Exploring 3D Spin Structures and Dynamics in Chiral Magnets with Advanced Synchrotron X-ray Techniques

Prof. **Thorsten Hesjedal**

16 October 2024

SPIN PHENOMENA INTERDISCIPLINARY CENTER

SPIN+X SFB/TRR 173 Kaiserslautern • Mainz

diamond

ISIS

UNIVERSITY OF OXFORD

Skyrmion Team & Collaborators

UNIVERSITY OF OXFORD

- *Shilei Zhang → ShanghaiTech*
- *Richard Brearton → Williams*
- *Jack Bollard*
- Ethan Arnold
- **Thorsten Hesjedal**

diamond

- *Adriana I Figueroa → Barcelona*
- David M Burn
- **Gerrit van der Laan**

TECHNISCHE UNIVERSITÄT DRESDEN

- *Marein Rahn → Augsburg*
- Moritz Winter
- **Bernd Rellinghaus**

Technische Universität München

- Christian Pfleiderer
- Andreas Bauer

ShanghaiTech University

- Keijing Ran
- Haonan Jin
- Xiaodong Xie
- **Shilei Zhang**

Institute of Physics CAS

- **Guoqiang Yu**

CHMFL

- **Haifeng Du**

KIT Karlsruher Institut für Technologie

- Markus Garst
- Amir Haghighirad

Funding:

2016-2023

THE SKYRMION PROJECT

The Skyrmion Project is a major national research programme designed to achieve a step-change in our understanding of skyrmions in magnetic materials and engineer them towards application.

EPSRC Engineering and Physical Sciences Research Council

Beamtime:

diamond

SOLEIL SYNCHROTRON

ALBA

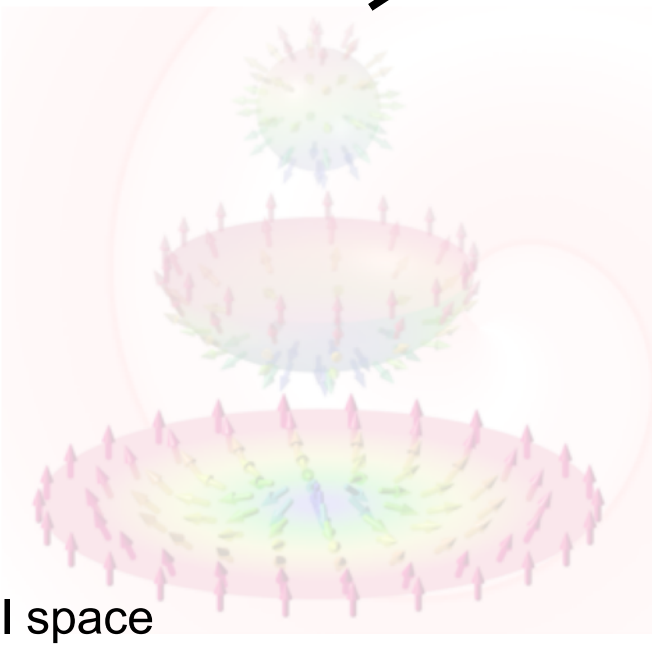
Science & Technology Facilities Council ISIS

HZB Helmholtz Zentrum Berlin

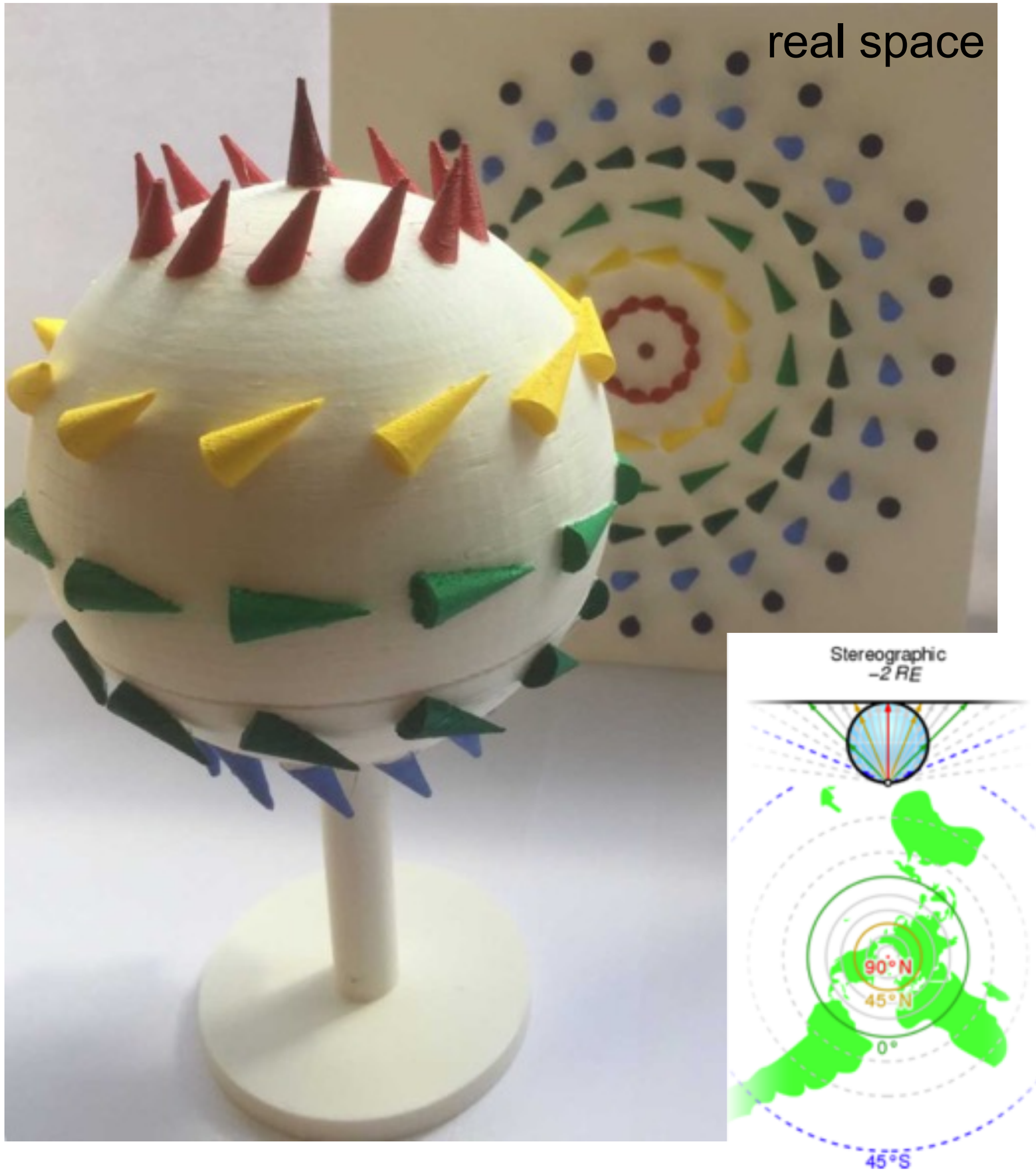
What are Magnetic Skyrmions?

vortices with a special knot ...

order parameter space



real space



Where Does the Name Come From?

Key idea:

Nucleons (fermions) as topological solitons in the (bosonic) pion fields

Baryon number (conserved) → winding number

Tony Skyrme (1922 – 1987)

- Part of Manhattan project
- Worked at Harwell (AERE) as head of the Theoretical Nuclear Physics Group
- Developed topological soliton model

distinct topological objects

properties of geometrical objects preserved through deformations

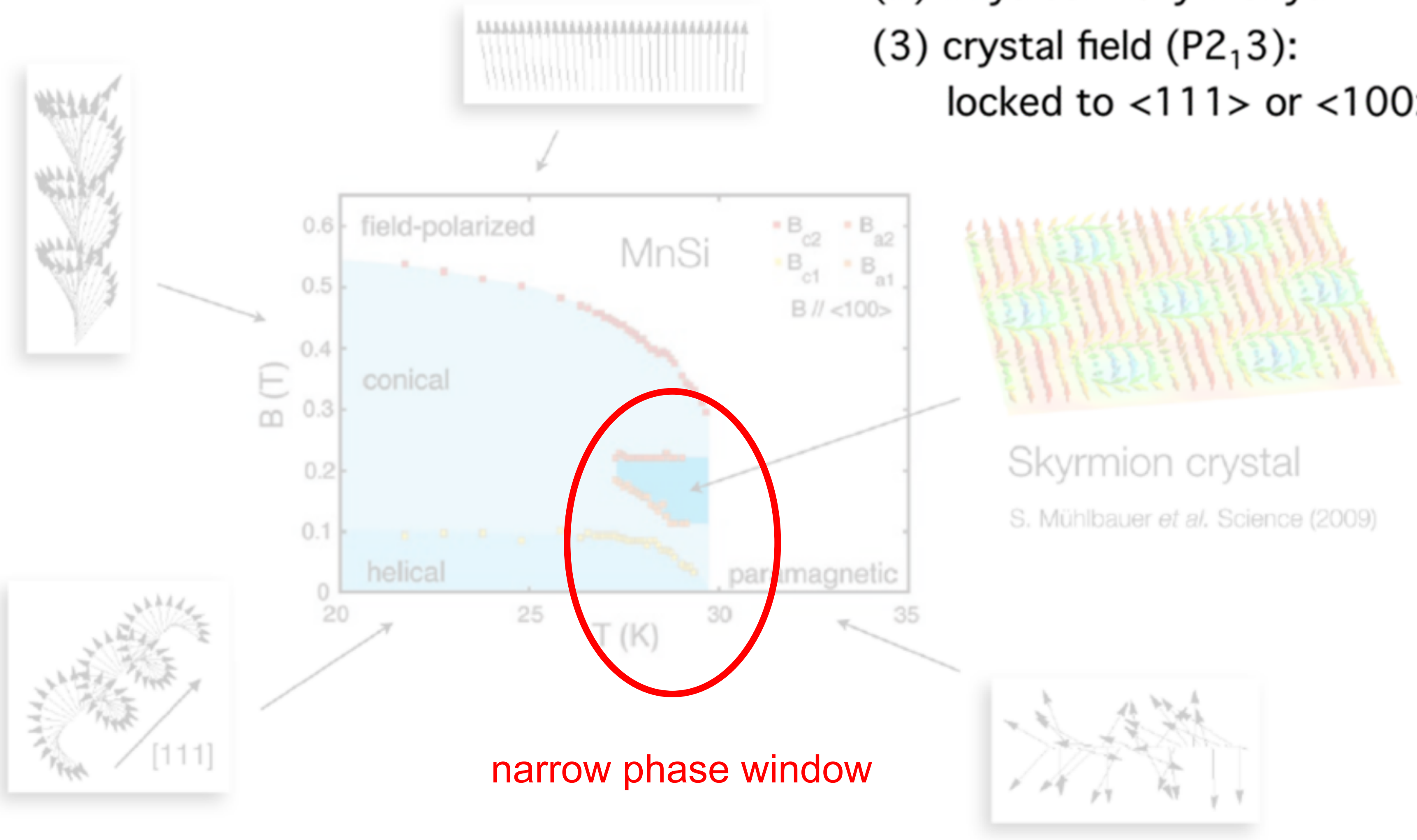
non-dispersing solitary wave

‘Universal’ Phase Diagram for Chiral B20 Crystals

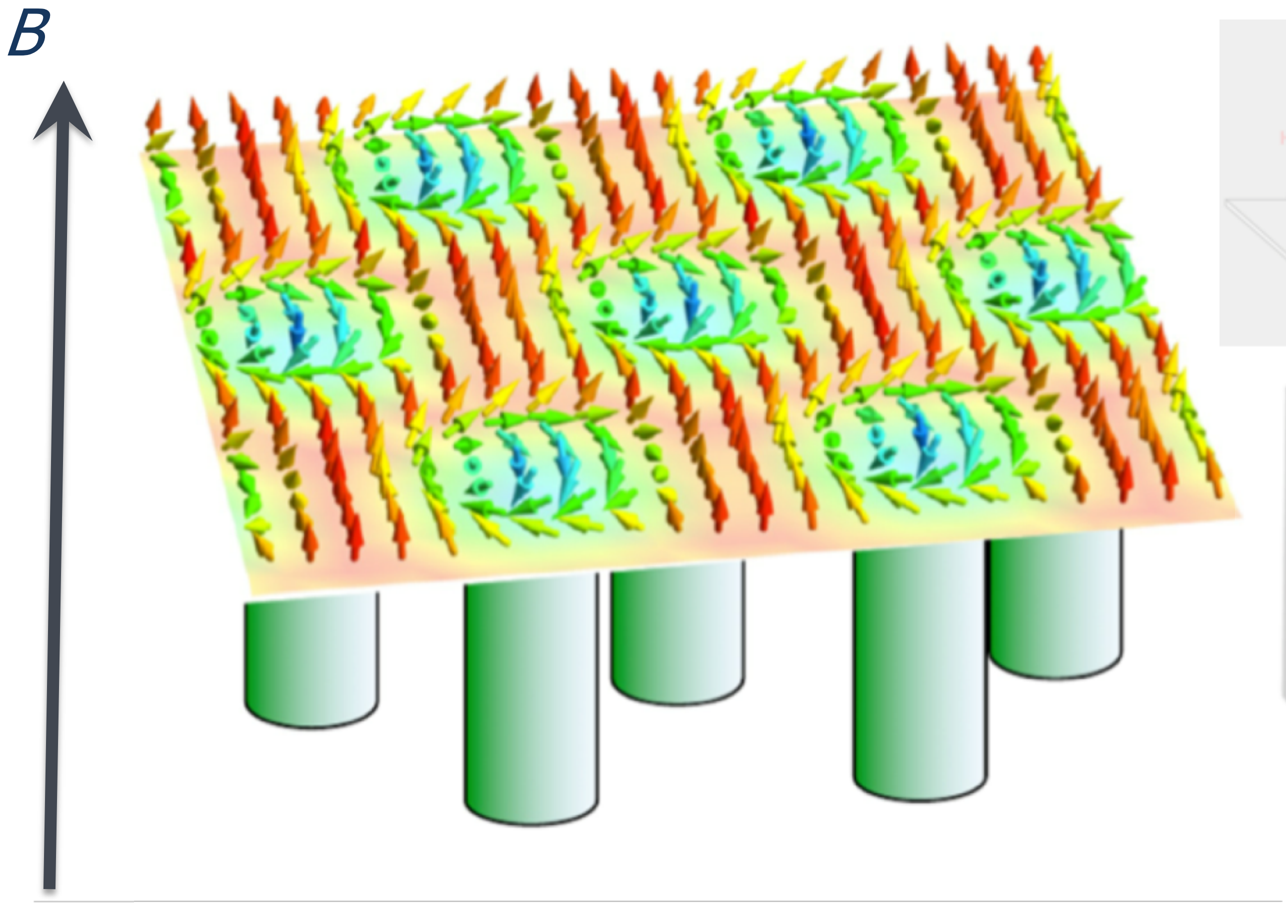
(1) ferromagnetism

(2) Dzyaloshinsky-Moriya

(3) crystal field ($P2_13$):
locked to <111> or <100>

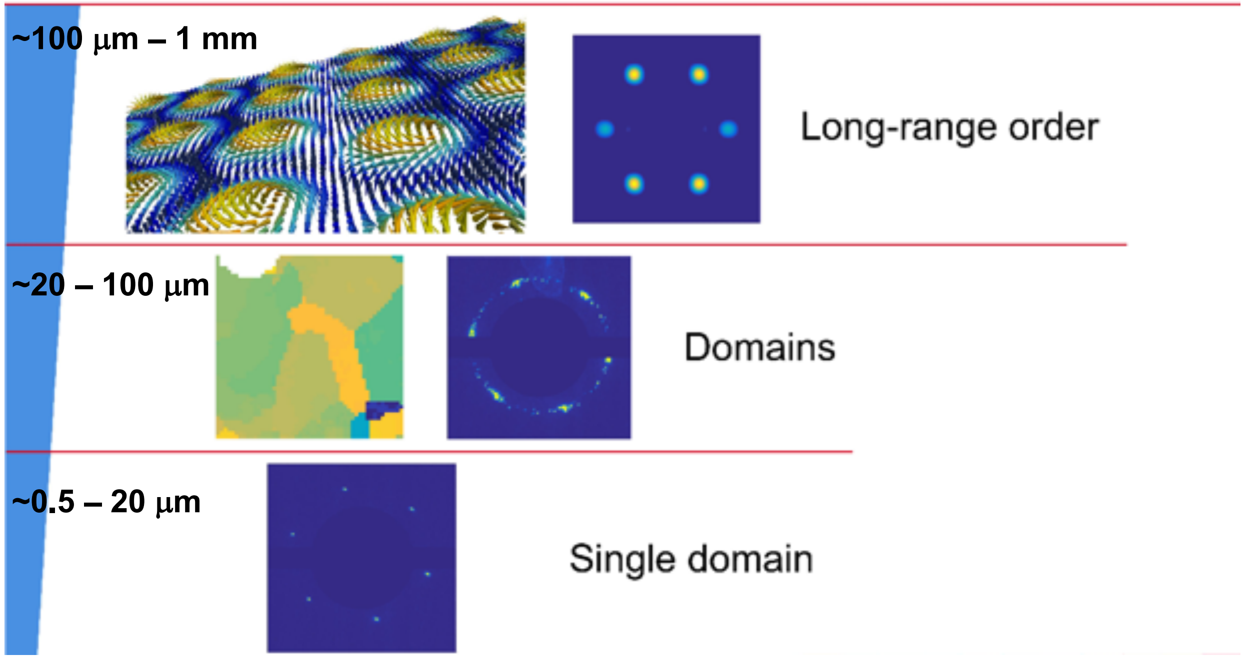


3D Skyrmion Lattice State – Skyrmion Tubes or Strings



H.S. Park et al., Nat. Nanotech. **9**, 337 (2014)

Overview of Lengthscales and Relevant Properties



PRB 93, 214420 (2016)

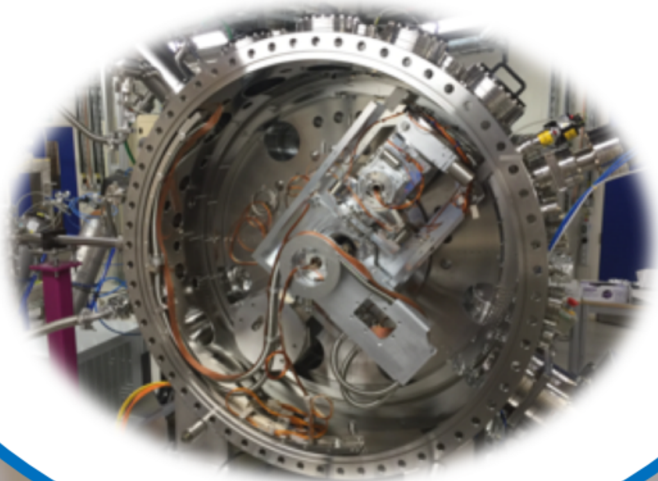
APL 109, 192406 (2016)

Nano Lett 16, 3285 (2016)

REXS on skyrmion lattices

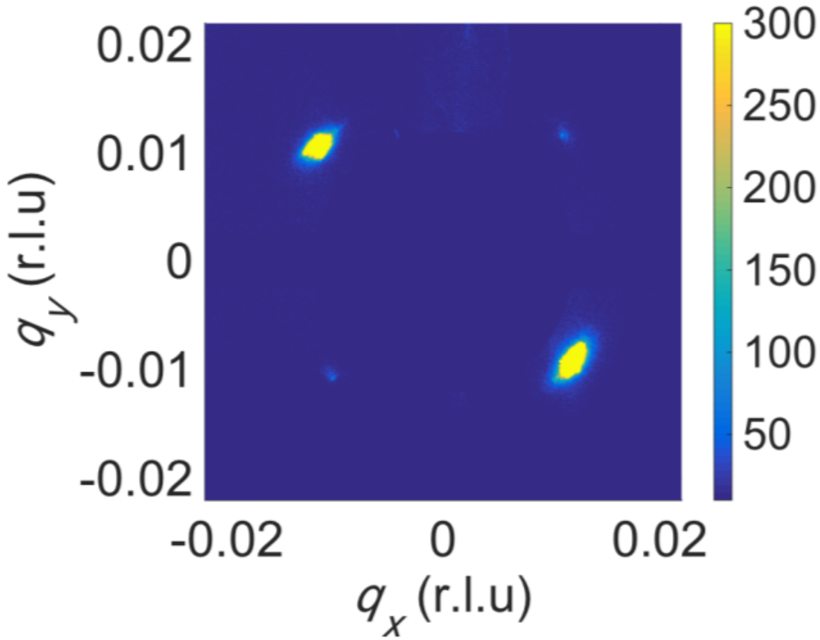
Resonant X-ray Magnetic Scattering Has Gone Soft

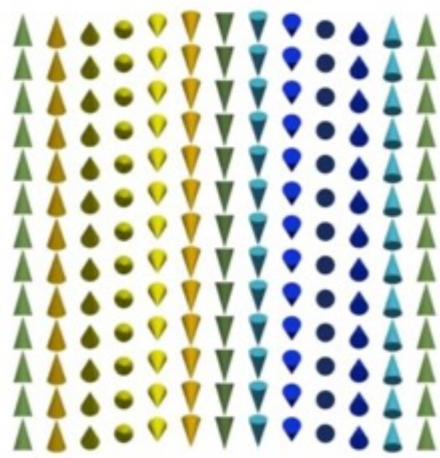
GERRIT VAN DER LAAN
Daresbury Laboratory, Warrington WA4 4AD, UK

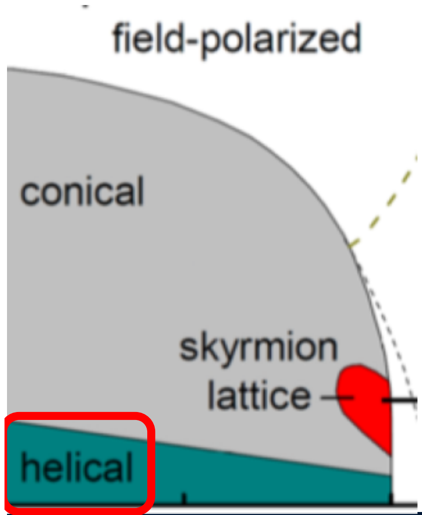


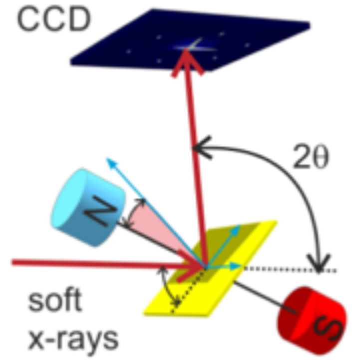
S. L. Zhang et al., Phys. Rev. B 93, 214420 (2016)

Cu_2OSeO_3 single crystal



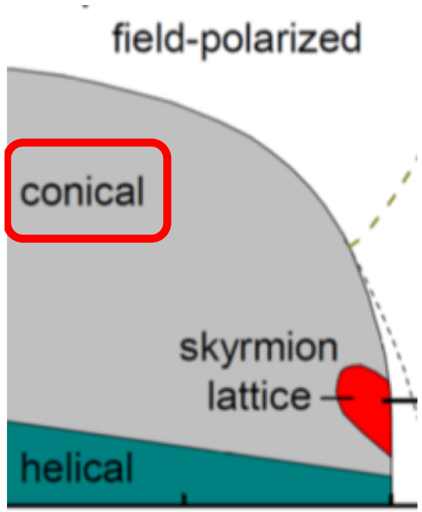


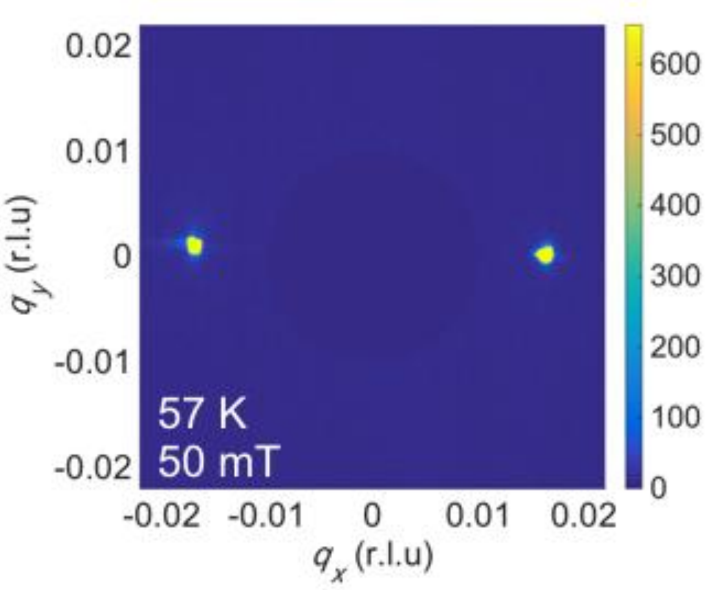


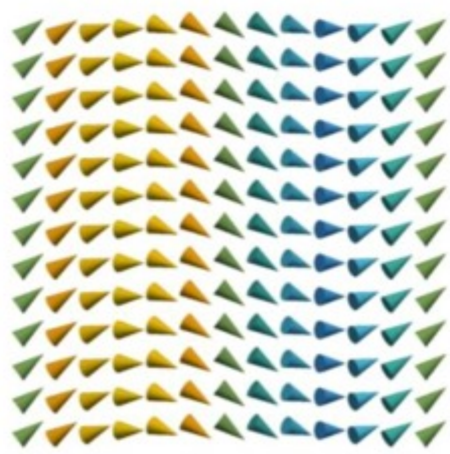


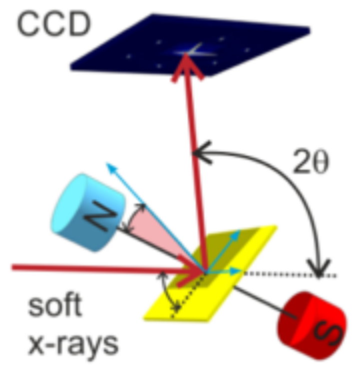
S. L. Zhang et al., Nano Lett. 16, 3285 (2016)

Cu_2OSeO_3 *single crystal*



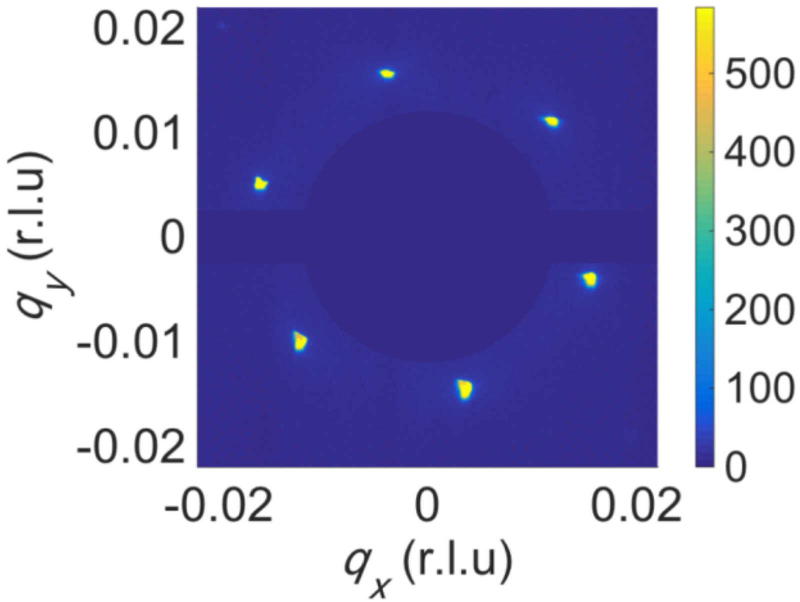


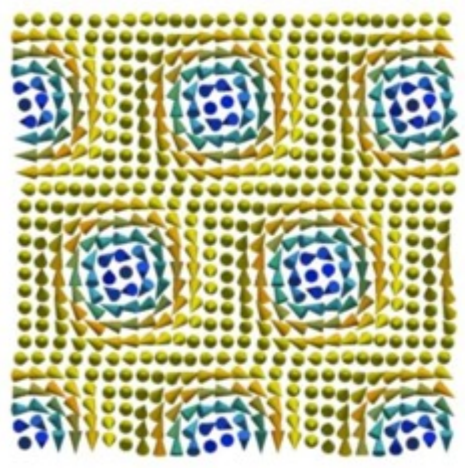


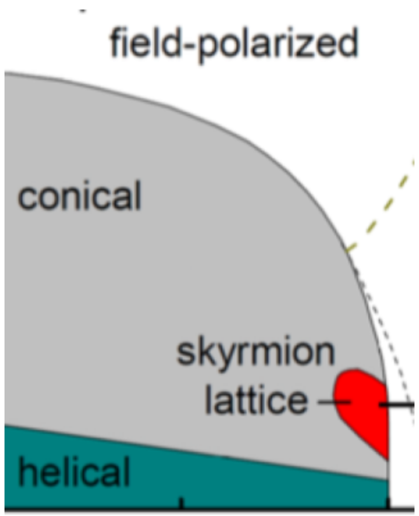


S. L. Zhang et al., Nano Lett. 16, 3285 (2016)

Cu_2OSeO_3 single crystal

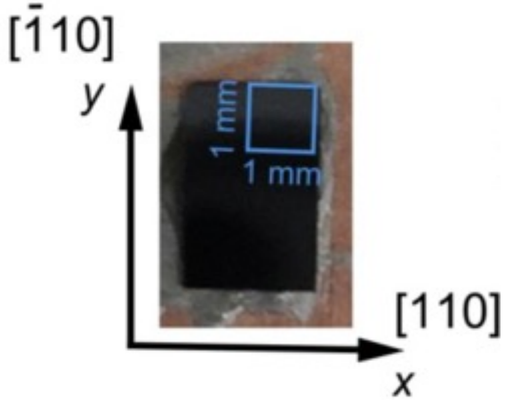


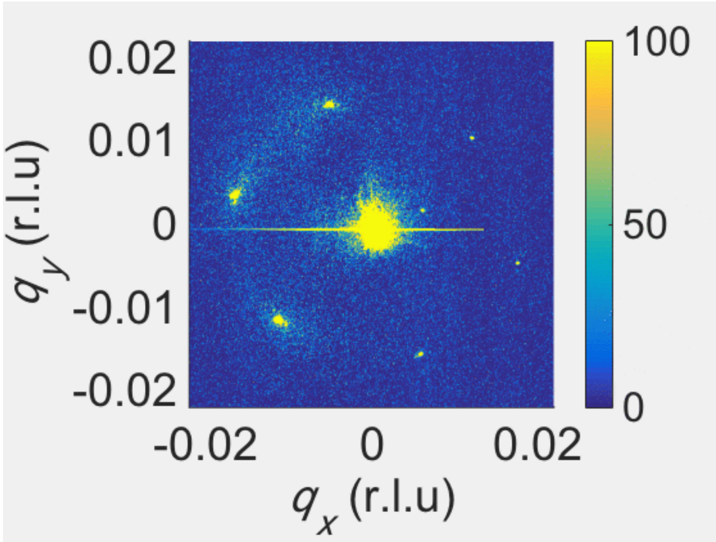




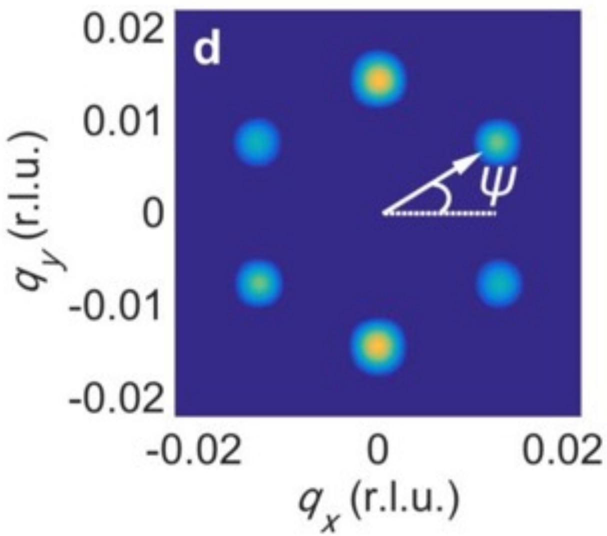
S. L. Zhang et al., Nano Lett. 16, 3285 (2016)

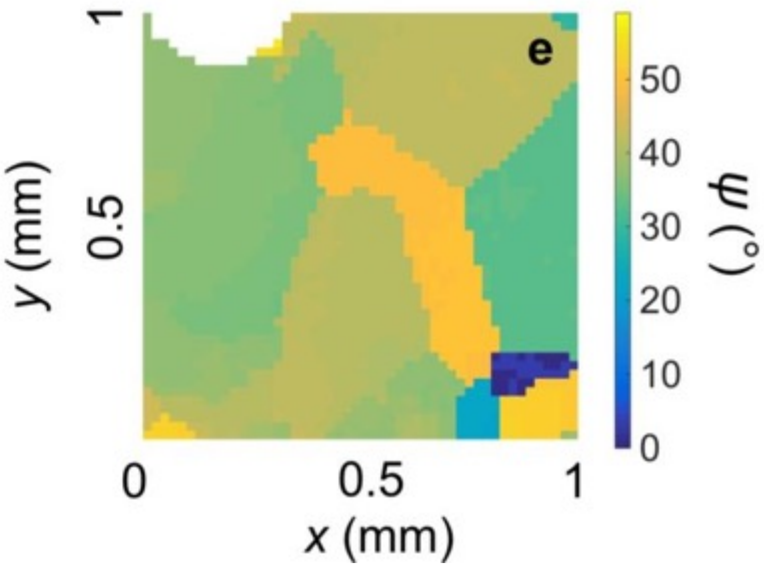
2D Lateral Raster Scan



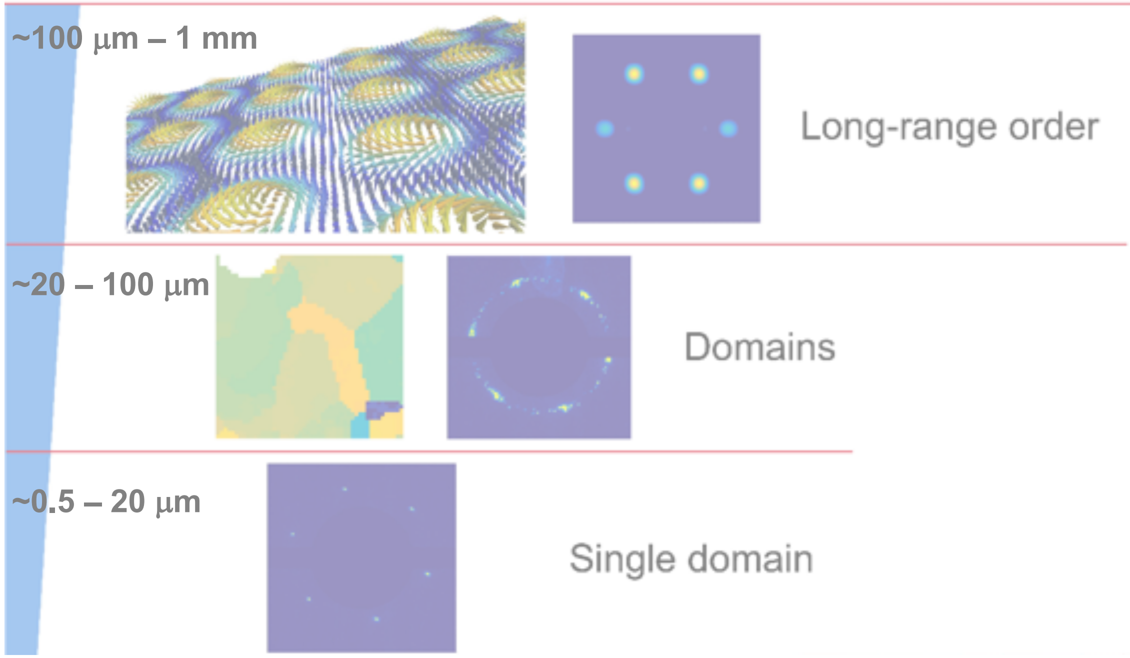


$\gamma = 10^\circ$





Overview of Lengthscales and Relevant Properties



… but what about the microscopic structure of a skyrmion?

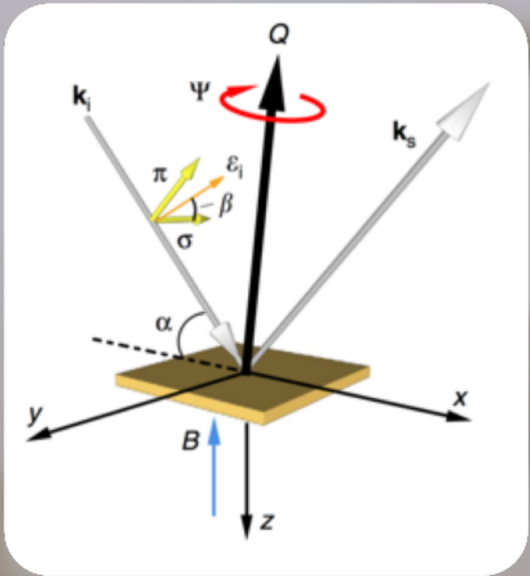
… what are the parameters to describe it?

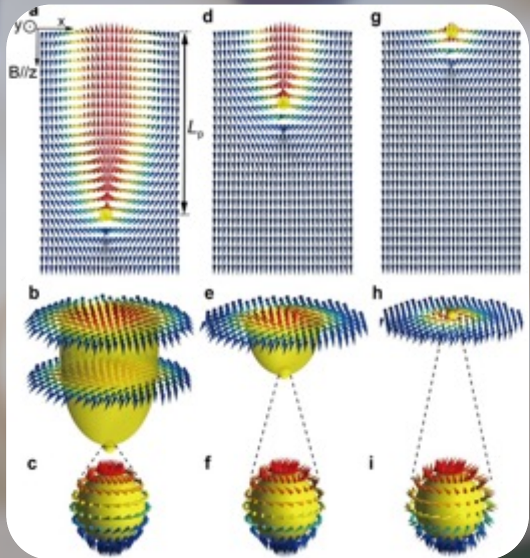
… and, most importantly, how can we measure it?

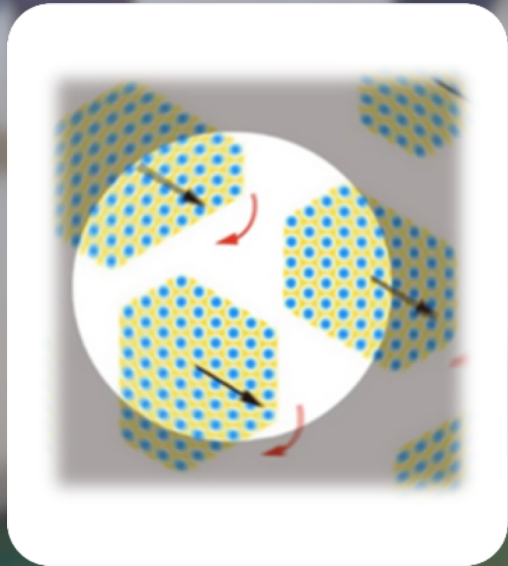
Advanced REXS Techniques

Exploration of 3D Spin Textures

Dynamics Across Different Timescales







Advanced REXS Techniques

Exploration of 3D Spin Textures

Dynamics Across Different Timescales

1. **Space** – *How to 'visualize' 3D magnetic structures?*

2. **Time** – *How to 'visualize' ps-scale spin dynamics?*

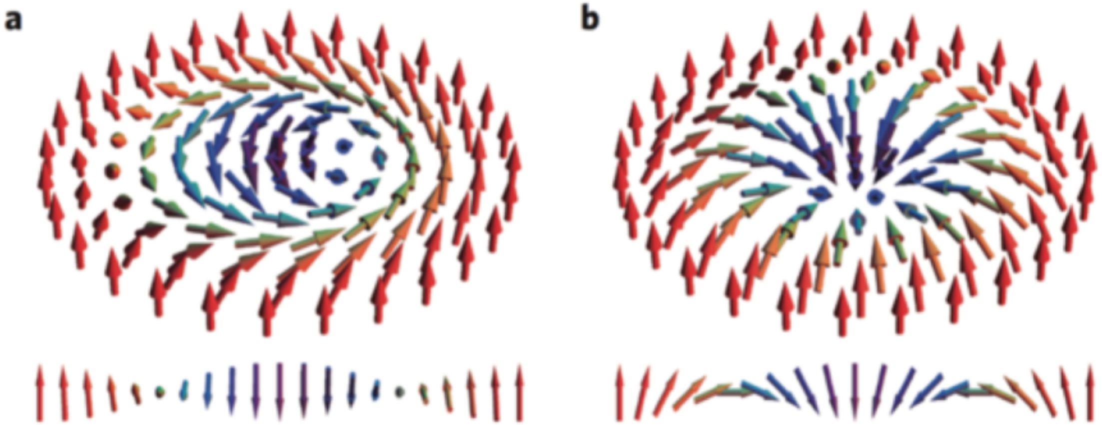
Skyrmion Texture: *Helicity χ and Winding Number N*

$$m_1(\rho, \psi) = M_S \sin[\theta(\rho)] \cos[N(\psi + \chi)]$$

$$m_2(\rho, \psi) = M_S \sin[\theta(\rho)] \sin[N(\psi + \chi)]$$

$$m_3(\rho, \psi) = M_S \lambda \cos[\theta(\rho)]$$

$\chi = \pm 90°$ $\chi = 0°, 180°$



Bloch Néel

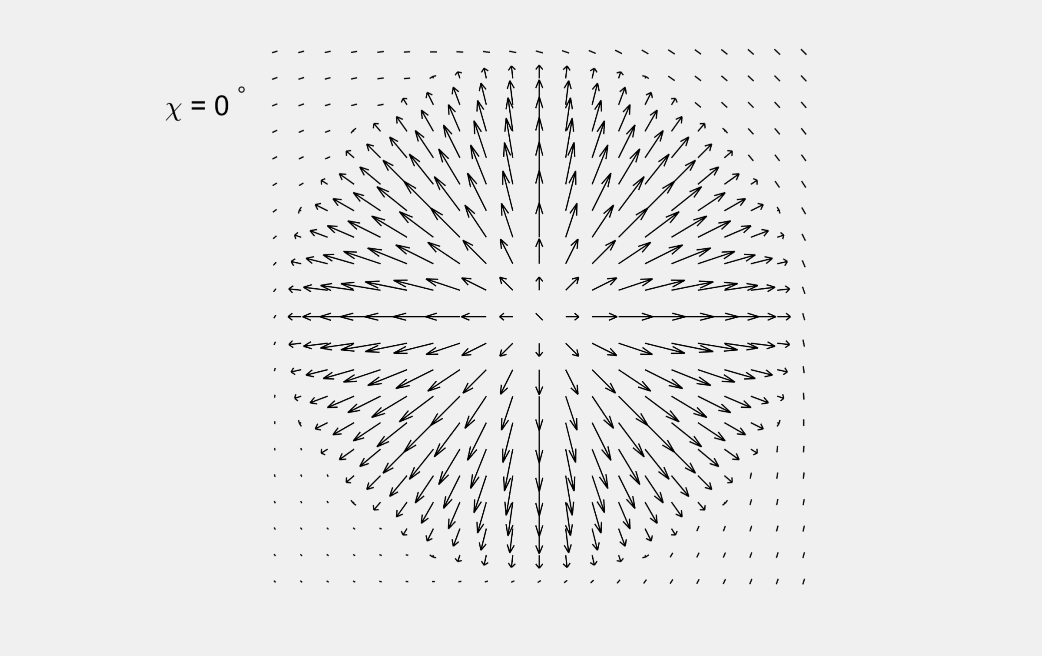
Skyrmion Texture: *Helicity χ and Winding Number N*

$$m_1(\rho,\psi) = M_S \sin[\theta(\rho)] \cos[N(\psi+\chi)]$$

$$m_2(\rho,\psi) = M_S \sin[\theta(\rho)] \sin[N(\psi+\chi)]$$

$$m_3(\rho,\psi) = M_S \lambda \cos[\theta(\rho)]$$

The helicity angle is a continuous variable

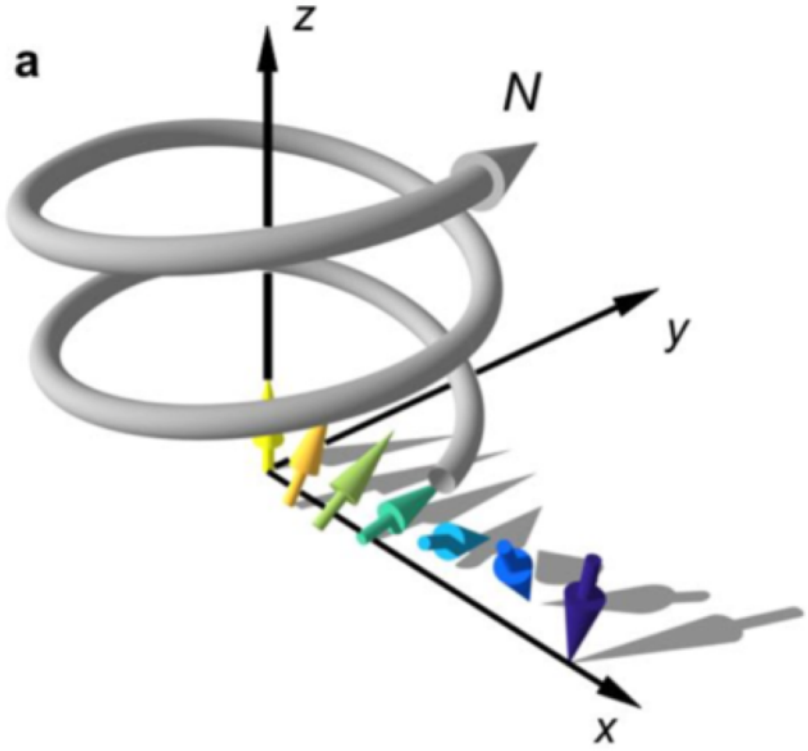


Description of a skyrmion

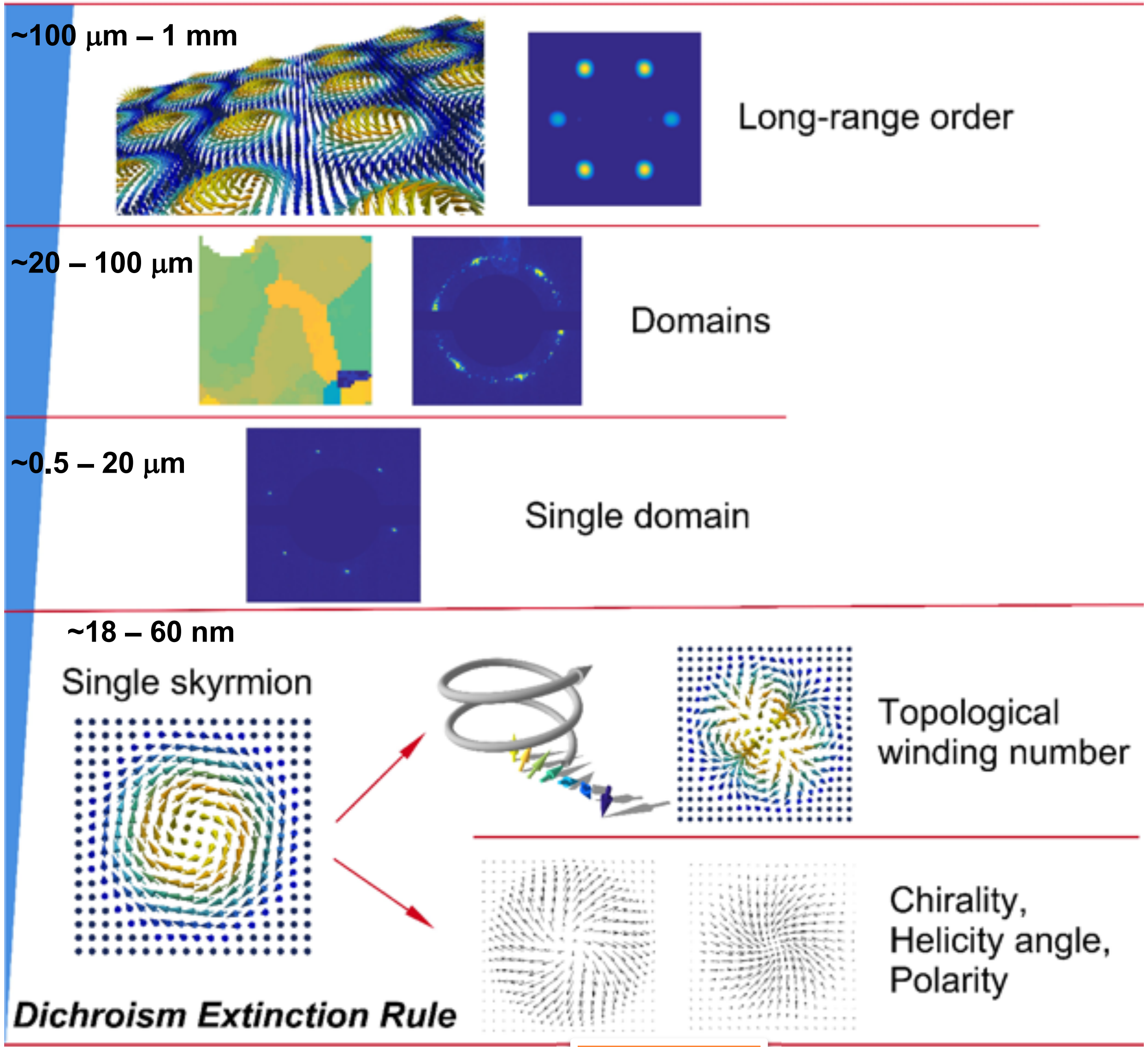
$$m_1(\rho,\psi) = M_S \sin[\theta(\rho)] \cos[N(\psi+\chi)]$$

$$m_2(\rho,\psi) = M_S \sin[\theta(\rho)] \sin[N(\psi+\chi)]$$

$$m_3(\rho,\psi) = M_S \lambda \cos[\theta(\rho)]$$



Overview of Lengthscales and Relevant Properties



CD-REXS

PRB 93, 214420 (2016)

APL 109, 192406 (2016)

Nano Lett 16, 3285 (2016)

Nat Com 8, 14619 (2017)

PRB 96, 094401 (2017)

PNAS 115, 6386 (2018)

PRL 120, 227202 (2018)

Measurement of the Microscopic Information:

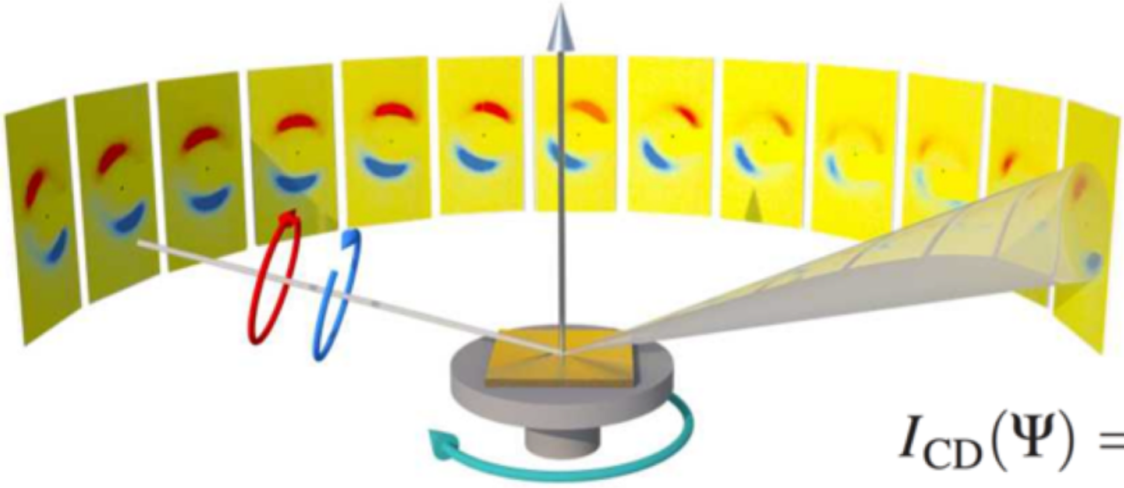
Circular Dichroism & REXS

Dichroism Extinction Rule (DER)

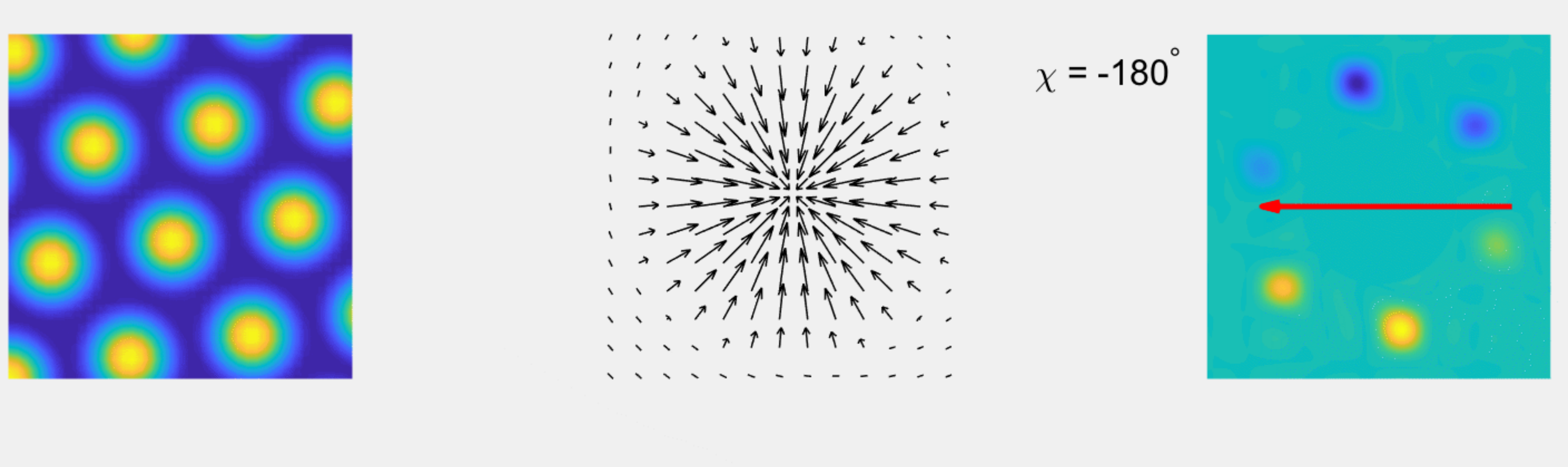
$$I_{\mathrm{CD}}(\Psi) = \lambda Y \sin(\Psi + \chi)$$

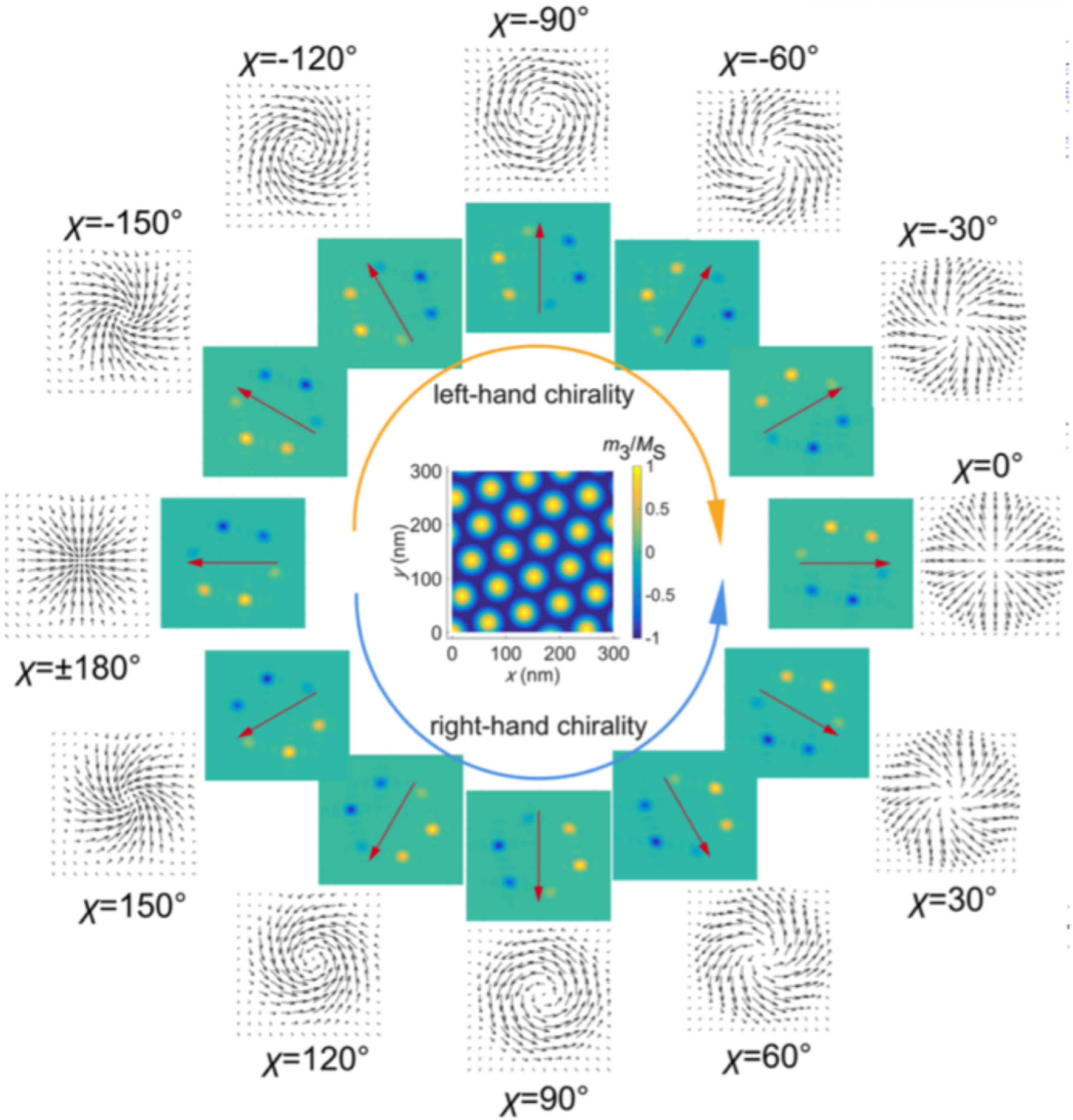
S. L. Zhang et al., Phys. Rev. B 93, 214420 (2016)

CD-REXS



$$I_{\mathrm{CD}}(\Psi) = \lambda Y \sin(\Psi + \chi)$$

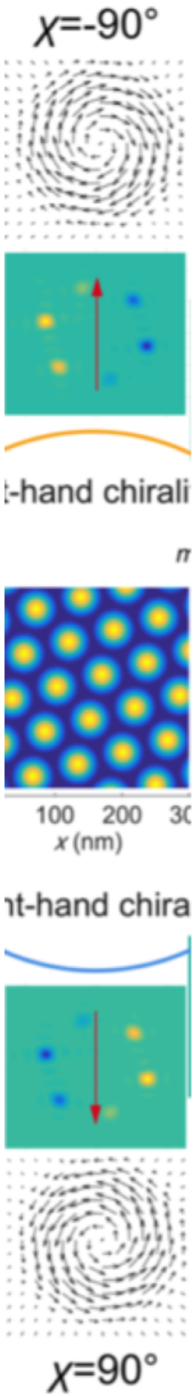



χ=-90°
χ=-120°
χ=-60°
χ=-150°
χ=-30°
left-hand chirality
m_3/M_S
χ=0°
χ=±180°
right-hand chirality
χ=150°
χ=30°
χ=120°
χ=60°
χ=90°
y (nm)
x (nm)

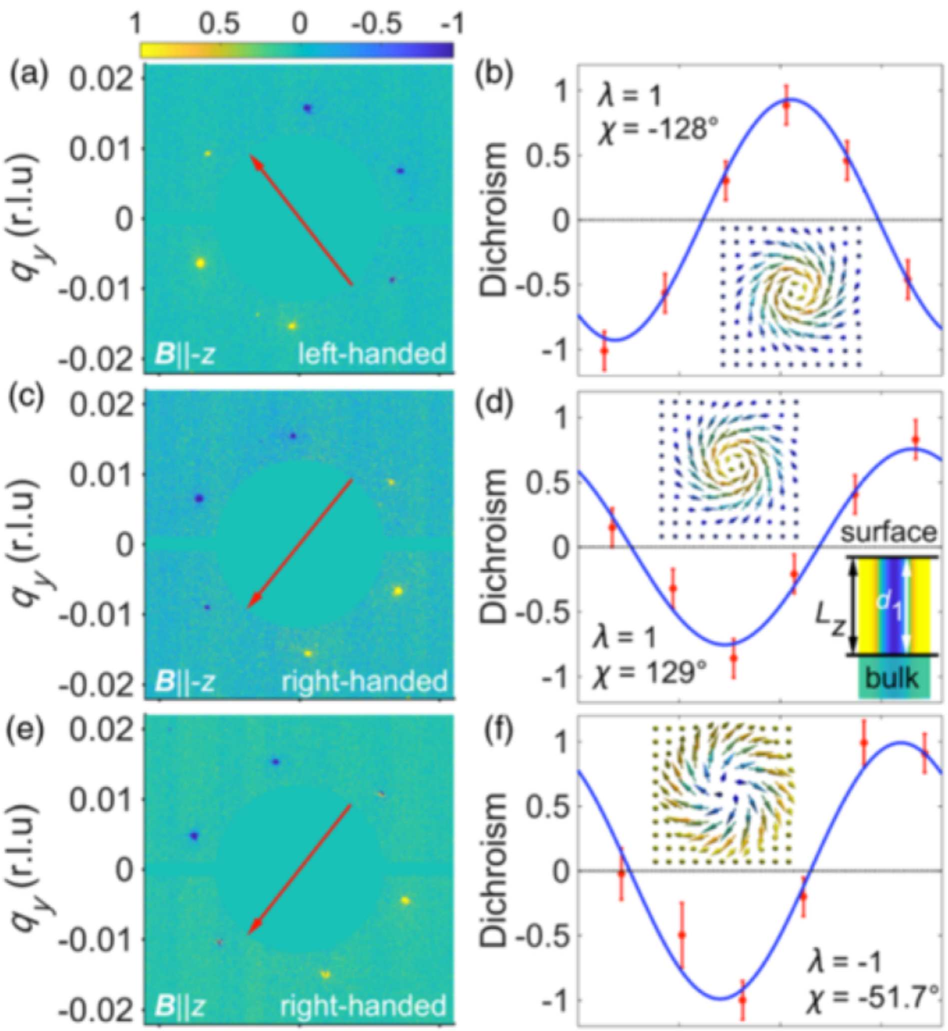
Expected behavior for Bloch-type skyrmions

=

Skyrmions in B20 systems



Dichroism Extinction Rule: Experiments on CSO



Left-handed crystal

Right-handed crystal

Right-handed crystal/
field reversed

S. L. Zhang et al., *Phys. Rev. Lett.* **120**, 227202 (2018)

Summary: *Dichroism Extinction Rule*

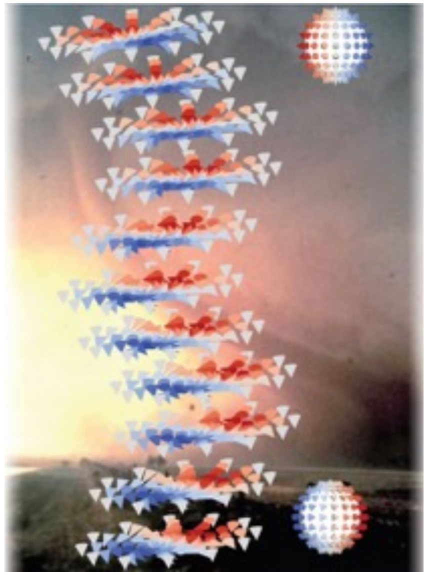
- **Topological winding number** = (Number of extinctions)/2
- **Chirality** → sign of slope around extinction points

Angles

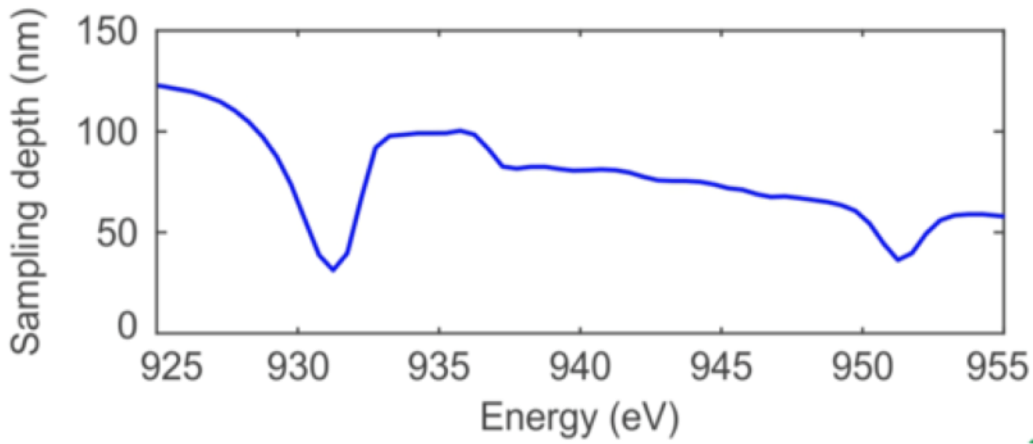
- Azimuthal canting angle of the spins → azimuthal angles of extinction points
- Tilting angle of the spins → asymmetry of the measured profile
- Conical spin angle → magnitude of the circular dichroism signal

3D Skyrmions

The Surface Twist Effect



Energy Scan → Depth-Dependent CDREXS



CD amplitude as a function of azimuthal angle Ψ

- 2D skyrmion $I(\Psi) = Y\sin(\Psi + \chi)$ *Y*: constant (scat. geometry; photon energy)

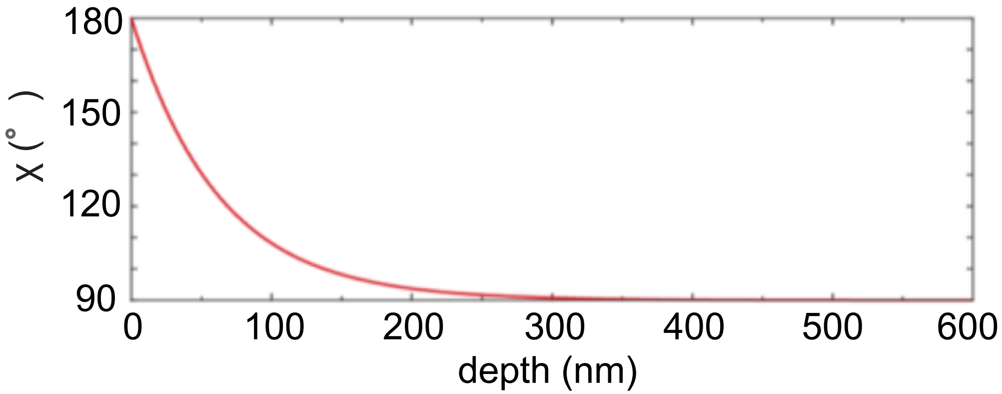
- 3D system → nonuniform χ(*z*) measured I_m = average w/ weighing factor *b*(*z*)

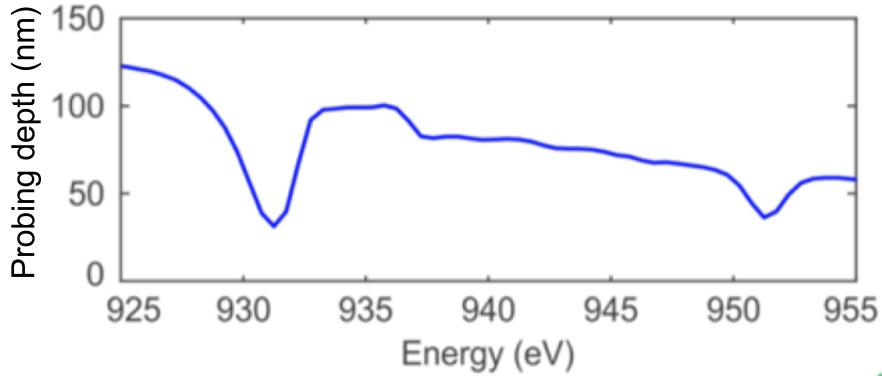
$$I_m(\Psi) = Y\sum_{z=0}^{\infty}\sin[\Psi + \chi(z)]b(z)$$

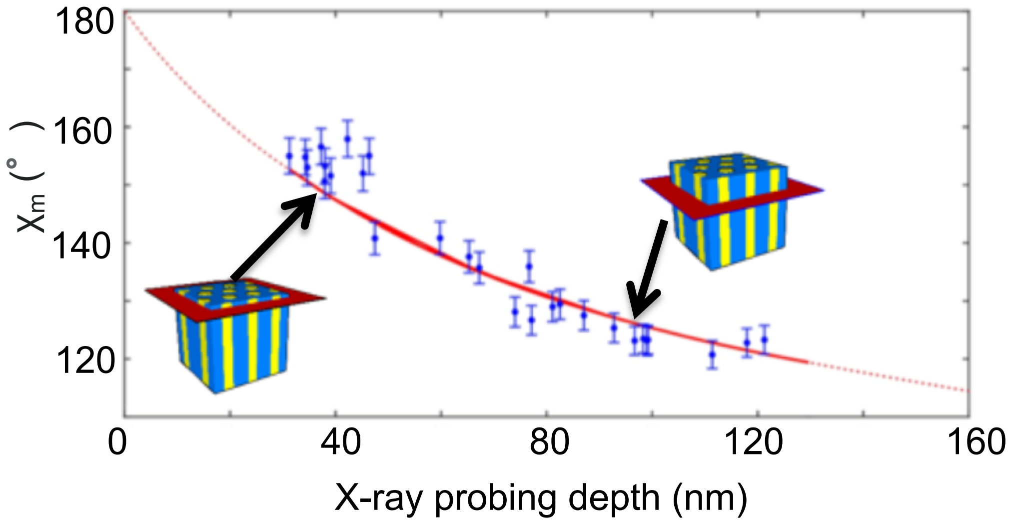
$$b(z) = e^{-2z\sec\alpha/\Lambda(\hbar\omega)}/\Lambda(\hbar\omega)$$

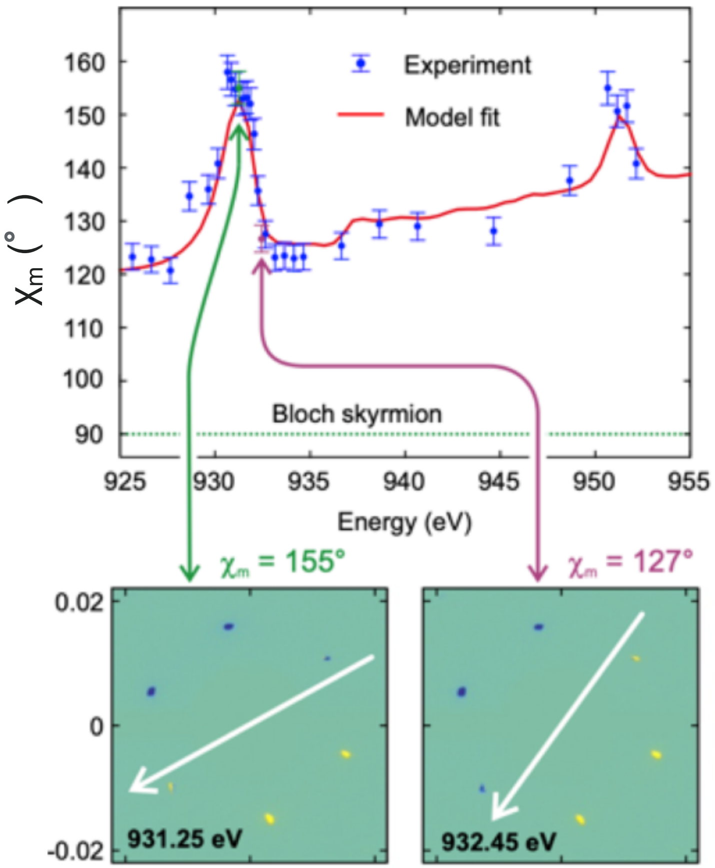
Λ – x-ray penetration length (photon energy)
α – incident angle wrt surface normal

Depth Dependence



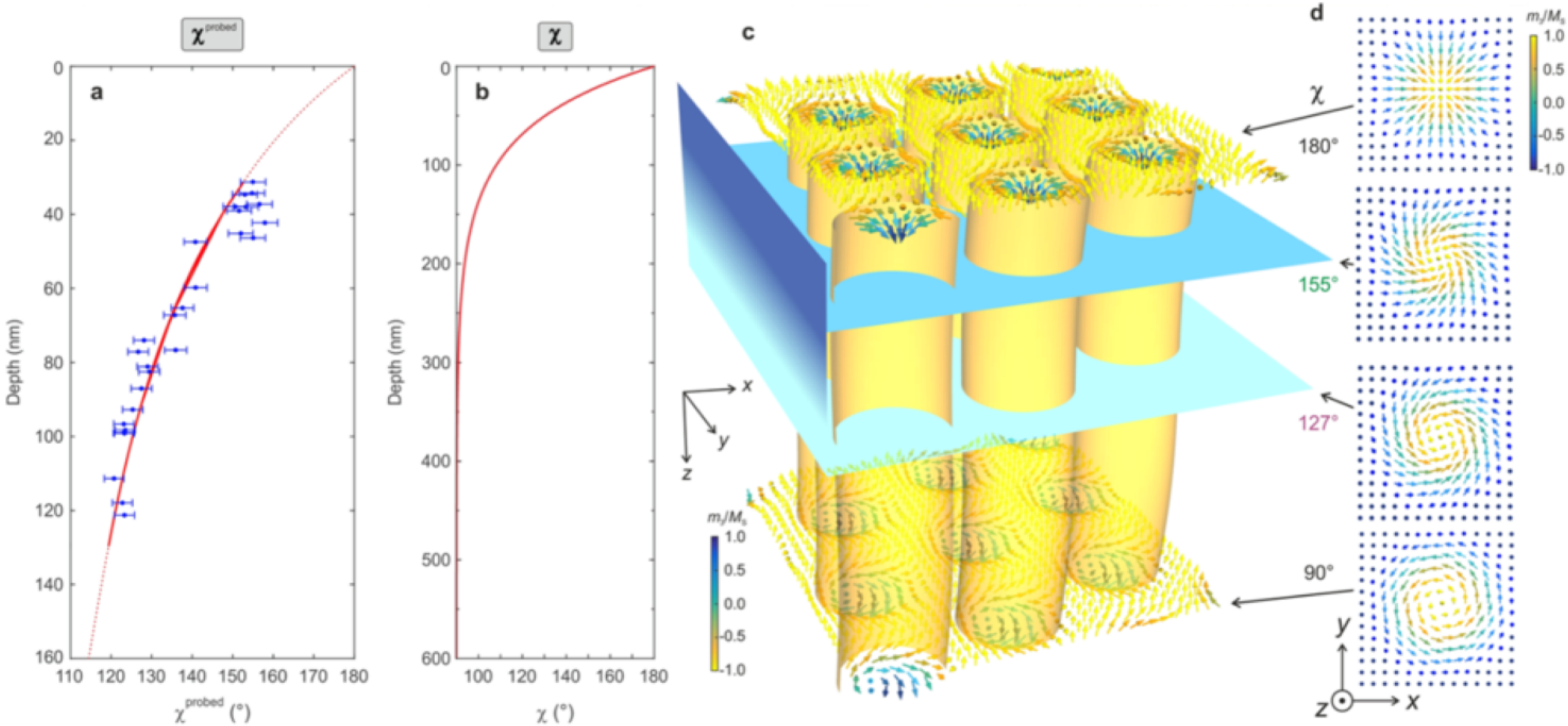






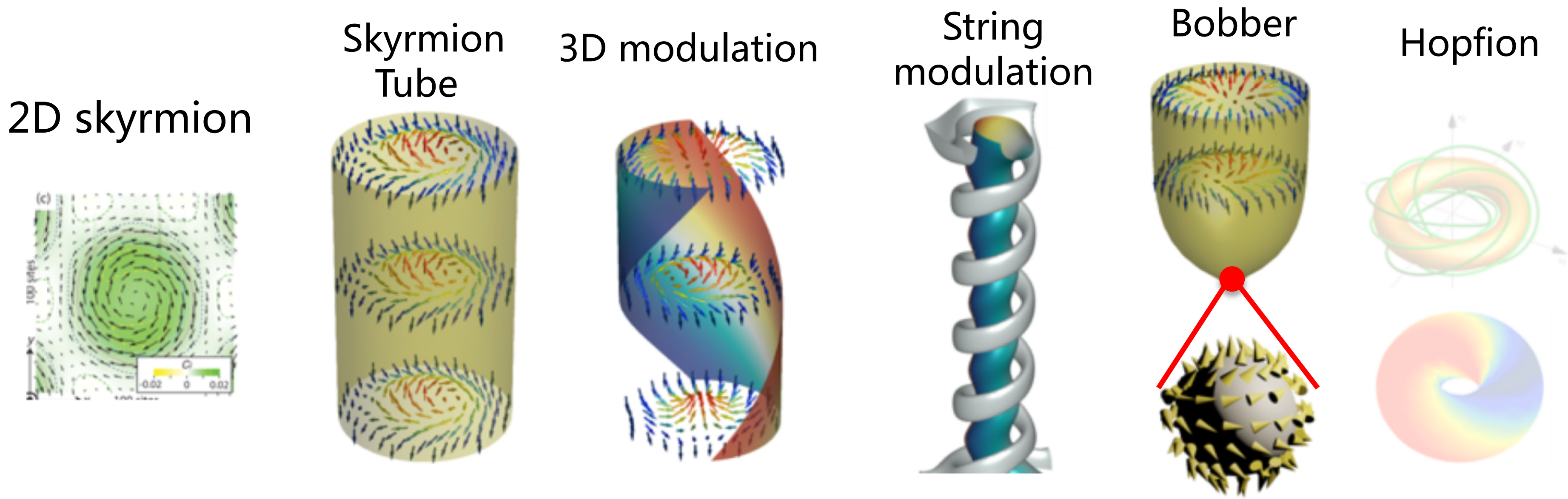
S. L. Zhang et al., *PNAS* **115**, 6386 (2018)

Surface State

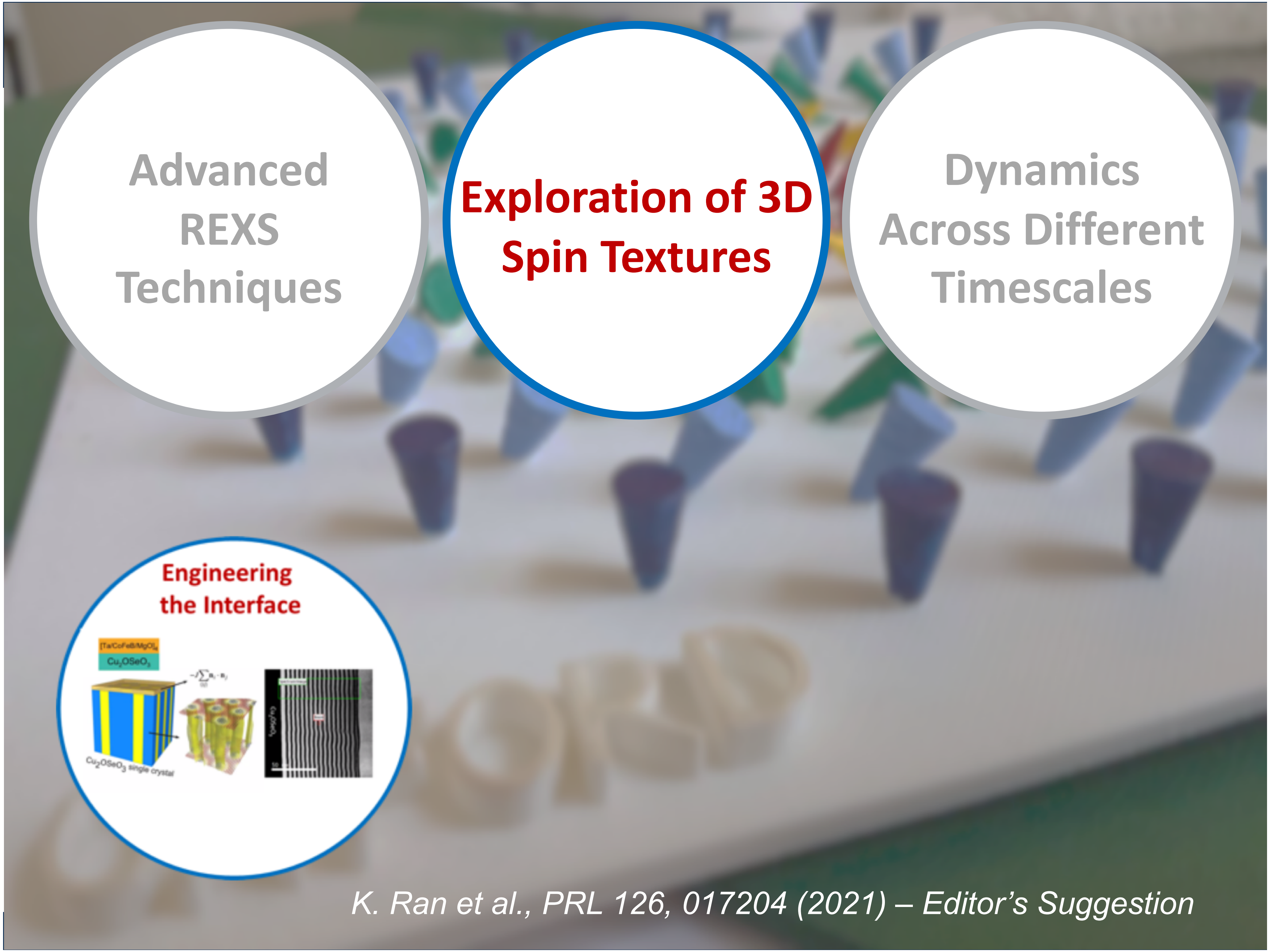


S. L. Zhang et al., *PNAS* **115**, 6386 (2018)

3D Skyrmion Structures in Chiral Magnets

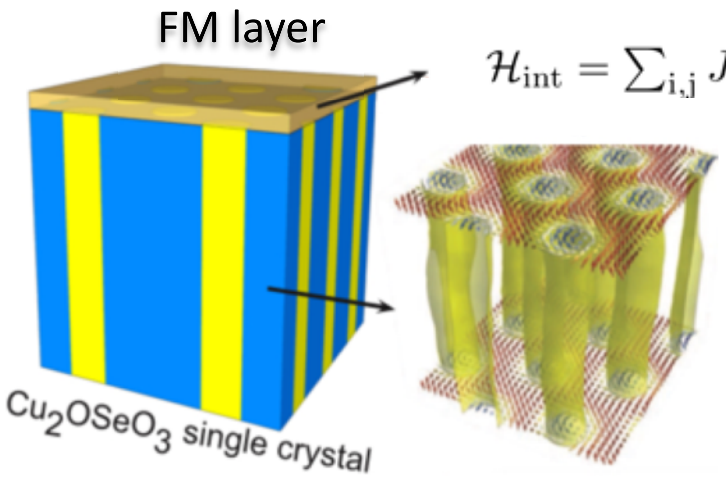


- **How can we stabilize these textures?**
- **And how can they be experimentally studied?**


Advanced REXS Techniques
Exploration of 3D Spin Textures
Dynamics Across Different Timescales
Engineering the Interface
[Ta/CoFeB/MgO]4
Cu2OSeO3
Cu2OSeO3 single crystal
K. Ran et al., PRL 126, 017204 (2021) – Editor's Suggestion

Interface Engineering – Chiral Bulk Magnet + Heterostructure

Magnetic heterostructure → provision of an extra exchange mechanism

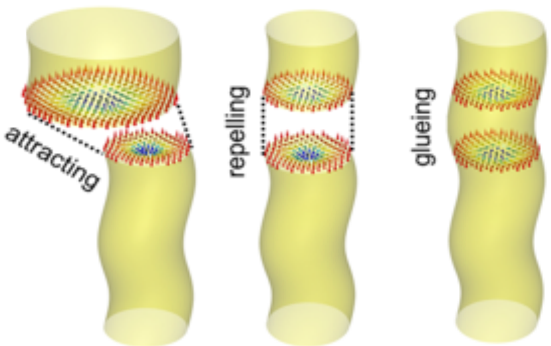


$\mathcal{H}_{\mathrm{int}} = \sum_{\mathrm{i,j}} J_{\mathrm{RKKY}}(\mathbf{S}_i \cdot \mathbf{S}_j)$ ⟺ interfacial exchange

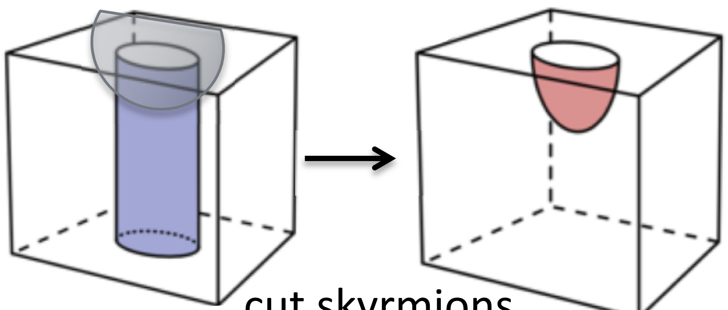
⇩

- locks the positions of the skyrmions
- aligns them with the bulk skyrmions

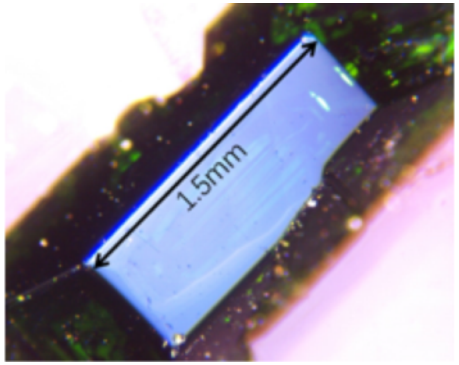
1. Imprinting effect



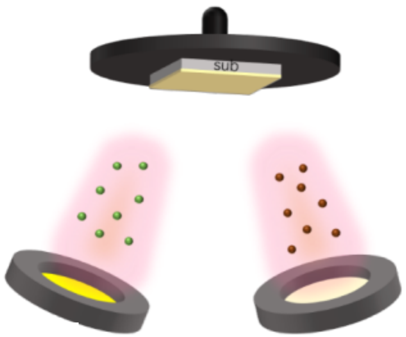
2. Effective field distribution



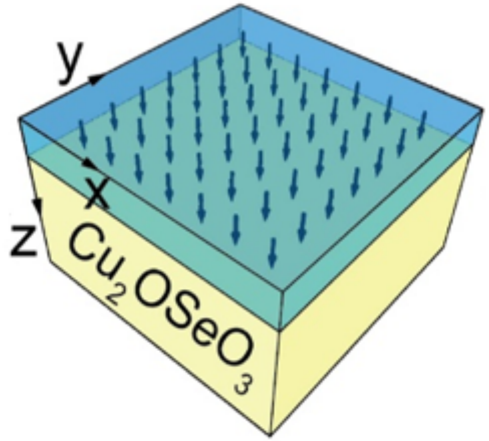
cut skyrmions
into bobbers



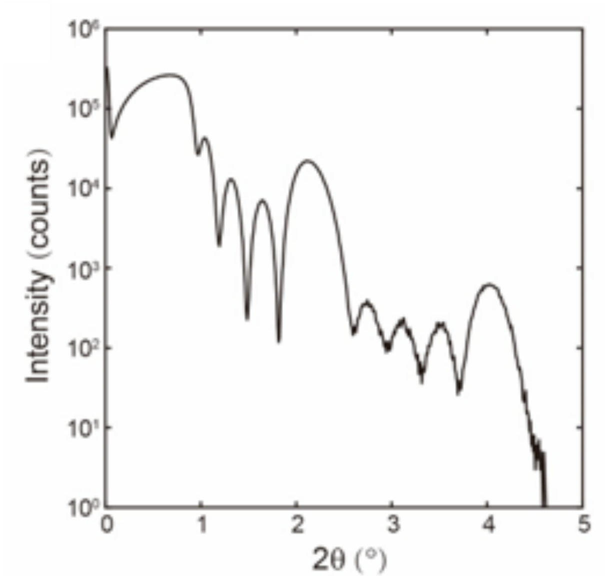
Polished single crystal

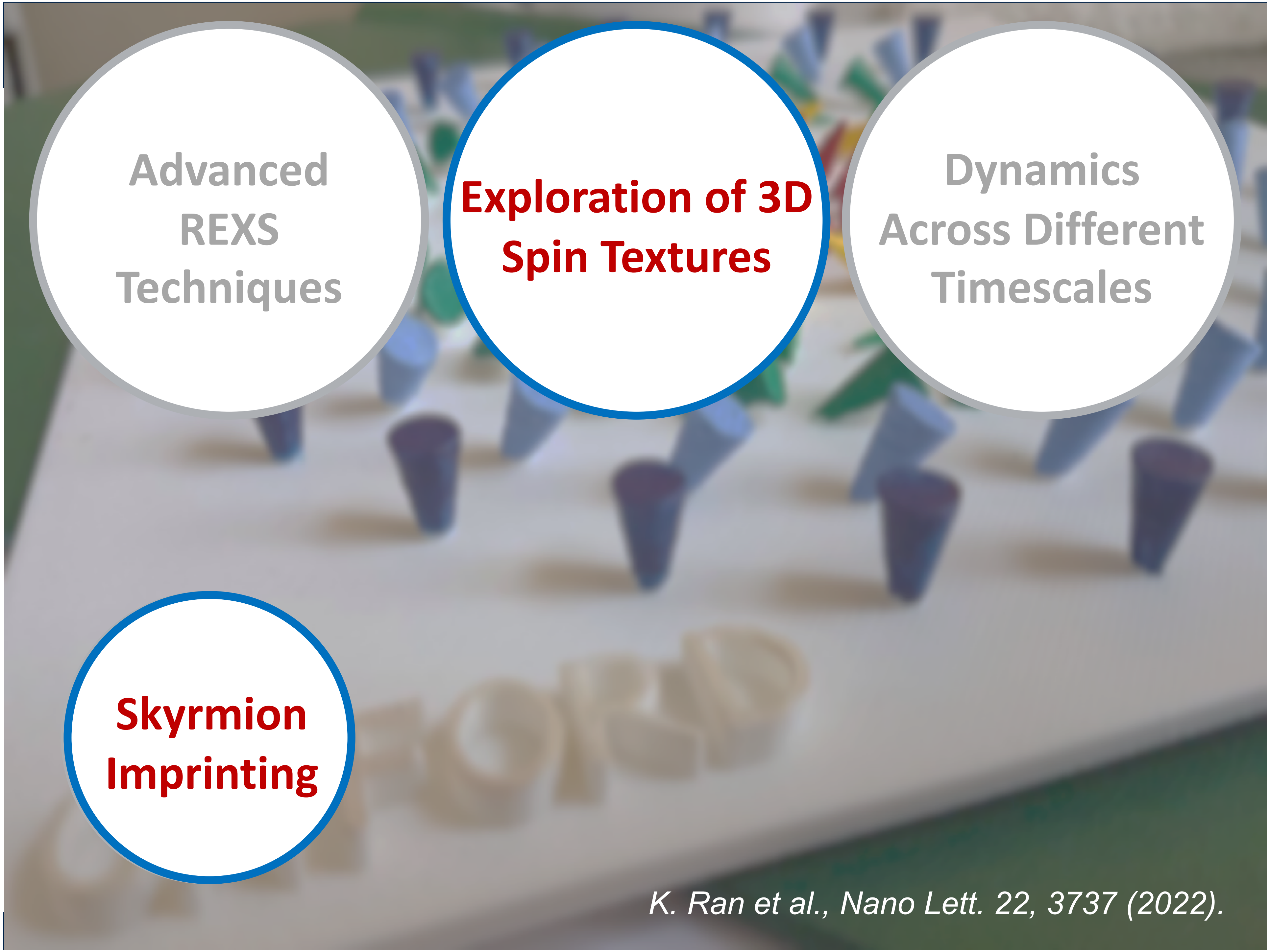


Magnetron sputtering



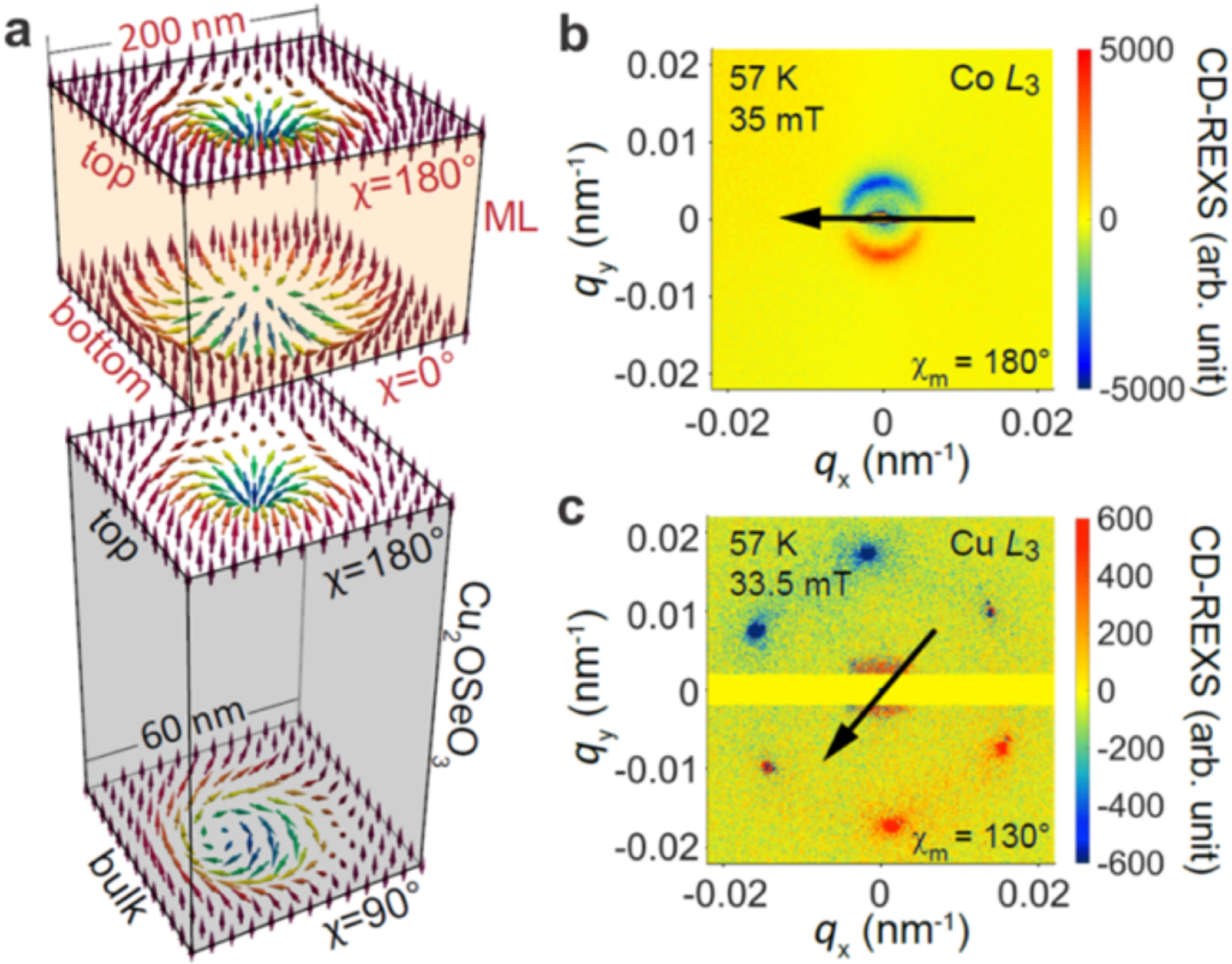
Heterostructure





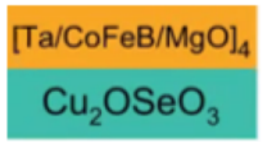
K. Ran et al., Nano Lett. 22, 3737 (2022).

Joining Skyrmion Systems: Chiral Bulk and Multilayer

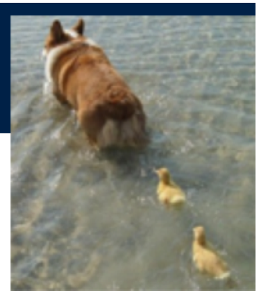


CDREXS measured in the skyrmion phase of the ML/Si

CDREXS measured in the skyrmion phase of Cu_2OSeO_3



Imprinting Effect

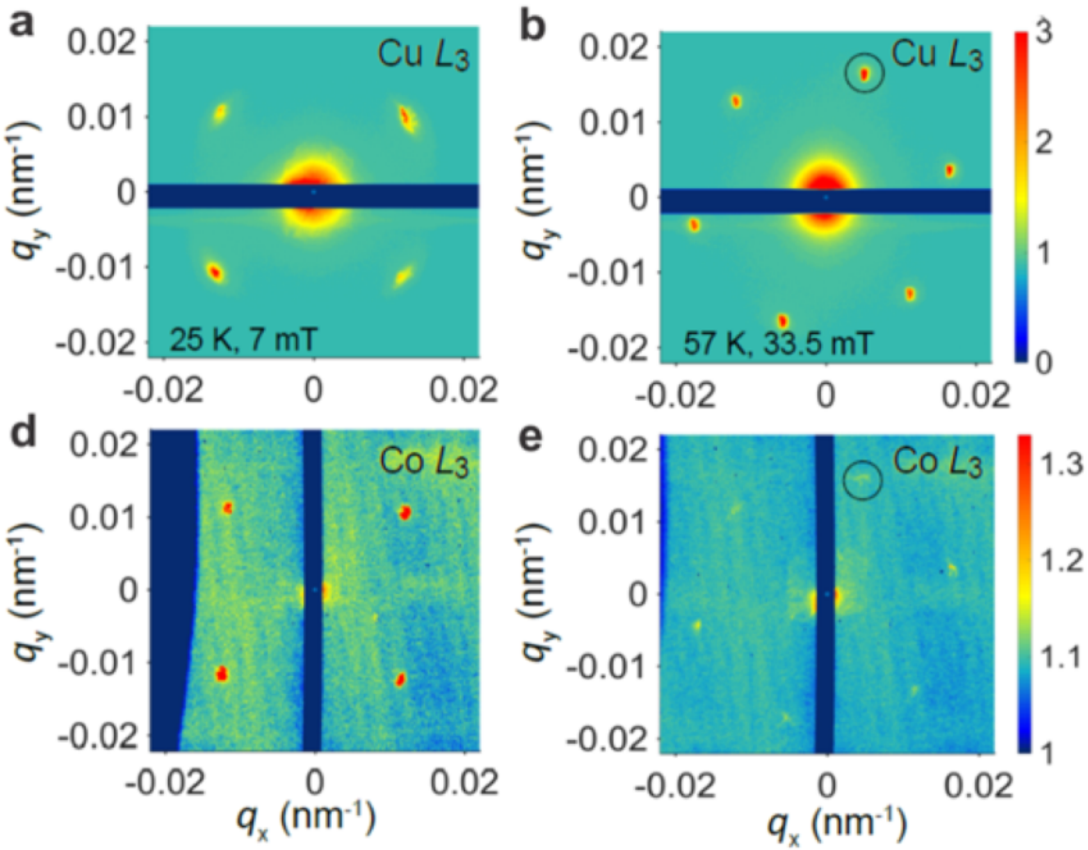


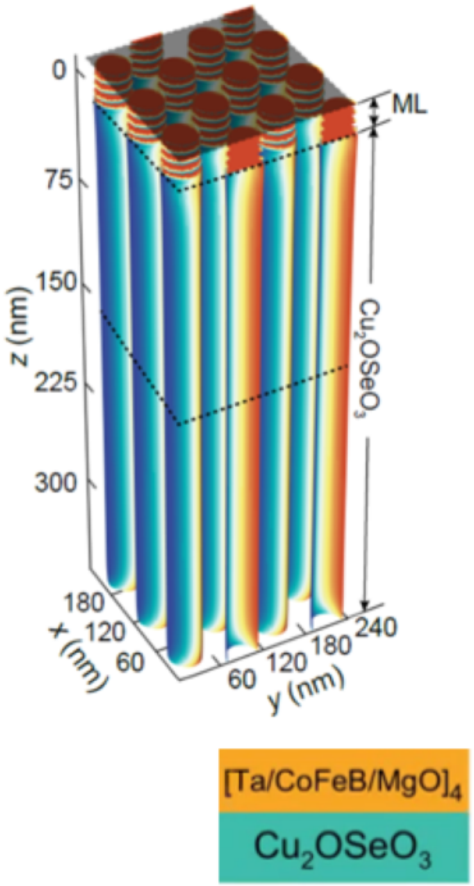
helical phase

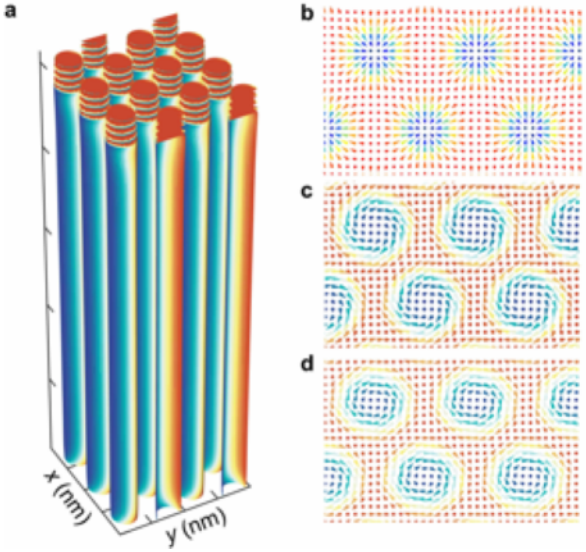
skyrmion phase

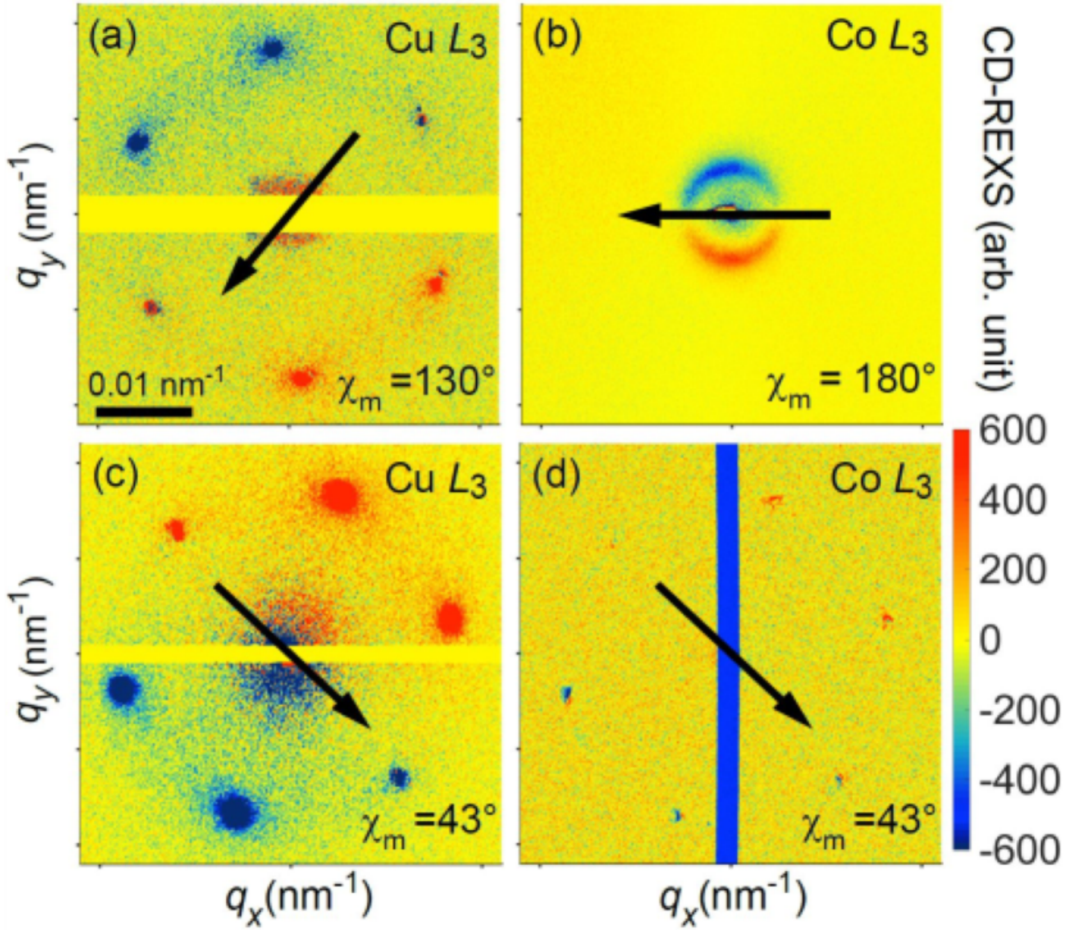
@ Cu edge

@ Co edge



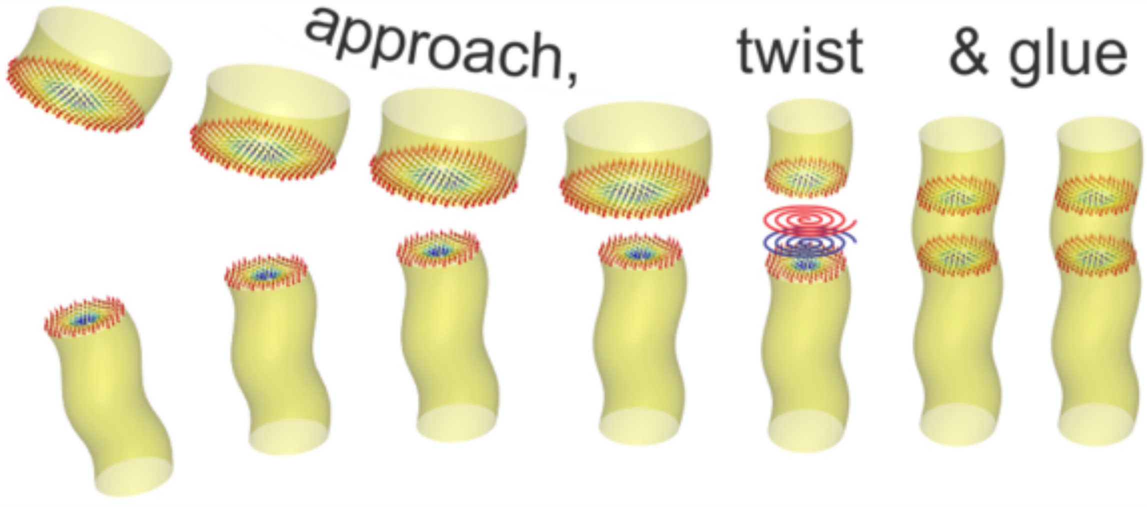


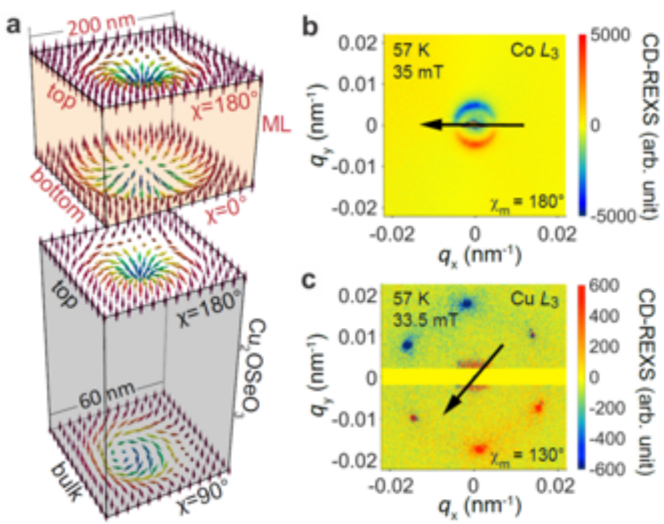


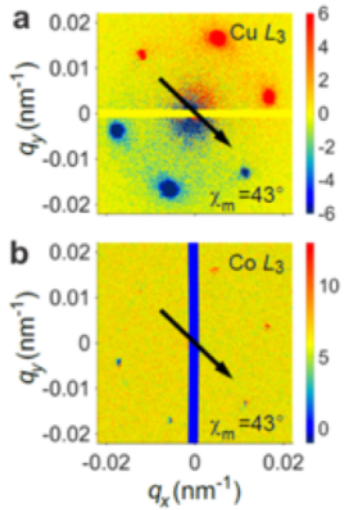


CD-REXS pattern measured in the skyrmion phase of Cu_2OSeO_3 on
(a) pristine Cu_2OSeO_3, (b) on Si/ML, and on
(b) Cu_2OSeO_3/ML at the (c) Cu and (d) Co L_3 edges.

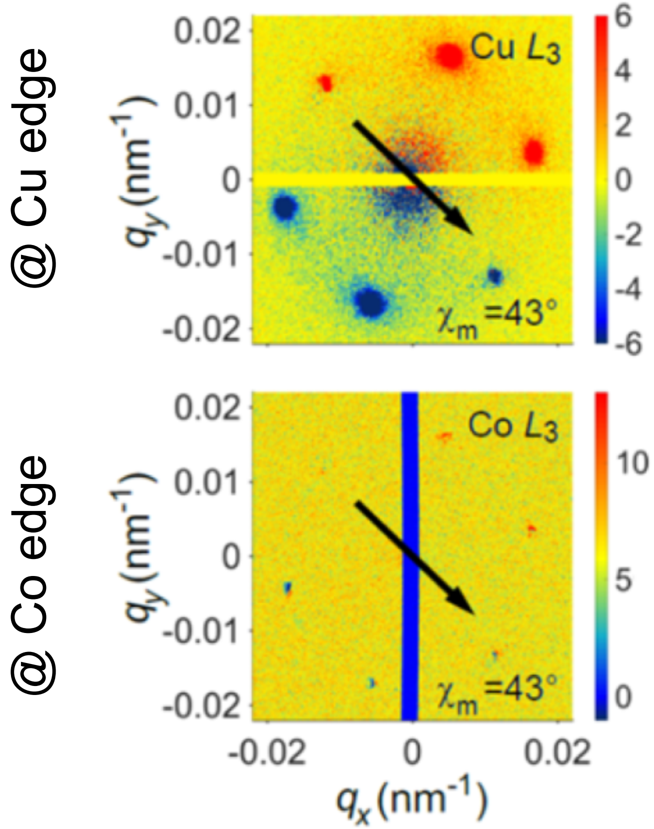
Approach, Twist & Glue

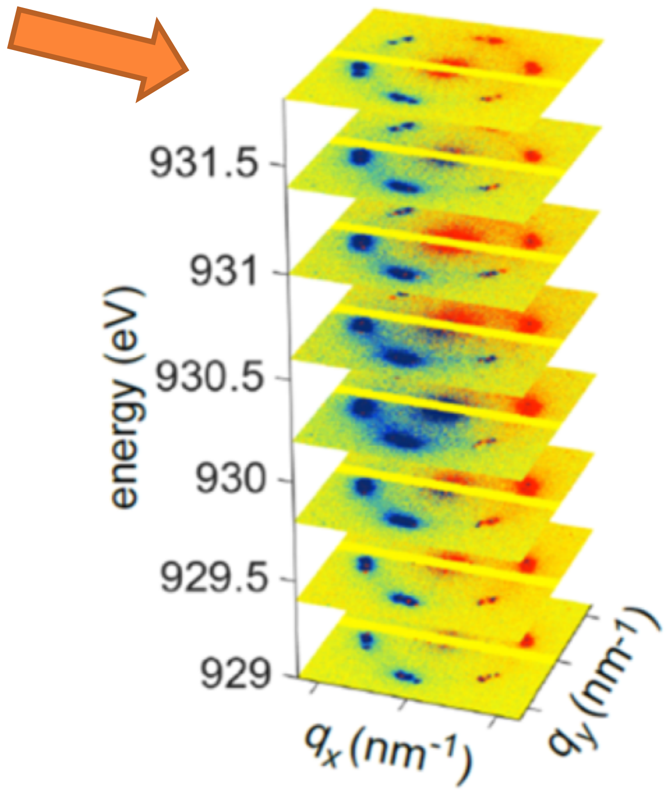






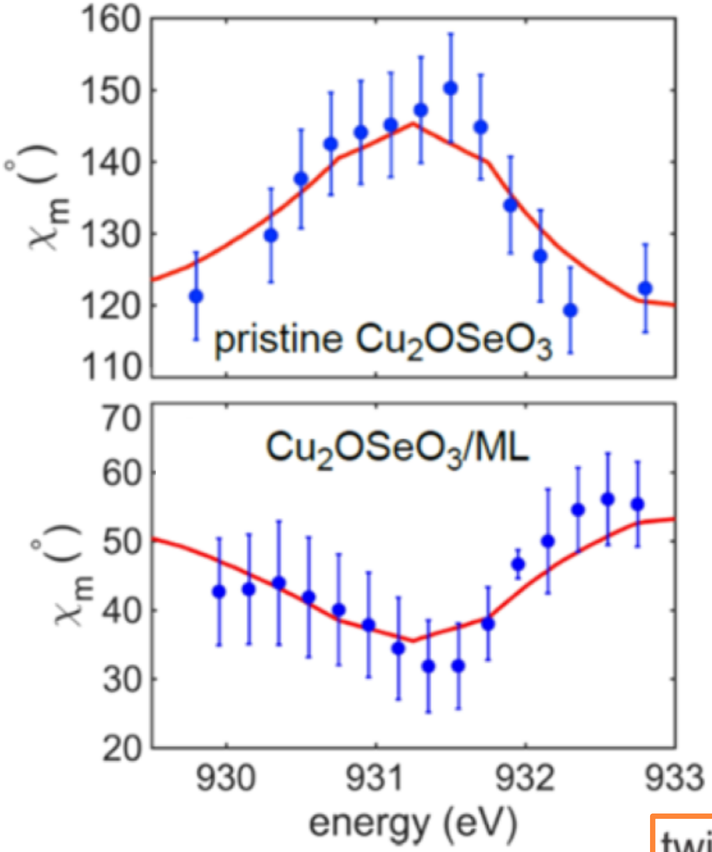
Depth-Resolved CD-REXS of Cu_2OSeO_3/ML

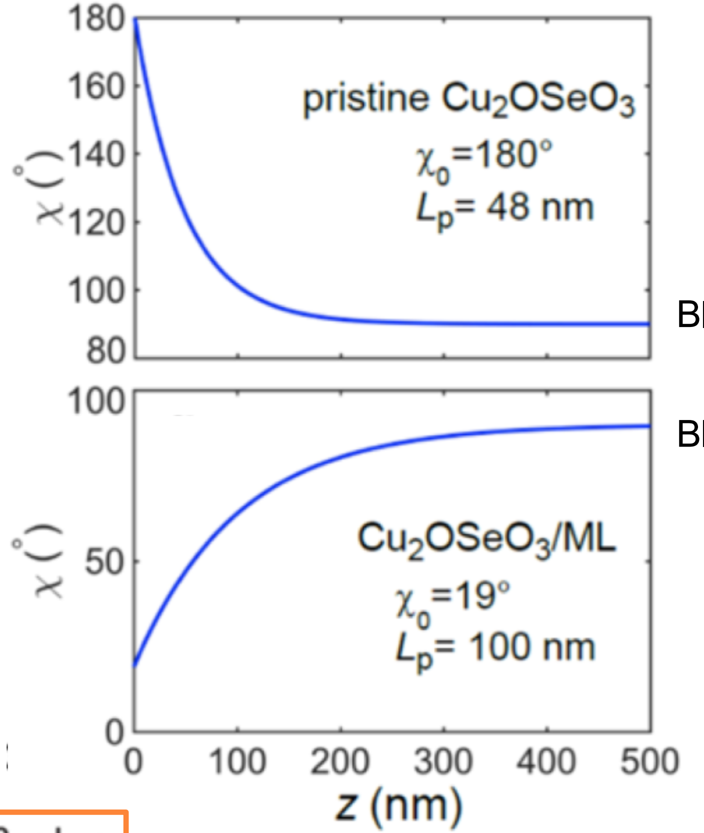




Depth-Resolved CD-REXS of Cu_2OSeO_3/ML

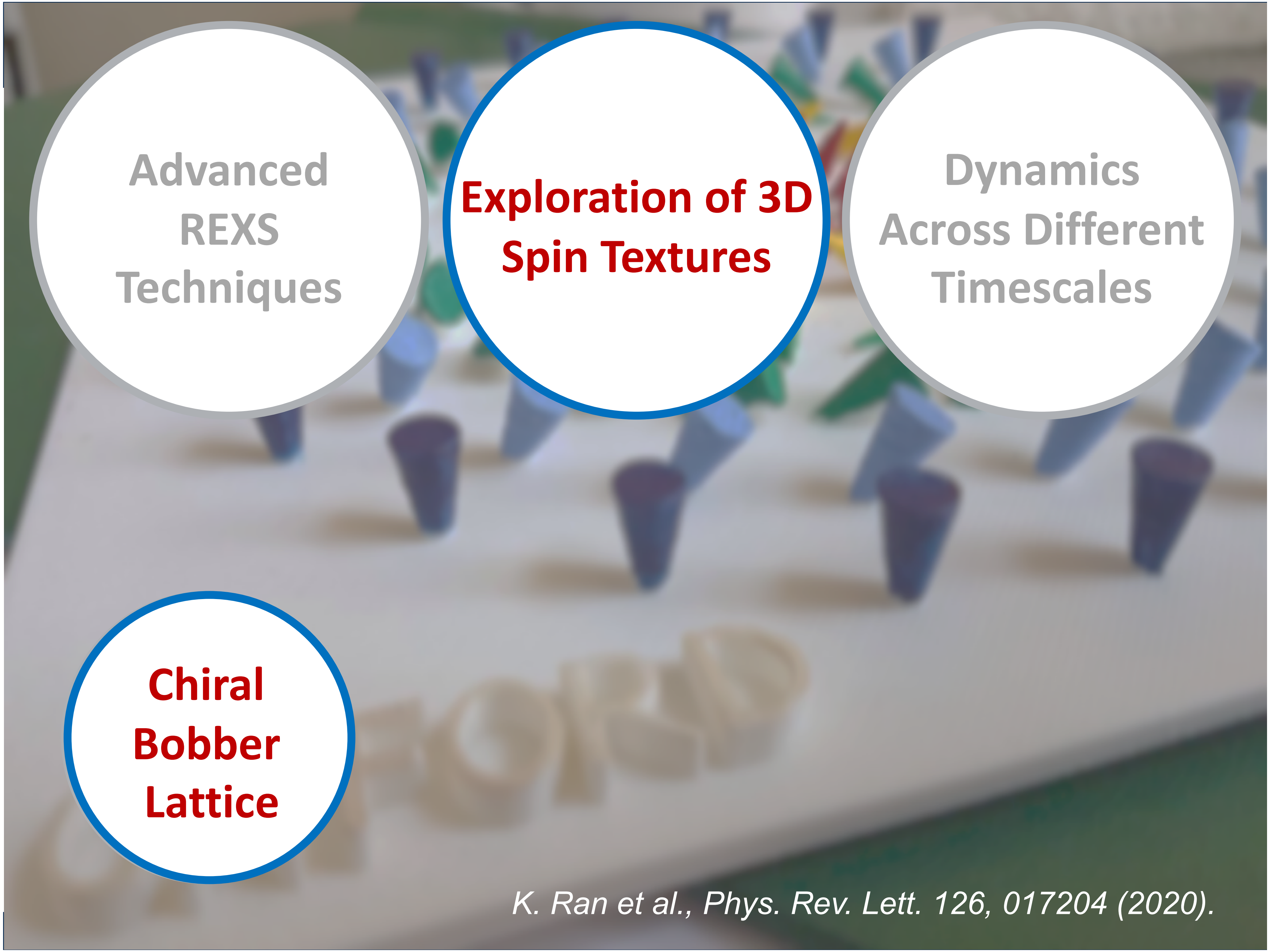
@ Cu edge





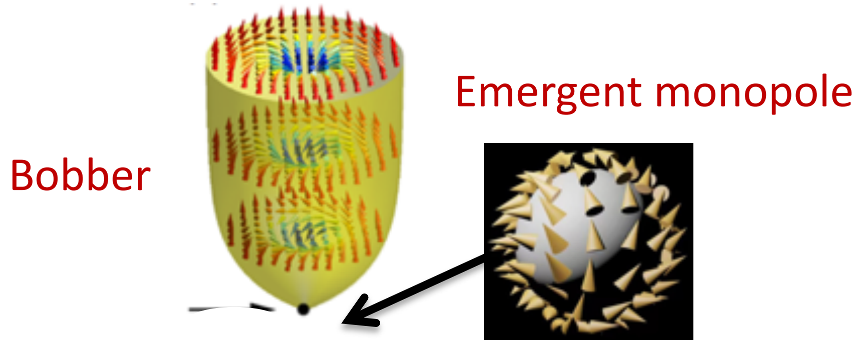
Bloch

Bloch


Advanced REXS Techniques
Exploration of 3D Spin Textures
Dynamics Across Different Timescales
Chiral Bobber Lattice
K. Ran et al., Phys. Rev. Lett. 126, 017204 (2020).

Chiral Bobbers – *An Elusive Species*

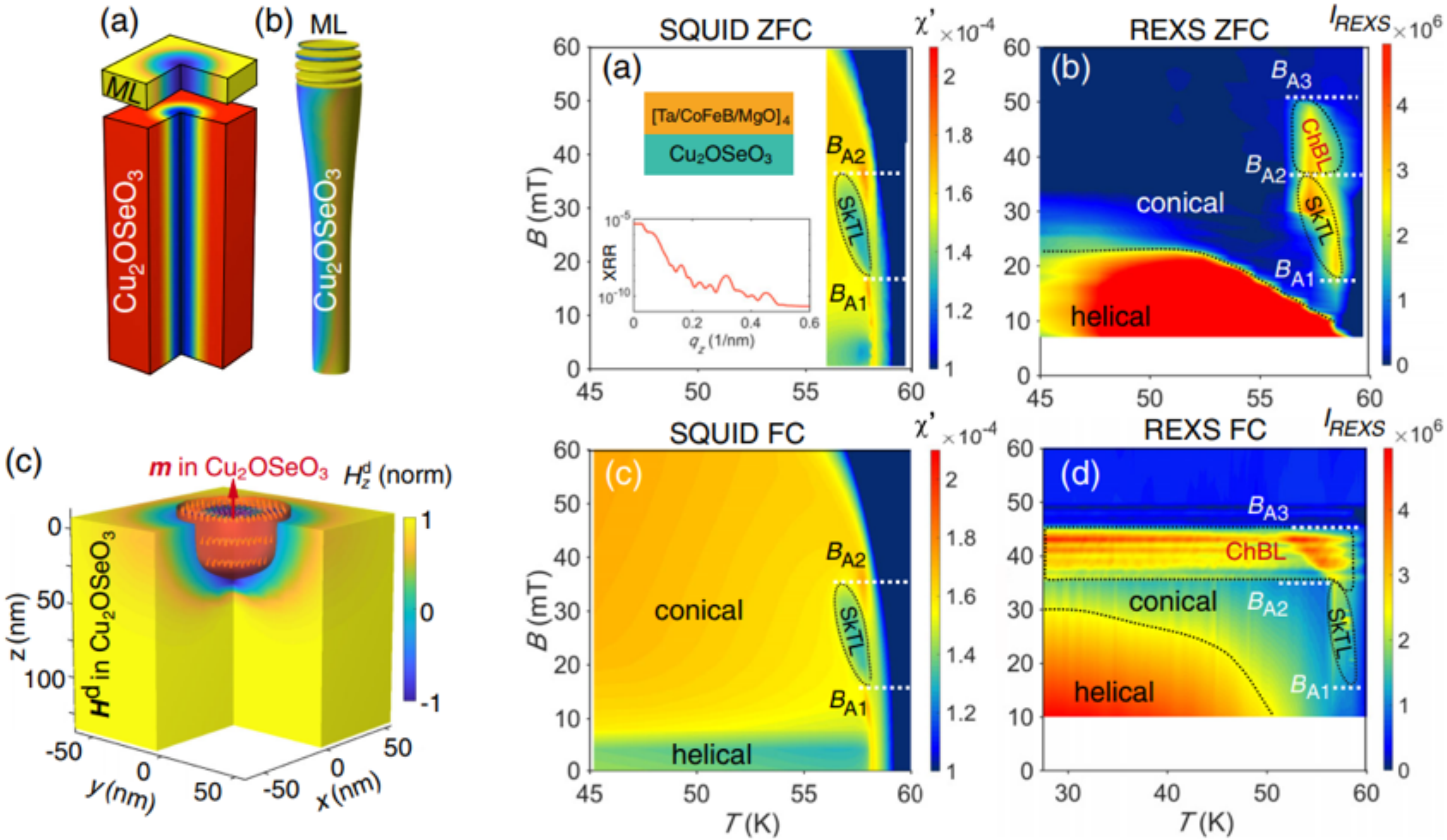
- Magnetic monopole science
- Exotic dynamic properties
- Exotic transport properties



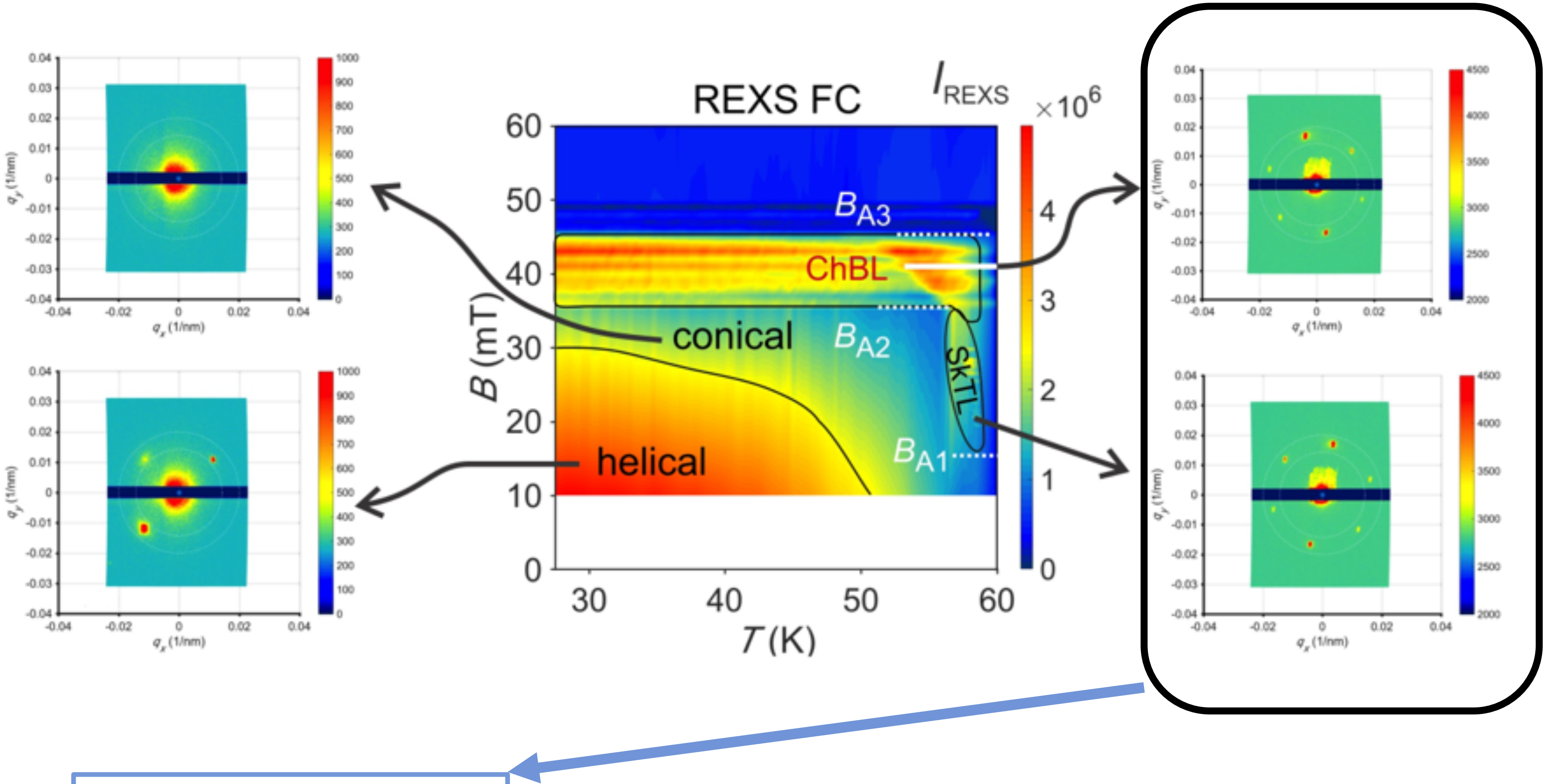
Metastable bobbers produced by either field-cooling or field-tilting in LTEM

F. Zheng et al., Nature Nanotech. 13, 451–455 (2018)

Magnetic Phase Diagrams – *Bulk vs Interface*



How to Identify Chiral Bobbers and Skyrmion Tubes?



What is needed:

Reconstruction of the detailed near-surface structure

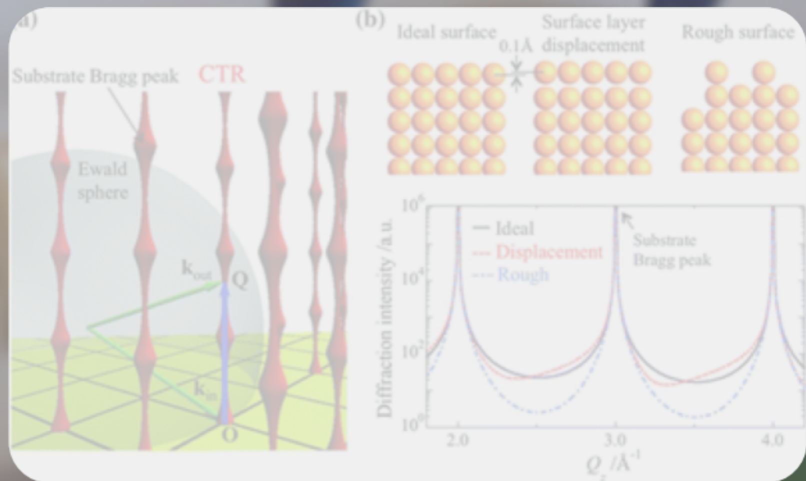
Advanced REXS Techniques

Exploration of 3D Spin Textures

Dynamics Across Different Timescales

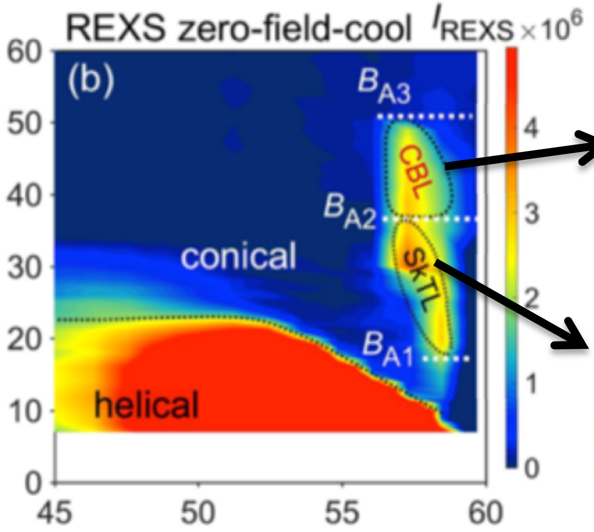
Folding Skyrmion Strings

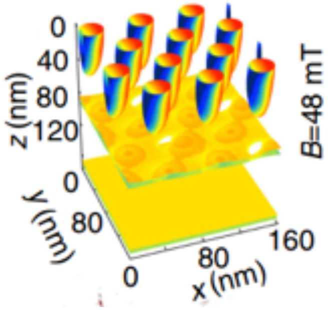
making use of magnetic truncation rods ...

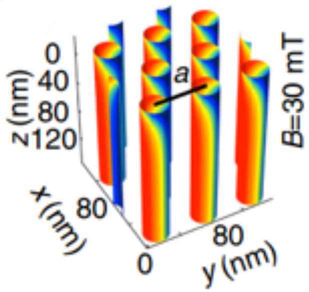


H. Jin et al., Nano Lett. 23, 5164 (2023).

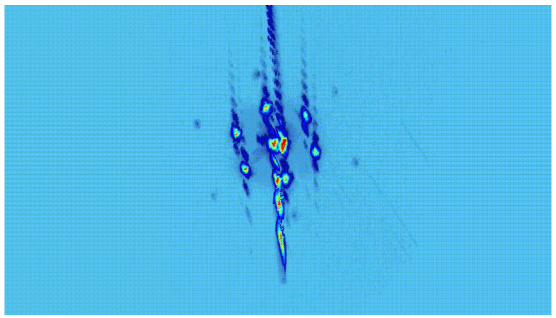
Magnetic Crystal Truncation Rods



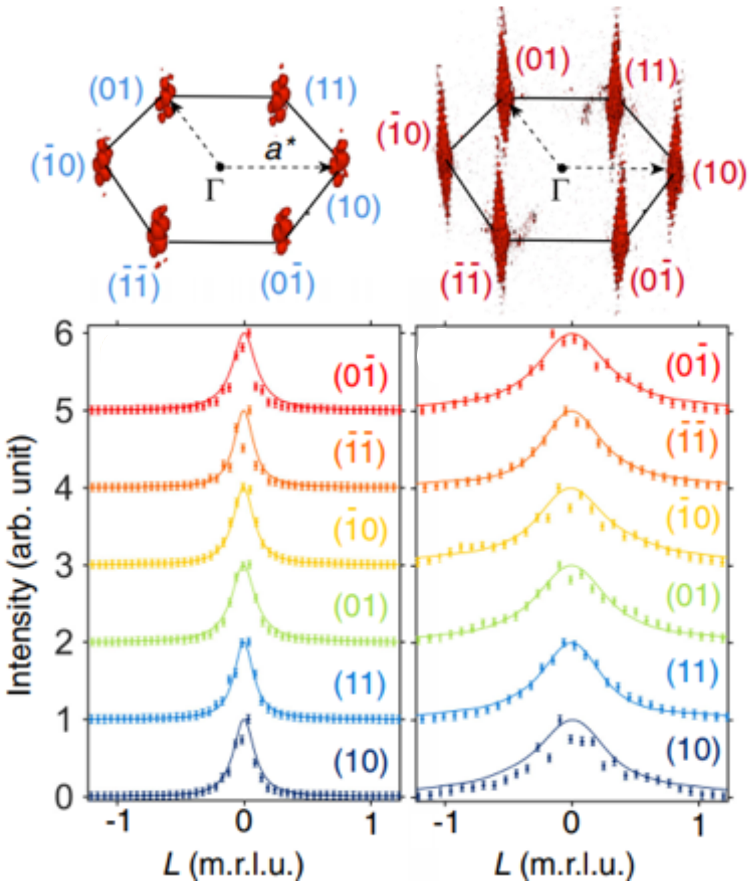




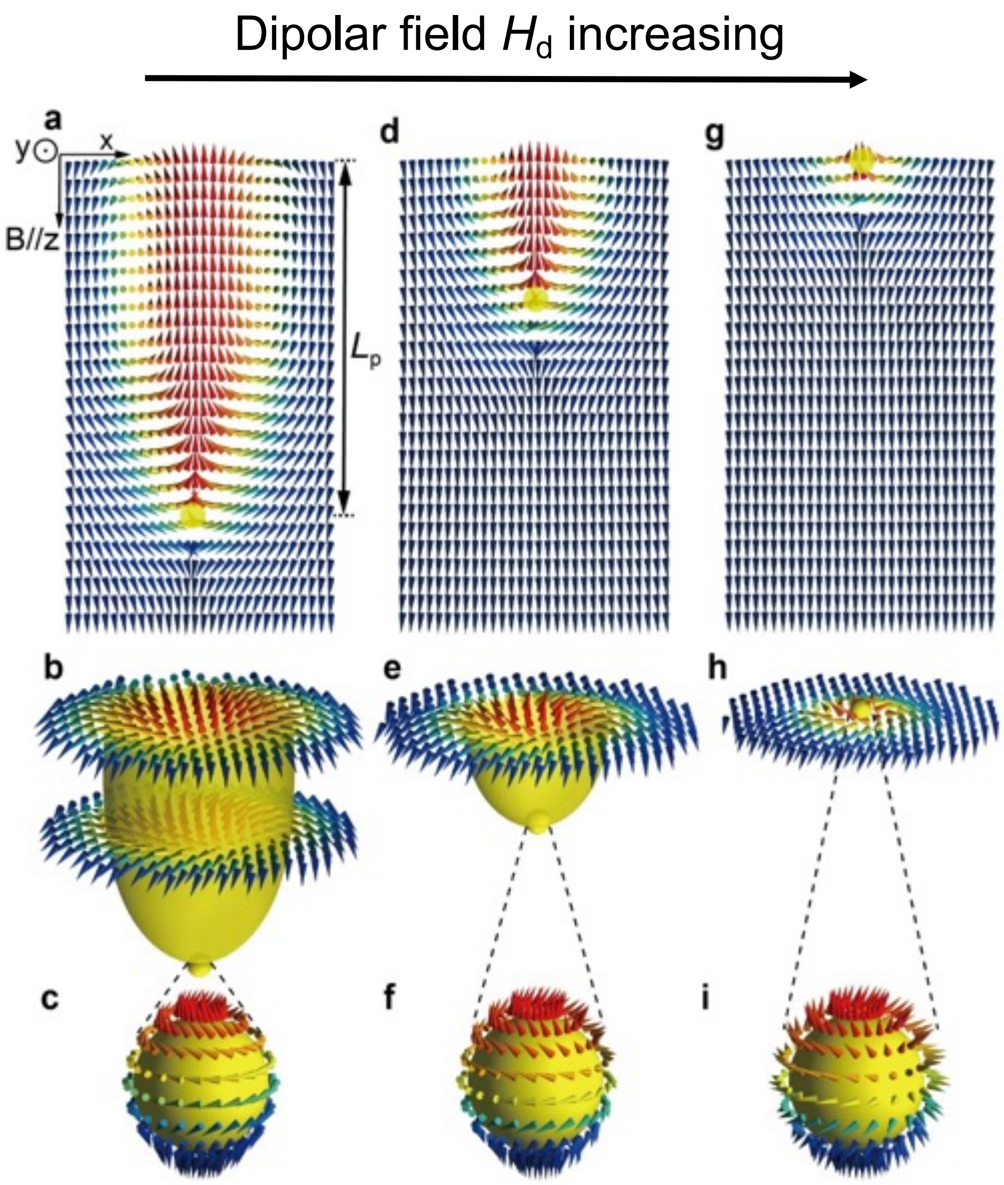
- ✓ Bobber depth L_P
- ✓ Shape d_{sk}

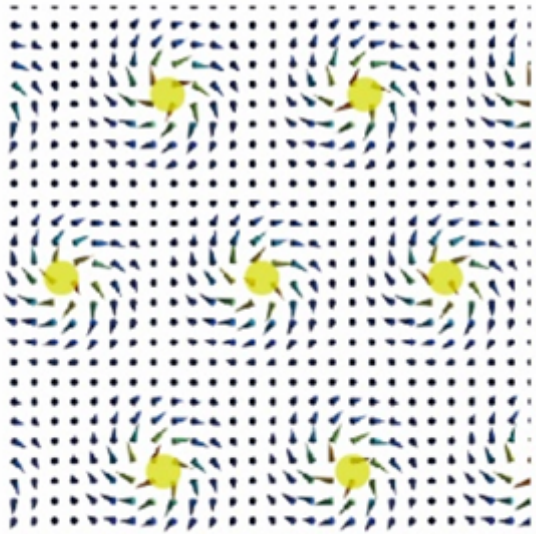


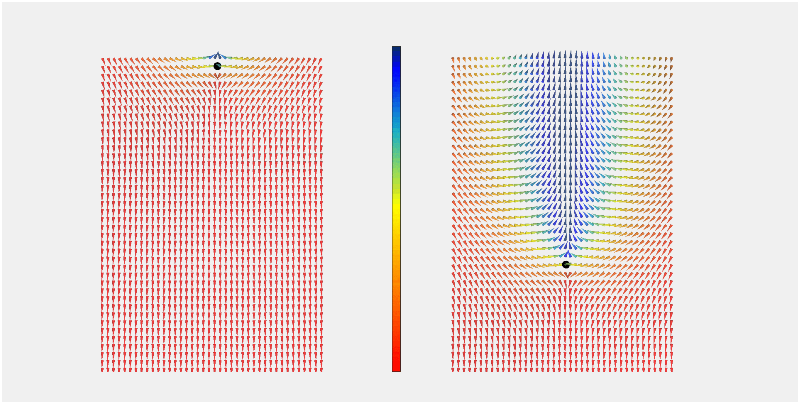
Skyrmion tube **Chiral Bobber**



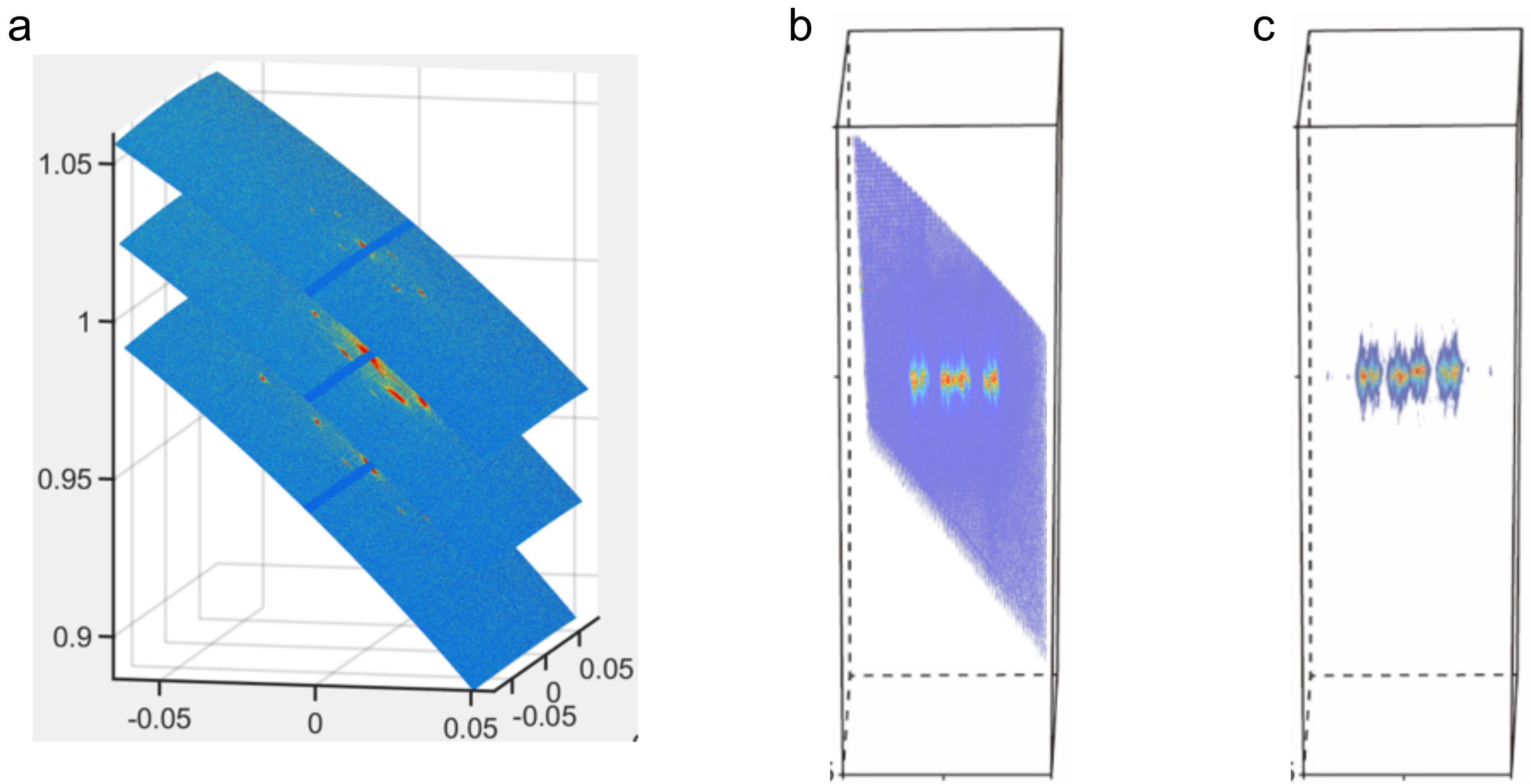
Chiral Bobbers Simulations – Tuning of the Coupling



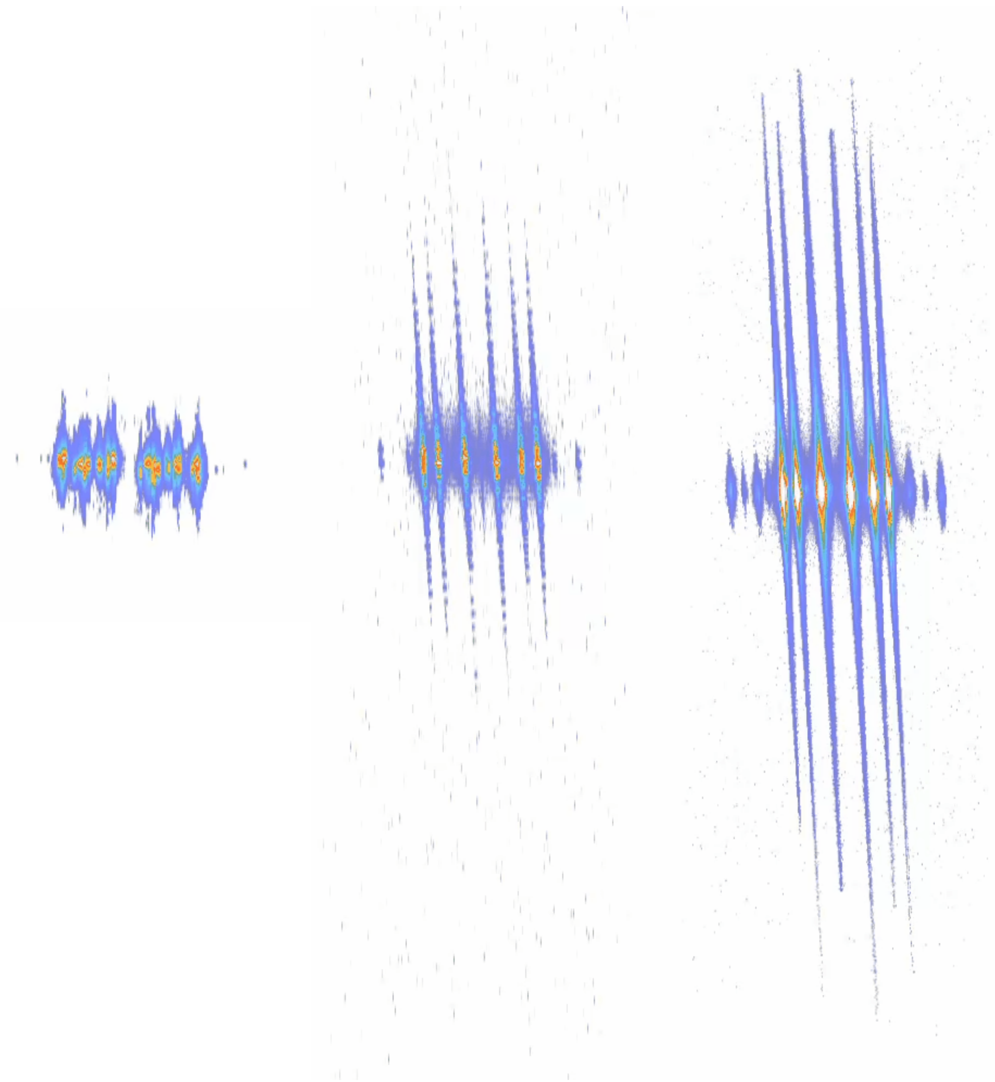


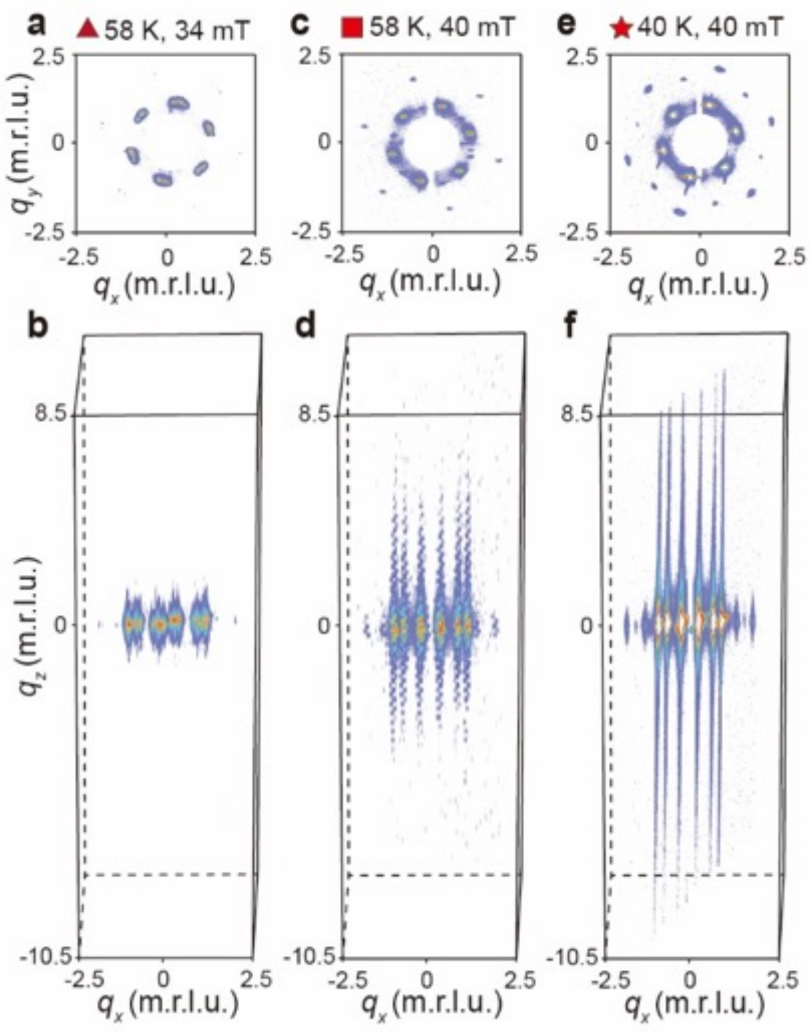


Reciprocal Space Mapping



RSM for the Different Magnetic Phases

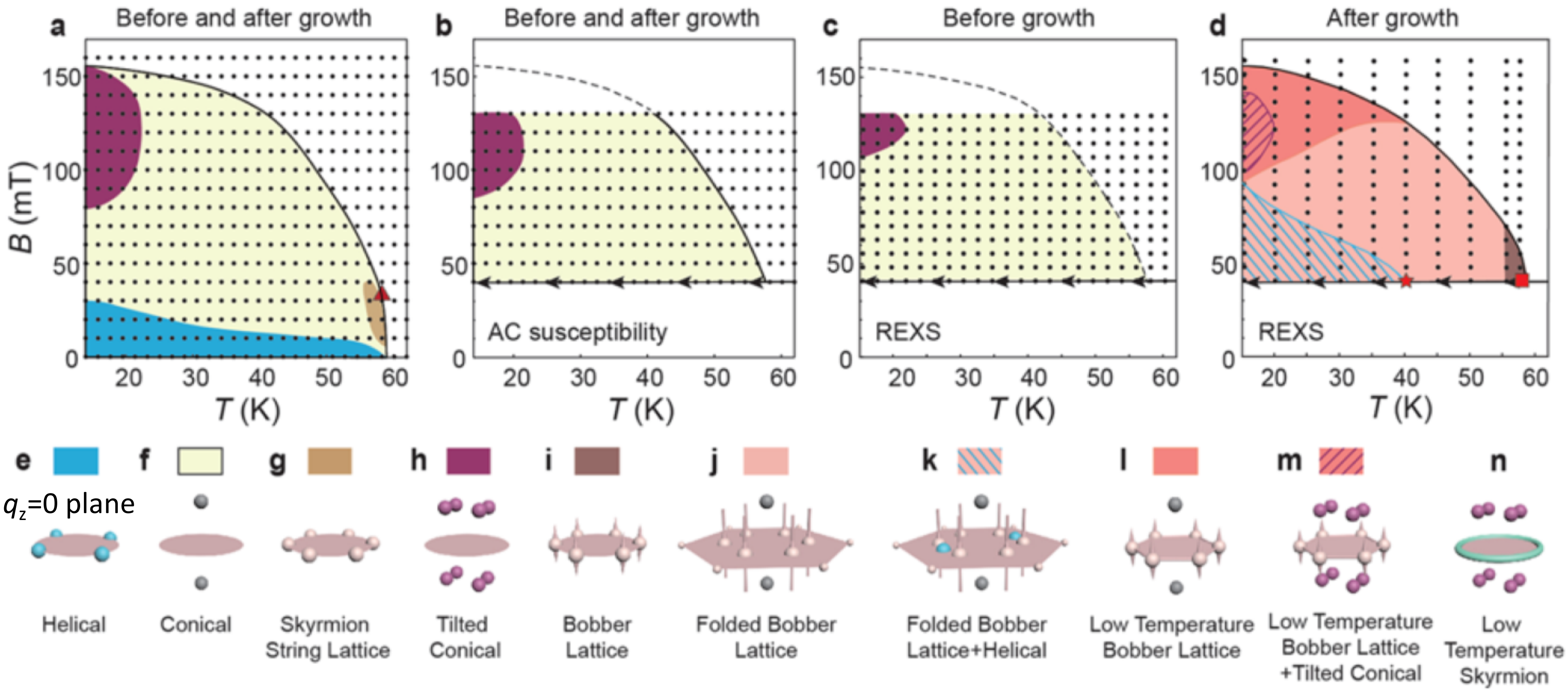




Phase Diagrams & Characteristic Magnetic Phases

AC susceptibility | AC susceptibility | REXS – CSO | REXS – CSO+ML

standard | *field-cool-field-sweep up with 40 mT cooling field*



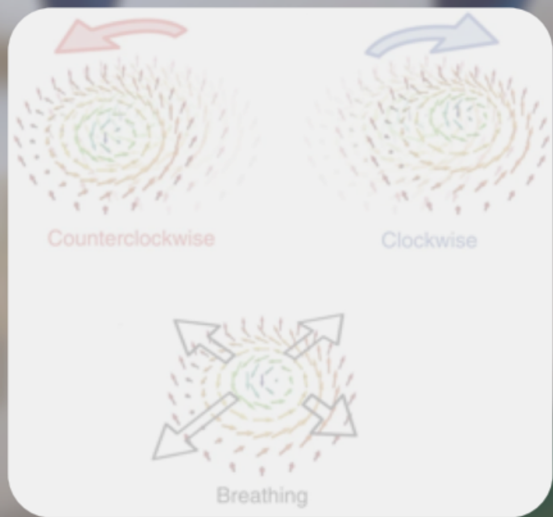
ML-induced phases

Advanced REXS Techniques

Exploration of 3D Spin Textures

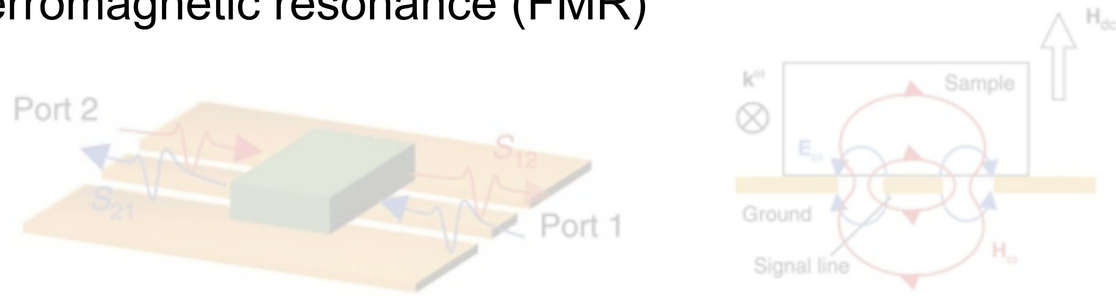
Dynamics Across Different Timescales

FAST **Internal Skyrmion Dynamics**

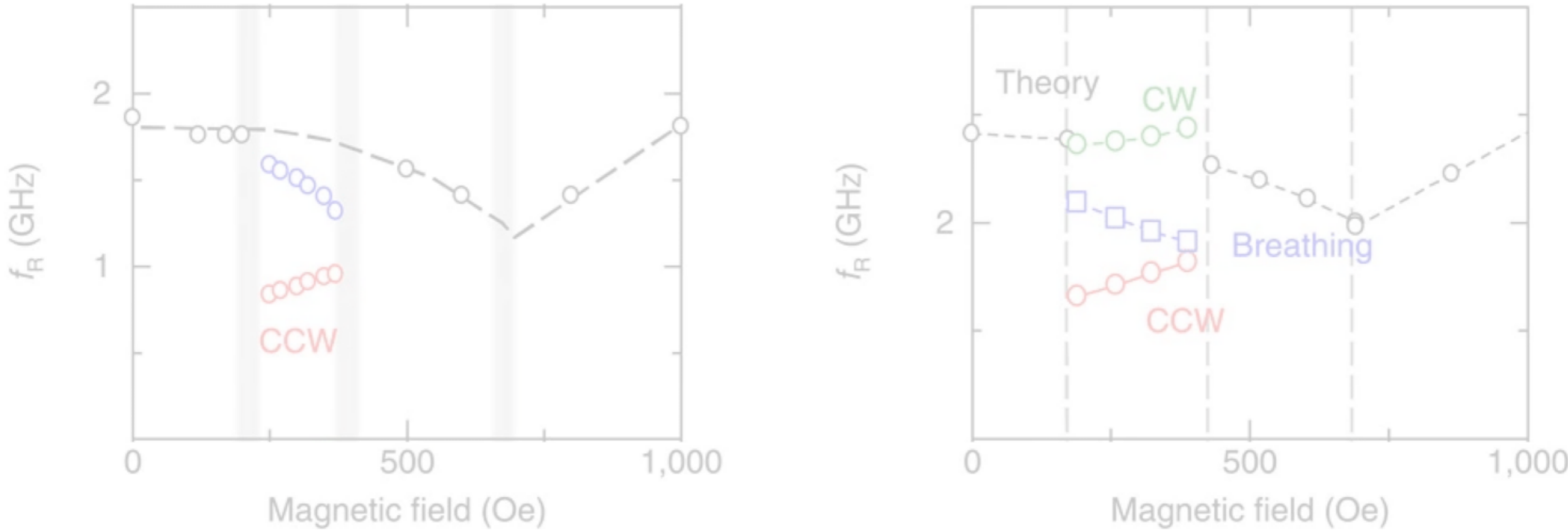


Resonant Dynamic Skyrmion Modes

Ferromagnetic resonance (FMR)



NO MODAL INFORMATION!



How Can Dynamic Modes be Measured and Identified?

Ferromagnetic resonance (FMR)

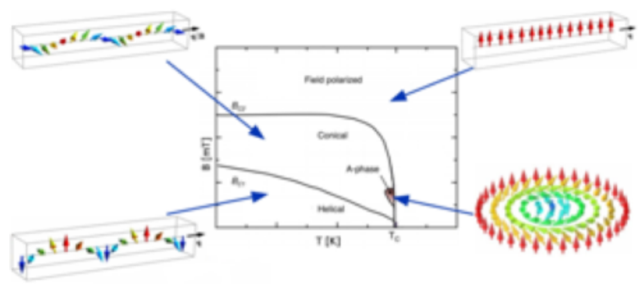
NO MODAL INFORMATION!

Q: How to perform mode-specific FMR in complex magnetic structures?

REXS → access to specific magnetic order

XFMR → access to element-specific dynamics

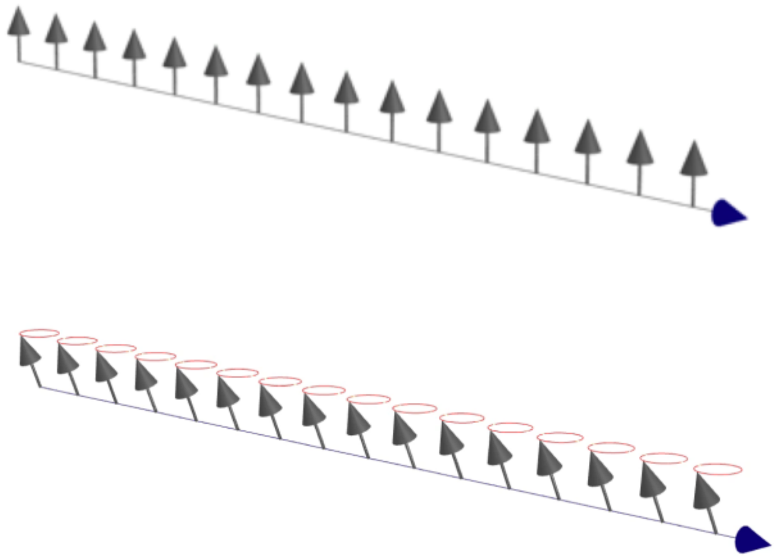
Complex dynamics!



→ Start with helical/conical phase

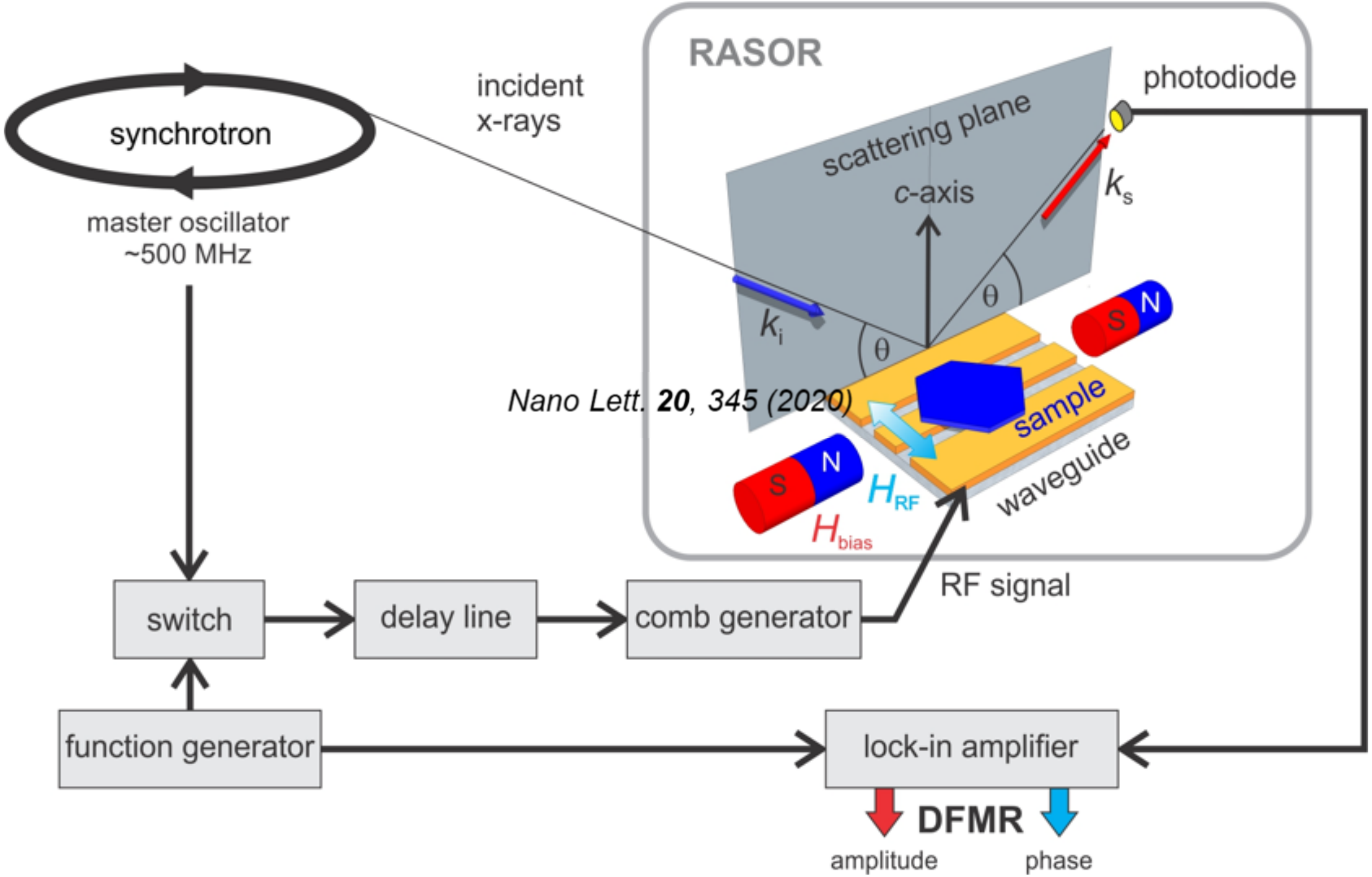
Chiral Magnetization Dynamics

Field-polarized state

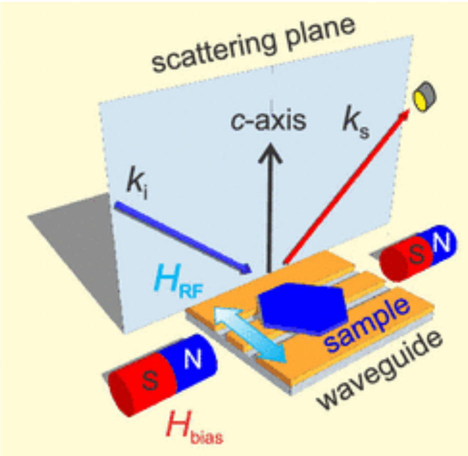


Helical state

Mode-Resolved Dynamic Probing – Diffractive FMR



How Can Dynamic Modes be Measured and Identified?



REXS + (X)FMR

AUTHORCHOICE

NANO LETTERS

Letter

Cite This: *Nano Lett.* 2020, 20, 345−352

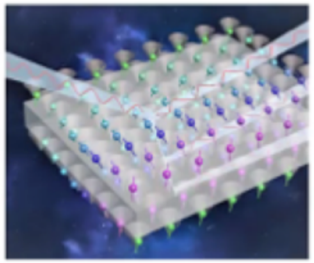
pubs.acs.org/NanoLett

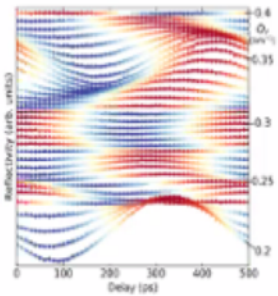
Mode-Resolved Detection of Magnetization Dynamics Using X-ray Diffractive Ferromagnetic Resonance

David M. Burn,[*,†] Shilei Zhang,[*,‡,#] Kun Zhai,[⊥,∇] Yisheng Chai,[§] Young Sun,[⊥,∇] Gerrit van der Laan,[*,†] and Thorsten Hesjedal[*,∥]

Reflectometry + (X)FMR

Phys. Rev. Lett. 125, 137201 (2020)





PHYSICAL REVIEW LETTERS **125**, 137201 (2020)

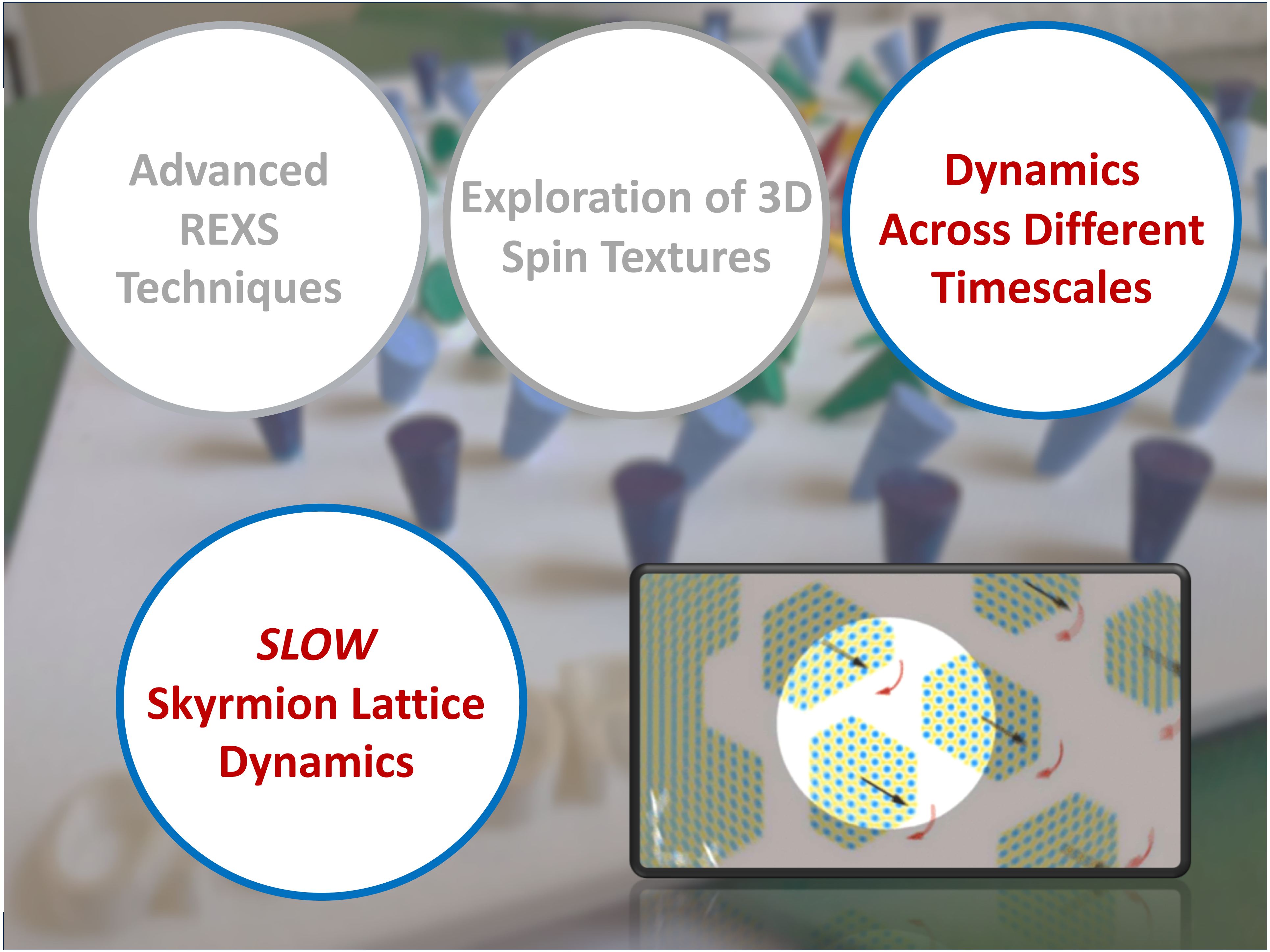
Editors' Suggestion | Featured in Physics

Depth-Resolved Magnetization Dynamics Revealed by X-Ray Reflectometry Ferromagnetic Resonance

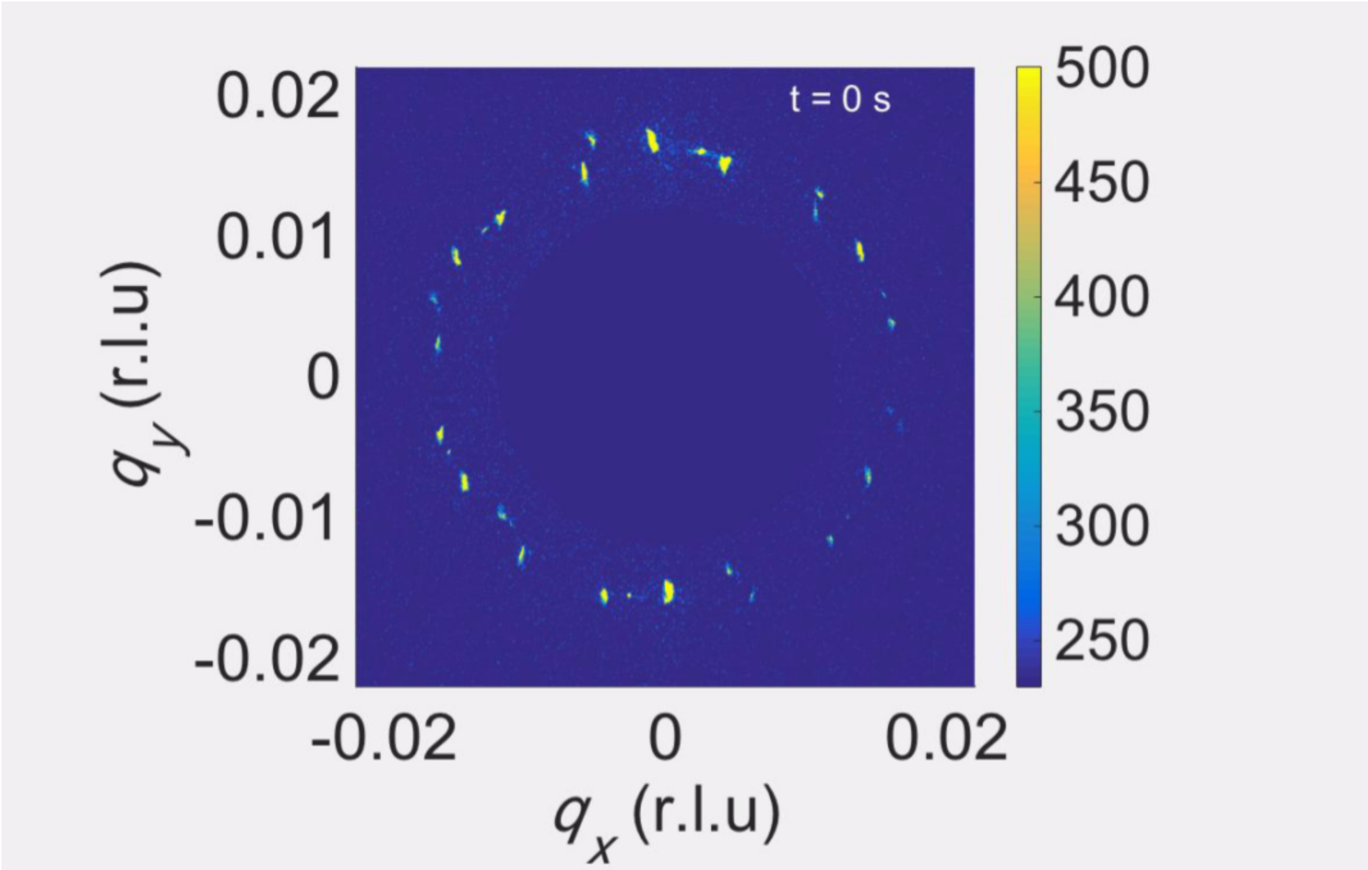
D. M. Burn,[1,*] S. L. Zhang,[2,3,†] G. Q. Yu,[4] Y. Guang,[4] H. J. Chen,[5] X. P. Qiu,[5] G. van der Laan,[1,‡] and T. Hesjedal[6]

[1]*Diamond Light Source, Harwell Science and Innovation Campus, Didcot, Oxfordshire OX11 0DE, United Kingdom*
[2]*School of Physical Science and Technology, ShanghaiTech University, Shanghai 200031, China*
[3]*ShanghaiTech Laboratory for Topological Physics, ShanghaiTech University, Shanghai 200031, China*
[4]*Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*
[5]*Shanghai Key Laboratory of Special Artificial Microstructure Materials and School of Physics Science and Engineering, Tongji University, Shanghai 200092, China*
[6]*Department of Physics, Clarendon Laboratory, University of Oxford, Oxford OX1 3PU, United Kingdom*

REVIEW: G. van der Laan and T. Hesjedal, Nucl. Instrum Methods Phys. Res. B **540**, 85 (2023).


Advanced REXS Techniques
Exploration of 3D Spin Textures
Dynamics Across Different Timescales
SLOW Skyrmion Lattice Dynamics

Rotation of the Skyrmion Lattice in a Field Gradient



Observations

- Bright spots → slow or even fixed
- Dimmer (small) spots → fast rotating
- Joining and separation of spots


nature COMMUNICATIONS

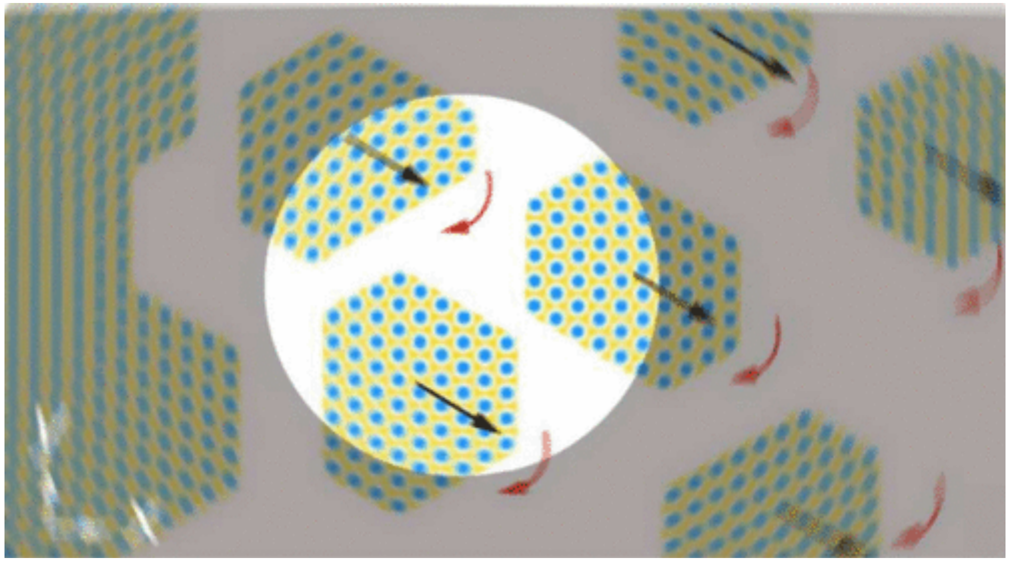
ARTICLE

OPEN

Manipulation of skyrmion motion by magnetic field gradients

S.L. Zhang[1], W.W. Wang[2], D.M. Burn[3], H. Peng[1], H. Berger[4], A. Bauer[5], C. Pfleiderer[5], G. van der Laan[3] & T. Hesjedal[1]

Rolling Motion of Rigid Skyrmion Crystallites



NANO LETTERS

Open Access

pubs.acs.org/NanoLett

Letter

Rolling Motion of Rigid Skyrmion Crystallites Induced by Chiral Lattice Torque

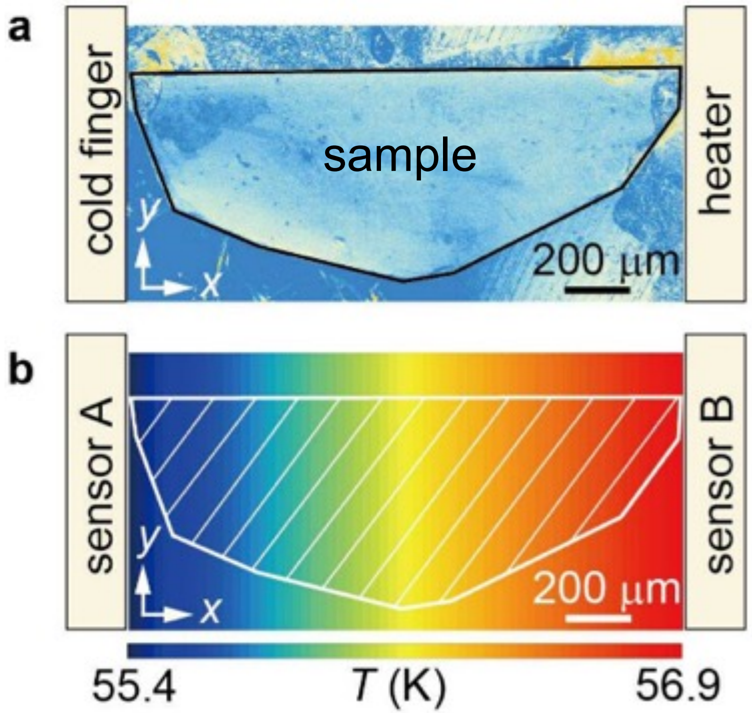
Haonan Jin, Jingyi Chen, Gerrit van der Laan, Thorsten Hesjedal, Yizhou Liu,* and Shilei Zhang*

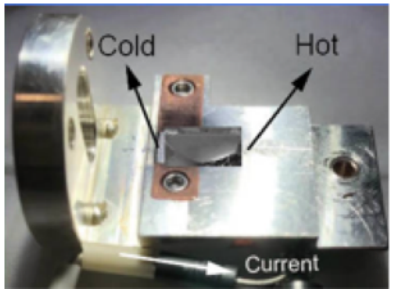
Cite This: *Nano Lett.* 2024, 24, 12226–12232

Read Online

Linear Temperature Gradient

SEM

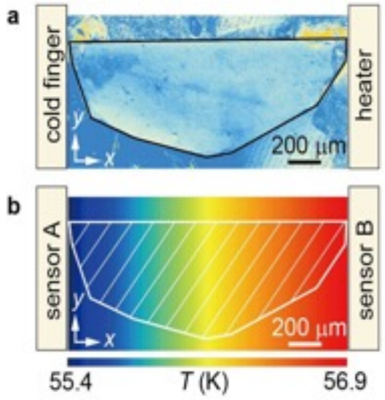


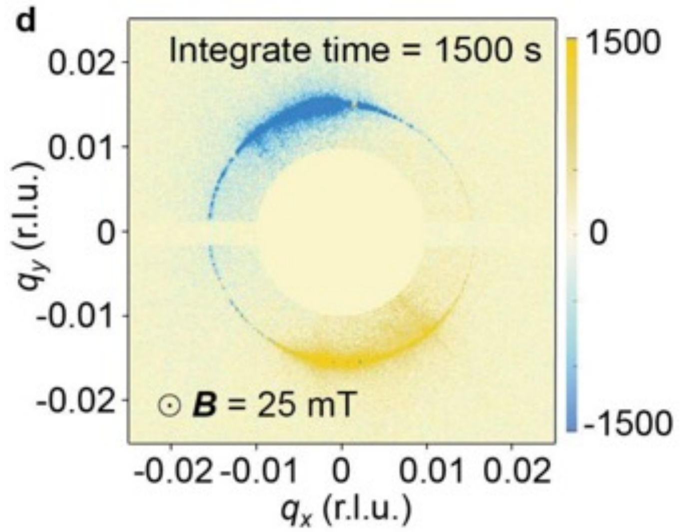


linear T gradient g_x = 0.94 K/mm

$\underline{T}$ = 56.15 K

Linear Temperature Gradient

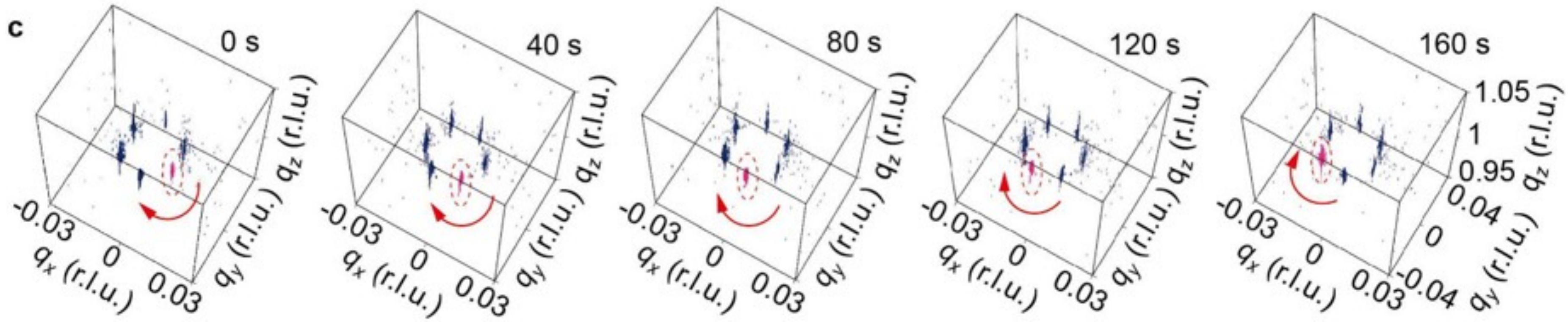




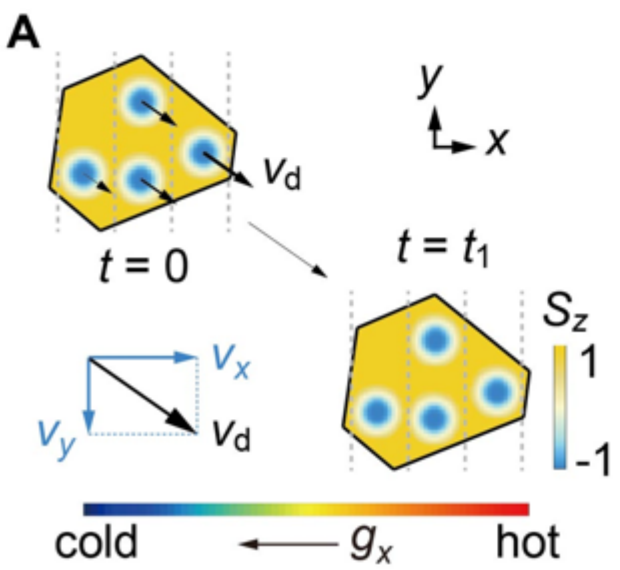
time-integrated measurement

→ *helicity angle*

3D-RSM patterns recorded in 40 s intervals (each measurement takes 2 ms)



Skyrmion Dynamics: Individual Skyrmions vs The Lattice



Ensemble of isolated skyrmions

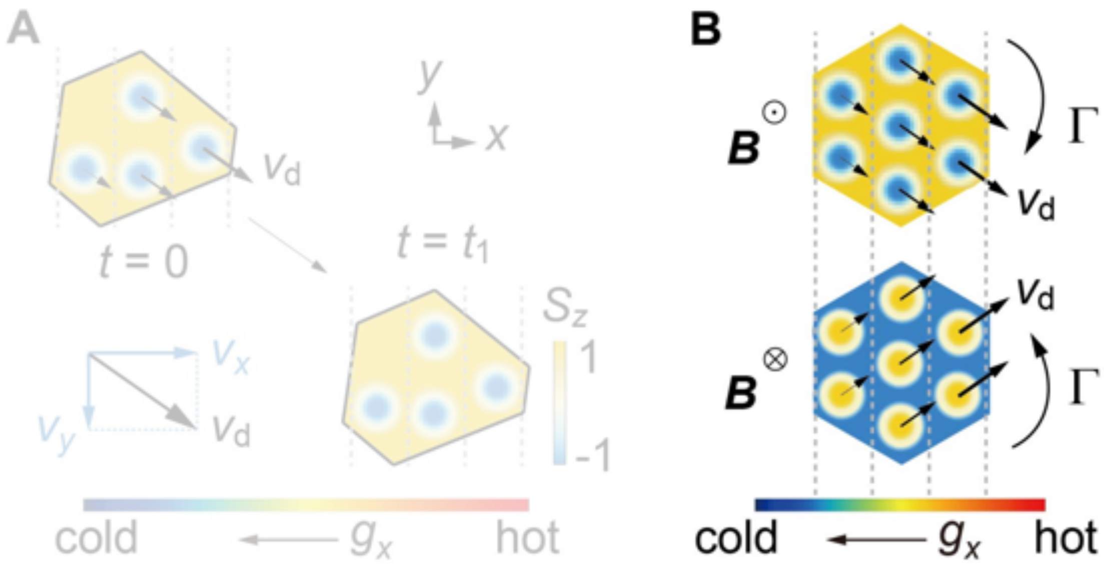
→ *translational motion*

$$\vec{v}_{drift} = \vec{v}_x + \vec{v}_y$$

|| gradient

Sk Hall effect (⊥ gradient)

Skyrmion Dynamics: Individual Skyrmions vs The Lattice



Ordered skyrmion lattice

→ *translational motion*

... with varying magnitude along x

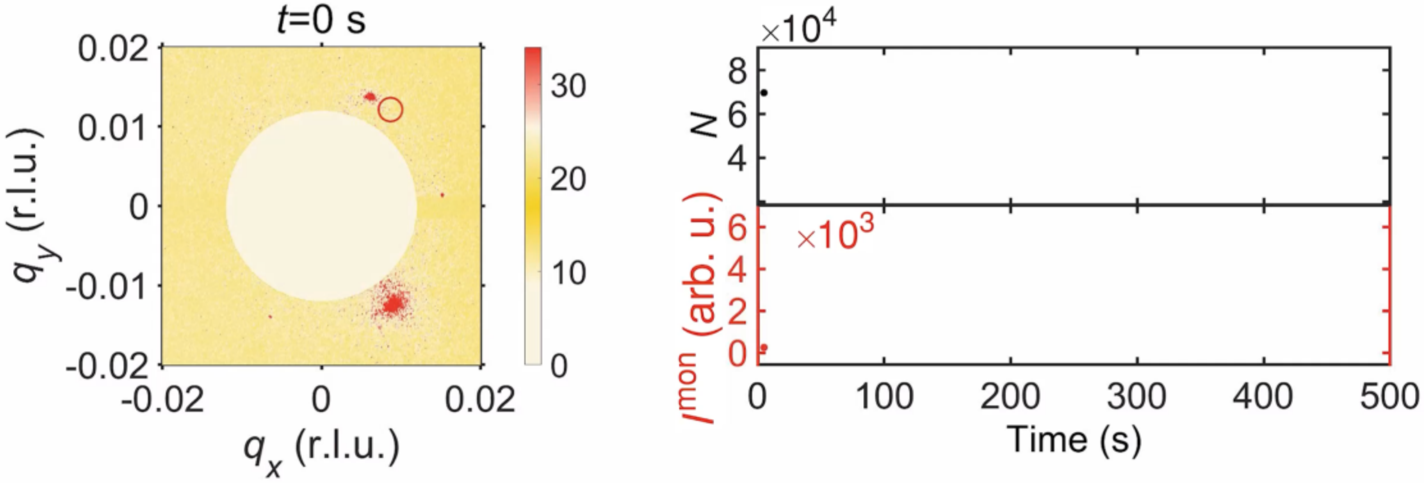
→ *rotation*

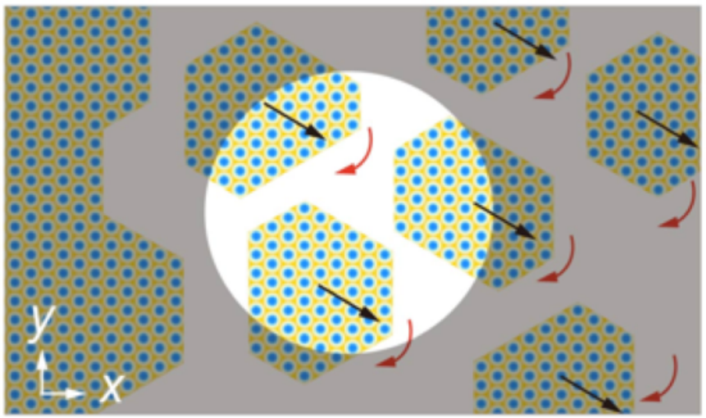
Isolated skyrmions

→ *translational motion*

- individual skyrmions experience different drift velocities (depending on their local g_x)
- resulting net torque drives the entire domain with a ***fixed chirality*** and ***uniform drift velocity***
- torque reverses sign when field is reversed

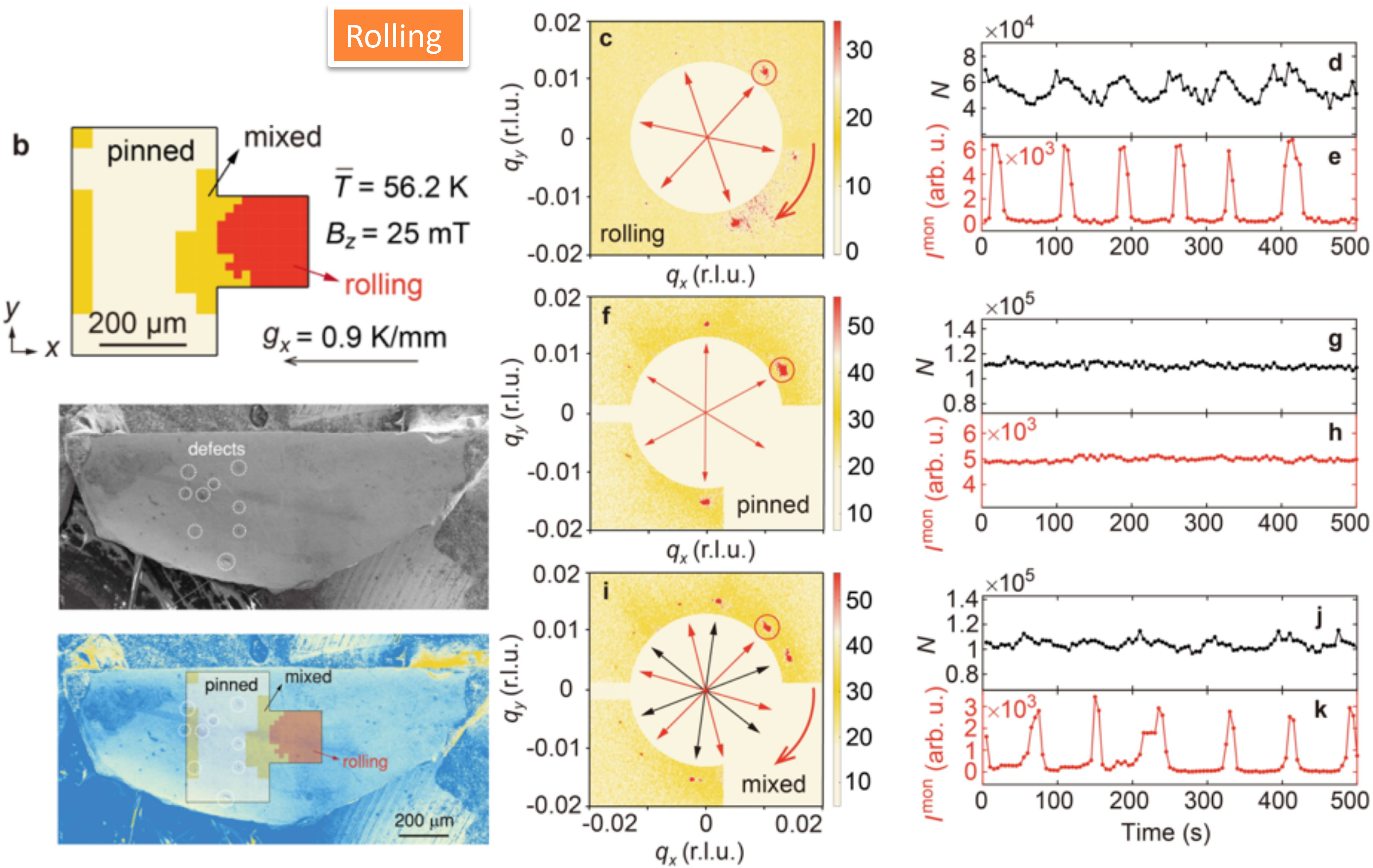
Spatially and Time-Resolved SkC Rolling Dynamics



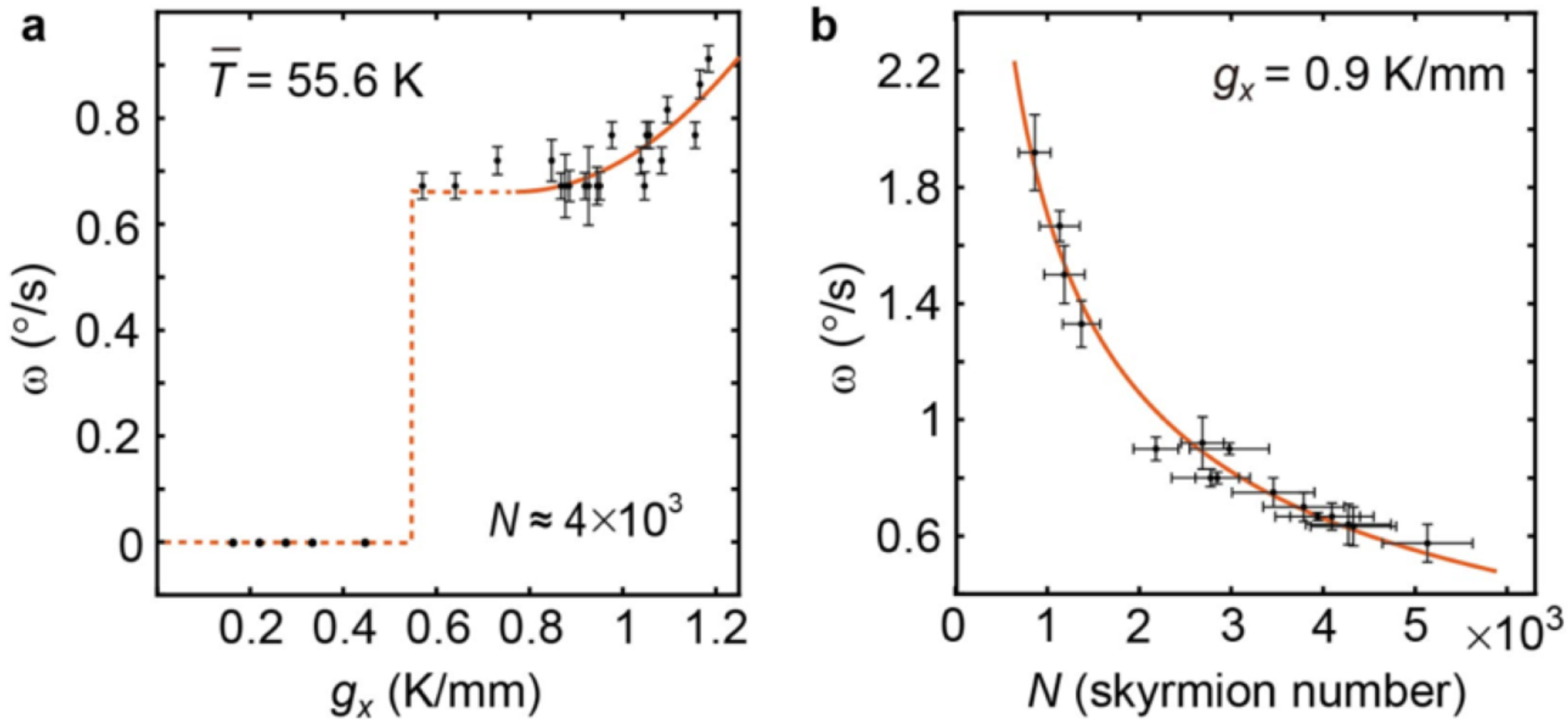


- X-ray probing area → local observation window for the dynamics
- ordered SkC patches consecutively enter and leave the monitored area, following a rolling motion.

Spatially and Time-Resolved Skyrmion Dynamics



Quantitative Chiral Lattice Torque Model Analysis

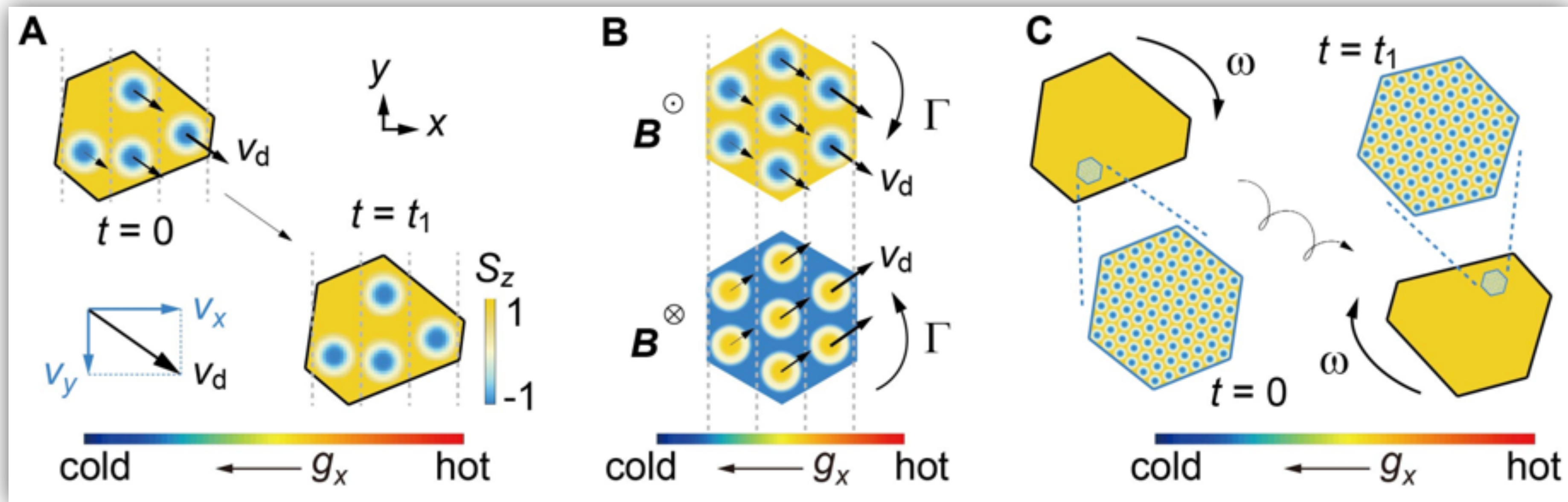


- SkC rotation angular velocity as a function of |g| (at $\underline{T}$ = 55.6 K)

- Skyrmion number fixed at ~4000 +/- 450

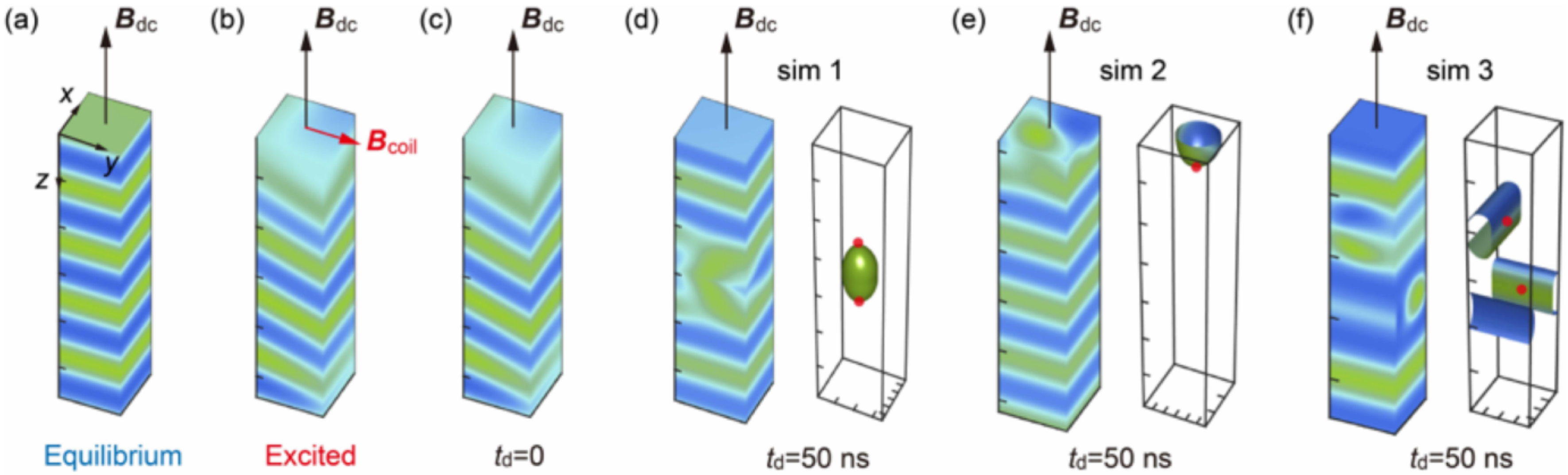
- Angular velocity as a function of skyrmion number for a fixed value of the gradient of 0.9 K/mm.

Summary: Individual Skyrmions vs The Lattice



- Under a linear temperature gradient, skyrmion crystallites exhibit a continuous rolling motion.
- The rolling motion is induced by a chiral lattice torque
- Speed can be controlled by the magnitude of the temperature gradient.
- The rotational sense can be controlled via the magnetic field.

Slow Equilibrium Relaxation Mediated by Topological Defects



PHYSICAL REVIEW LETTERS **VOL..XX,** 000000 (XXXX)

Slow Equilibrium Relaxation in a Chiral Magnet Mediated by Topological Defects

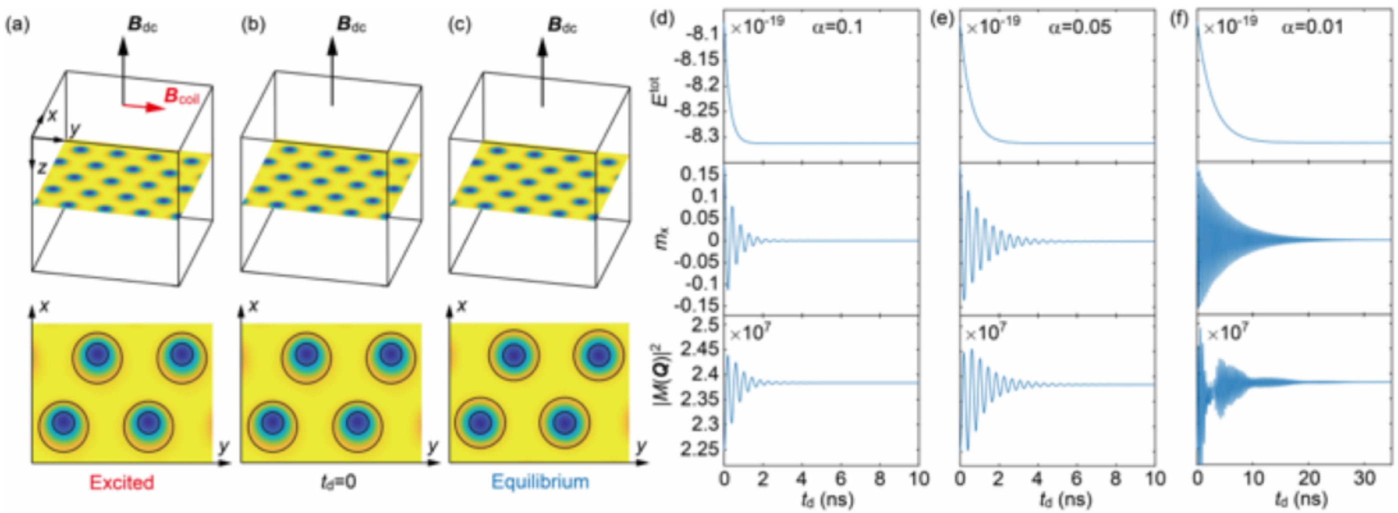
Chenhao Zhang,[1,*] Yang Wu,[1,*] Jingyi Chen,[1,*] Haonan Jin,[1,2] Jinghui Wang,[1,2] Raymond Fan,[3] Paul Steadman,[3] Gerrit van der Laan,[3] Thorsten Hesjedal,[4] and Shilei Zhang[1,2,5,†]

[1]*School of Physical Science and Technology, ShanghaiTech University, Shanghai 201210, China*
[2]*ShanghaiTech Laboratory for Topological Physics, ShanghaiTech University, Shanghai 201210, China*
[3]*Diamond Light Source, Harwell Science and Innovation Campus, Didcot OX11 0DE, United Kingdom*
[4]*Department of Physics, Clarendon Laboratory, University of Oxford, Oxford OX1 3PU, United Kingdom*
[5]*Center for Transformative Science, ShanghaiTech University, Shanghai 201210, China*

(Received 16 April 2024; revised 15 July 2024; accepted 18 September 2024)

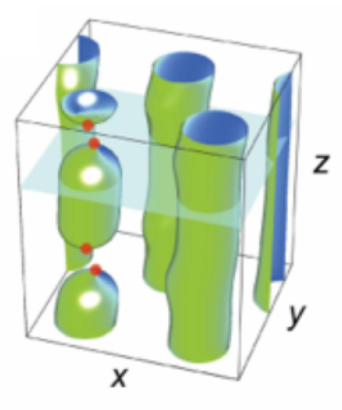
Fast vs Slow Skyrmion Dynamics

In general, dynamical processes in non-collinear magnetic systems should be fast (~ns).

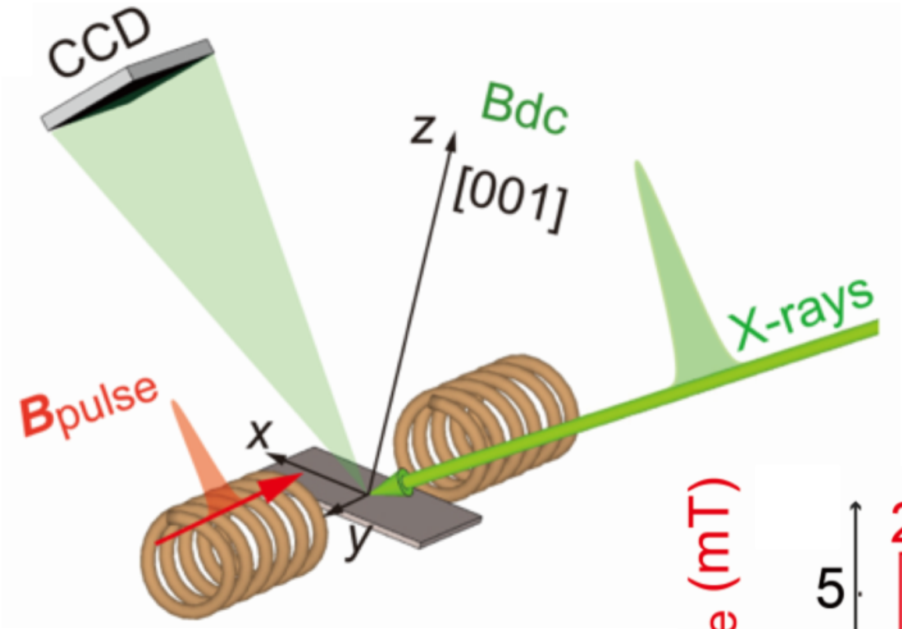


In chiral magnets, topological defects are commonly observed …

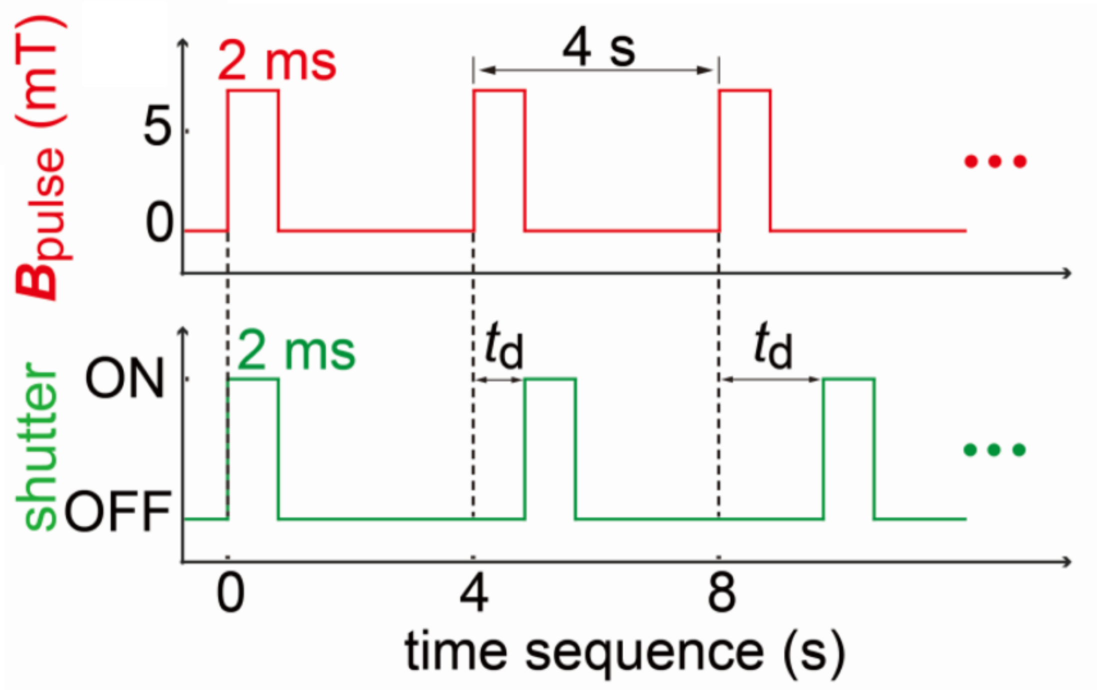
what is their role in the relaxation process?



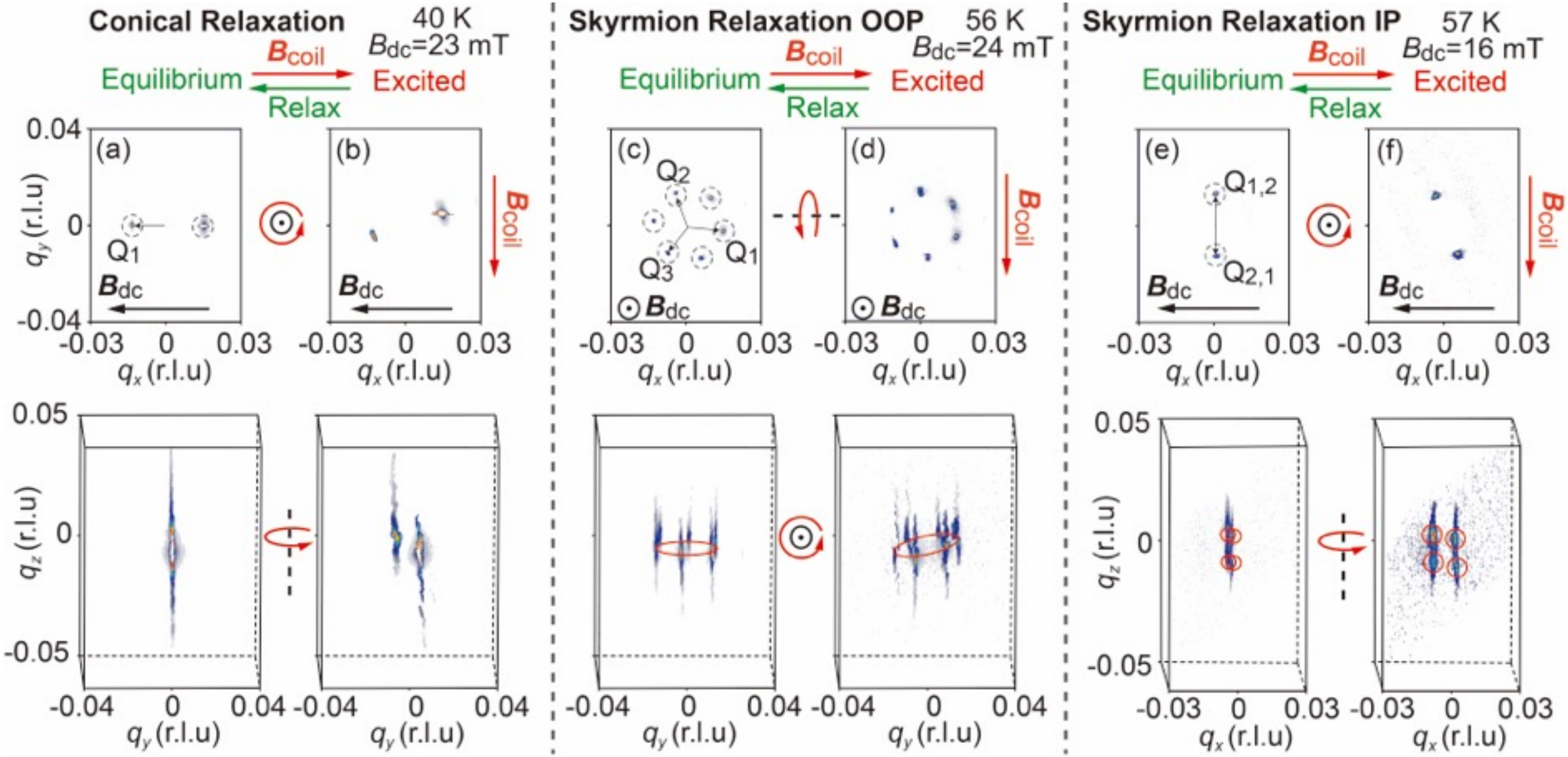
Experimental Setup for Studying Slow Dynamics



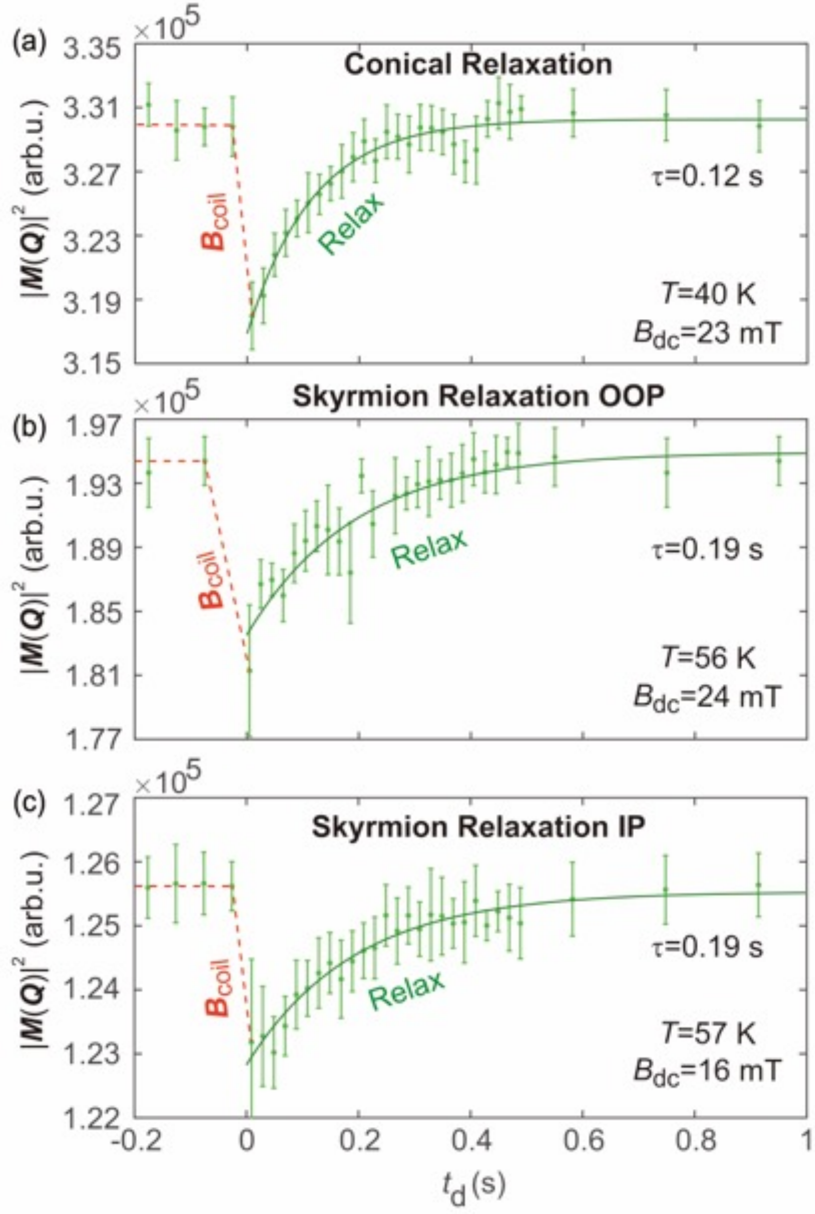
7 mT (peak) along *y*-axis



Static States in a Constant Tilted Field



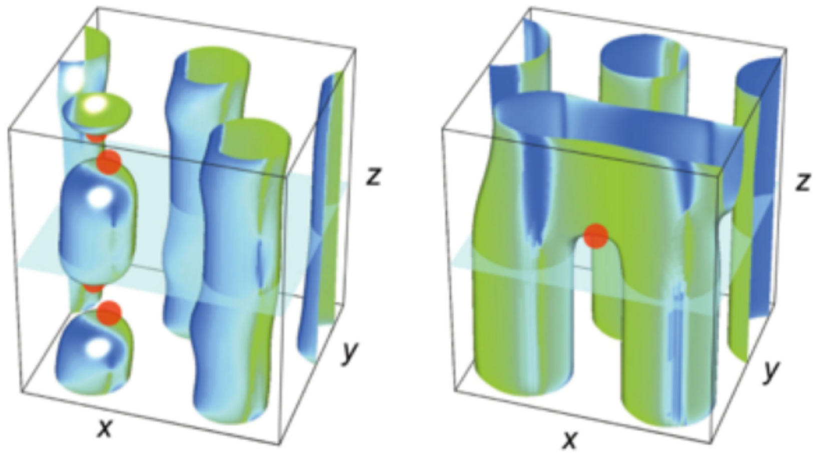
Slow Relaxation of the Magnetic Phases



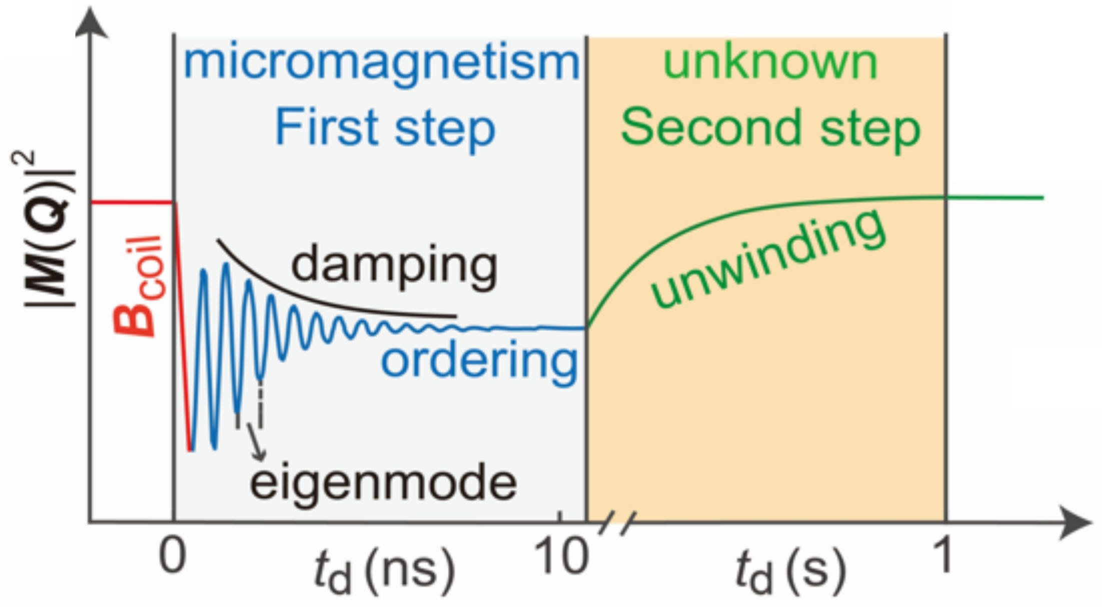
- relaxation time of ~s
- generally similar behavior of the magnetic phase

  → *similar mechanism*

Micromagnetic simulations of the formation of topological defects following the removal of the tilted field.



Summary – Slow Skyrmion Dynamics



- **Long Relaxation**: Unlike typical fast magnetic responses (ns), the system requires ~0.2 s.
- **Universal Mechanism**: Both conical and skyrmion lattice (SkX) phases exhibit similar slow relaxation behaviors, suggesting a common mechanism.
- **Topological Defects Drive Slow Dynamics**: The observed slow relaxation is attributed to topological defects, which gradually dissipate over time.
- **Relevance for Spintronics**: Understanding these slow relaxation processes is crucial for developing robust spintronic devices.

Advanced REXS Techniques

Exploration of 3D Spin Textures

Dynamics Across Different Timescales