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Jonathan White :: Laboratory for Neutron Scattering and Imaging :: Paul Scherrer Institute

Application of scattering techniques for the study of skyrmionics

13th May, 2019



Joint French-Swedish school on X-rays and Neutrons techniques for the study of functional materials for energy

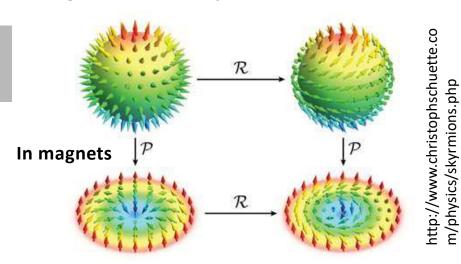
13-17 May 2019 Lund (Sweden)



- What are magnetic Skyrmions, and where do we find them?
 - Non-centrosymmetric crystals
 - Synthetic systems
- Application of scattering techniques
 - Neutron and resonant x-ray scattering
- Magnetic Skyrmions for Applications 'Skyrmionics'
 - Concepts, status and challenges
- Summary



In 'original' sense as a 'particle'



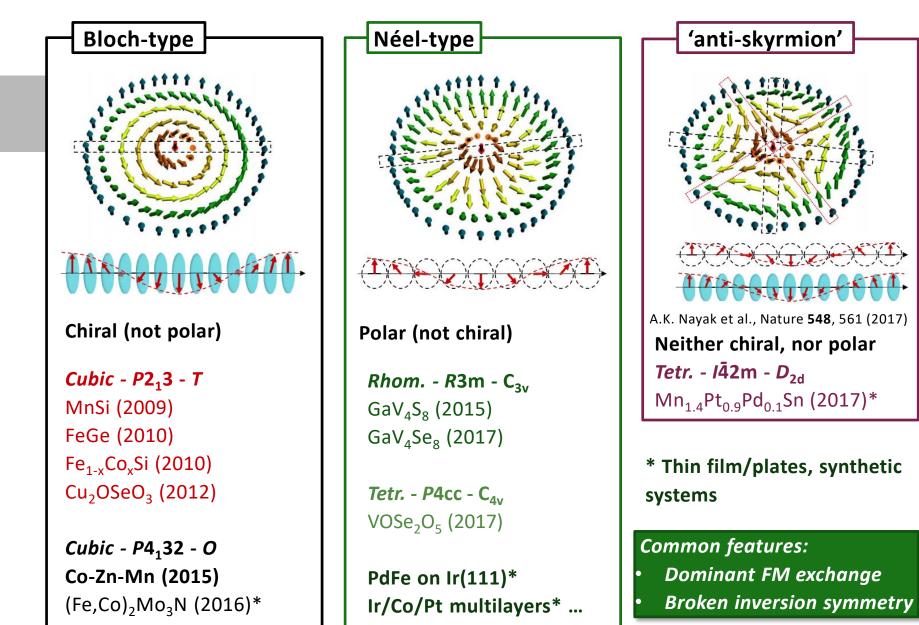
Intuitive picture for topological winding of skyrmion:

$$\int d^2 r \left(\frac{\partial \hat{n}}{\partial x} \times \frac{\partial \hat{n}}{\partial y} \right) \cdot \hat{n} = \pm 4\pi Q \quad |\mathbf{Q}| = 1$$

In magnets $\int \mu_0 H$ P. Milde *et al.*, Science **340**, 1076 (2013)

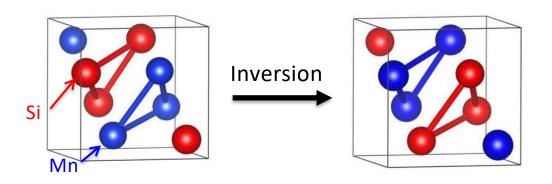
Topologically non-trivial (countable objects) Closed particle-like state (physical stability)

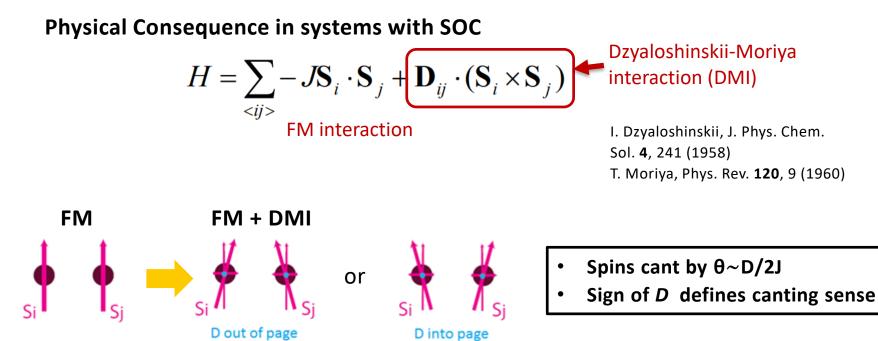






MnSi





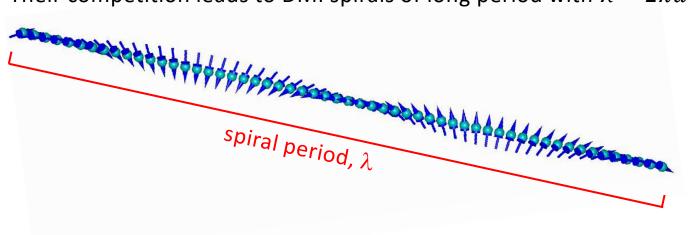


Long period spiral magnetism

$$H = \sum_{\langle ij \rangle} -J\mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$$

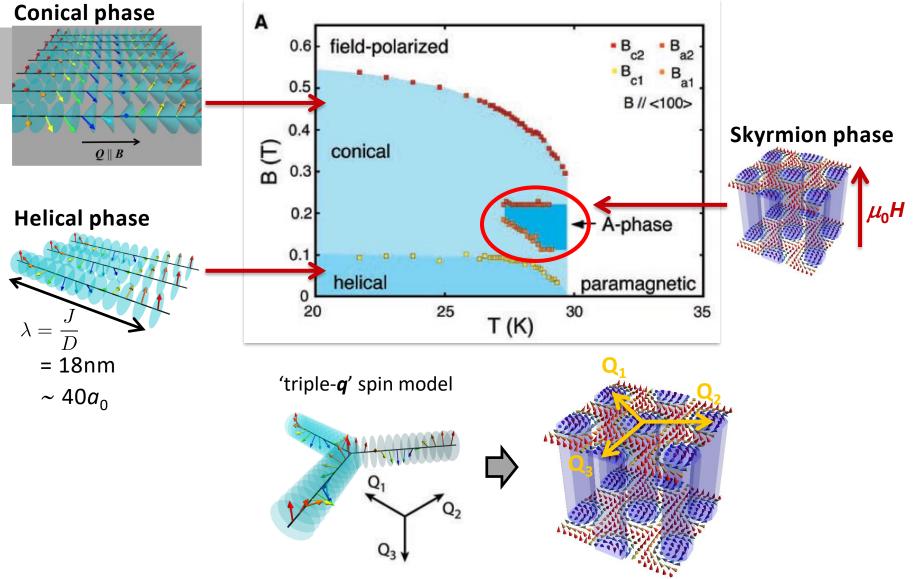
J favours parallel moments*D* favours perpendicular moments

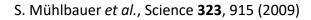
Their competition leads to DMI spirals of long period with $\lambda = 2\pi a \frac{J}{D}$



The DMI is a key ingredient of skyrmion formation

First observation in MnSi

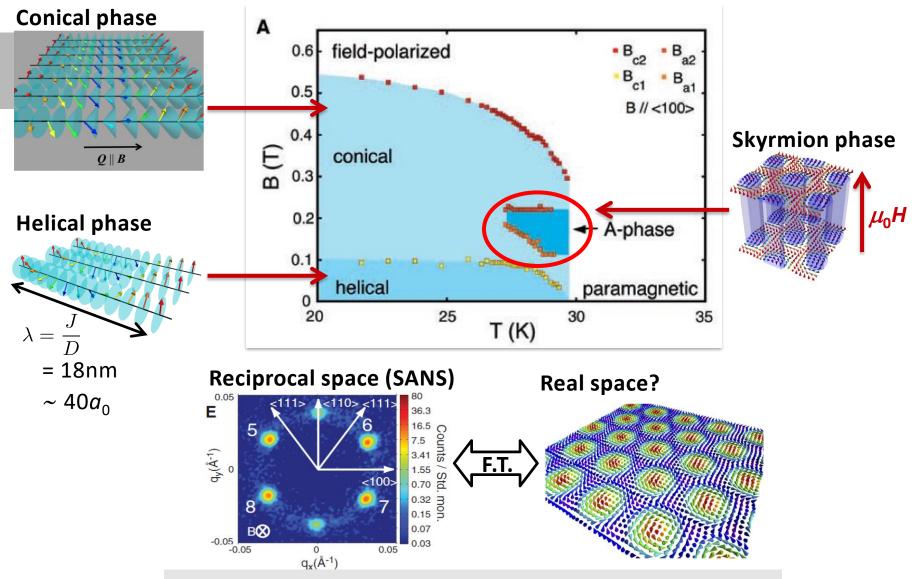






First observation in MnSi

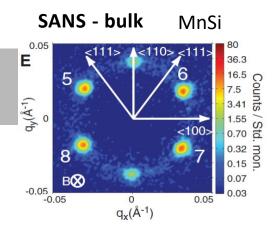
Phase diagram

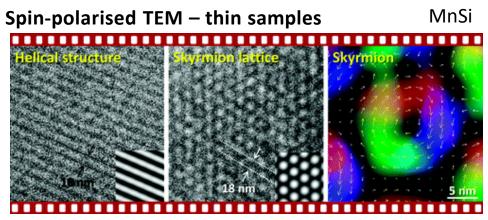


Neutron scattering does not directly 'see' individual skyrmions



Real-space imaging of the Skyrmion particle

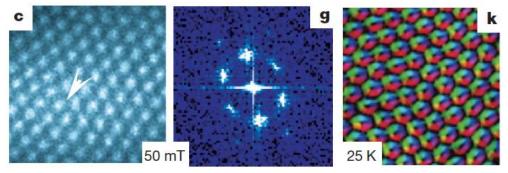




A. Tonomura et al., Nano Lett., 12, 1673 (2012)

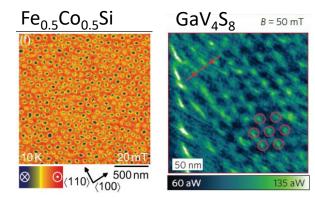
Lorentz force TEM – thin samples

Fe_{0.5}Co_{0.5}Si and FeGe



Z.X. Yu et al., Nature 465, 901 (2010), Nat. Mater. 10, 106 (2011)

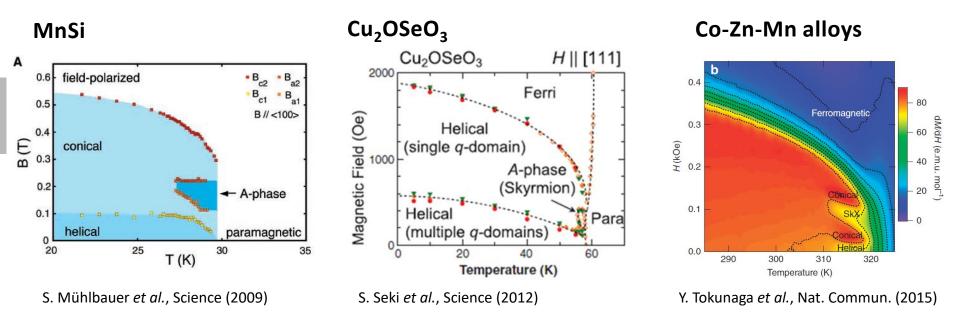
Magnetic tip AFM (MFM) – surfaces



P. Milde *et al.*, Science **340**, 1076 (2013)
I. Kézsmárki *et al.*, Nat. Mater. **14**, 116 (2015)

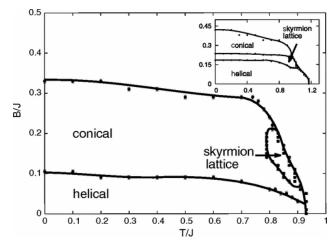


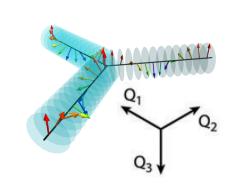
Skyrmion phase stability



In mean-field theory, J, DMI and Zeeman terms only give a helical + conical phases

Monte-Carlo simulations with thermal fluctuations





Skyrmions stabilized by DMI spiral magnetism + spiral mode-coupling + thermal fluctuations



Synthetic systems: Metallic multilayers

S₁ P₁₂ S₂

Interfaces break inversion symmetry!

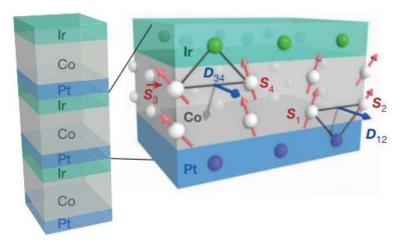
Interfacial DMI emerges between:

- Ferromagnetic layers, e.g. Co, CoFeB, Fe
- Heavy metal layers with SOC, e.g. Ir, Pt

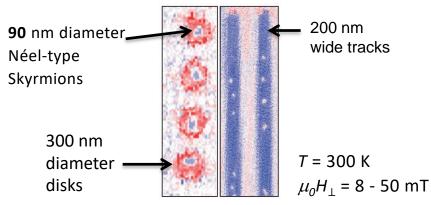
Tune effective strength of DMI:

- Vary number of interfaces, and layer thicknesses
- Additive interfacial chiral interactions → strong DMI at room temperature

Ir/Co/Pt multilayer [Ir(1 nm)/Co(0.6 nm)/Pt(1 nm)]₁₀



Scanning X-ray transmission microscopy



C. Moreau-Luchaire et al., Nat. Nanotech. 11, 444 (2016)

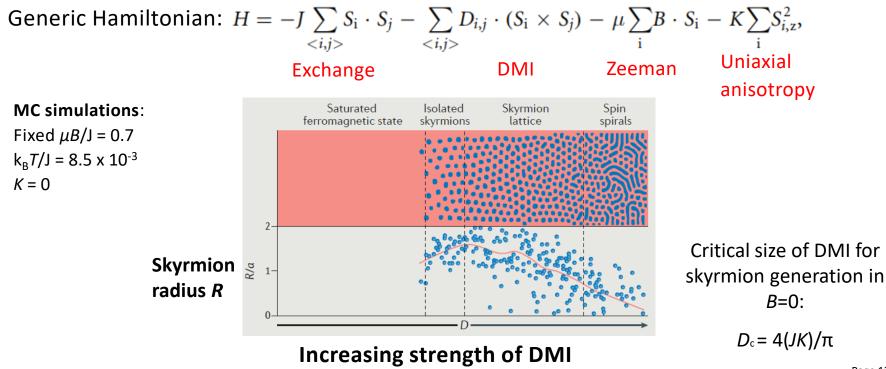


R. Wiesendanger, Nat. Rev. Mater. 1, 16044 (2016)

Systems	Temperature (K)	Magnetic field (mT)	Skyrmion size (nm)	Refs
Sputtered multilayers				
[Ta(5)/CoFeB(1.1) /TaO _x (3)]	300	0.5	700–2,000	72
[Pt(3)/Co(0.9)/Ta(4)] ₁₅	300	0–2	400–500	33
[Pt(4.5)/CoFeB(0.7)/MgO(1.4)] ₁₅	300	0-2	400-500	33
[lr(1)/Co(0.6)/Pt(1)] ₁₀	300	0–80	40–90	32

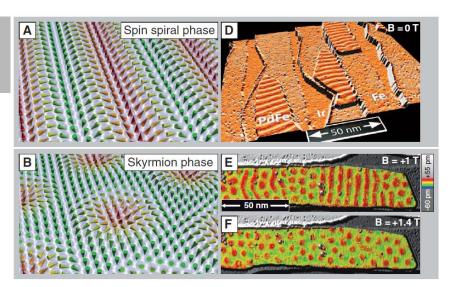
Energetics

A. Siemens, et al. New J. Phys. 18, 045021 (2016).

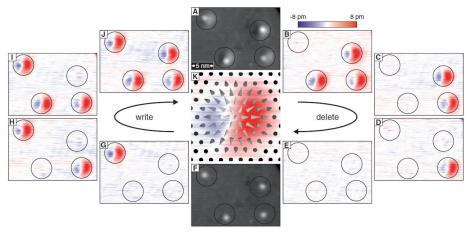




PdFe bilayer on Ir(111) – 3 nm diameter Skyrmions



Writing and erasing of individual Skyrmions SP-STM imaging



N. Romming *et al.*, Science **341**, 636 (2013) S. Heinze *et al.*, Nat. Phys., **7**, 713 (2011)

Systems	Temperature (K)	Magnetic field (mT)	Skyrmion size (nm)	Refs
Ultrathin epitaxial films and multilayers				
1-ML Fe/Ir(111)	11	0	1	13
1-ML Fe/Ir(111)/YSZ/Si(111)	26.4	0	1	56
1-MLPd/1-MLFe/lr(111)	2.2	1,500	3	12
3-ML Fe/Ir(111)	7.8	2,500	~3	34

R. Wiesendanger, Nat. Rev. Mater. 1, 16044 (2016)



Summary: Part 1

Skyrmions are topologically non-trivial magnetic objects described by an integer winding number.

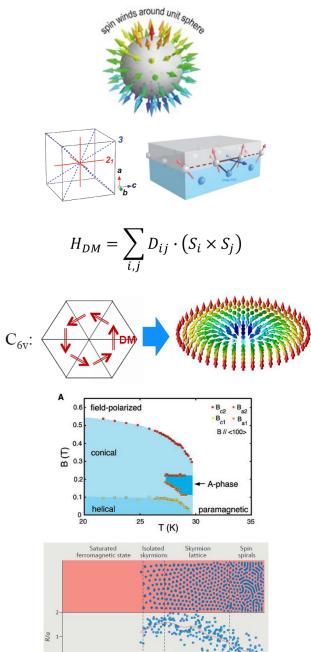
Found in some non-centrosymmetric crystals and various synthetic systems (metallic multilayers)

The Dzyaloshinskii-Moriya interaction (DMI) is a common aspect behind skyrmion formation.

The skyrmion type is determined by the local symmetries; this determines the pattern of DMIs.

In non-centrosymmetric magnets, the skyrmion phase is stabilized by mode coupling and fluctuations just below to T_{c} .

In multilayers, skyrmions are stabilized by balance between exchange, DMI, Zeeman, and anisotropy energies. Balance is tunable by sample synthesis.



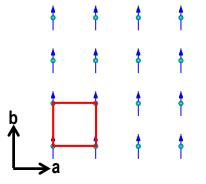


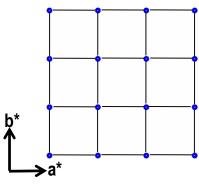
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Reminder : magnetic order in reciprocal space

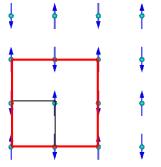
Ferromagnet

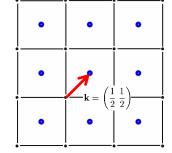




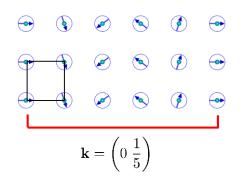
Magnetic scattering overlaps nuclear scattering in reciprocal space.

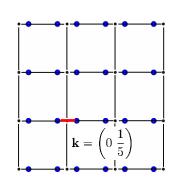
Commensurate antiferromagnet





Incommensurate structures





Magnetic scattering separated from nuclear scattering by ${f k}$.

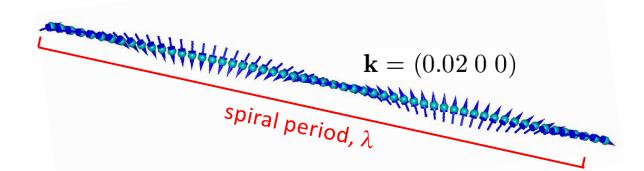
Magnetic scattering is separate from nuclear scattering.

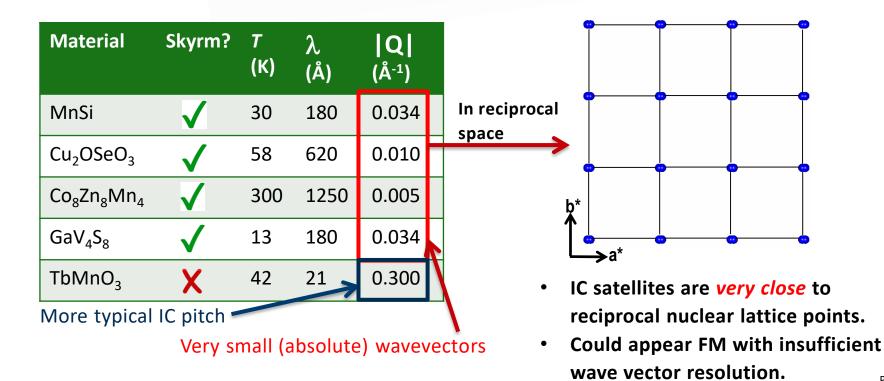
Each node of the nuclear reciprocal lattice is flanked by a pair of satellites due to $\mathbf{k} \& -\mathbf{k}$.



Spirals in non-centrosymmetric magnets

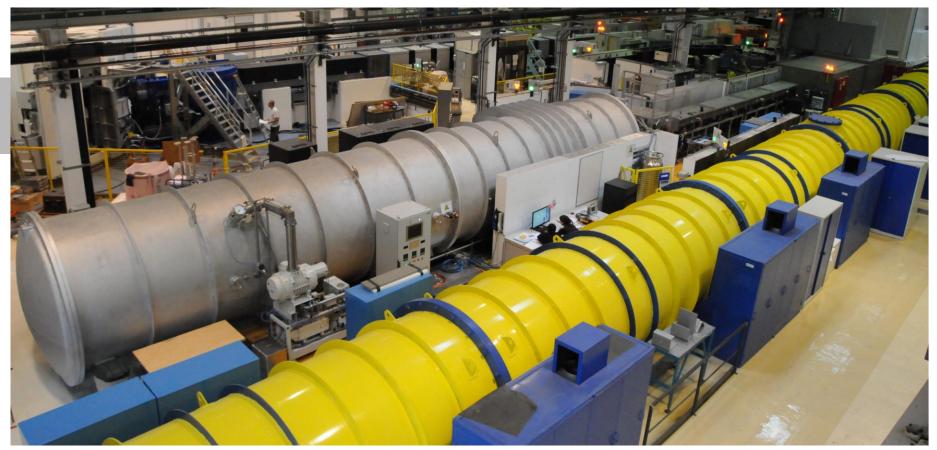
Non-centrosymmetric magnets host long period spirals and skyrmions.







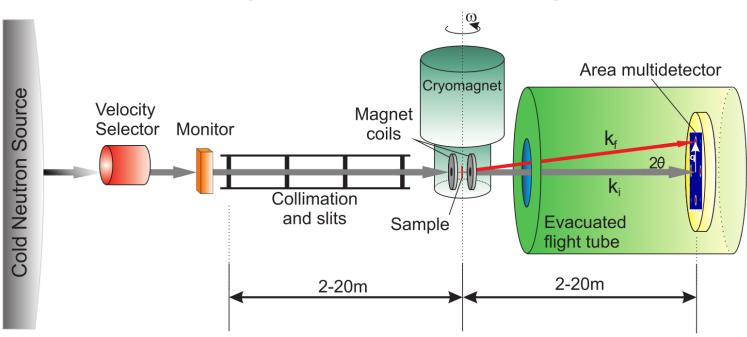
D33 SANS at the Institut Laue-Langevin (ILL), France



D33 @ ILL (Courtesy C. Dewhurst)

- SANS provides the sufficient wavevector resolution at low momentum transfers.
- Neutron scattering is an established and sensitive probe of magnetic structures.
- Diffraction in transmission geometry

Small-angle neutron scattering (SANS)

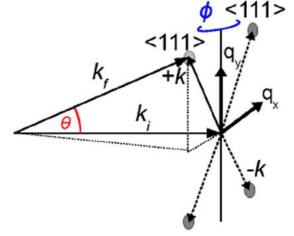


• Length of instrument: 4-40 m

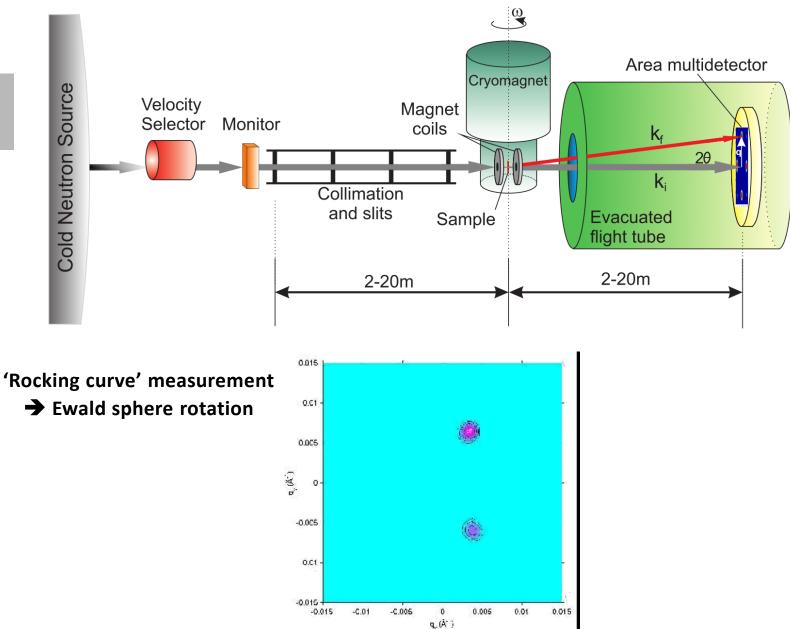
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- Large period structures (nm to μ m) \rightarrow low q
- Low q scattering range : 0.002 to 0.5 Å⁻¹.
- Small scattering angles: ~1-5° $\lambda=2d{
 m sin} heta$
- Neutron spin polarisation analysis

Magnetic peaks in reciprocal Space



Small-angle neutron scattering (SANS)

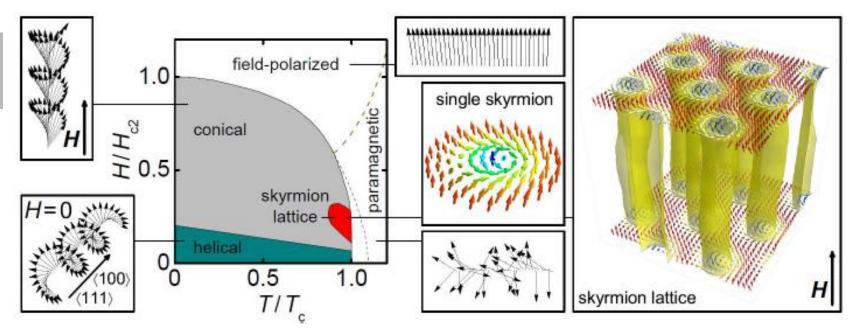


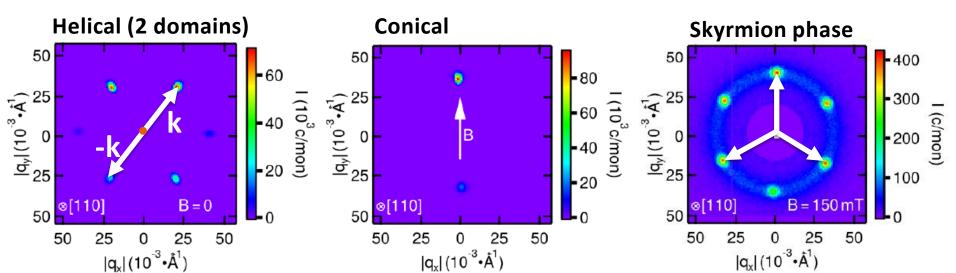


Bloch-type skyrmions in chiral magnets

e.g. MnSi, FeGe, Cu₂OSeO₃, Co₈Zn₈Mn₄..

Slide courtesy J. Kindervater







$$f_n \left(\mathbf{k}_{i}, \mathbf{k}_{f}, \hbar \omega \right) = f_n^{(\text{charge})} \left(\mathbf{Q} \right) + f_n^{(\text{non-res})} \left(\mathbf{Q}, \mathbf{k}_{i}, \mathbf{k}_{f} \right) + f_n^{(\text{res})} \left(\mathbf{Q}, \mathbf{k}_{i}, \mathbf{k}_{f} \right)$$

$$Atomic electrons (Thomson scat.) Sensitive to magnetism$$

Few unpaired electrons contribute to the magnetic scattering vs. all electrons in the Thomson scattering.

Non-resonant magnetic signal is weak: Intensity ratio between charge and non-resonant magnetic scattering

$$\left(rac{\hbar\omega}{mc^2}
ight)^2\sim 10^{-4}\,$$
 for 10keV x-rays

Help 1: Modern, high brilliance synchrotron sources (>10¹² ph/mm²/s)



$$\begin{array}{l} \textbf{Help 2: X-ray beam polarization analysis} \\ \hline \textbf{Help 2: X-ray beam polarization analysis} \\ \hline \textbf{Mon-resonant scattering amplitude} \\ f_n^{(non-res(mag))} \propto ir_0 \left(\frac{\hbar\omega}{mc^2}\right) \begin{bmatrix} \frac{1}{2} \textbf{L}(\textbf{Q}) \cdot \textbf{A} + \textbf{S}(\textbf{Q}) \cdot \textbf{B} \end{bmatrix} \\ \hline \textbf{Orbital} \\ \textbf{Spin density} \\ \textbf{A} = 2 \left(1 - \hat{\textbf{k}}_i \cdot \hat{\textbf{k}}_f\right) (\hat{\epsilon}' \times \hat{\epsilon}) - (\hat{\textbf{k}}_i \times \hat{\epsilon}) (\hat{\textbf{k}}_i \cdot \hat{\epsilon}') + (\hat{\textbf{k}}_f \cdot \hat{\epsilon}') (\hat{\textbf{k}}_f \cdot \hat{\epsilon}) \\ \hline \textbf{B} = (\hat{\epsilon}' \times \hat{\epsilon}) + (\hat{\textbf{k}}_f \times \hat{\epsilon}') (\hat{\textbf{k}}_f \cdot \hat{\epsilon}) - (\hat{\textbf{k}}_i \times \hat{\epsilon}) (\hat{\textbf{k}}_i \cdot \hat{\epsilon}') - (\hat{\textbf{k}}_f \cdot \hat{\epsilon}') \times (\hat{\textbf{k}}_i \cdot \hat{\epsilon}) \\ \hline \textbf{C} \end{array}$$

•S/L separation (unlike neutrons)

•Distinction between magnetic and charge scattering

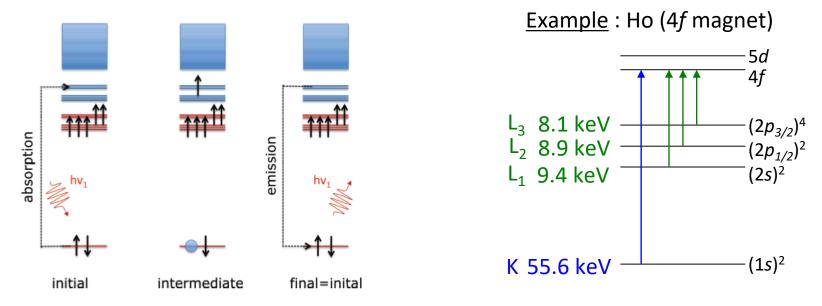
Help 3: <u>Wide x-ray energy selectivity</u>

Tune the x-ray energy to match atomic transitions \rightarrow resonance effects \rightarrow <u>leads to large magnetic intensities</u>



Resonant X-ray scattering

'Virtual transition' process



- Photon excites a core-level electron into a partly filled shell.
- Spontaneously decays through the emission of an elastically scattered photon with particular polarization.
- Scattering amplitude dependent on the relative directions of the local *E*-field (incoming polarization) and local quantization axes (magnetic state of level probed)

Enhanced sensitivity to magnetism and elemental sensitivity

Most common transition is electric dipole $\Delta\ell=\pm 1$, e.g. 2p to 3d:

$$f_n^{(\text{res})}\left(E1\right) = F^{(0)}\left(\hat{\epsilon}\cdot\hat{\epsilon}'\right) - iF^{(1)}\left(\hat{\epsilon}'\times\hat{\epsilon}\right)\cdot\boldsymbol{\mu}_j + F^{(2)}\left(\hat{\epsilon}'\cdot\boldsymbol{\mu}_j\right)\left(\hat{\epsilon}\cdot\boldsymbol{\mu}_j\right)$$

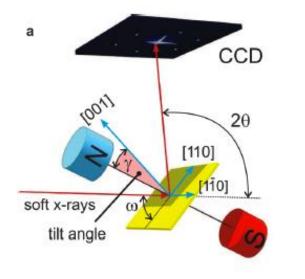


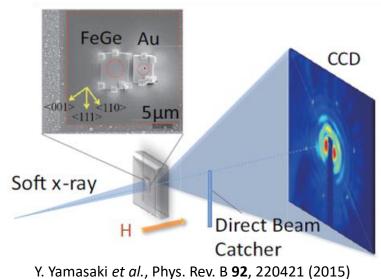
Resonant magnetic soft x-ray scattering

- For Bloch-type skyrmion materials excite 2p to 3d transitions of the TM element (the L edge), e.g. Fe in FeGe or Cu in Cu₂OSeO₃
- At the Fe $L_{2,3}$, x-ray energy is in the 'soft' range 480 950 eV, $\lambda \sim 15$ -20 Å
- Avoid air absorption of x-ray beam by placing the entire diffractometer in vacuum (10⁻⁸ torr).
 - Very restricted wavevector range at soft x-ray energies.
 - Rare to be able to reach *any* Bragg spots, including structural peaks.
 - But possible to measure spiral/skyrmion peaks at low wavevectors (like SANS).

Reflection geometry (bulk crystals)

SAS transmission geometry (~100nm thick) No pol. anal!







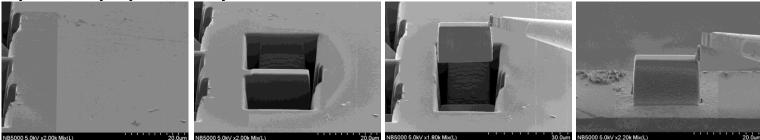
Sample preparation for soft X-ray experiment

- Sample size is limited by the attenuation length of soft X-ray beam (\sim 100 1000 nm thick for TM) and size of the beam (aperture size of 5-10 μ m)
- Method: thin plates prepared by Focused Ion Beam (FIB) milling

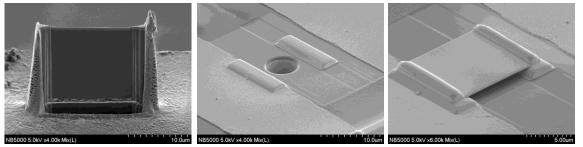
Membrane preparation Si₃N₄ membrane: thickness of 200 nm (transparent for soft X-rays)

F. Büttner et al., Optics Express 21, 30563 (2013).

Specimen preparation by FIB



Bulk single crystal (FeGe)



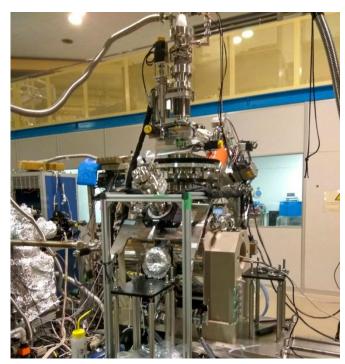
Thinning process (100-200 nm) Making an aperture and specimen attachment to SiN membrane

Slide: courtesy V. Ukleev



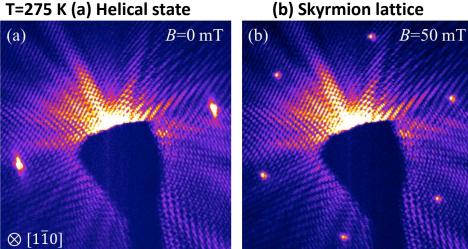
Resonant soft x-ray scattering from FeGe

Slide: courtesy V. Ukleev

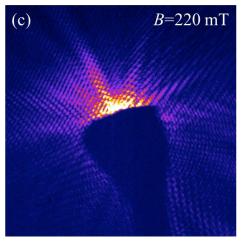


RSXS endstation, Photon Factory, BL-16A

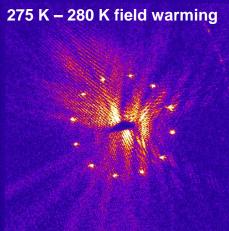
Y. Yamasaki *et al.*, Phys. Rev. B **92**, 220421 (2015)
V. Ukleev *et al.*, Quantum Beam Science **2**, 3 (2018)



(c) Field-polarized / conical



(d) Sk. lattice field-warming



3 s per exposure c.f. SANS pattern takes 10-30 mins!



Summary: Part 2

Compare between x-rays and neutrons for magnetism

Resonant X-rays

- Improved sensitivity due to high flux
- High spatial resolution
- Spatial and temporal beam coherence
 - Real-space imaging
 - Pump-probe
- Element specificity
- Electron shell selective
- S/L separation
- Tiny crystals (< 1mg) /surfaces/thin films
- Identification of magnetic ions
- Wide x-ray energy tunability
- Polarimetry in reflection geometry
- Resonance edge may not accessible
- Hard x-rays (indirect probe states)
- Soft x-rays (very limited Ewald sphere)
- Data modelling is complex
- Full magnetic structure refinements not possible by resonant x-rays.
- Heat loading can prevent low *T*s.
- For thin samples, unavoidable strain effects with *T* variation.

Neutrons

- High magnetic sensitivity
- Polarization techniques
- Highly penetrating probe of bulk samples
- Highly penetrating probe of complex sample environments
- Magnetic structure calculations simpler
- Bulk crystals/thin film stacks
- Strain free bulk measurements.

- Flux limited
- Insensitive to tiny magnetic moments
- Low spatial resolution
- Some elements are too neutron absorbing to study e.g. Dy, Gd, B
- Magnetic scattering not element specific
- Needs larger sample masses (> 2 mg)
- Challenging to study surfaces and thin films.
- Low temporal resolution



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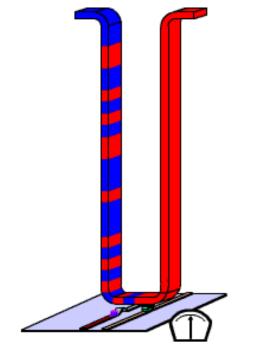
Magnetic Skyrmions for Applications – 'Skyrmionics'

Skyrmionics:

Demonstrate the motion, creation, annihilation of small (< 10 nm) skyrmions in confined geometries, using electric currents (or electric fields) at room *T*.

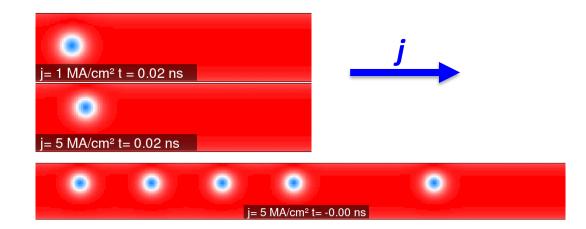
Concept: Racetrack Memory

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



No mechanical parts High domain densities

Skyrmion racetrackA. Fert *et al.*, Nat. Nanotech. 8, 152 (2013)Theory: skyrmions move under very low current densities!



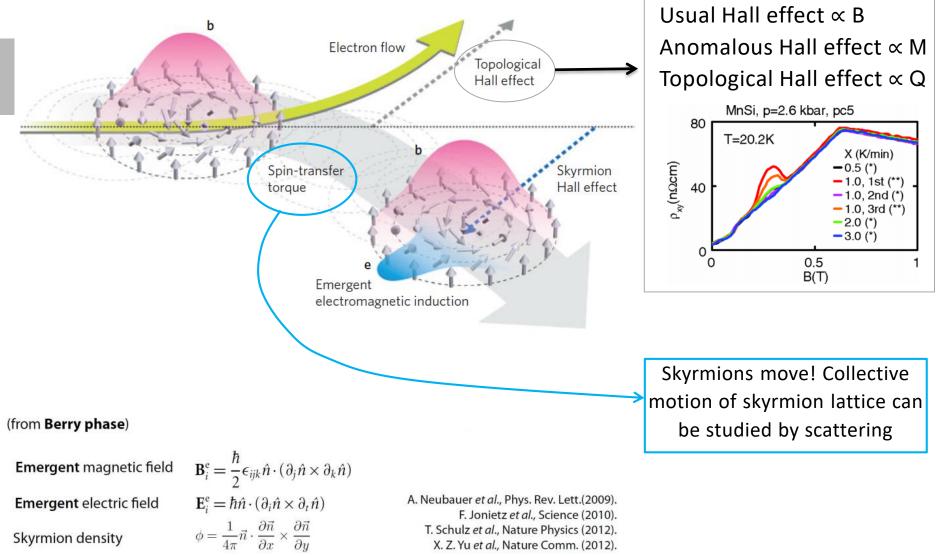
Potential skyrmion applications

- build non-volatile computer memories
- superfast devices with low dissipation
- low power-dissipation transistors
- efficient GHz oscillators
- other exotic ideas out there..



Conduction electrons and skyrmions

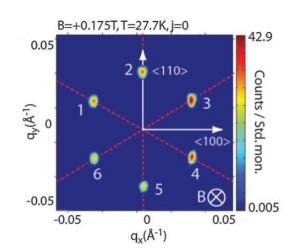
N. Nagaosa and Y. Tokura, Nat. Nanotech. 8, 899 (2013)

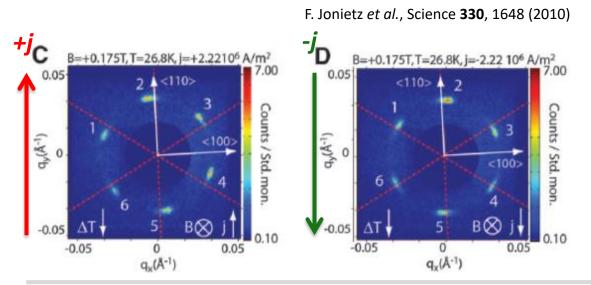


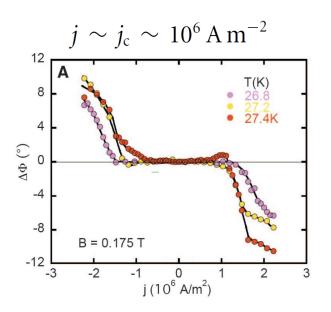


Spin transfer torques and SANS

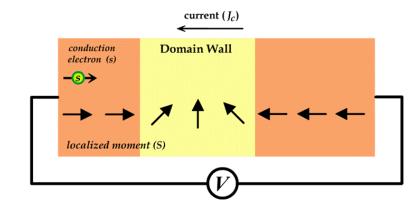
MnSi at low temperature







Compare: Current pulses drive the movement of FM domain walls by STTs: $j\sim j_{\rm c}\sim 10^{11}{\rm A~m}^{-2}$

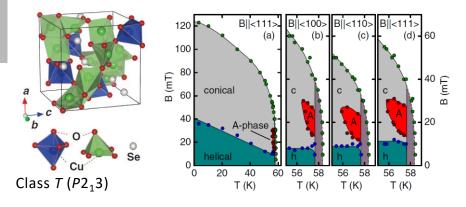


Tiny current densities needed to drive skyrmions compared with FM domains – energy efficient! Pa

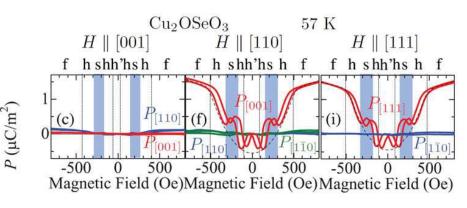


Magnetoelectric insulator Cu₂OSeO₃

- S. Seki, et al., Science **336**, 198, (2012)
- T. Adams et al., Phys. Rev. Lett. **108**, 237204 (2012)



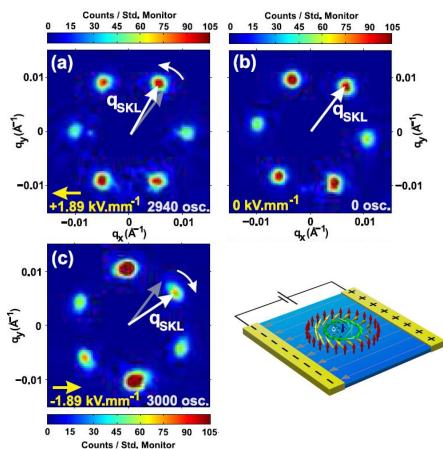
Magnetoelectric coupling in Skyrmion phase



Symmetry of ME coupling: *d-p* hybridization model

Electric field control of the Skyrmion lattice

JSW et al., Phys. Rev. Lett. 113, 107203 (2014)



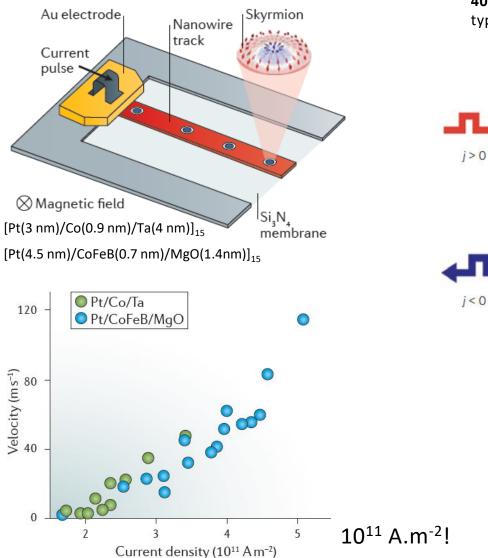
Skyrmion lattice rotations achieved with no STTs – i.e. no current or magnon flows

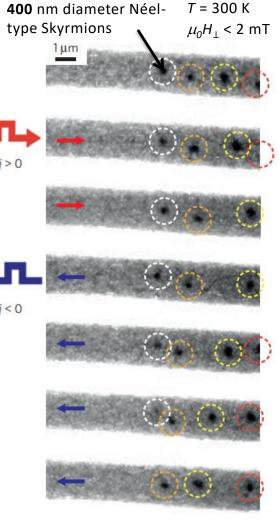
Microscopic explanation for rotation still missing!



Room temperature, current pulse-driven motion of skyrmions at high speeds

S. Woo et al., Nat. Mater. 15, 501 (2016)







Summary: Part 3

Skyrmionics:

Demonstrate the motion, creation, annihilation of small (< 10 nm) skyrmions in confined geometries, using electric currents (or electric fields) at room *T*.

Initial challenges for skyrmionics:

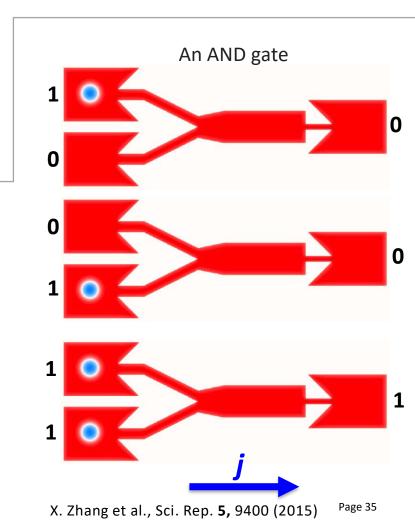
- stability: skyrmions at room temperature
- creation/destruction of skyrmions
- control of skyrmions in bulk / nanostructures

Present challenges:

- More materials and systems
- creation /demonstration of functional / integrated devices
- materials synthesis and technology

most promising materials: multilayer systems BUT: problems with defects induced by synthesis.

• Identify "killer applications" that are properly competitive (e.g. for memories)





- What are magnetic Skyrmions, and where do we find them?
 - Non-centrosymmetric crystals
 - Synthetic systems
- Application of scattering techniques
 - Neutron and resonant x-ray scattering
- Magnetic Skyrmions for Applications 'Skyrmionics'
 - Concepts, status and challenges
- Summary

Funding: Swiss National Science Foundation

Sinergia project 'NanoSkyrmionics'



Nan Skymi

https://skyrmions.epfl.ch/