Advancing magnetic soft x-ray microspectroscopy towards the quantum regime

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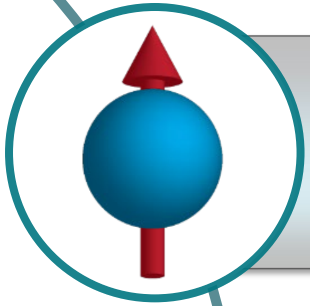
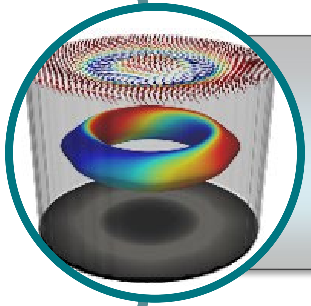
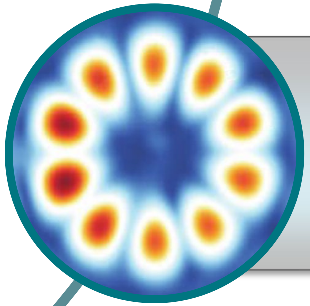
Outline



Nanomagnetism – From classical to quantum



Topological (quantum) spin textures



X-ray vortex beams | OAM beams

From Classical to Quantum

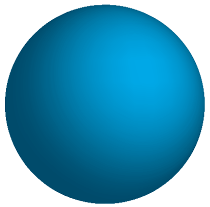
- tunneling
- superposition
- entanglement
- quantum phases, e.g. BE condensates
- quantum criticality, e.g. probing phase transitions at quantum boundaries
- quantum coherence
- bridging quantum mechanics and topological physics
- potential applications in quantum computing

From basic materials science to quantum computing

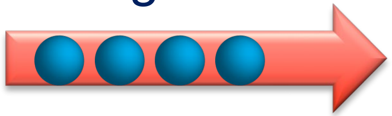
electron

charge

spin

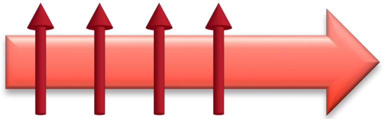


ELECTRONICS
only charge degree of freedom
charge currents



J_C

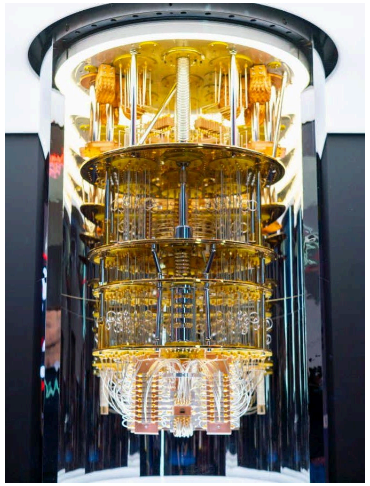
SPINTRONICS
(only) spin degree of freedom
spin currents



J_S

SC qubits

ATLANTIC QUANTUM



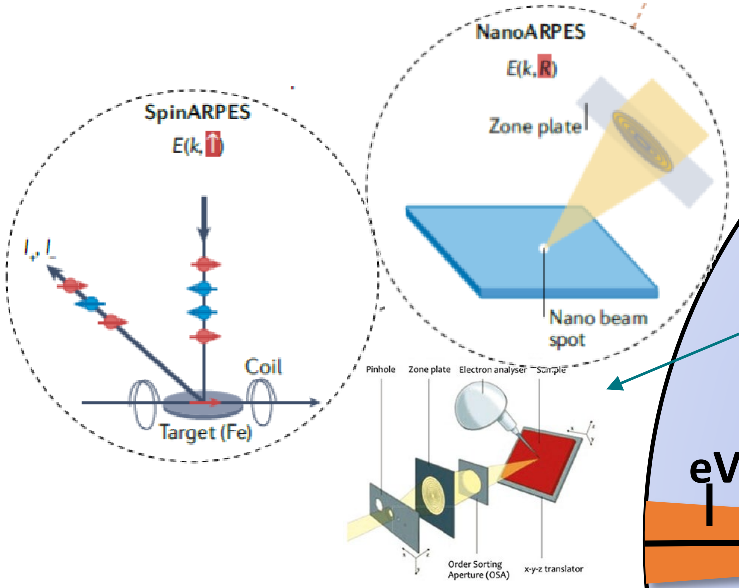
IQM

OQC

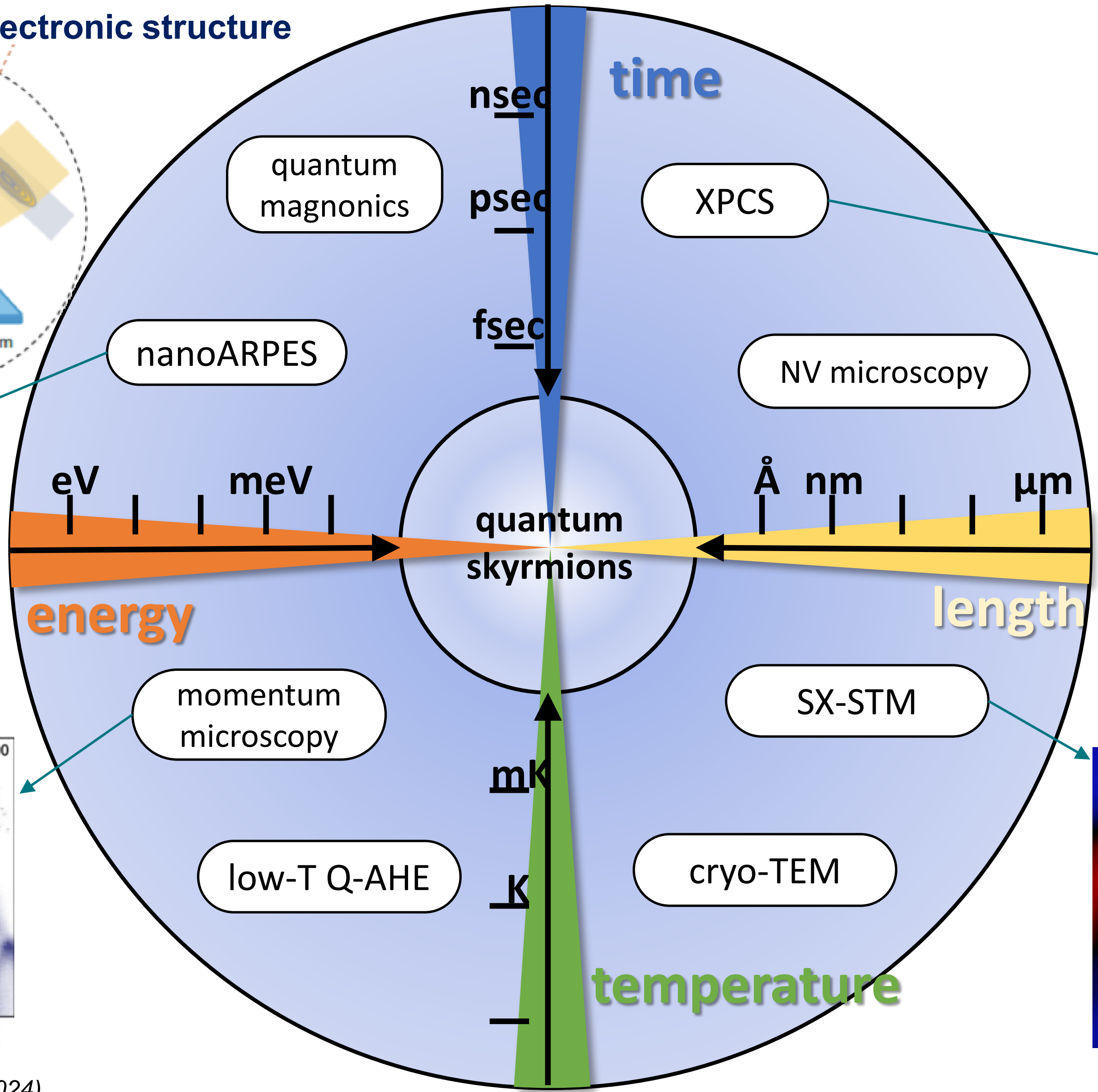
Skyrmion qubits

Experimental challenges towards the quantum regime

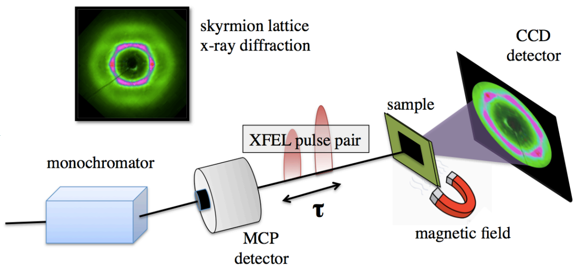
- **spatiotemporal dependence of electronic structure**



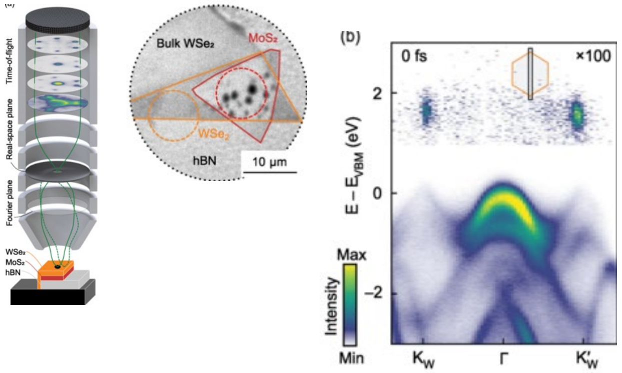
CY Lim, et al, Current Applied Physics 60 43 (2024)



- **Ultrafast nanoscale spin fluctuations**

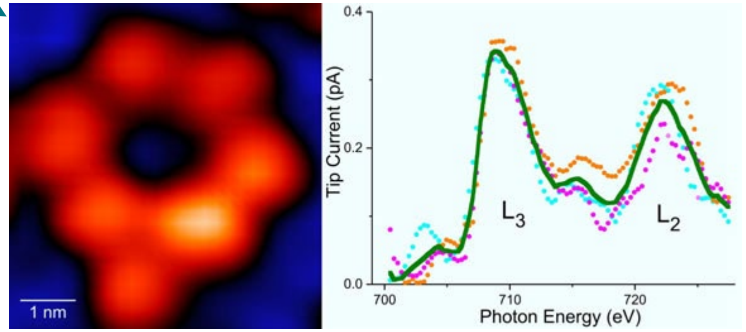


M.H. Seaberg et al, Phys Rev Lett 119 067403 (2017)



F. Boschini et al, Rev. Mod. Phys. 96, 015003 (2024)

- **Characterization of just one atom using synchrotron X-rays**

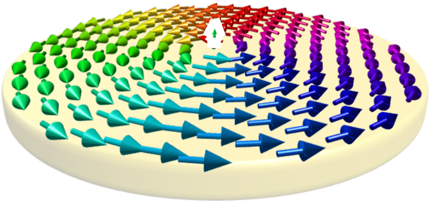


T. M. Ajayi et al, Nature 618 69 (2023)

A. Petrovic et al, (2025) arXiv.2410.11427

Topological spin textures: From 2D/3D to quantum

Vortex



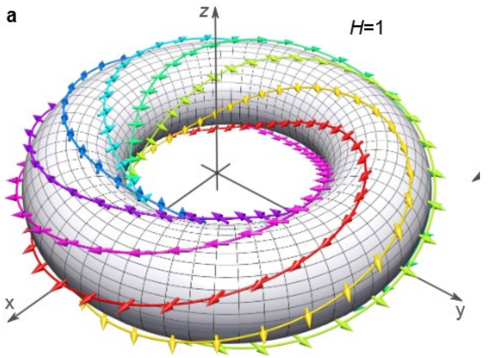
- polarity $p=\pm 1$
- chirality(circularity) $n=\pm 1$

Classical Skyrmion

- **Skyrmion** number: winding number

$$n = \frac{1}{4\pi}\int \vec{M}\cdot\left(\partial_x\vec{M}\times\partial_y\vec{M}\right)dxdy$$

Hopfion



F. Rybakov, et al. APL Mater. 10, 111113 (2022)

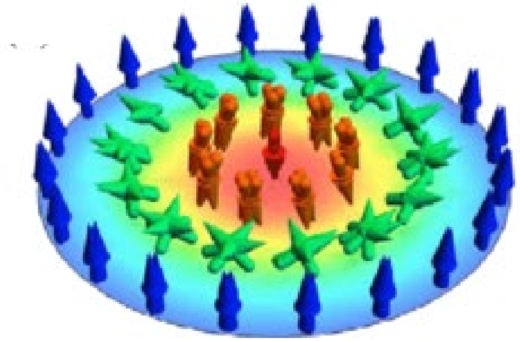
- **Hopf** number: linking number

$$Q_H = -\int B\cdot A\, dr$$

$$B_i = \frac{1}{8\pi}\epsilon_{ijk}s\cdot\left(\nabla_j s\times\nabla_k s\right)$$

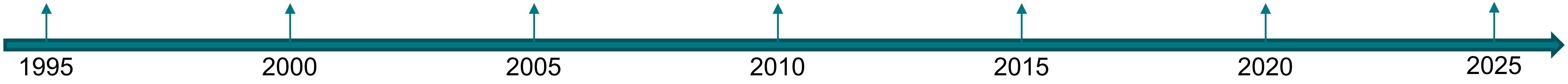
*Y. Liu et al, Phys Rev B **98**, 174437 (2018)*

Quantum Skyrmion



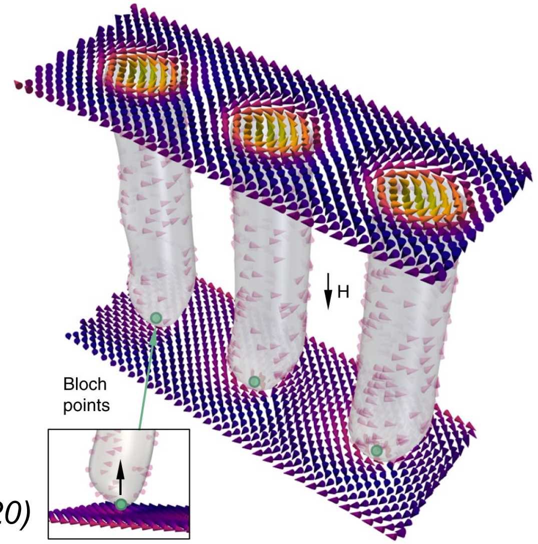
- tunneling between skyrmion and antiskyrmion states
- superpositions of classical configurations
- expectation values of the local spin vector operator $\langle \mathbf{S}(\mathbf{r})\rangle$

V. Lohani et al, (2019) Phys. Rev. X 9, 041063
A. Petrovic et al, (2025) arXiv.2410.11427



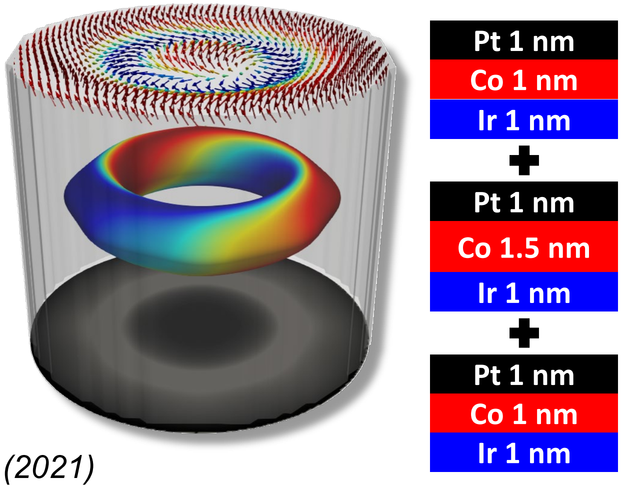
Experimental validation of unconventional topological spin textures

- Imaging of **skyrmion tubes** in a lamella of FeGe using resonant magnetic x-ray diffraction and holography



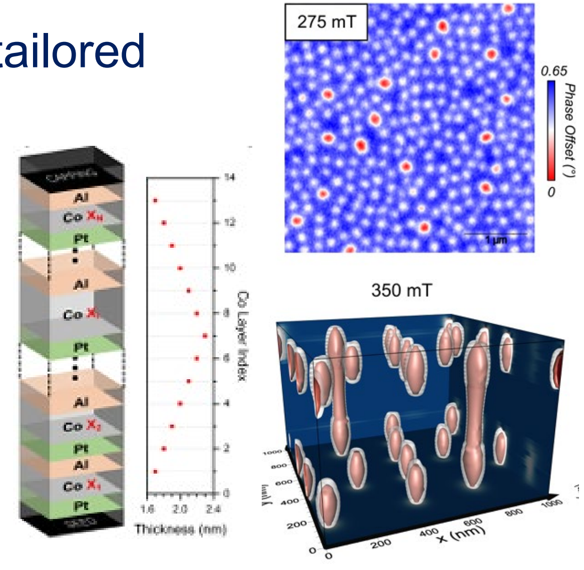
M.T. Birch et al., Nature Comm 11 1726 (2020)

- Experimental verification of magnetic **Hopfions** in magnetic multilayers with a vertically graded PMA by surface/bulk sensitive X-PEEM/MTXM imaging



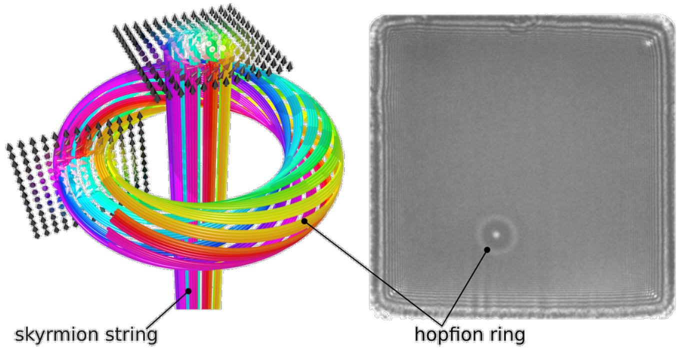
N. Kent et al., Nature Comm 12 1562 (2021)

- 3D skyrmionic **cocoons** in tailored (single gradient) magnetic multilayers with a characteristic ellipsoidal shape confirmed by signatures in MFM images



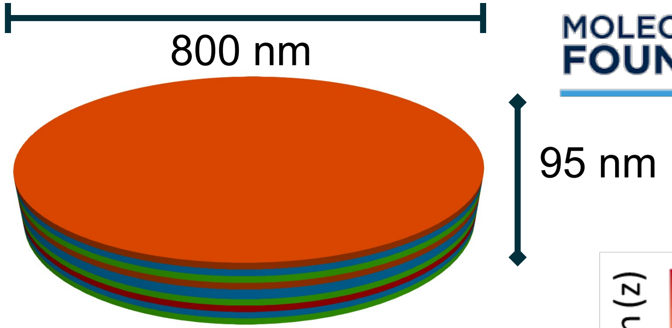
M. Grelier et al., Nature Comm 13, 6843 (2022)

- Tailored nucleation and direct observation of **Hopfions** forming coupled states with skyrmion strings in B20-type FeGe crystals using L-TEM and electron holography

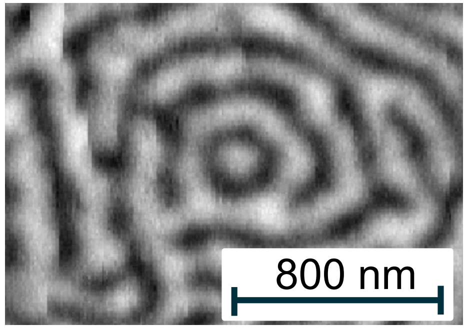


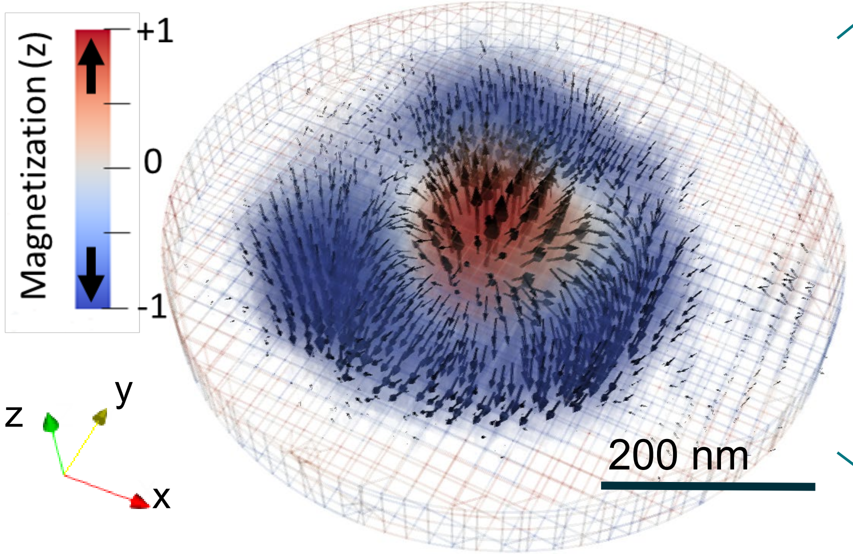
F. Zheng et al., Nature 623 718 (2023)

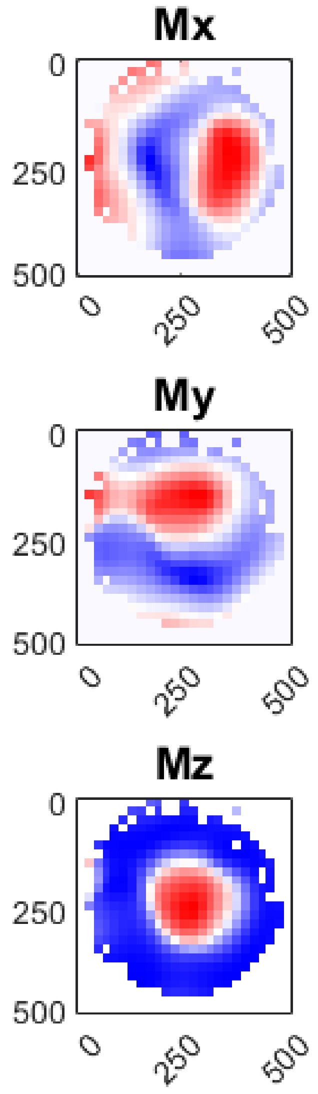
Full 3D reconstruction of the spin texture in a skyrmion

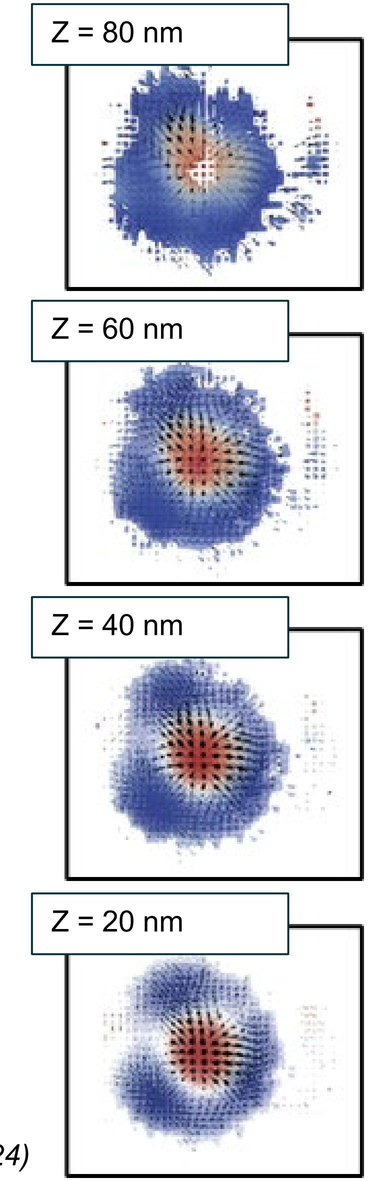


10x[Ir(1nm)/**Co(1nm)**/Pt(1nm)] /
10x [Ir(1nm)/**Co(1.5nm)**/Pt(1nm) /
10x[Ir(1nm)/**Co(1nm)**/Pt(1nm)]





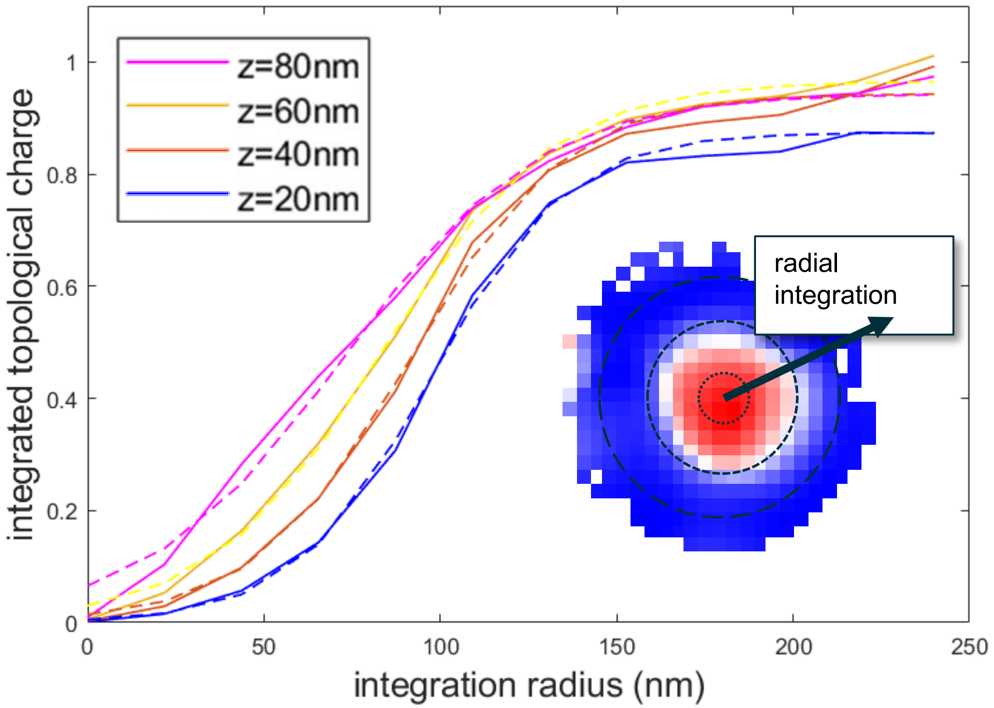


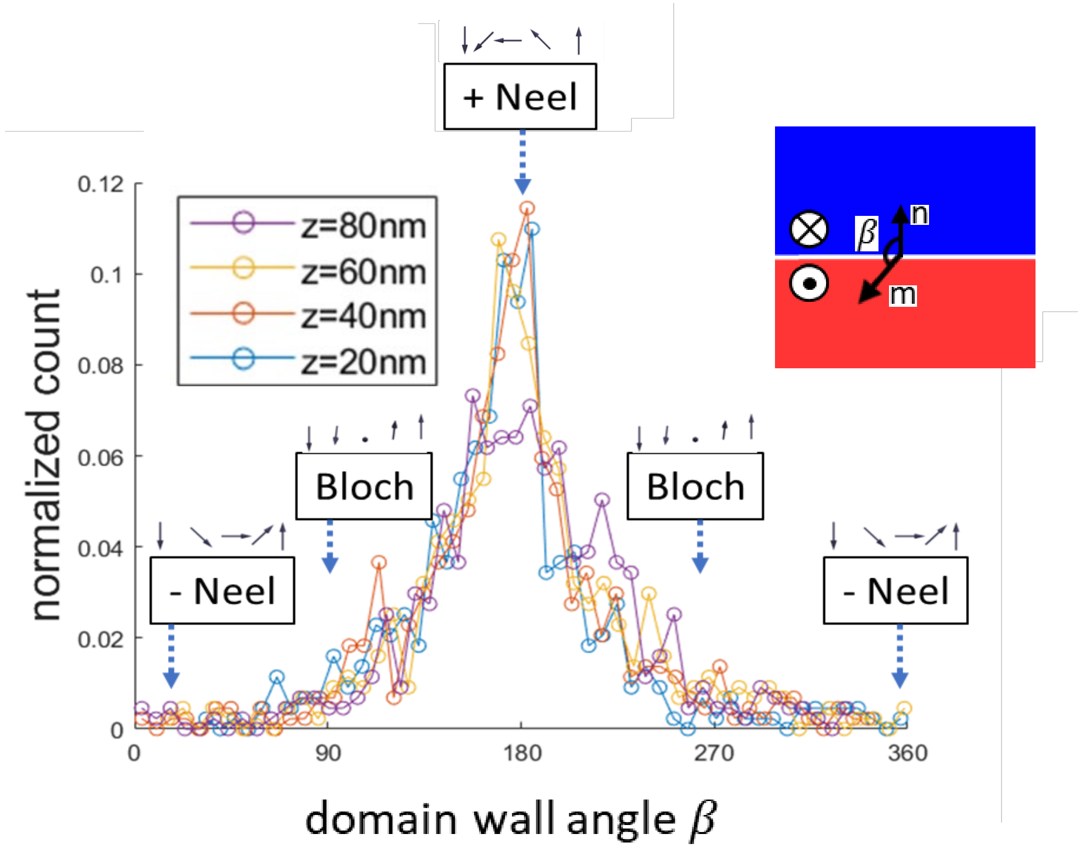


- MFM shows target skyrmion
- Resolution is determined by Fourier shell correlation analysis
- FSC curve obtained for each magnetization component

D. Raftrey, S. Finizio, R.V. Chopdekar, S. Dhuey, T. Bayaraa, P. Ashby, J. Raabe, T. Santos, S. Griffin, P. Fischer, Science Advances 10, eadp8615 (2024)

Depth-dependent topology and chirality

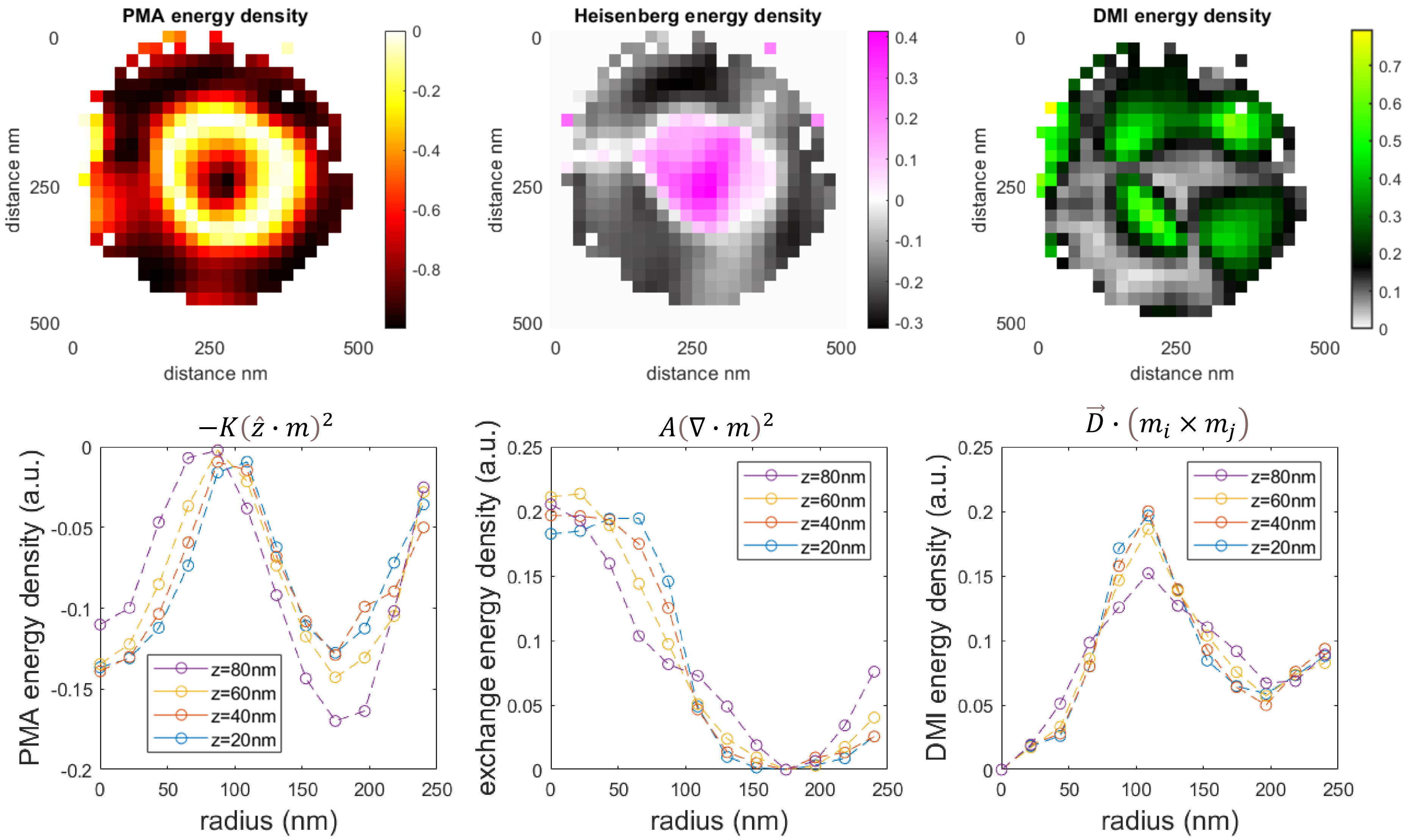




- Depth dependent radially integrated topological charge profile indicates a “barrel-like” structure similar to vortex cores in disks *(see P. Fischer, M.-Y. Im, S. Kasai, K. Yamada, T. Ono, A. Thiaville, PRB Brief Report 83 212402 (2011))*
- Layer-resolved histogram of the chirality of the domain wall, indicating a positive Neél profile across the depth

D. Raftrey, S. Finizio, R.V. Chopdekar, S. Dhuey, T. Bayaraa, P. Ashby, J. Raabe, T. Santos, S. Griffin, P. Fischer, Science Advances 10, eadp8615 (2024)

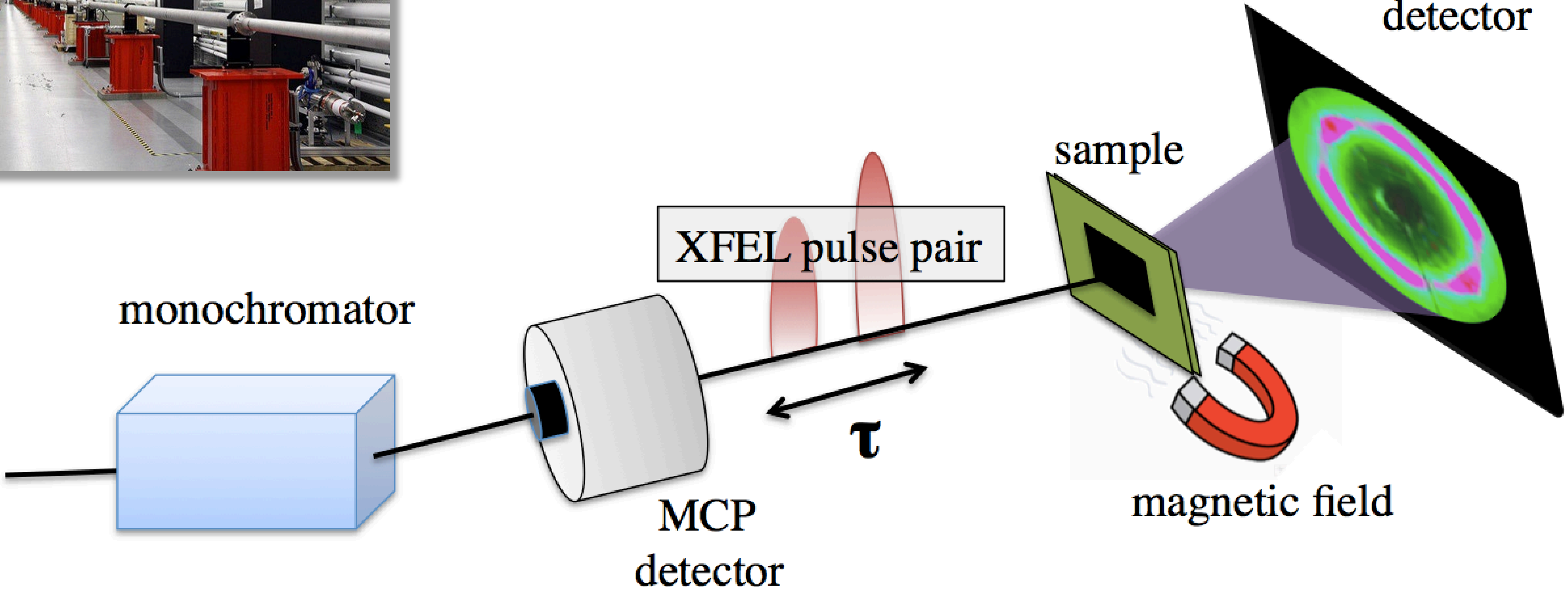
Quantification of fundamental magnetic parameters in 3D

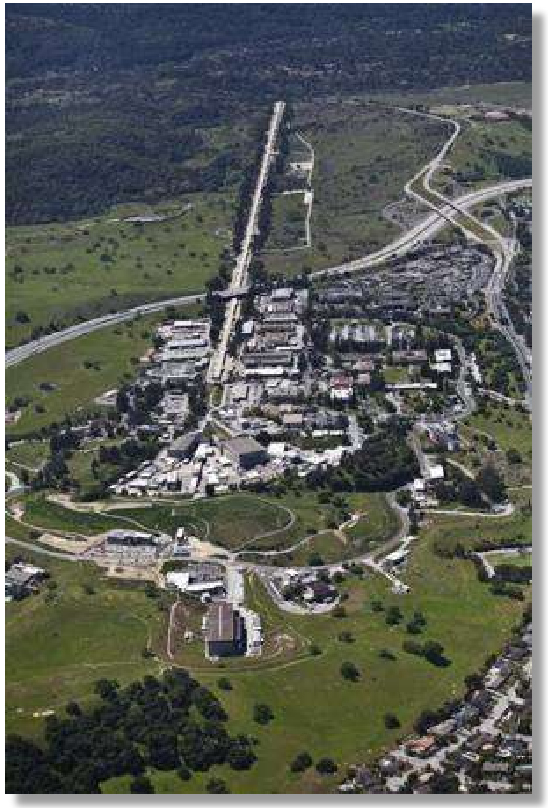


D. Raftrey, S. Finizio, R.V. Chopdekar, S. Dhuey, T. Bayaraa, P. Ashby, J. Raabe, T. Santos, S. Griffin, P. Fischer, Science Advances 10, eadp8615 (2024)

Nanosecond X-ray Photon Correlation Spectroscopy

SLAC

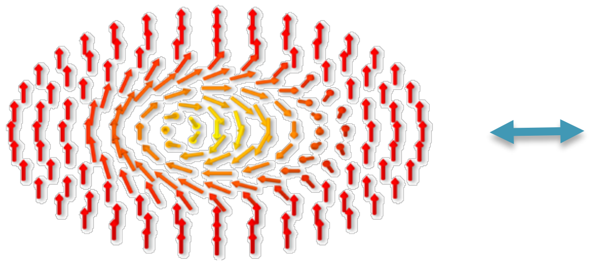


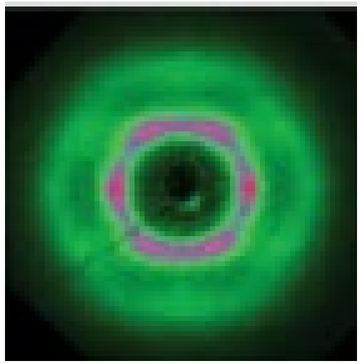


- **New technique at LCLS using the high coherent flux and short pulse length**
- Pairs of fsec pulses at LCLS enable measuring fluctuations down to τ = 350 ps
- Recording the sum of speckle patterns allows to calculate the autocorrelation

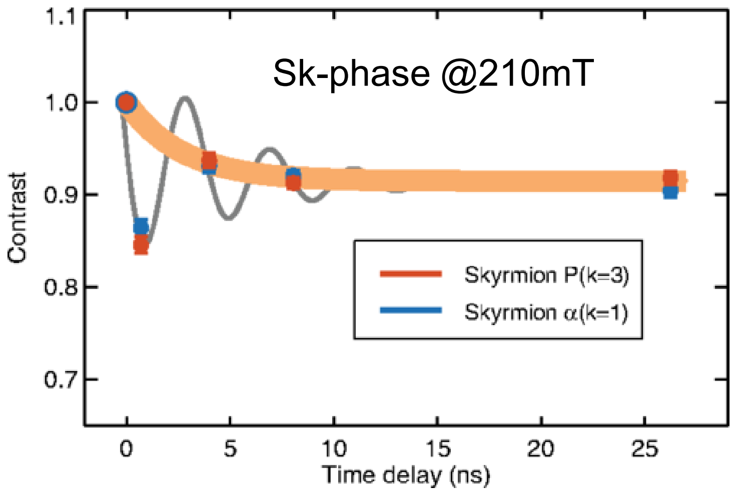
M. H. Seaberg, B. Holladay, J. C. T. Lee, M. Sikorski, A. H. Reid, S. A. Montoya, G. L. Dakovski, J. D. Koralek, G. Coslovich, S. Moeller, W. F. Schlotter, R. Streubel, S. D. Kevan, P. Fischer, E. E. Fullerton, J. L. Turner, F.-J. Decker, S. K. Sinha, S. Roy, J. J. Turner, Phys Rev Lett 119 067403 (2017)

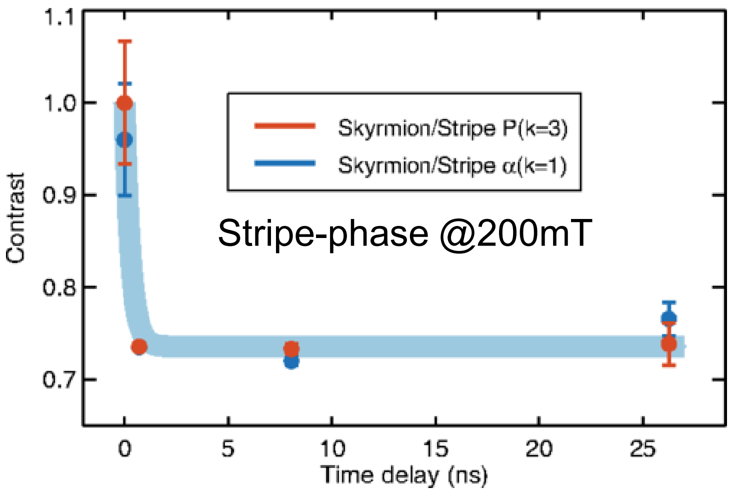
Ultrafast skyrmion fluctuations





Skyrmion lattice in x-ray diffraction





- Sample: Fe/Gd MLs that exhibit ordered stripe and skyrmion lattice phases
- Found different correlation times of the fluctuations in these phases

M. H. Seaberg, B. Holladay, J. C. T. Lee, M. Sikorski, A. H. Reid, S. A. Montoya, G. L. Dakovski, J. D. Koralek, G. Coslovich, S. Moeller, W. F. Schlotter, R. Streubel, S. D. Kevan, P. Fischer, E. E. Fullerton, J. L. Turner, F.-J. Decker, S. K. Sinha, S. Roy, J. J. Turner, Phys Rev Lett 119 067403 (2017)

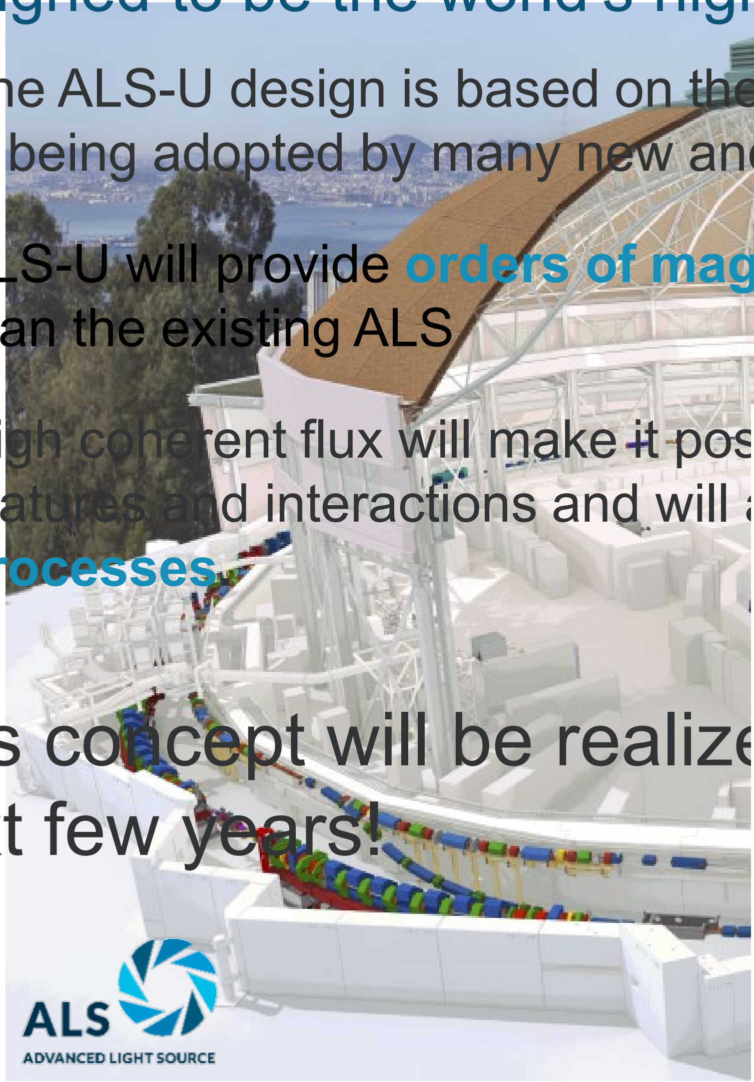
The next generation (ALS-U) - DLSR in Berkeley

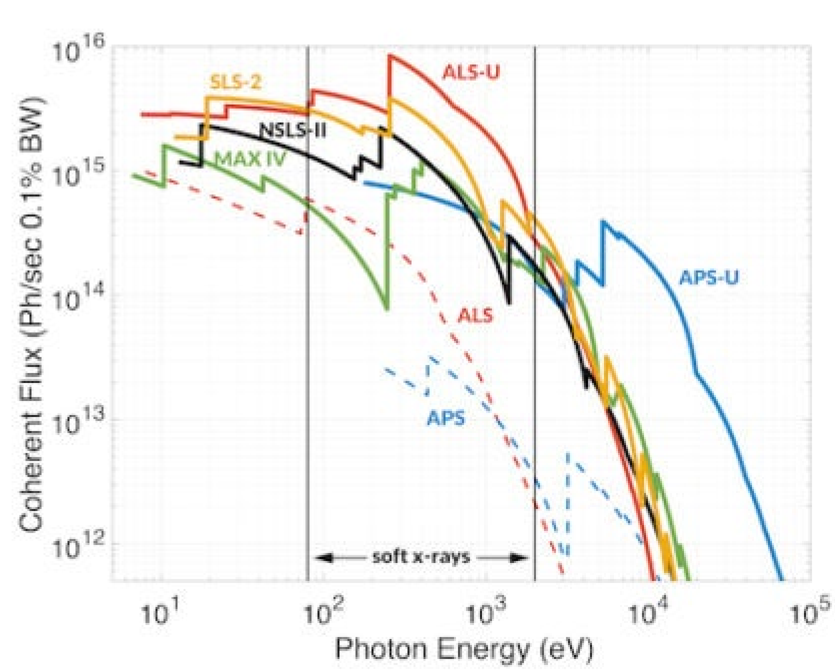
BERKELEY LAB

Designed to be the world's highest coherent flux soft x-ray synchrotron light source

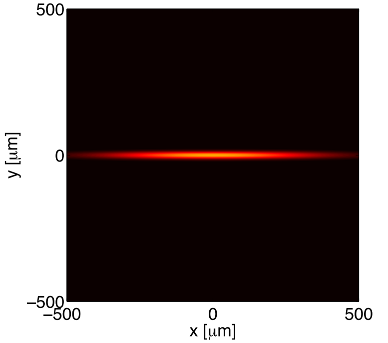
- The ALS-U design is based on the **multibend achromat lattice** that is being adopted by many new and upgraded facilities.
- ALS-U will provide **orders of magnitude more coherent soft x-ray flux** than the existing ALS
- High coherent flux will make it pos features and interactions and will a **processes**

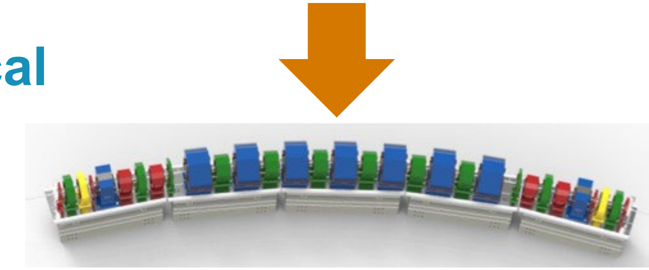
This concept will be realize next few years!

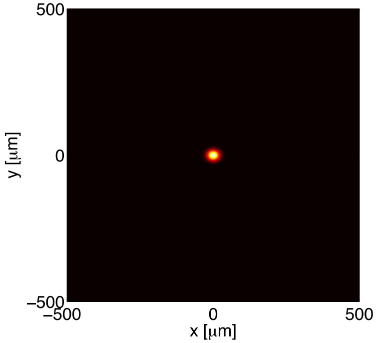




cal

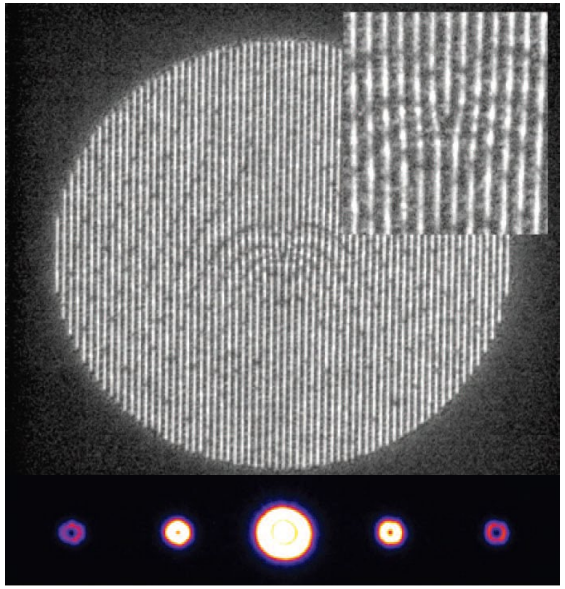




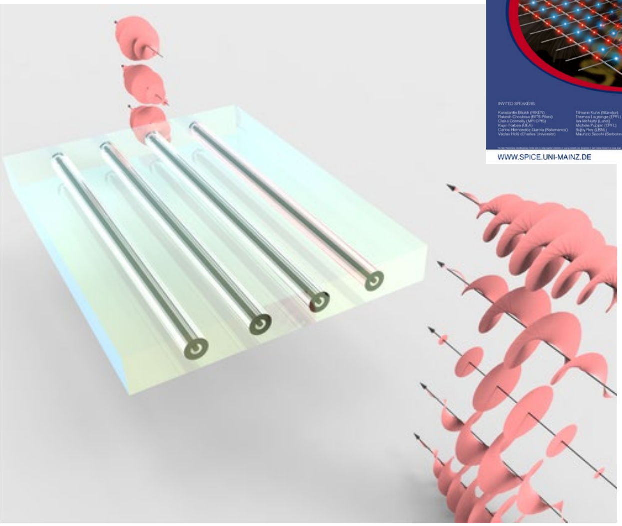


https://als.lbl.gov/als-u/overview/

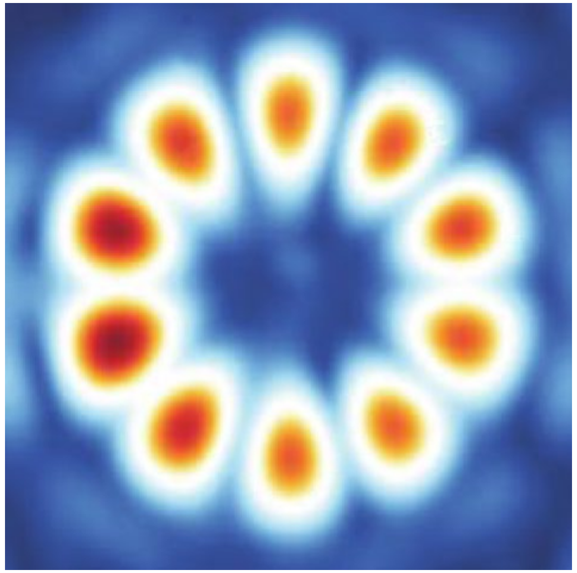
Coherence enables OAM X-ray beams



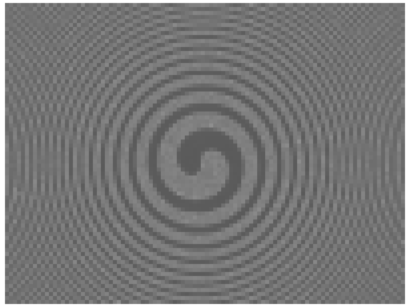
- Coherent e-beams in a TEM with helical wavefronts carry large OAM
  B. McMorran et al., Science 331 191 (2011)

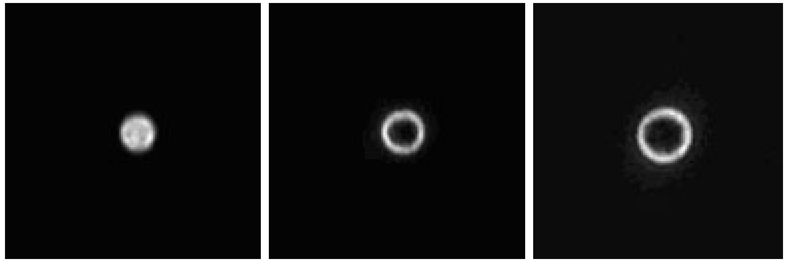


- Twisted light in a photonic chip
  Y. Chen et al PRL 121 233602 (2018)



- Generating X-ray vortices
  J. C.T. Lee et al. Nature Photonics **13** 205 (2019)
- OAM X-ray beams (= phase structured beams) with >1 ħ/photon
- New spectroscopy, scattering, imaging modalities

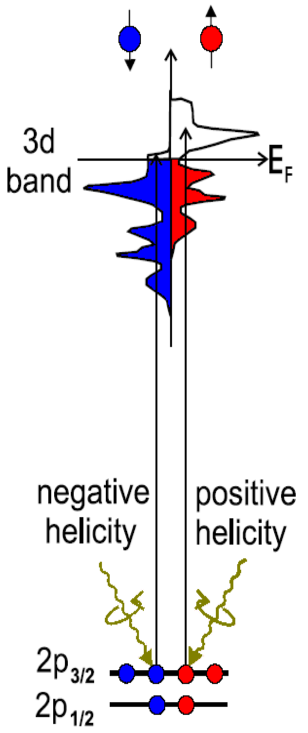




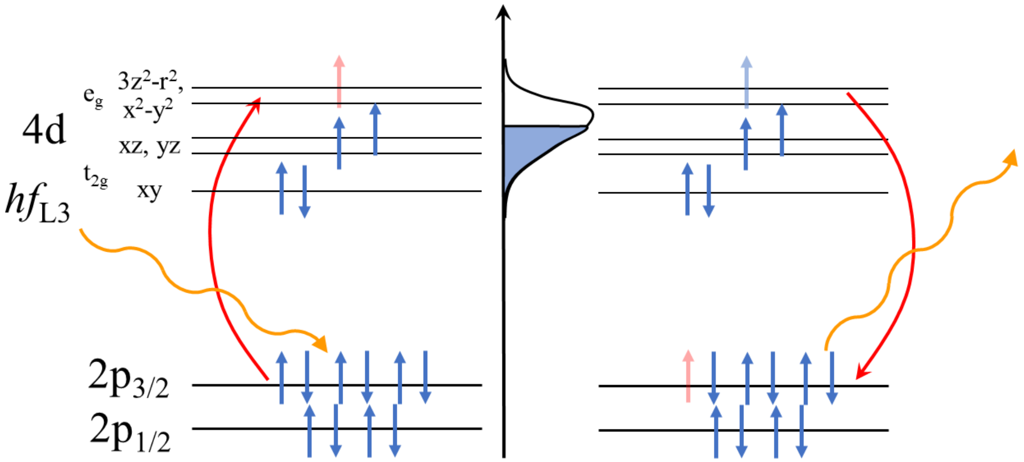
A. Sakdinawat et al., Optics Letters ***32 2635 (2007)***

X-ray spectroscopy, scattering, microscopy with OAM beams

- How to distinguish quantum behavior from classical effects in complex materials?
- Entanglement mapping?



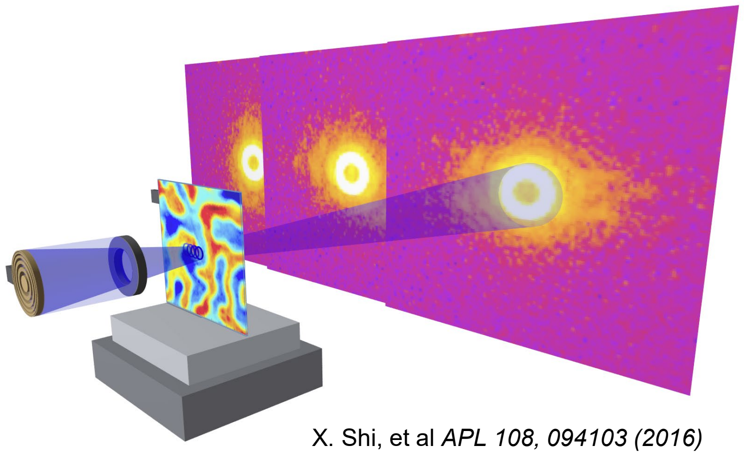
$$\mu(E) \propto |M_{if}|^2 \rho(E)$$



$$f = f^0 + f' + if''$$

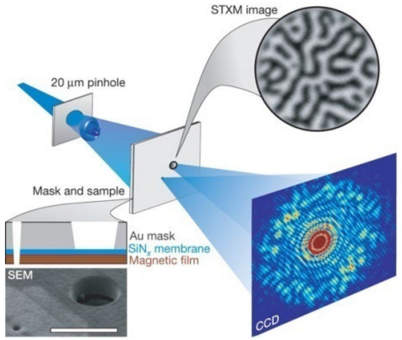
https://www.diamond.ac.uk/Instruments/Magnetic-Materials/I16/Techniques/rxs.html

X-ray ptychography



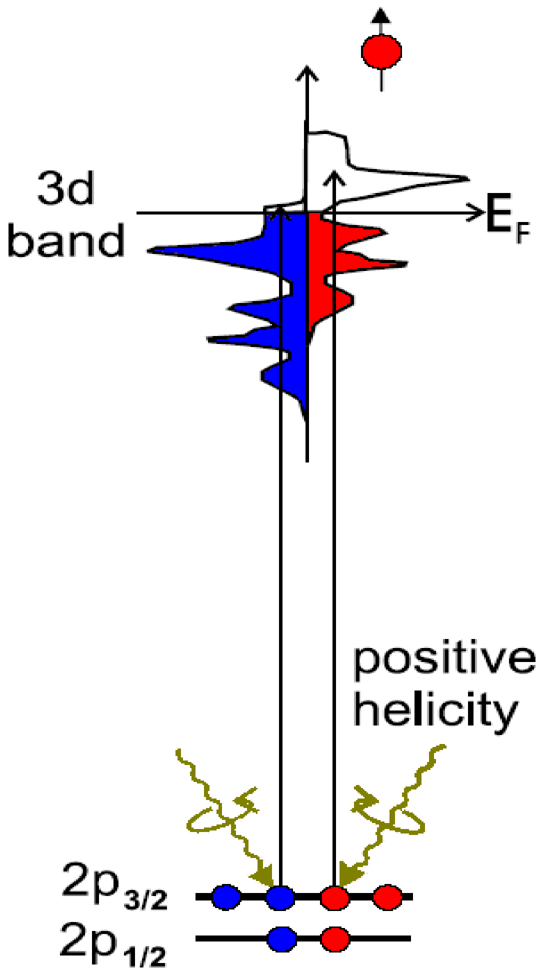
X. Shi, et al *APL 108, 094103 (2016)*

X-ray holography



S. Eisebitt, et al. Nature ***432****, 885 (2004)*

X-ray absorption spectroscopy - basics



J. Stöhr, H.C. Siegmann, Magnetism (Springer)

Fermi's Golden Rule

$$\mu(E) \propto |M_{if}|^2 \rho(E)$$

Conservation Laws

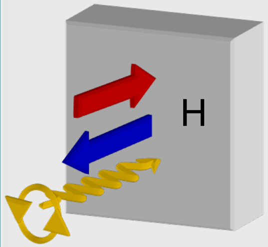
- Energy: $E=E_\gamma-E_B$
- Linear momentum: (for small e-energies) in direction of E-vector
- Orbital momentum (symmetry)

TODAY:
since one photon carries only 1 ħ (SAM), dipolar approximation is justified
Δl=+1 (transition Δl =-1 weak)

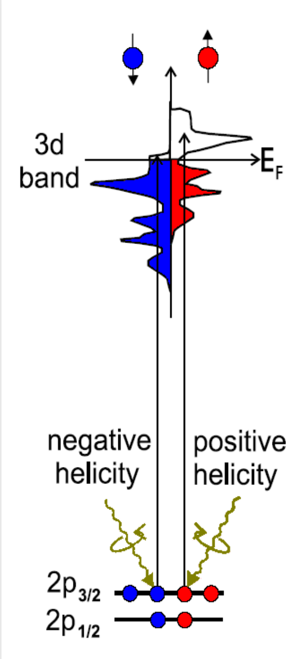
Next generation SR facilities
"Diffraction-Limited Storage Ring (DLSR)"
Orbital Angular Momentum (OAM) X-ray beams (= phase structured beams) with >1 ħ/photon !!!

X-rays as a powerful [element-specific] tool for magnetism

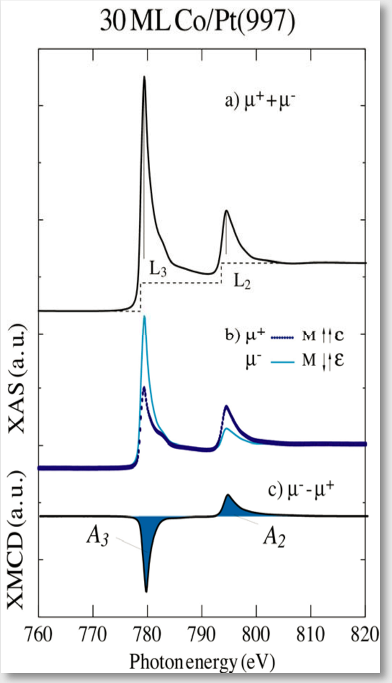
X-ray Magnetic Circular Dichroism (XMCD)



circularly polarized



J. Stöhr, H.C. Siegmann, Magnetism (Springer)

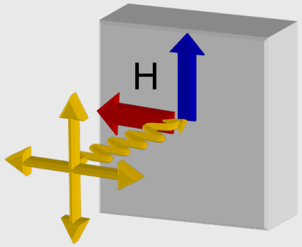


- $I_{XMCD} = I_{\uparrow\downarrow} - I_{\uparrow\uparrow} \propto \langle m \rangle$
  $\langle m \rangle$ = EV of atomic magnetic moment
- quantitative information
- **Ferro-** and **Ferri**magnets

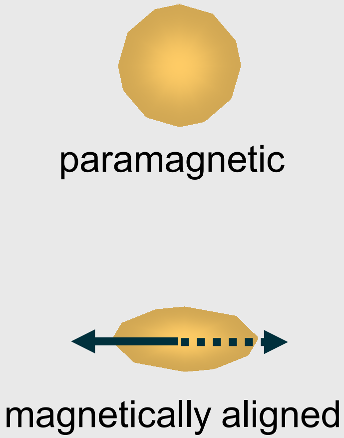
Spin moment
$m_S = -3 n_h \mu_B (A_3 - 2A_2) + m_T$
Orbital moment
$m_L = -2 n_h \mu_B (A_3 + A_2)$

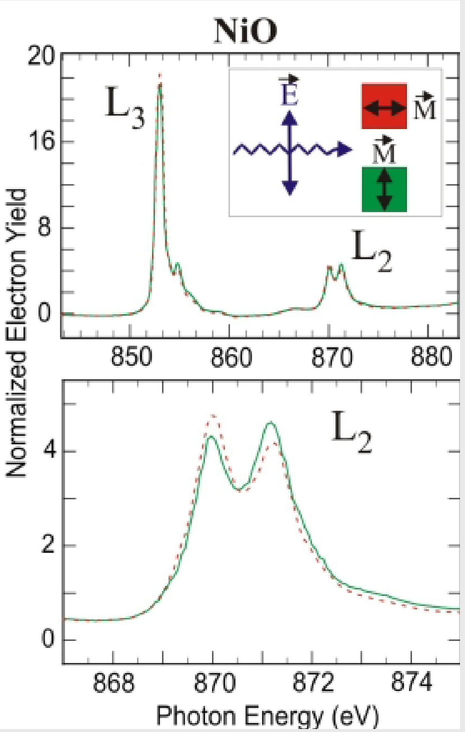
B. Thole et al., PRL 68, 1943 (1992)
P. Carra et al., PRL 70, 694 (1993)

X-ray magnetic linear dichroism (XMLD)



linearly polarized

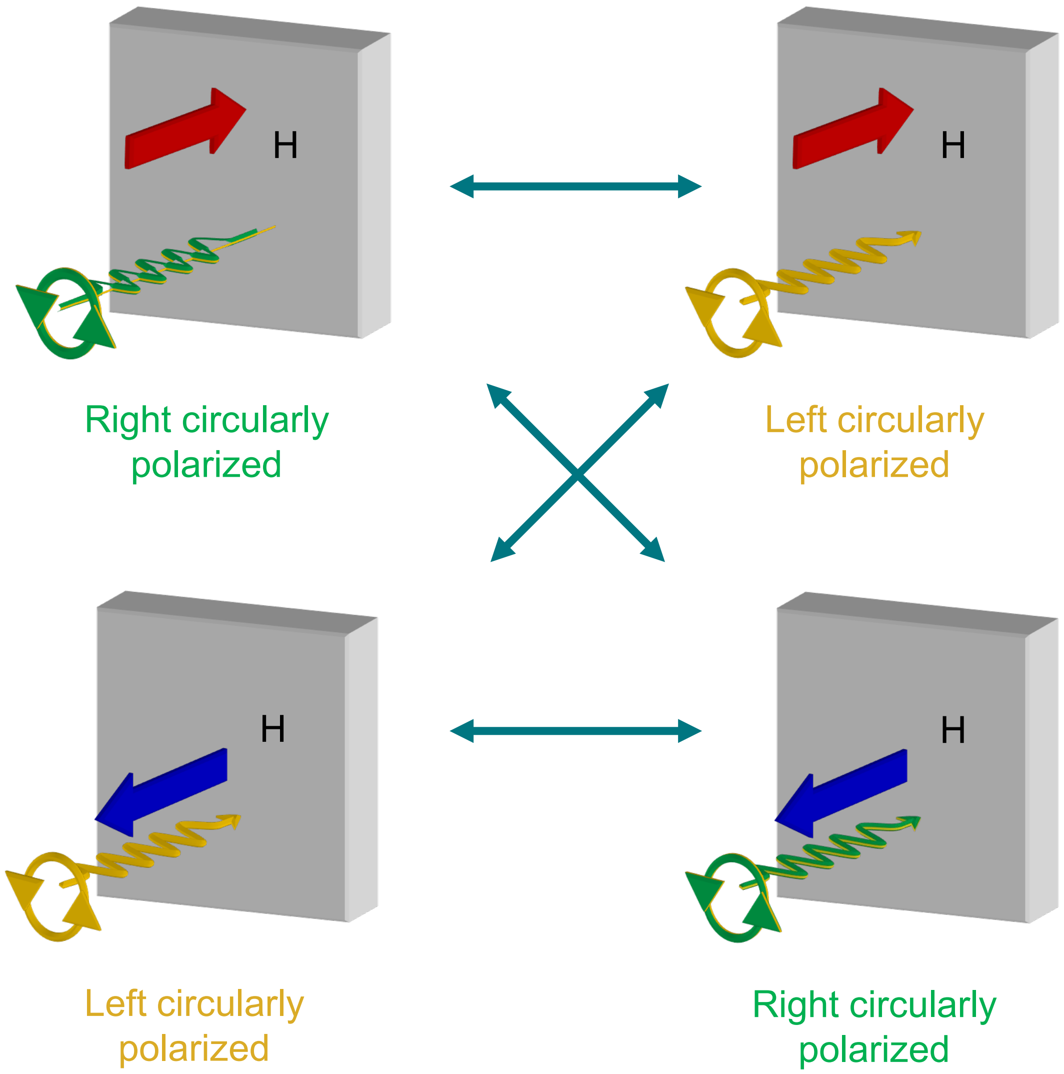




- XMLD arises from non-spherical charge distribution
- $I_{XMLD} = I_{\parallel} - I_{\perp} \propto \langle m^2 \rangle$
  $\langle m^2 \rangle$ = EV of square of atomic magnetic moment
- **Antiferro**magnets (to some degree also **Ferro**- and **Ferri**magnets)

Courtesy E. Arenholz and https://www-ssrl.slac.stanford.edu/stohr/xmcd.htm

Beyond XMCD with OAM beams



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Outlook: Open Questions and Next Steps

Skyrmion Qubits

Integrating skyrmions with quantum computing architectures. Scalability remains a key challenge.

Material Engineering

Designing materials for stable quantum skyrmion states. Requires precision at atomic scales.

Real-Time Measurements

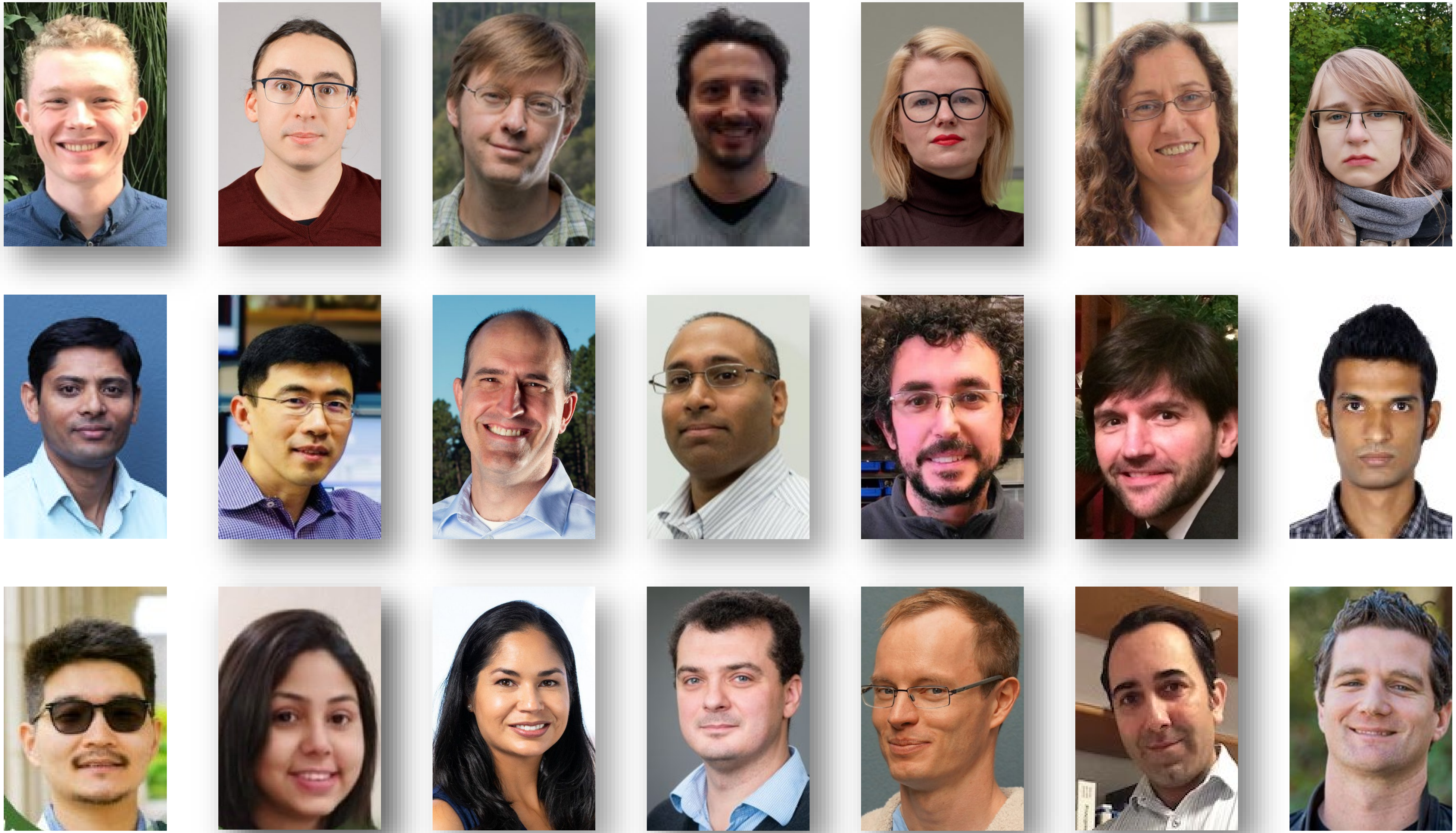
Developing techniques for tracking skyrmion quantum dynamics. Higher temporal resolution needed.

Quantum Networks

Exploring skyrmion entanglement for quantum communication. Could enable novel quantum protocols.

Acknowledgements

BERKELEY LAB



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