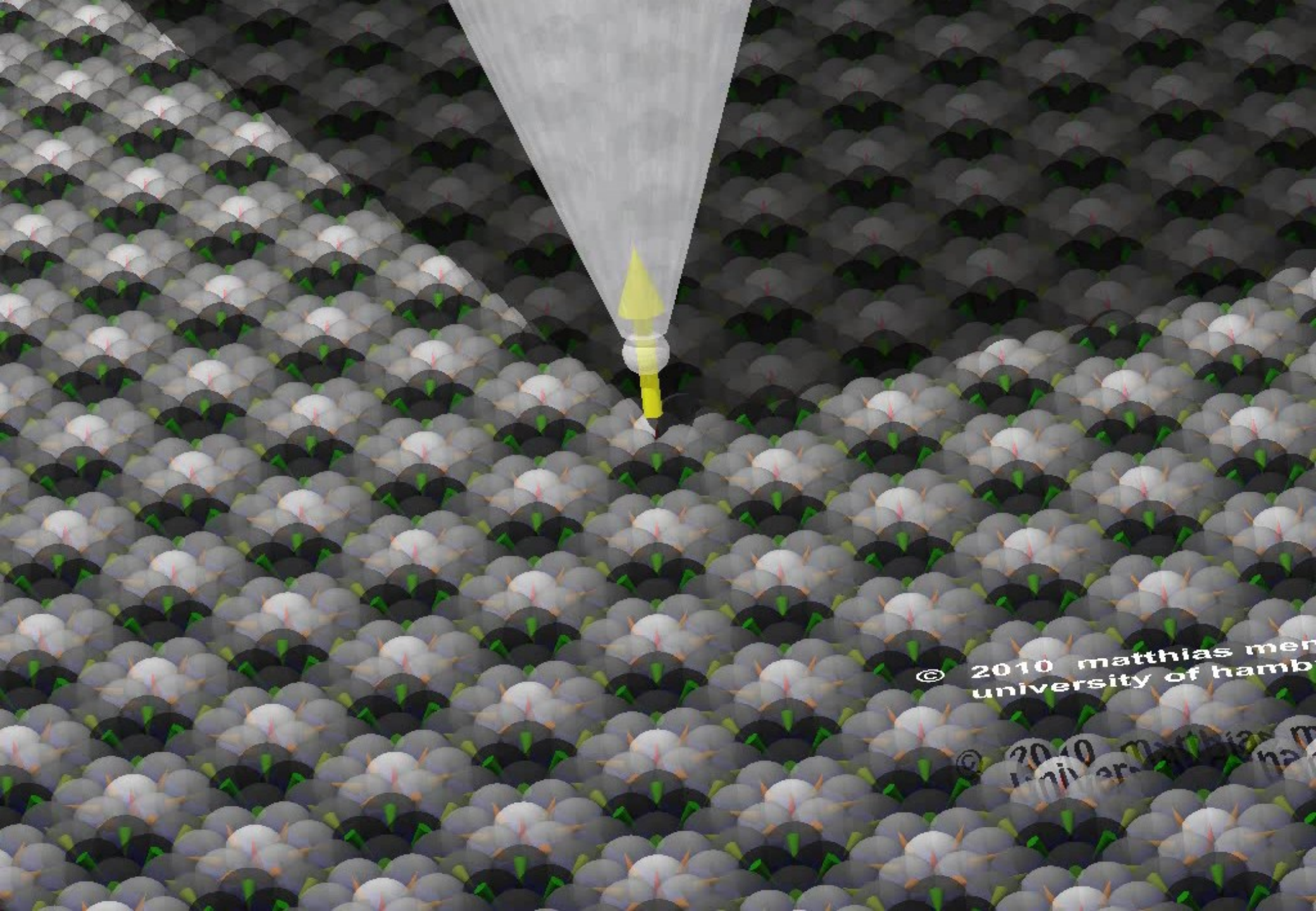
## Top View of the Tip



Alignment of the  
tip-magnetization  
in the external field



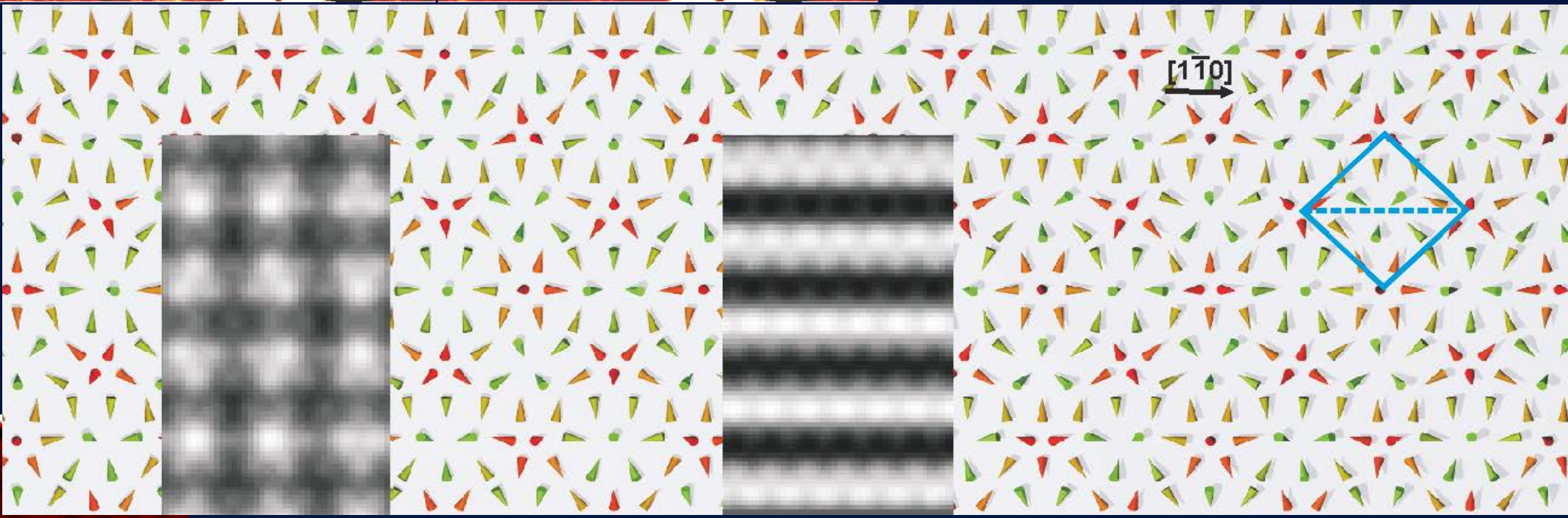
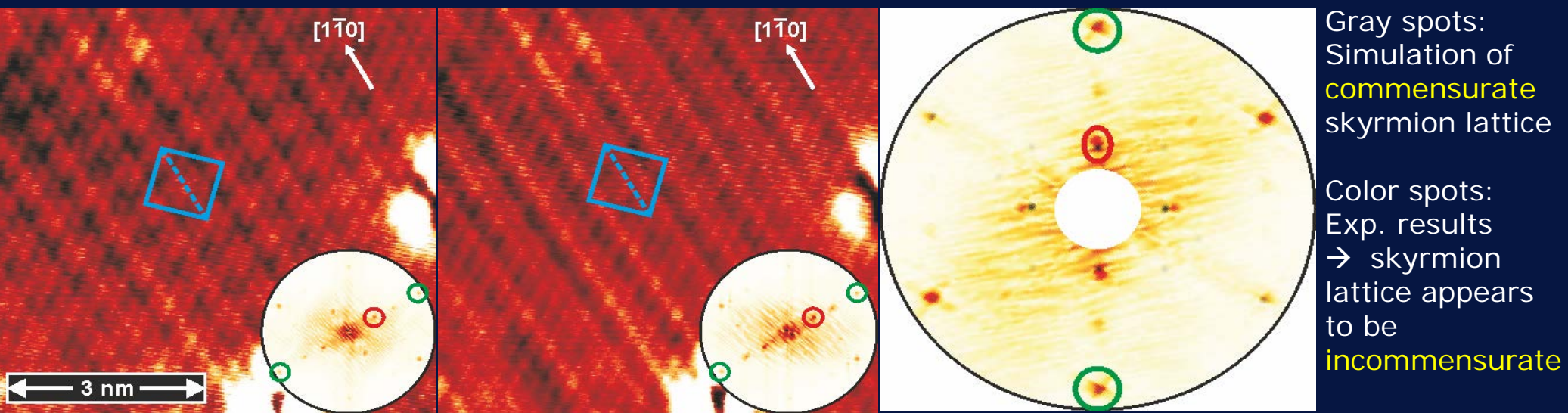
Investigation of the  
absolute magnetization  
direction in the sample is possible



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university of hamb

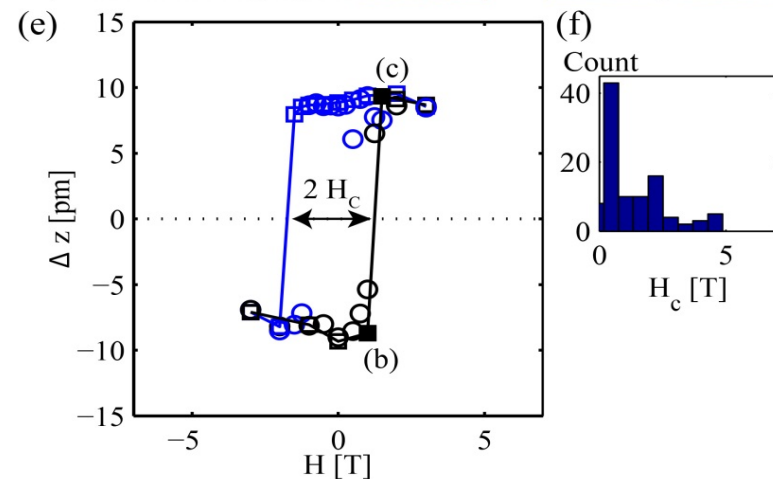
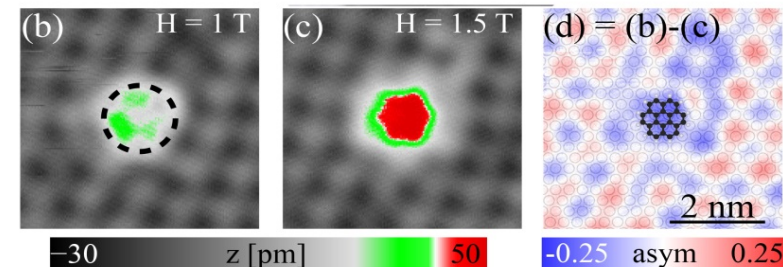
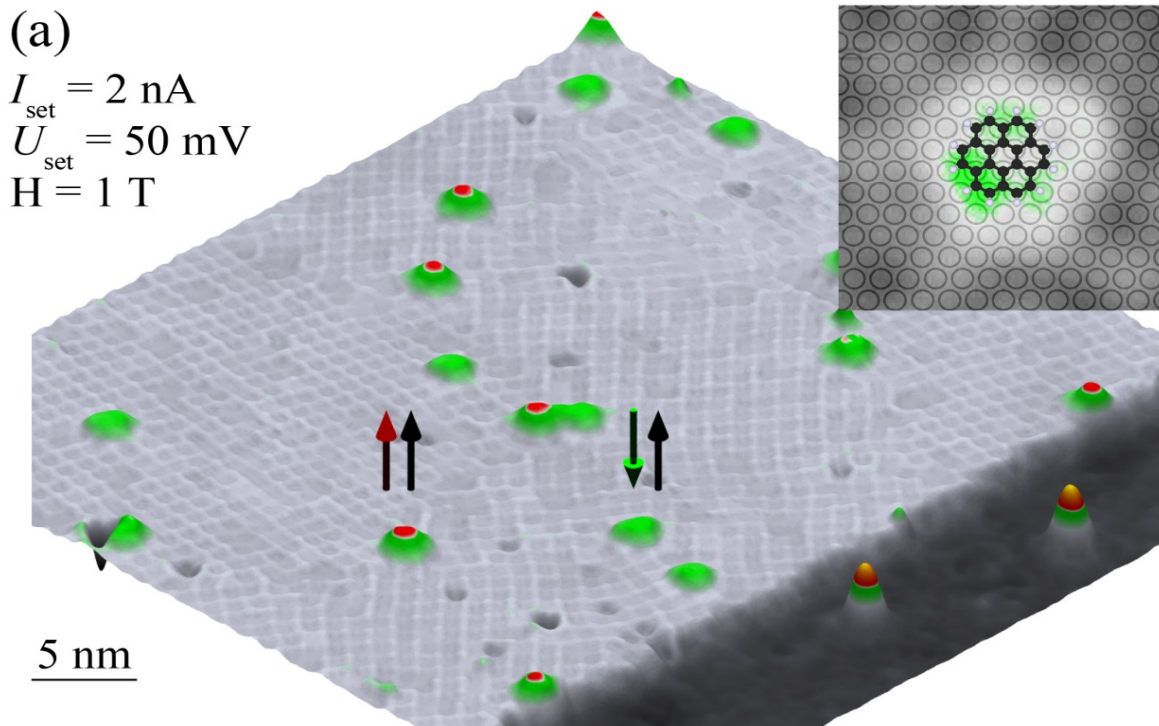
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# Atomic Resolution for Nanoskyrmion Lattice of 1 ML Fe on Ir(111)





# Magnetic Properties of Coronene Molecules on Fe / Ir(111)

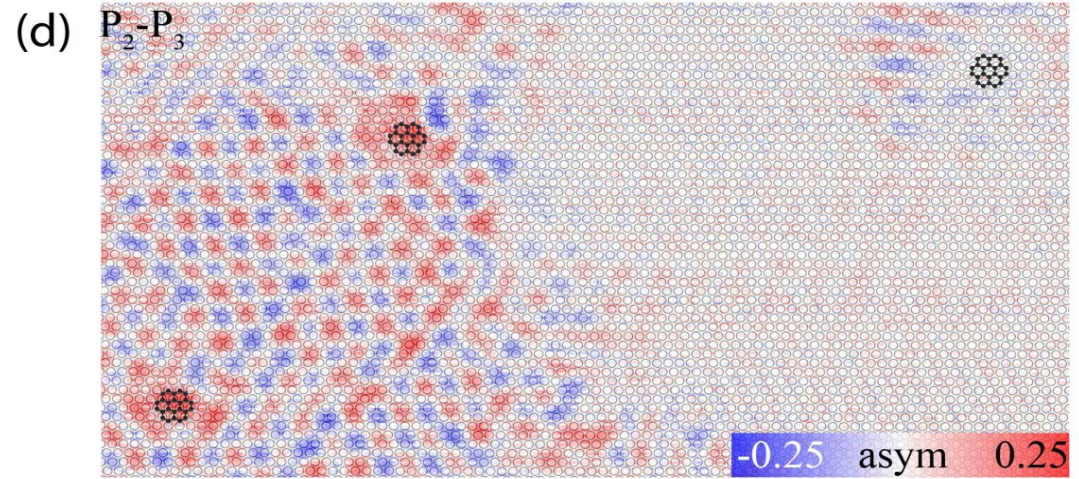
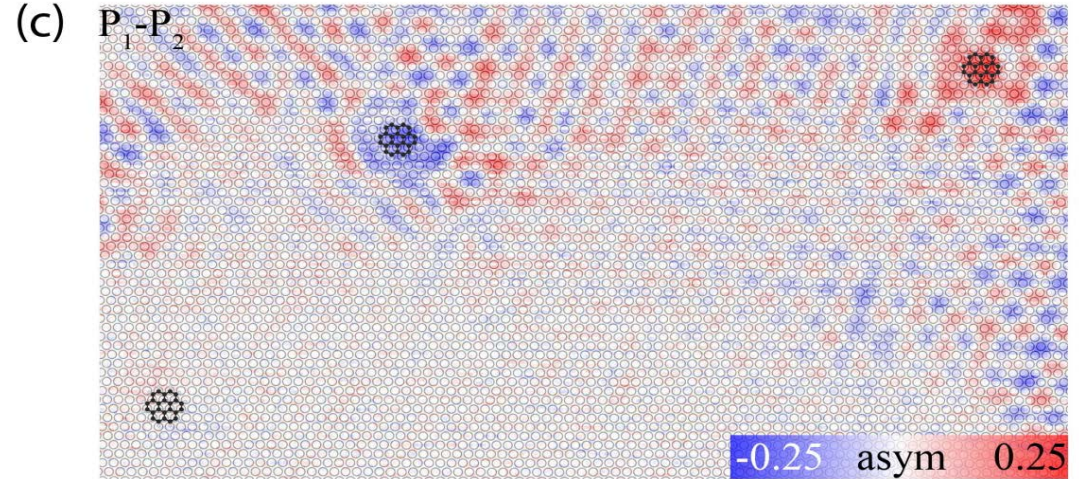
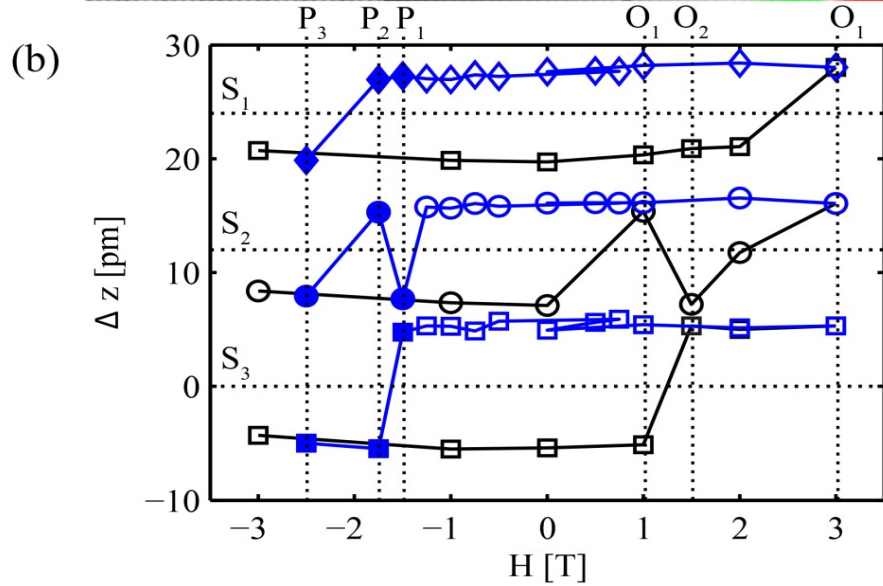
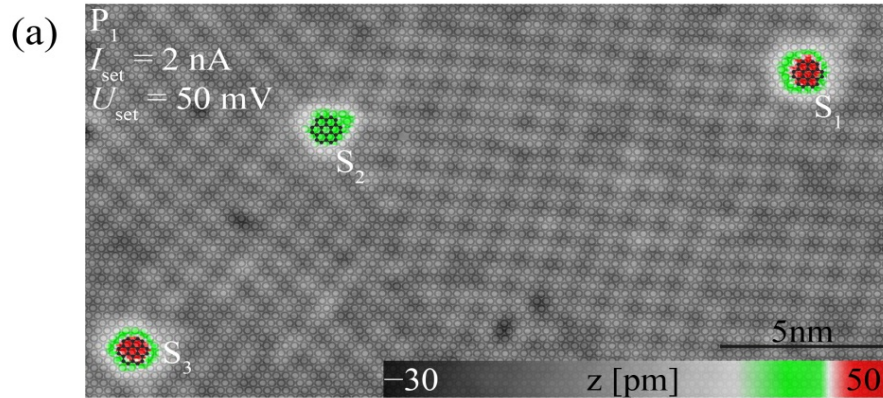


→ high coercivity of coronene / Fe / Ir(111) system (1.5 - 2 T)

→ magnetic hardening effect due to organic molecule on top of Fe layer



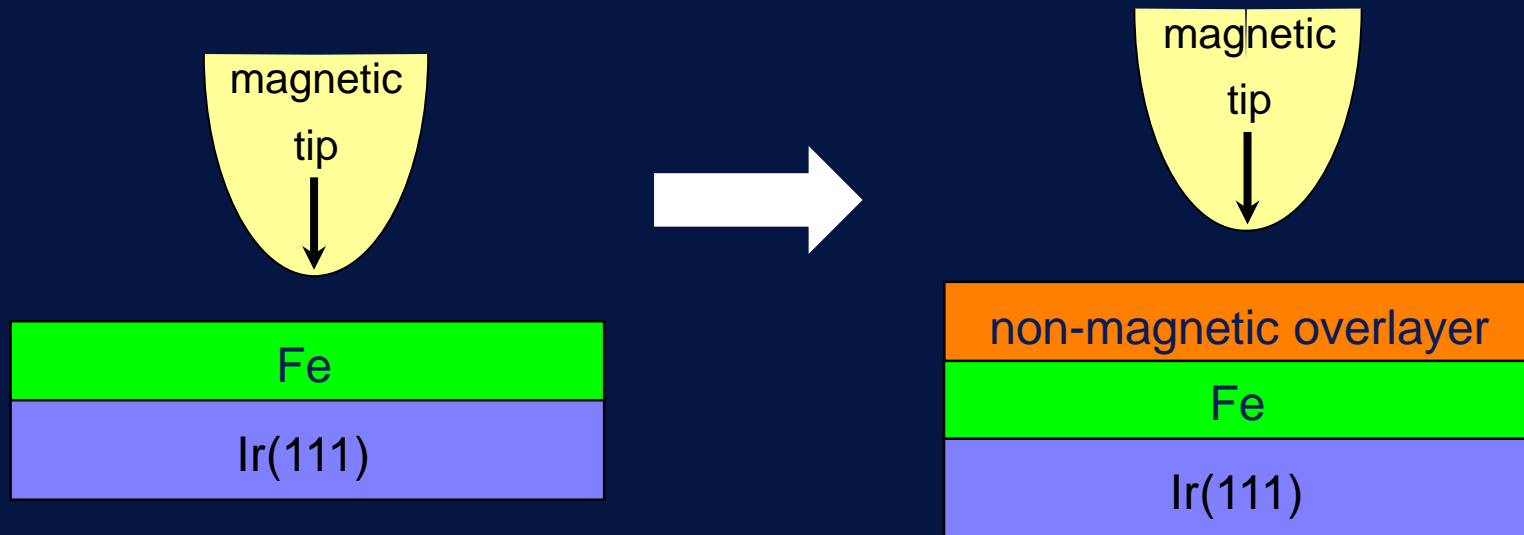
# Spin State Information Transmission via a Skyrmion Lattice: Coronene Molecules on Fe/Ir(111)



J. Brede et al., Nature Nanotechnol. **9**, 1018 (2014)

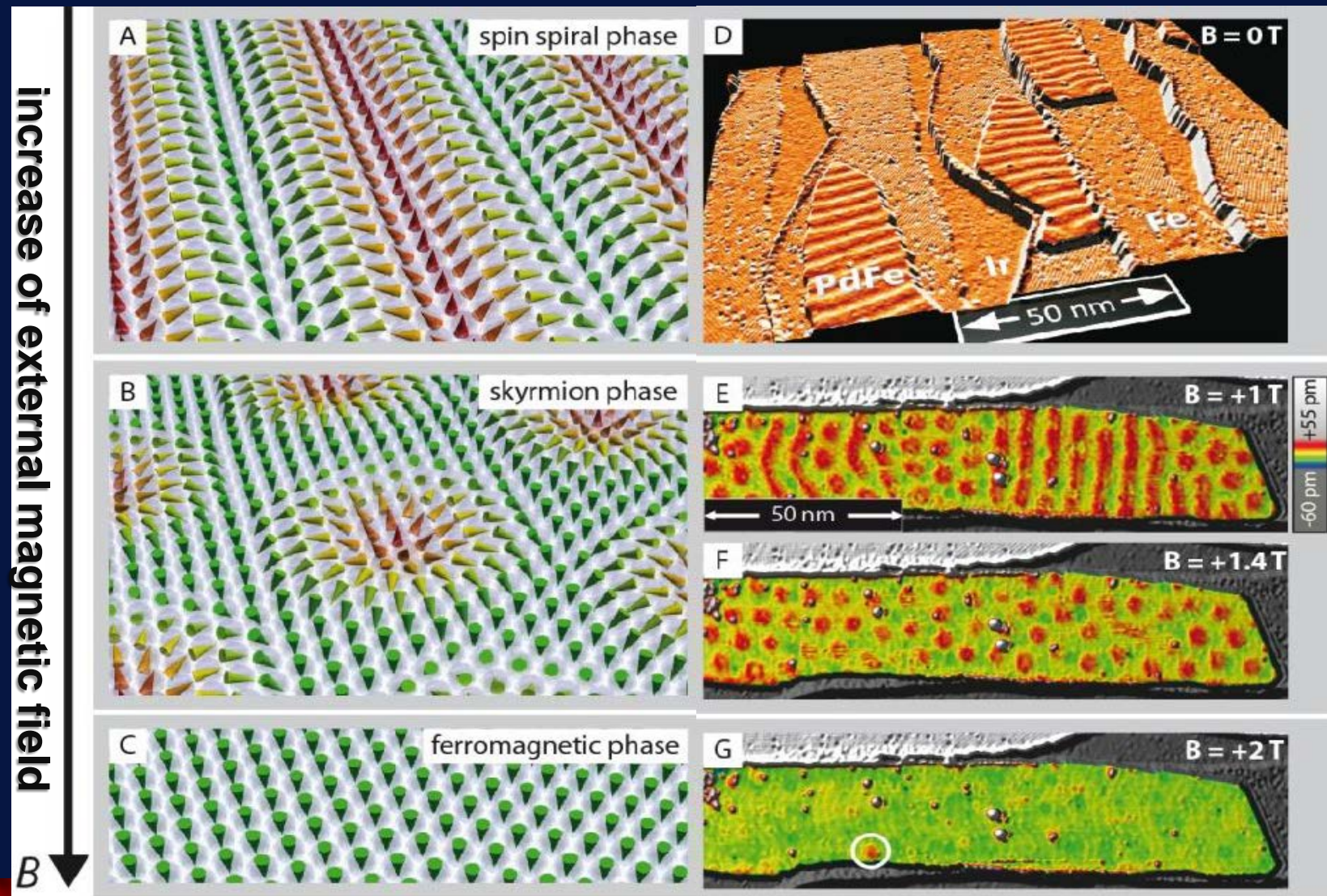
M. Cinchetti, Nature Nanotechnol. **9**, 965 (2014): „Topology communicates“

# Tailoring Skyrmionic States by Multiple Interface Engineering



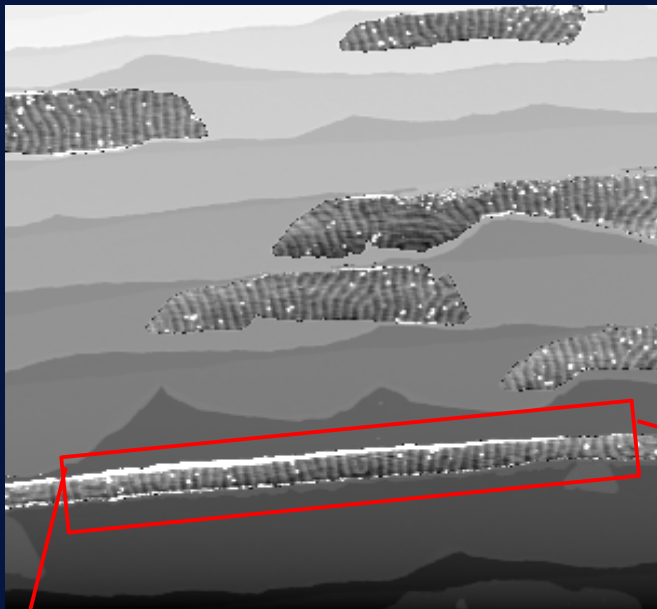
- Introducing a second interface by a non-magnetic overlayer
- Tuning of magnetic anisotropies and spin-orbit coupling via the second interface
- Tailoring the overall magnetic state of the hybrid structure

# Pd / Fe / Ir(111): From Spin Spirals to Skyrmions to Ferromagnetic State

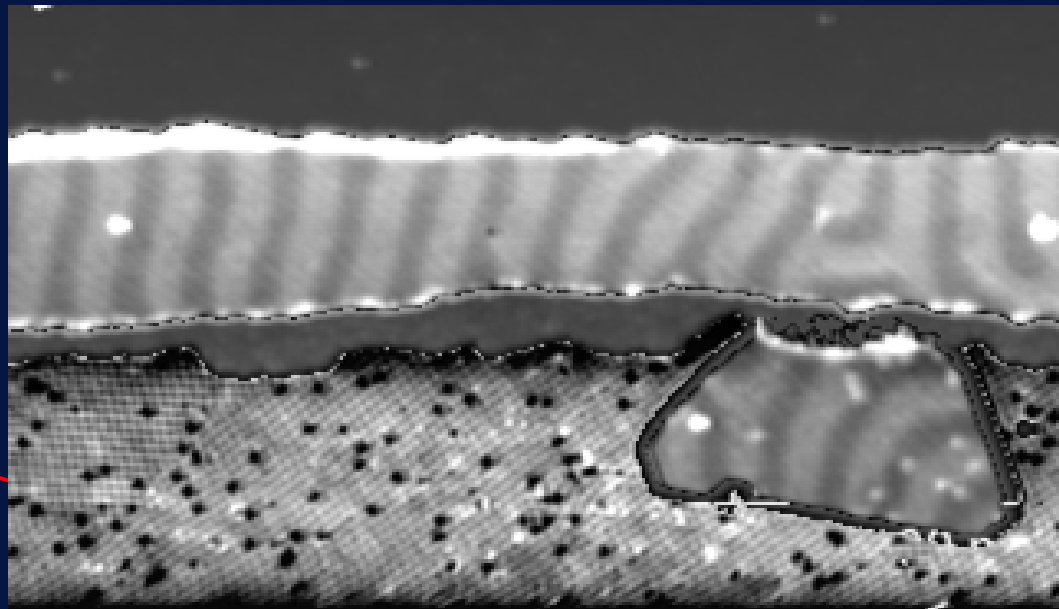


$T = 8\text{ K}$ ,  $U = +0.05\text{ V}$ ,  $I = 0.2\text{ A}$ , Cr-bulk tip

# Pd monolayer on fcc-Fe / Ir(111) in zero field

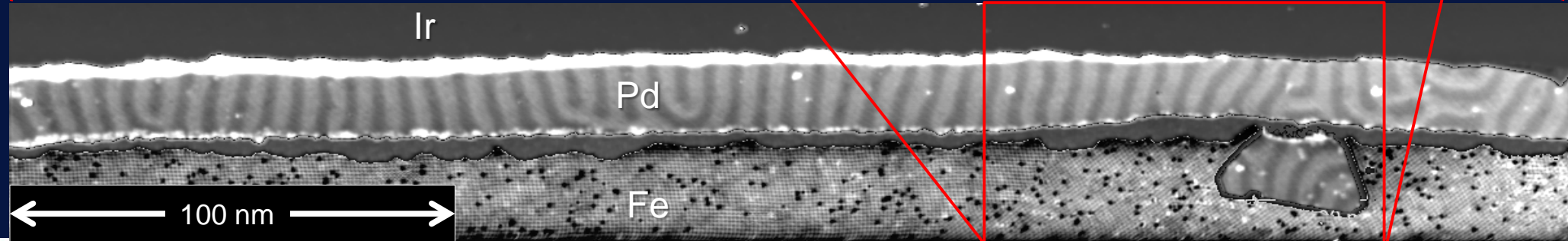


Ir  
Fe  
Pd

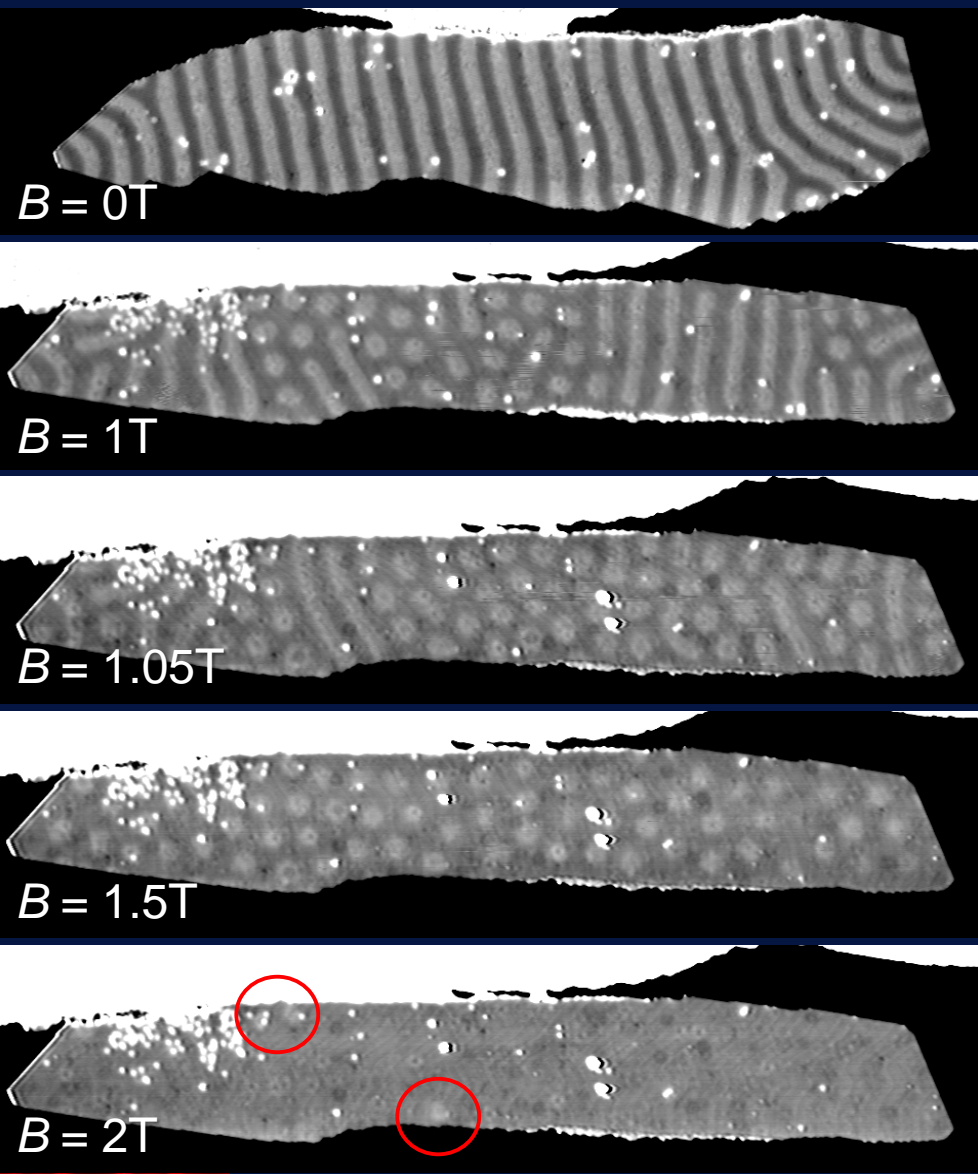


$U = +50 \text{ mV}, I = 200 \text{ pA}, T = 8 \text{ K}$

- uniaxial spin spiral with  $\sim 7 \text{ nm}$  periodicity
- spin spiral aligns perpendicular to edges of islands
- periodicity is one order of magnitude larger than Fe

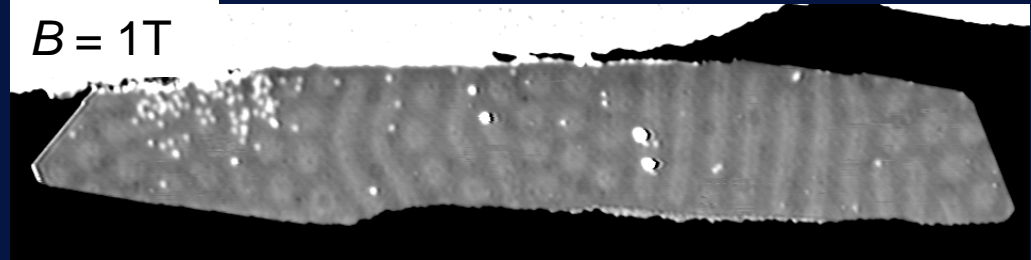
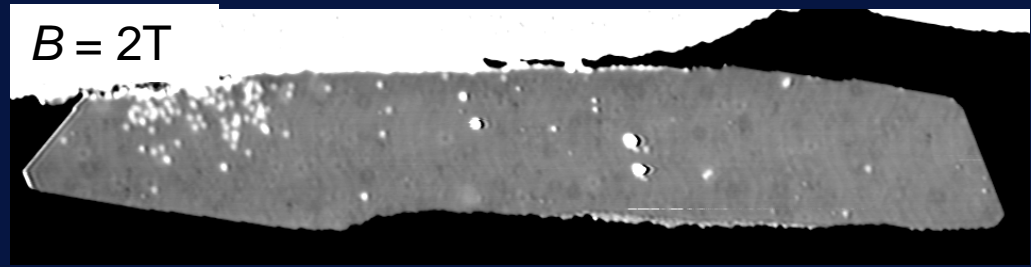
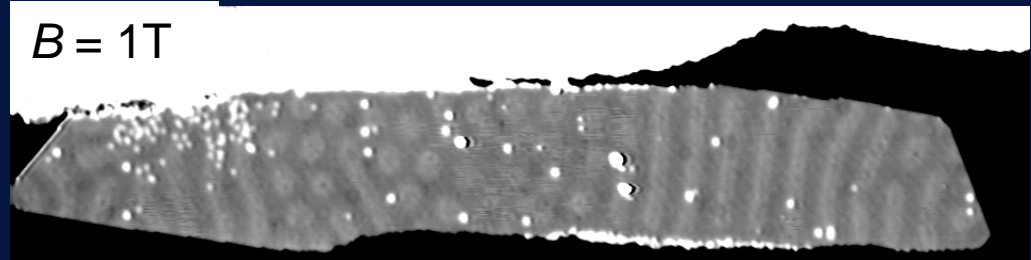


# B-field dependence of Pd/Fe bilayer on Ir(111)



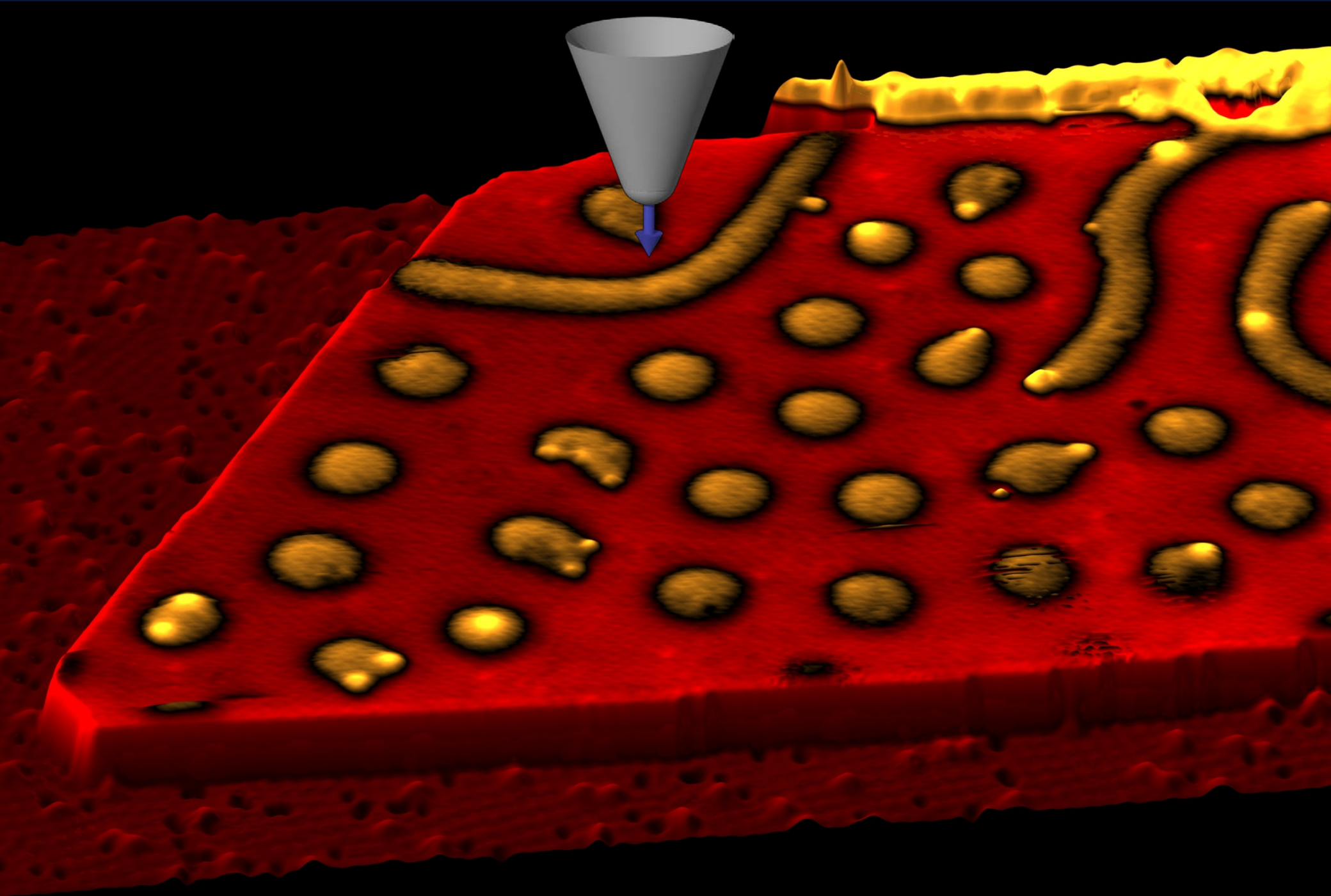
- five different phases:
- $B < 0.8T$ : spin spiral phase (ground state)
  - $0.8 < B < 1T$ : mixed phase (skymions & spiral)
  - $1T < B < 1.5T$ : skymion crystal
  - $1.5T < B < 2T$ : mixed phase (skymions & FM)
  - $B > 2T$ : saturated ferromagnetic phase

Reversible behaviour at  $T = 8 K$ :



$U = +50 mV, I = 200 pA, T = 8 K$



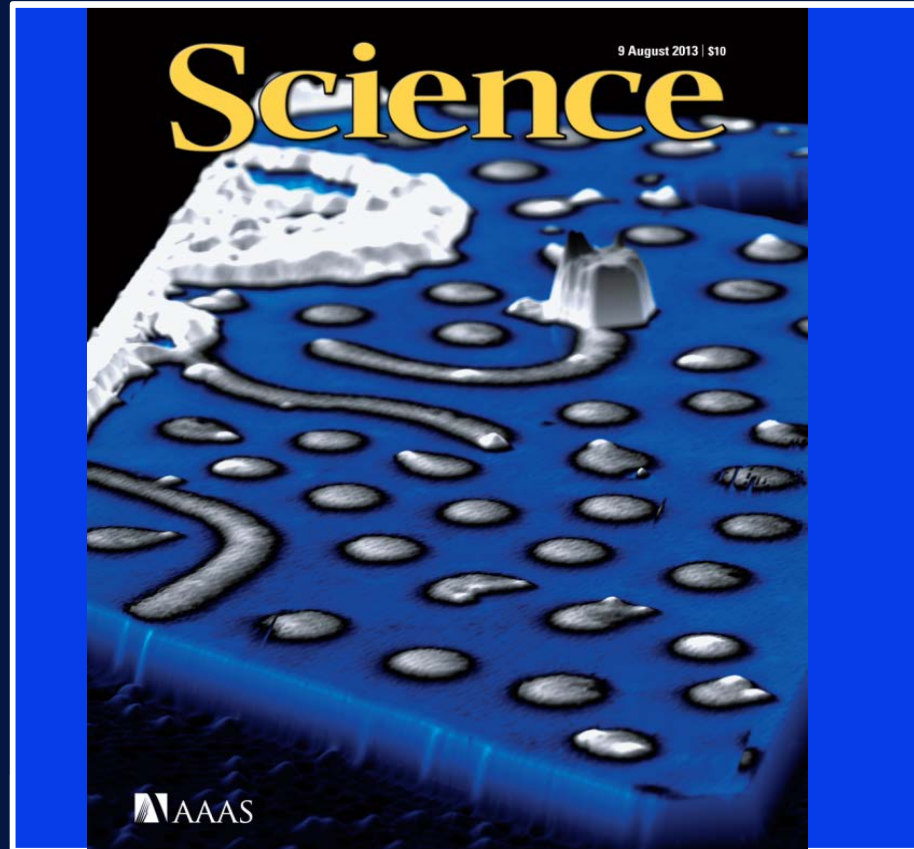


# First report on isolated chiral magnetic skyrmions !

$U = +200 \text{ mV}$   
 $I = 1.0 \text{ nA}$   
 $T = 2.2 \text{ K}$   
 $B = -1.5 \text{ T}$

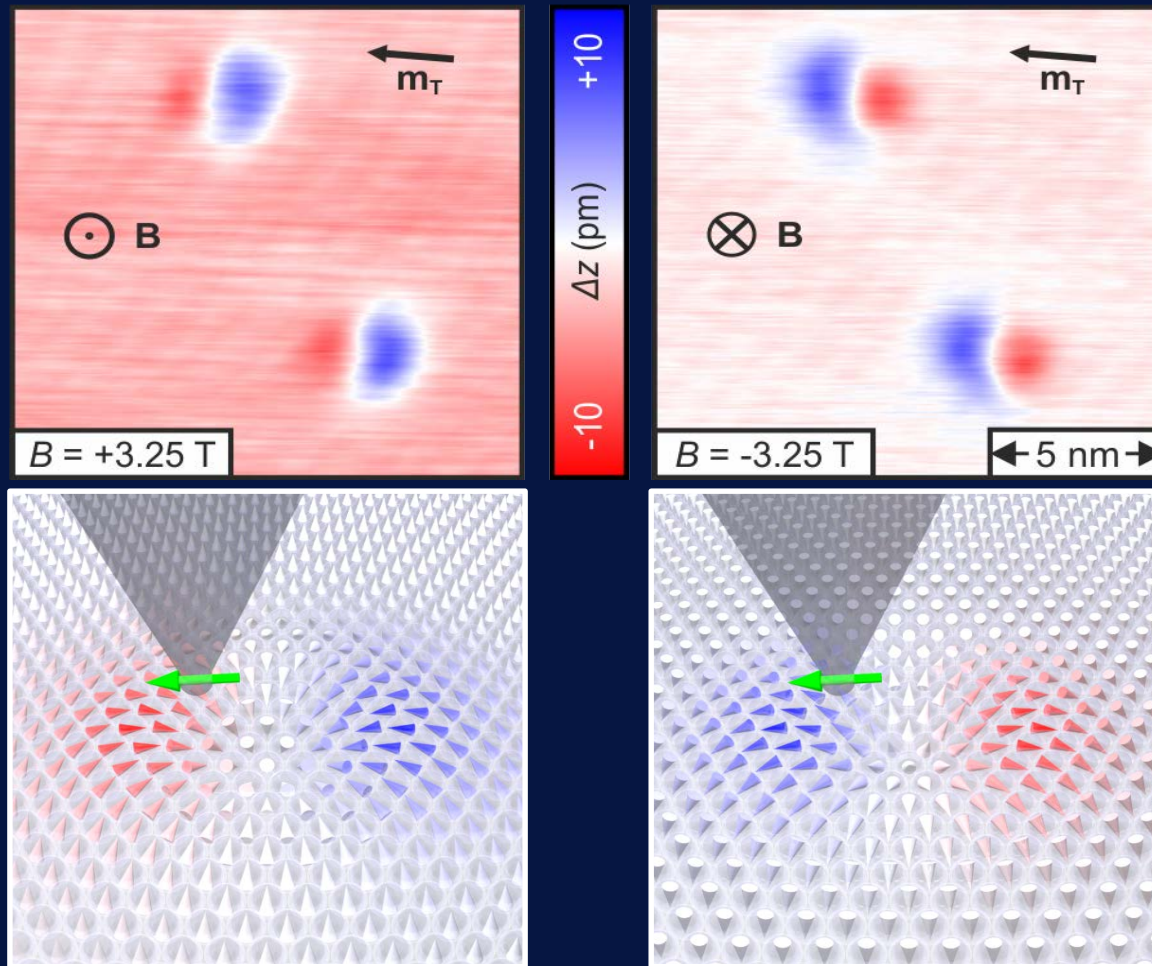


→ skyrmion looks axisymmetric with out-of-plane-tip





# Skyrmions in Opposite Magnetic Fields



- canted SPSTM tips lead to an asymmetric appearance of skyrmions
- skyrmions with the same rotational sense look different in opposite magnetic fields

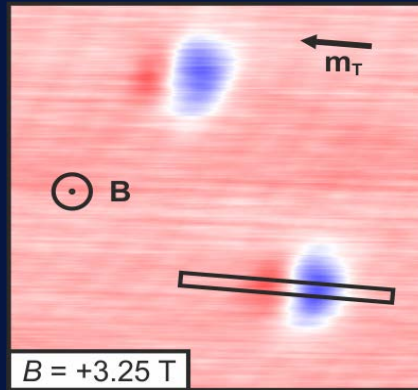


# Atomic-Scale Profile of a Single Skyrmion: Comparison of Experiment with Theory

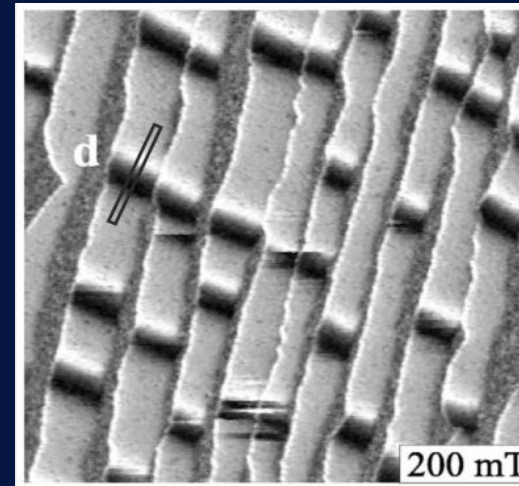
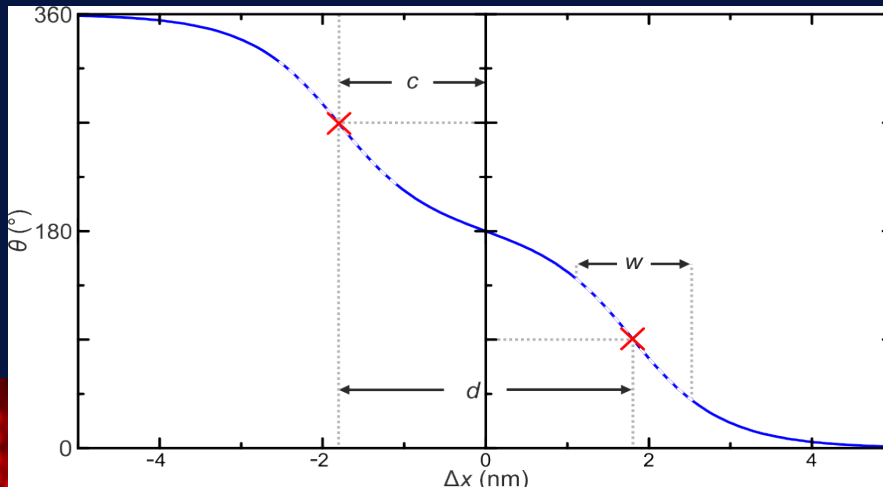
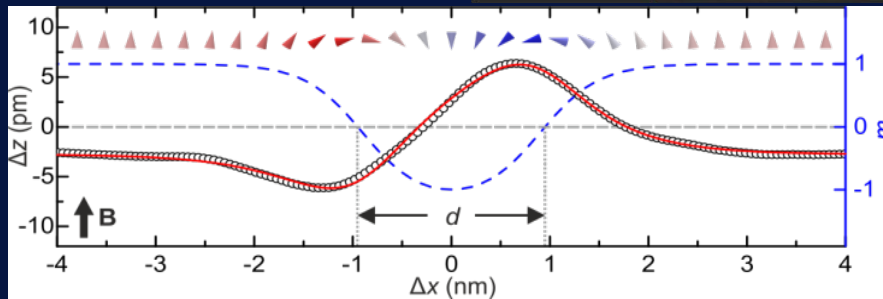
Chiral skyrmions  
in Pd/Fe/Ir(111)

N. Romming *et al.*:  
Science **341**, 636 (2013)

N. Romming *et al.*:  
PRL **114**, 177203 (2015)



$B = +3.25 \text{ T}$



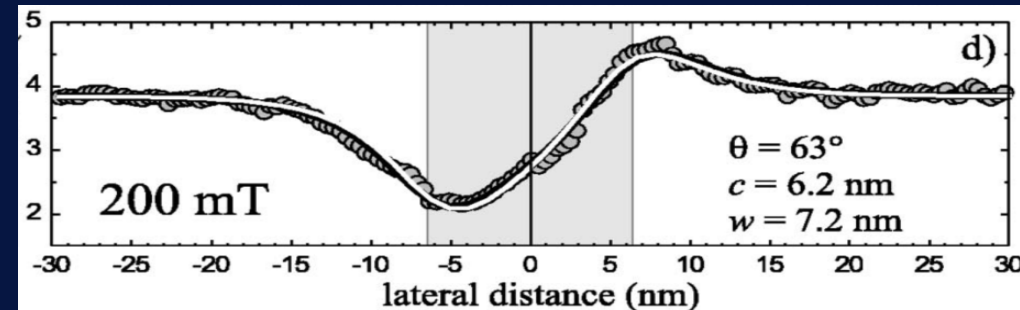
Chiral 360°-domain  
walls in DL Fe/W(110)

O. Pietzsch *et al.*:  
Science **292**, 2053 (2001)

A. Kubetzka *et al.*:  
PRL **88**, 057201 (2002)

A. Kubetzka *et al.*:  
Phys. Rev. **B67**, 020401 (2003)

E. Y. Vedmedenko *et al.*:  
Phys. Rev. **B 75**, 104431 (2007)

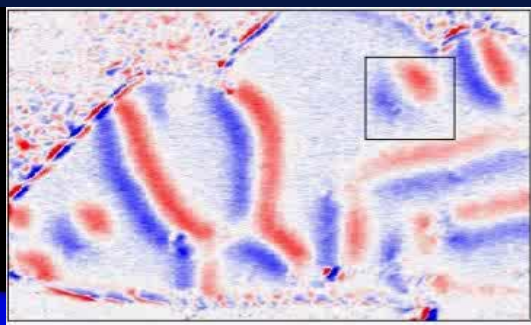
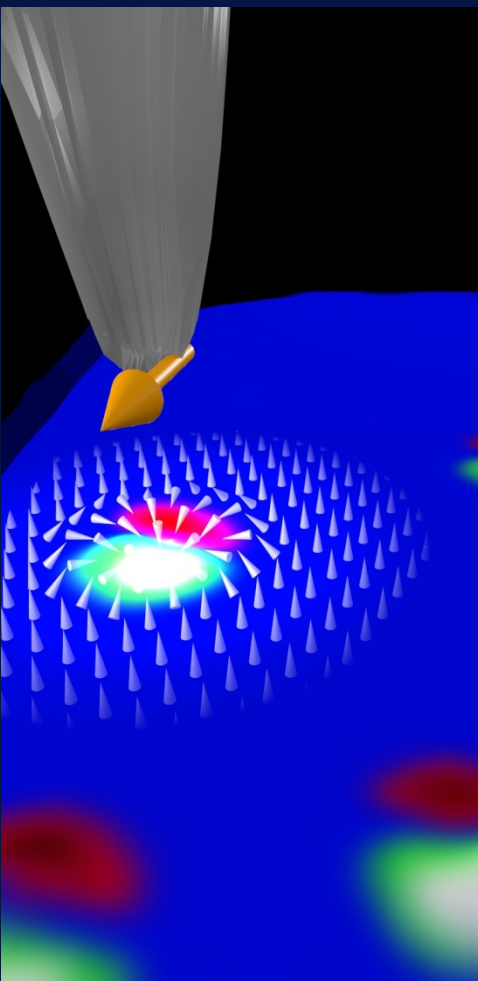


Analytical formula for two 180° domain walls:

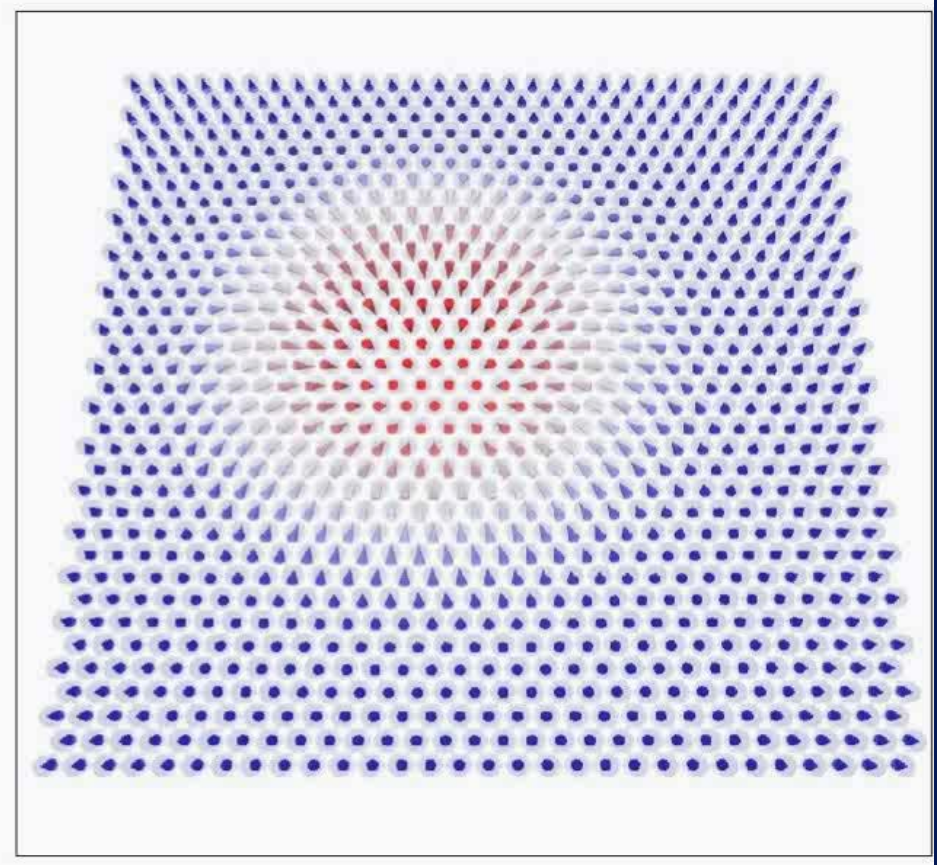
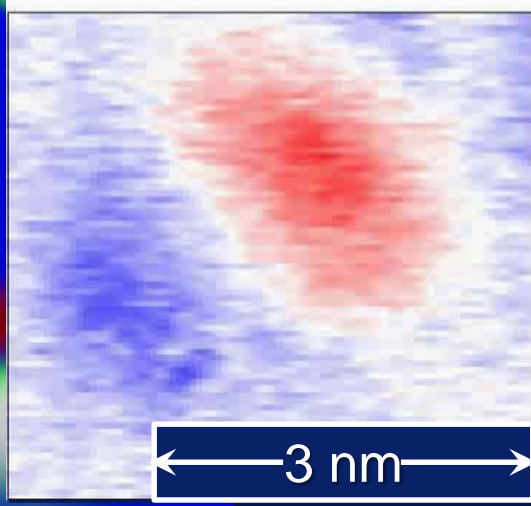
$$\theta(\rho, c, w) = \begin{cases} \sum_{+,-} \left[ \arcsin \left( \tanh \frac{-\rho \pm c}{w/2} \right) \right] + \pi & |B_z| > 0 \\ \sum_{+,-} \left[ \arcsin \left( \tanh \frac{-\rho \pm c}{w/2} \right) \right] & |B_z| < 0 \end{cases}$$

N. Romming *et al.*, PRL **114**, 177203 (2015)

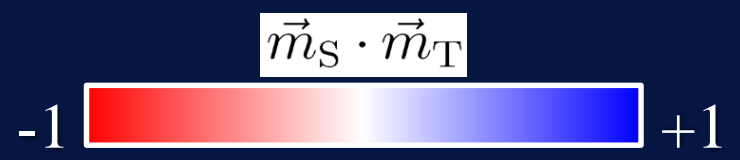
# Field-Dependent Skyrmion Diameter



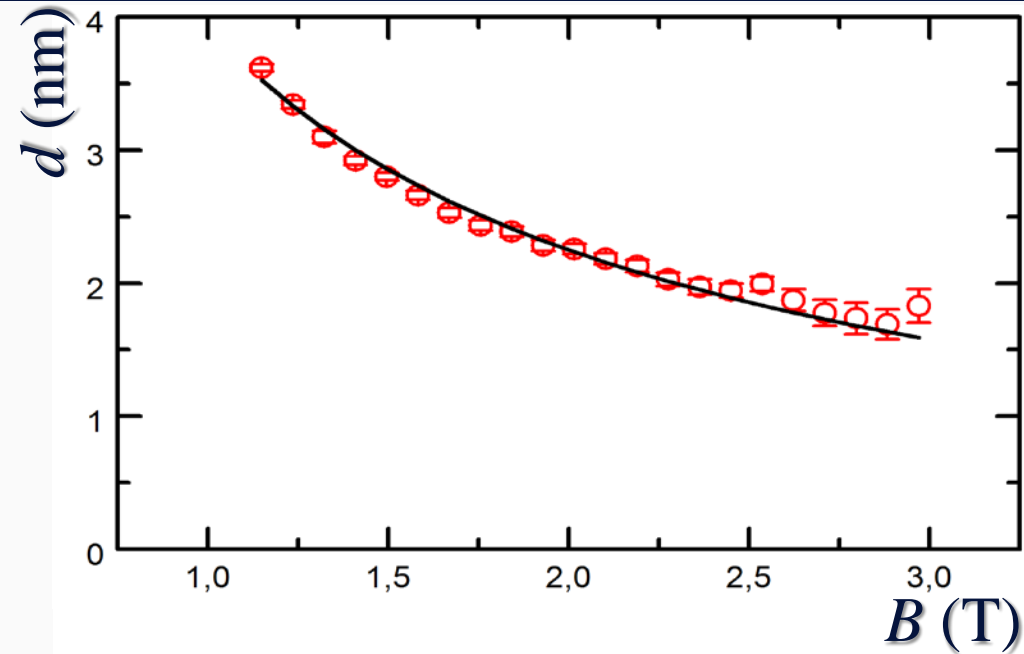
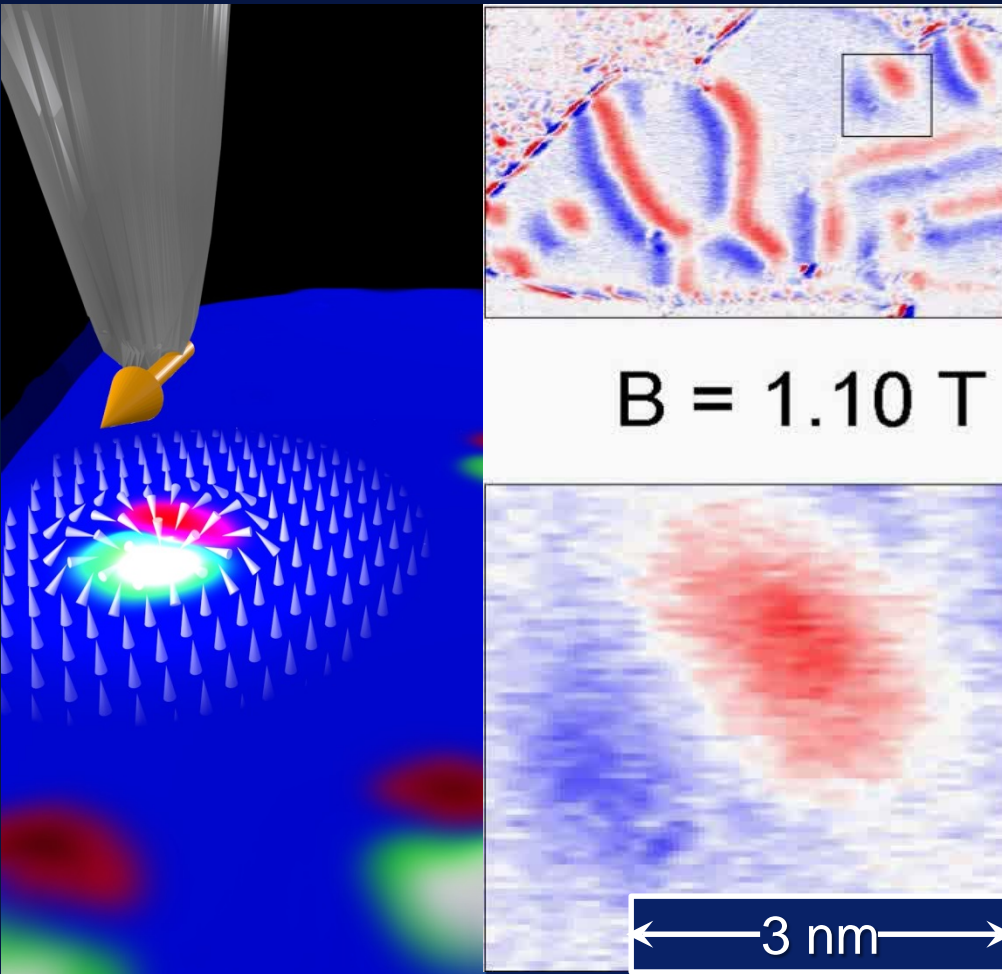
$B = 1.10 \text{ T}$



Difference SP-STs data taken during field-sweep from  $B = -3\text{T}$  to  $+3\text{T}$



# Field-Dependent Skyrmion Diameter



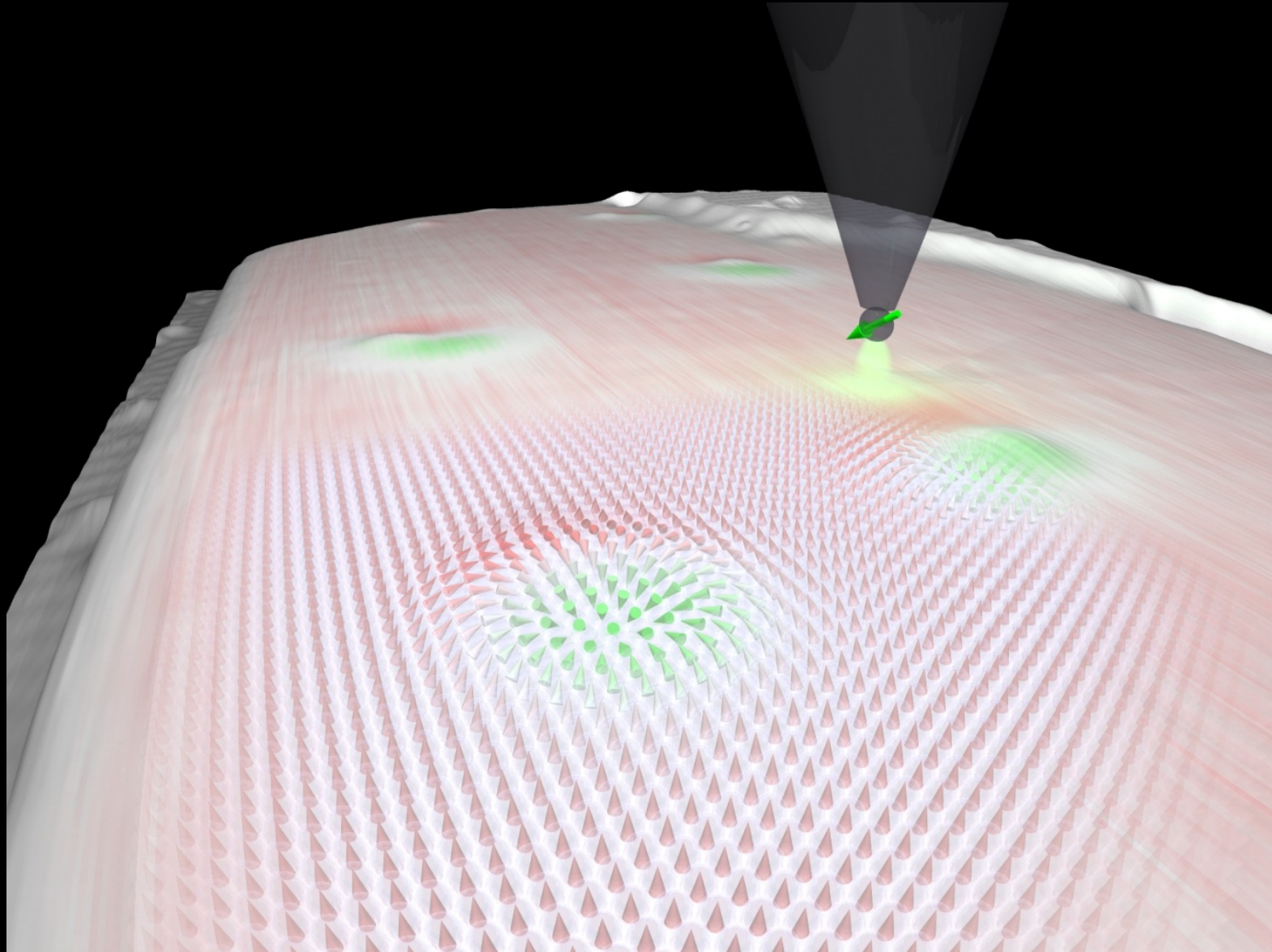
→ Skyrmion size and shape change with magnetic field can be reproduced by micromagnetic simulations with fitting parameters  $A$ ,  $D$ , and  $K$ .

Difference SP-STS data taken during field-sweep from  $B = -3\text{T}$  to  $+3\text{T}$

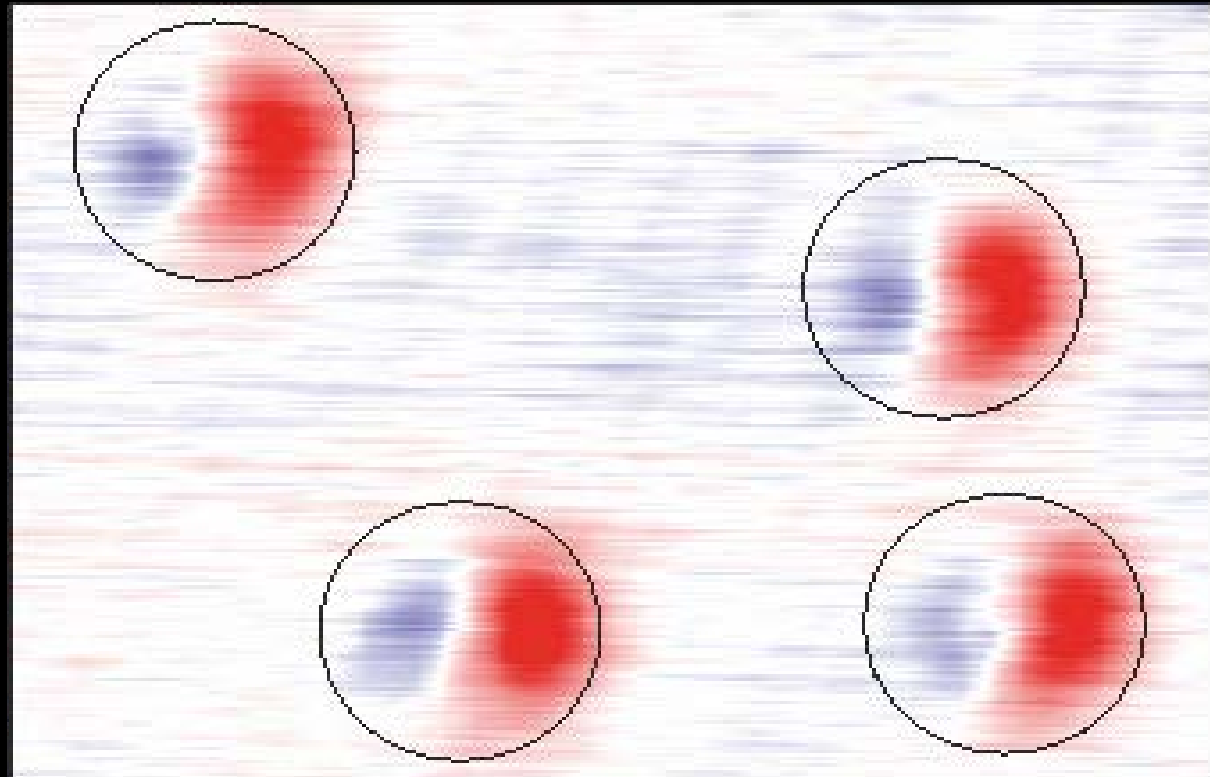
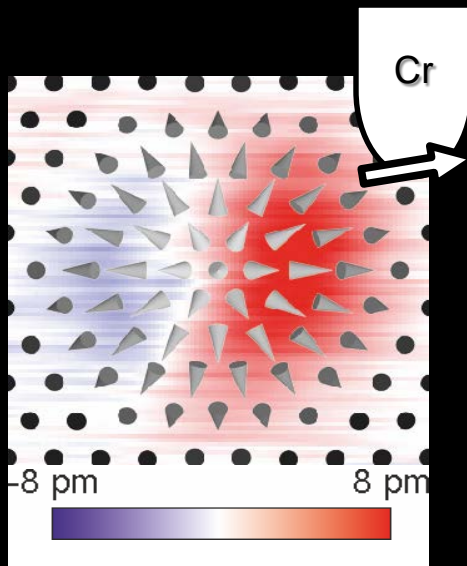
Saturation magnetization:  $M_s = 1.1 \text{ MA/m}$   
 Exchange constant:  $A = 2.0 \text{ pJ/m}$   
 DMI constant:  $D = 3.9 \text{ mJ/m}^2$   
 Anisotropy constant:  $K = 2.5 \text{ MJ/m}^3$



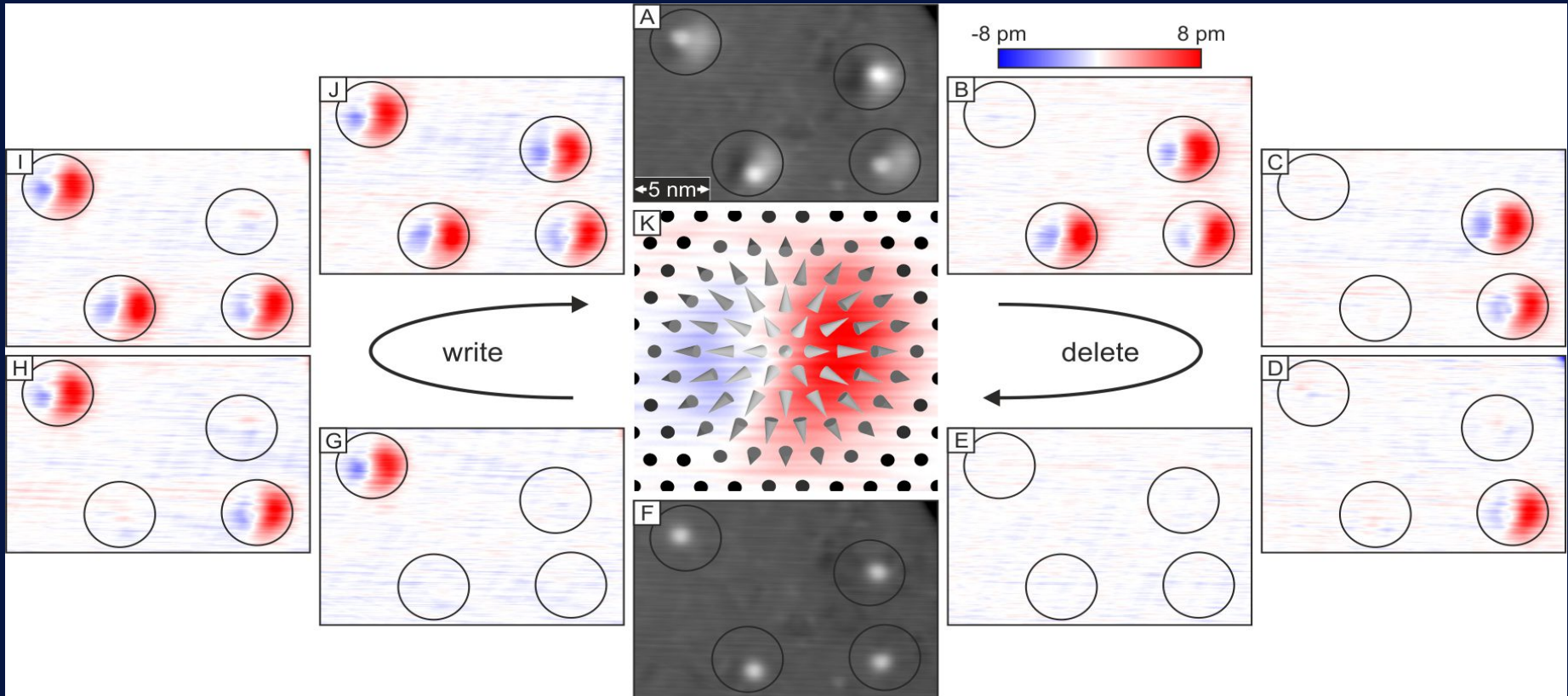
# From Imaging of Individual Skyrmions to Local Manipulation by Spin-Polarized Current Injection



# Writing and Deleting Single Skyrmions



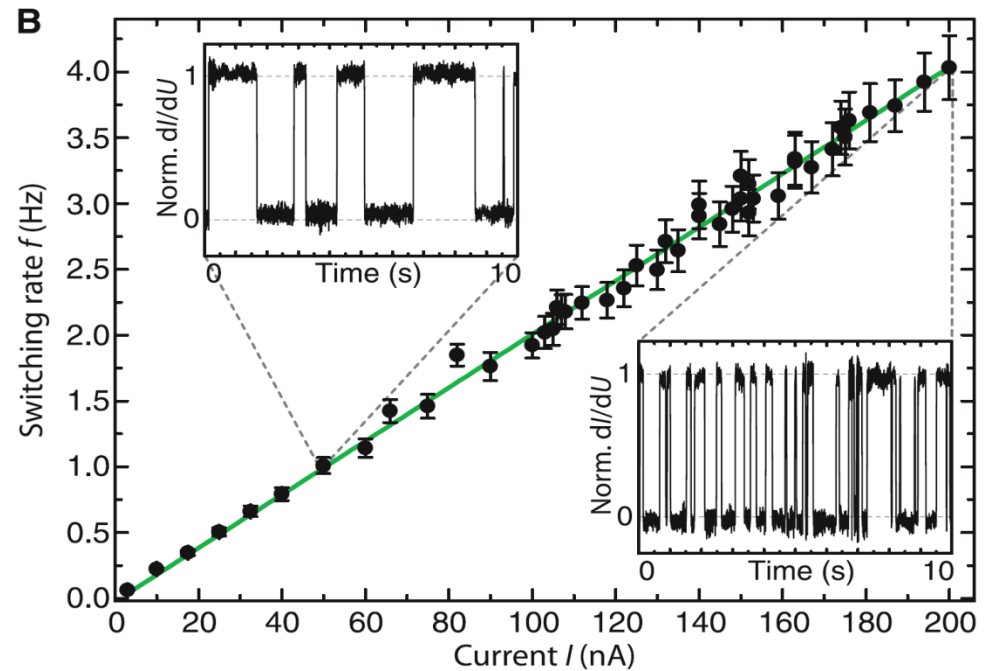
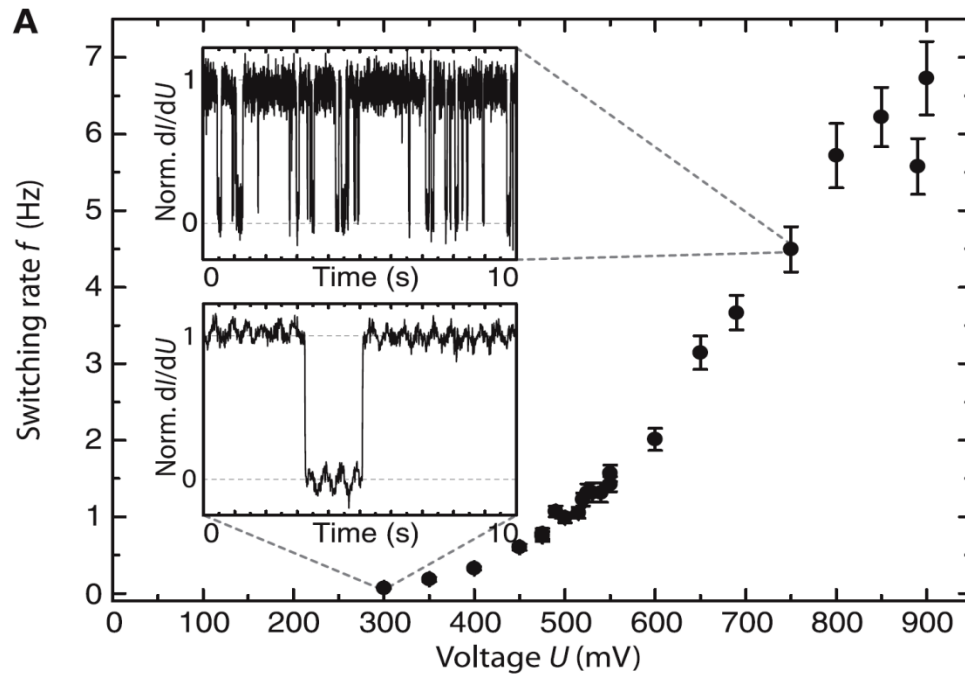
# Writing and Deleting Single Skyrmions by a SP-STM tip



- Writing and deleting of single skyrmions by localized spin current injection
- Imaging of individual skyrmions by in-plane sensitive SP-STM probe tip



# Switching Mechanism



low bias: non-perturbing imaging  
higher bias: increased switching frequency  
due to non-thermal excitations  
by injected electrons

→ switching rate increases  
linearly with current

At low  $U, I$ : no switching

→ thermally activated switching does not play a role !

At constant power: switching rate still depends critically on  $U$

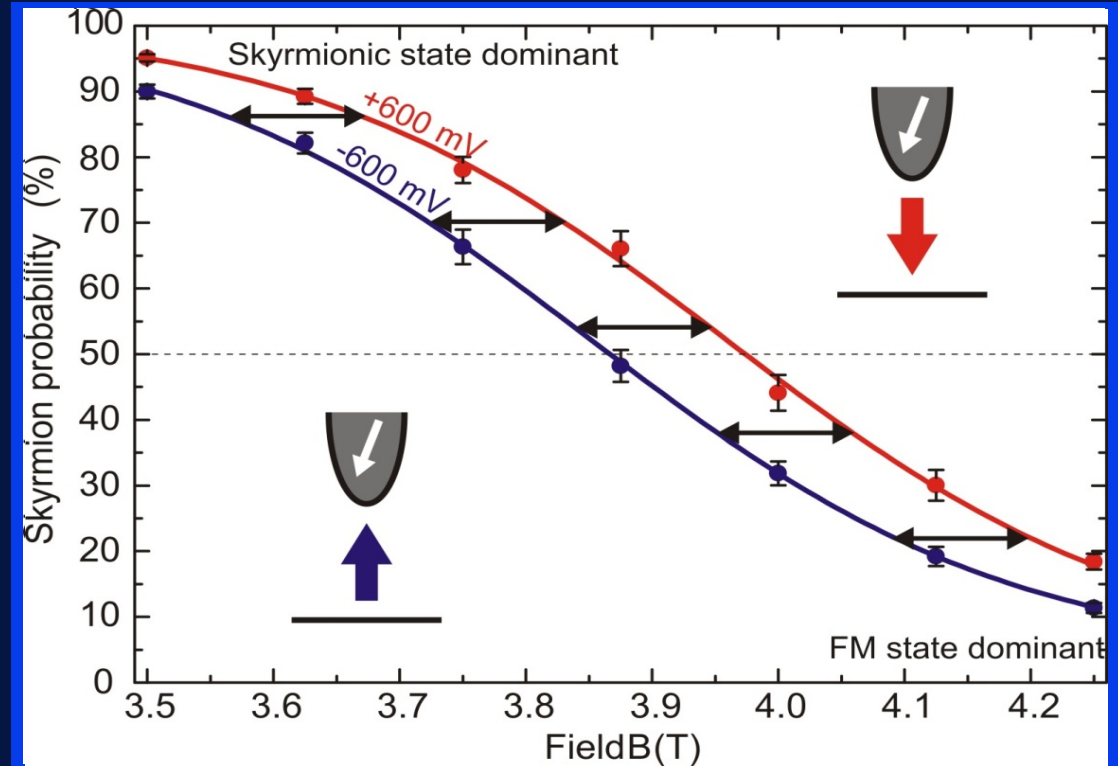
→ local (Joule-)heating does not play a decisive role !





# Switching Mechanism

- (i) ~~thermal noise~~
- (ii) ~~Joule heating~~
- (iii) non-thermal excitations from injected electrons
- (iv) spin transfer torque

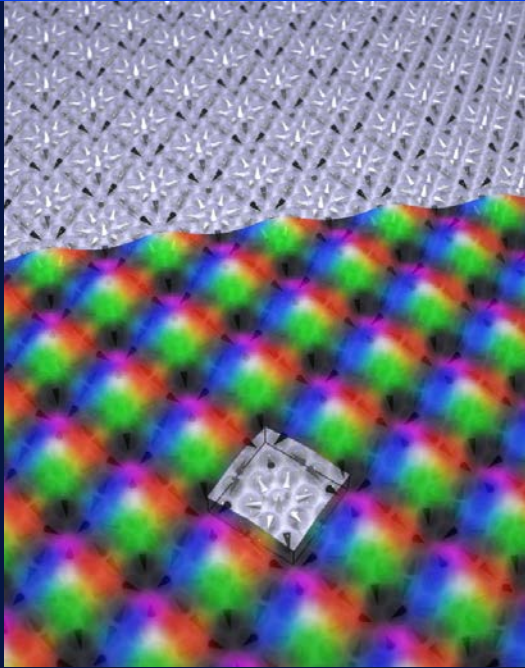


spin transfer torque leads to a shift of  $\sim 100$  mT  
→ induces directionality to the switching !

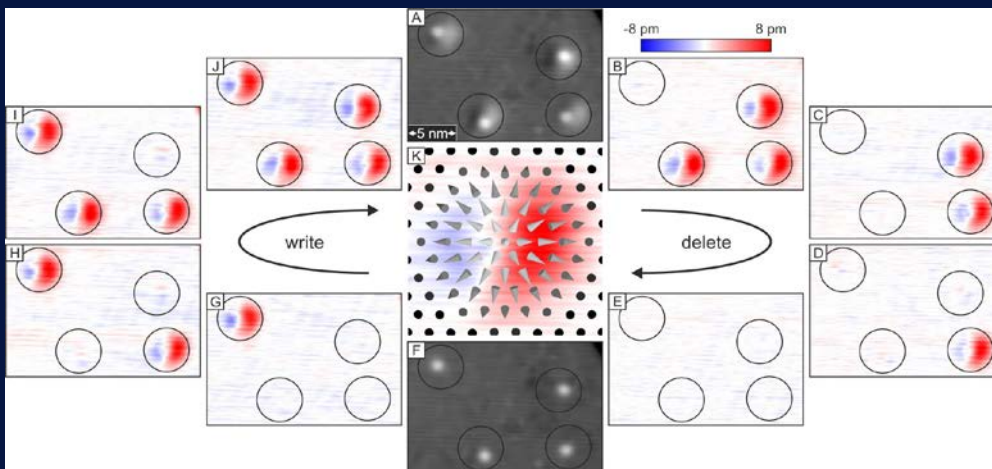
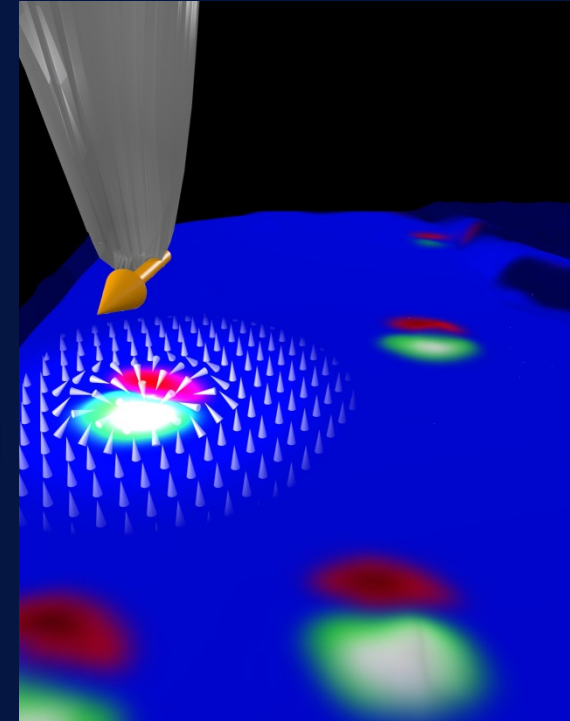
S. Krause et al., Science **317**, 1537 (2007)  
A.A. Khajetoorians et al., Science **339**, 55 (2013)



# Summary

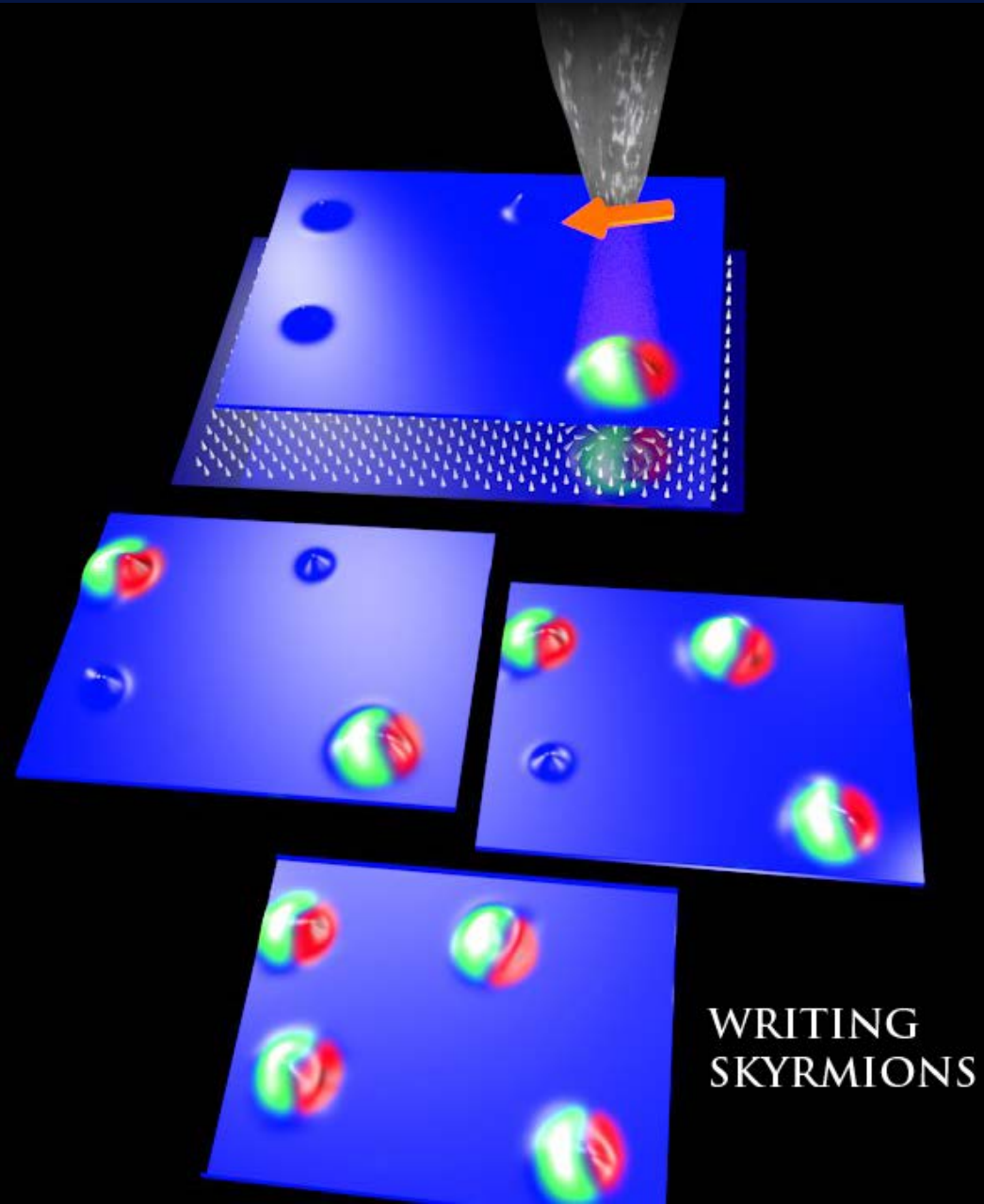


- **Interface-driven skyrmionic lattices** have been discovered in ultrathin magnetic films and atomically resolved by SP-STM
- The **interaction between individual magnetic adatoms and molecules with skyrmionic lattices** has directly been observed in real space by SP-STM
- **Individual skyrmions** were observed in bilayer films of Fe and Pd



- **Individual skyrmions** can be created and deleted by local spin-polarized current injection from a spin-polarized STM tip

# New SCIENCE & Technology with Skyrmions



# Acknowledgements



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**Matthias Menzel**



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**Kirsten von Bergmann**



**Elena Vedmedenko**

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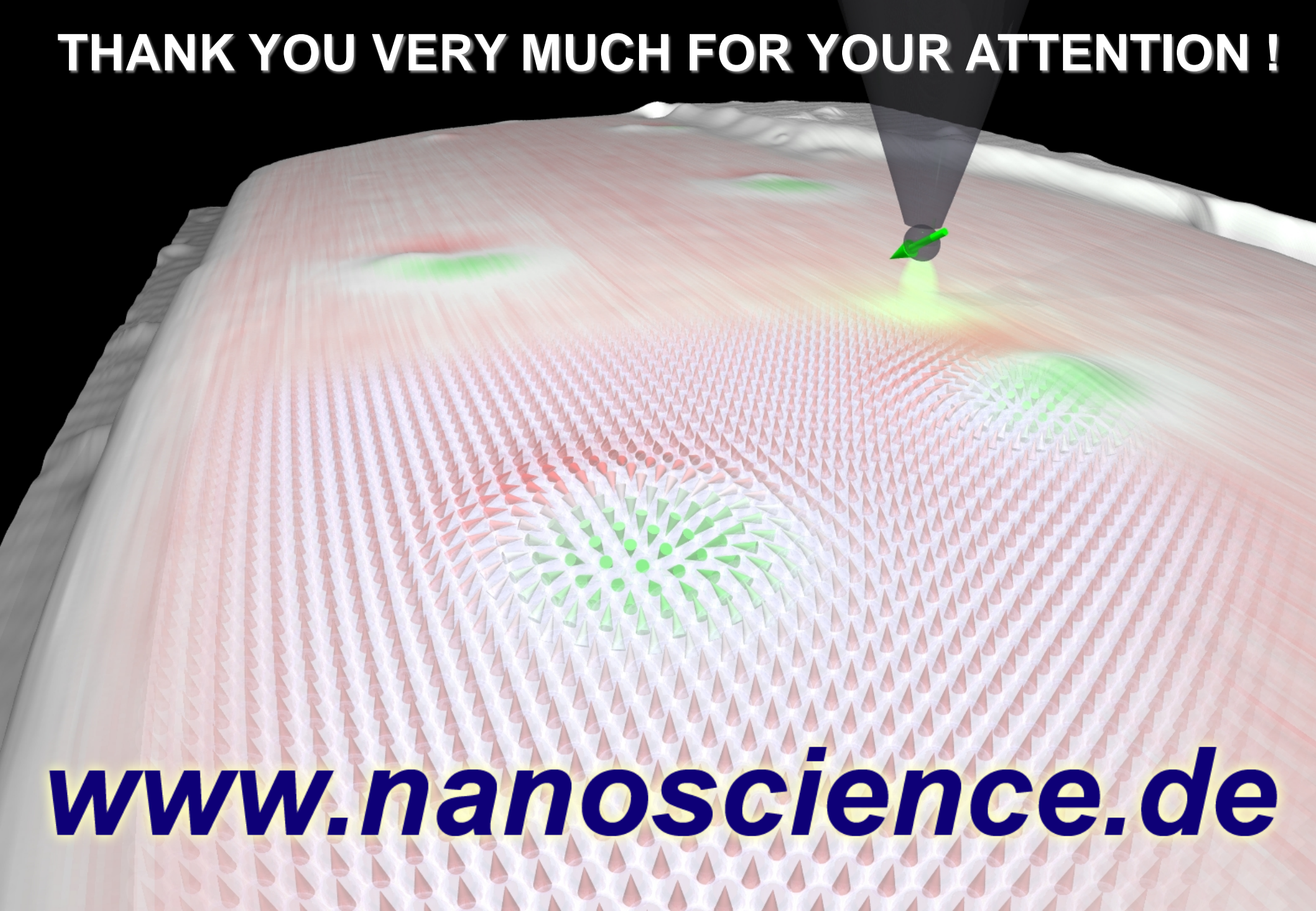


**Stefan Heinze**  
**University of Kiel**



**Stefan Blügel**  
**IFF, FZ Jülich**

**THANK YOU VERY MUCH FOR YOUR ATTENTION !**



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