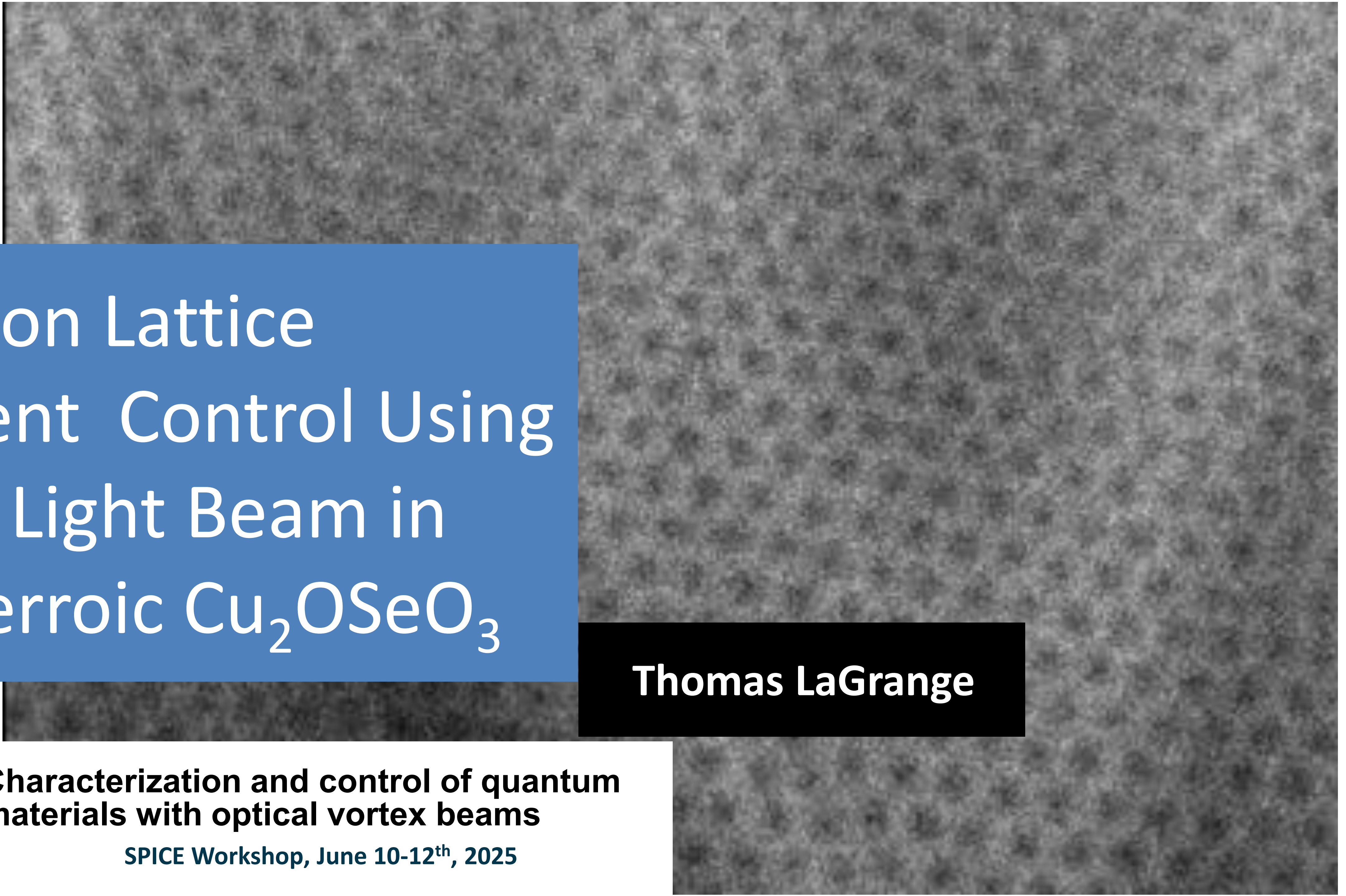

# Skyrmion Lattice Coherent Control Using Vortex Light Beam in Multiferroic $Cu_2OSeO_3$

Thomas LaGrange

EPFL
LUMES
École polytechnique fédérale de Lausanne

**Characterization and control of quantum materials with optical vortex beams**

SPICE Workshop, June 10-12th, 2025

# Acknowledgements

EPFL







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Post Docs: Phoebe Tengdin, Ivan Madan,Alexey Sapozhnik,

Fabrizio Carbone

Collaboration with EFPL:

- Henrik Ronnow, Dirk Grundler, P. Huang, A. J. Kruchkov, T. Schonenberger, P. Che, M. Cantoni, A. Magrez,

External Collaborators

- Achim Rosch, University of Koln
- Angel Rubio, Hamburg

# Coherent Control Experiments at EFPL

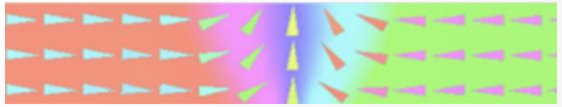
EPFL

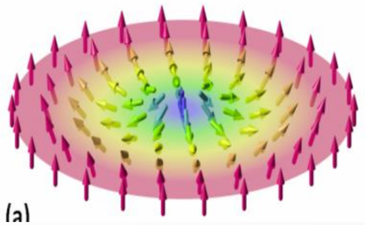
## Motivation

We want to explore how low-energy excitations couple to coherent-resonant magnetic dynamics

**Magnons**

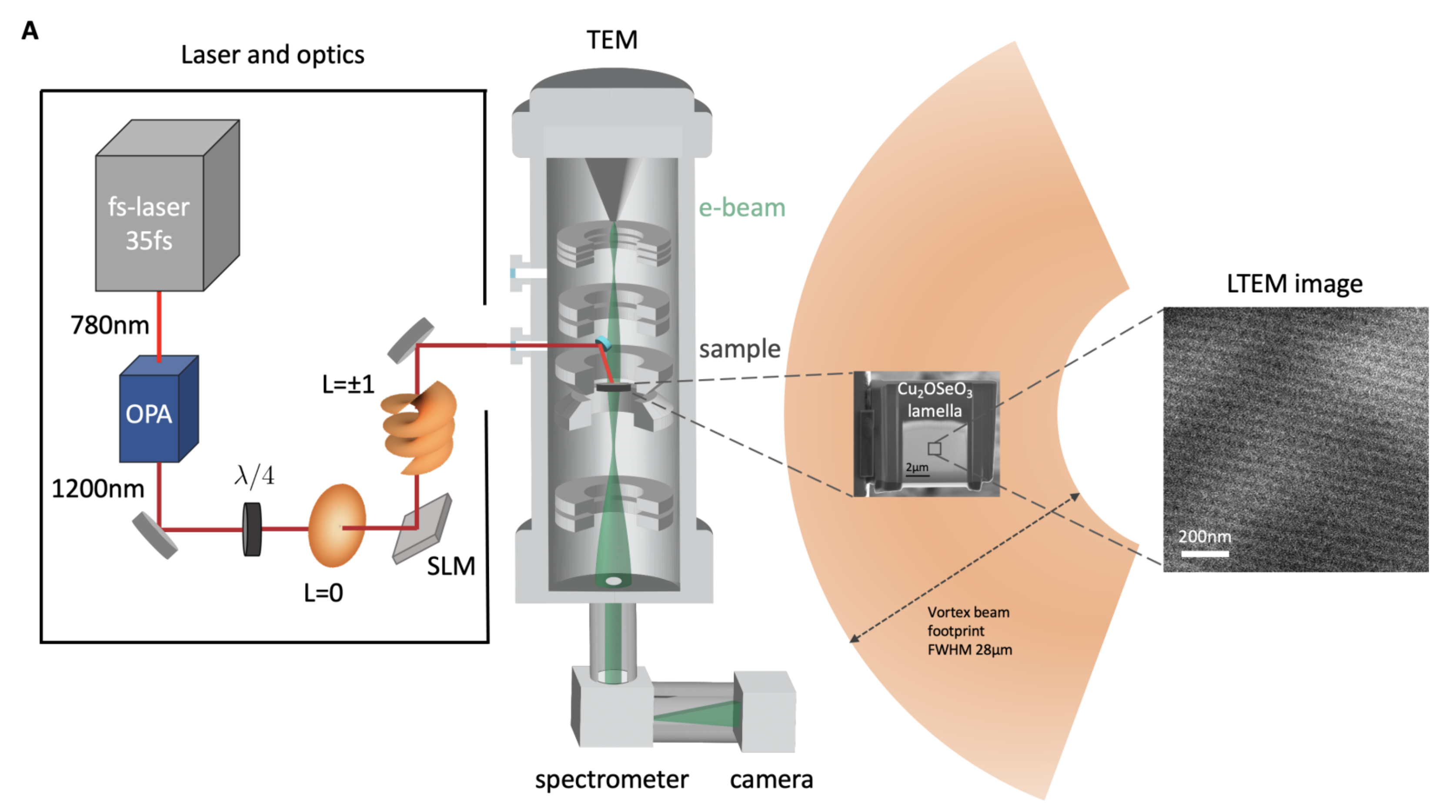
**Domain walls**






**Topological phases and textures**

## TEM Setup





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# Outline

- What are Magnetic Skrymions?
  - Magnetic Phase Diagrams
  - Topology vs. Thermodynamics
  - $Cu_2OSeO_3$ (COSO)
  - Lorentz TEM imaging of Skyrmions
- Coherent Control using Circular Polarized Light
  - Rotation from single pulse pumping
  - Rotation from two pulse pumping
  - Inverse Faraday Effect
- Coherent control from Vortex Light (OAM beams)
  - 1200nm light
  - 1030nm light

PHYSICAL REVIEW X **12,** 041030 (2022)

Featured in Physics

**Imaging the Ultrafast Coherent Control of a Skyrmion Crystal**

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Exotic magnetic textures emerging from the subtle interplay between thermodynamic and topological fluctuation have attracted intense interest due to their potential applications in spintronic devices. Recent advances in electron microscopy enable the imaging of random photogenerated individual skyrmions. However, their deterministic and dynamical manipulation is hampered by the chaotic nature of such fluctuations and the intrinsically irreversible switching between different minima in the magnetic energy landscape. Here, we demonstrate a method to coherently control the rotation of a skyrmion crystal by discrete amounts at speeds which are much faster than previously observed. By employing circularly polarized femtosecond laser pulses with an energy below the band gap of the Mott insulator $Cu_2OSeO_3$, we excite a collective magnon mode via the inverse Faraday effect. This triggers coherent magnetic oscillations that directly control the rotation of a skyrmion crystal imaged by cryo-Lorentz transmission electron microscopy. The manipulation of topological order via ultrafast laser pulses shown here can be used to engineer fast spin-based logical devices.

DOI: 10.1103/PhysRevX.12.041030

Subject Areas: Condensed Matter Physics, Magnetism Spintronics



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# What are Magnetic Skyrmions?

**The Dzyaloshinskii-Moriya Interaction (DMI) favors the** canting of antiparallel spins. Promotes weak ferromagnetism within an antiferromagnetic background

$$\mathcal{H}_{\mathrm{DMI}} = \sum_{i,j} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j),$$

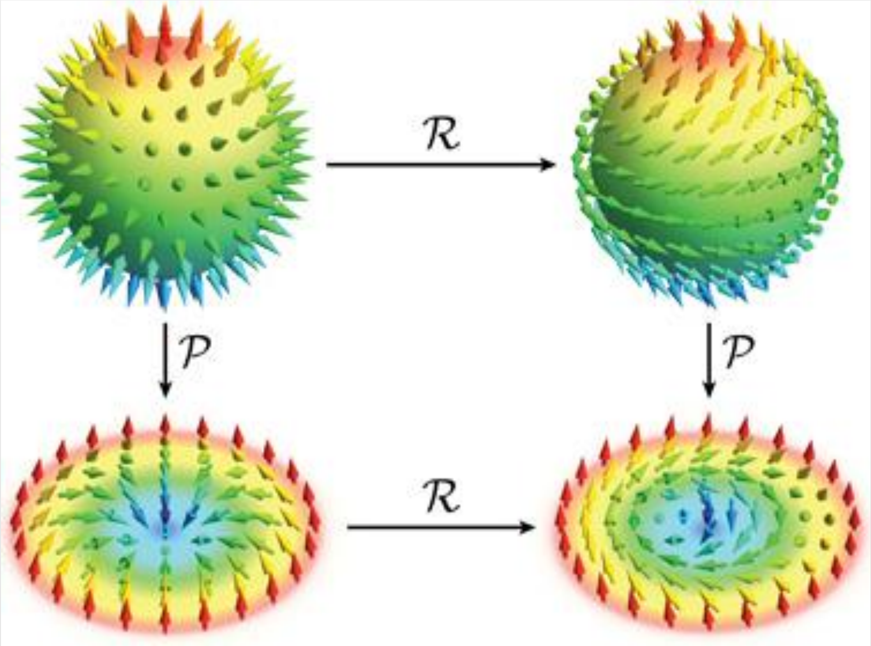
$$\mathcal{H}_{\mathrm{DMI}}^{\mathrm{Bloch}} = -D \sum_{i} \mathbf{S}_i \times \mathbf{S}_{i+\hat{x}} \cdot \hat{x} + \mathbf{S}_i \times \mathbf{S}_{i+\hat{y}} \cdot \hat{y},$$

$$\mathcal{H}_{\mathrm{DMI}}^{\mathrm{Néel}} = -D \sum_{i} \mathbf{S}_i \times \mathbf{S}_{i+\hat{x}} \cdot \hat{y} - \mathbf{S}_i \times \mathbf{S}_{i+\hat{y}} \cdot \hat{x},$$

**Topological charge of a skyrmion:**

$$Q = \frac{1}{4\pi} \int dxdy\ \mathbf{m} \cdot \left( \frac{\partial \mathbf{m}}{\partial x} \times \frac{\partial \mathbf{m}}{\partial y} \right),$$

$$\mathbf{m}(x, y) = \mathbf{M}(x, y) / |\mathbf{M}|$$




Néel skyrmion Bloch skyrmion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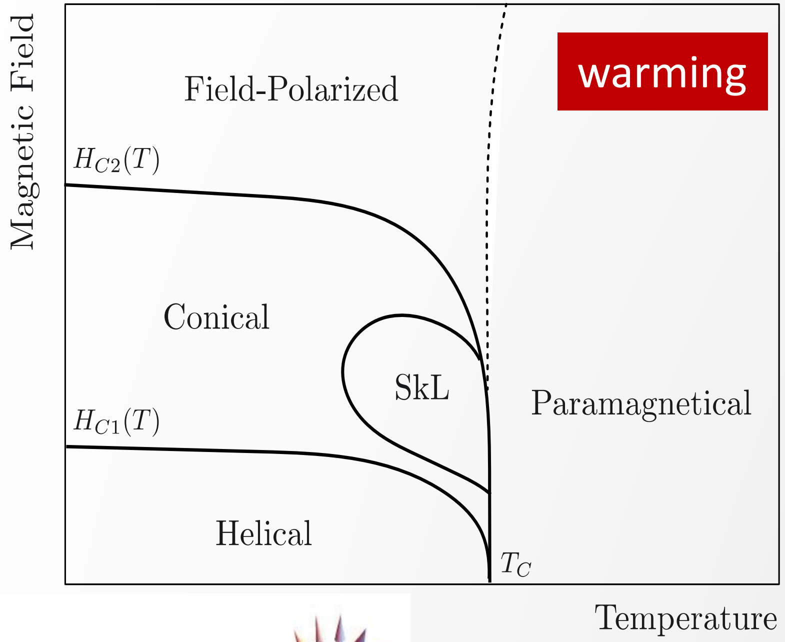
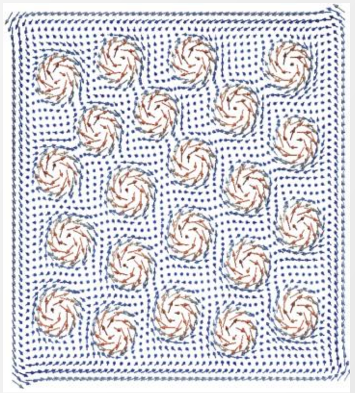
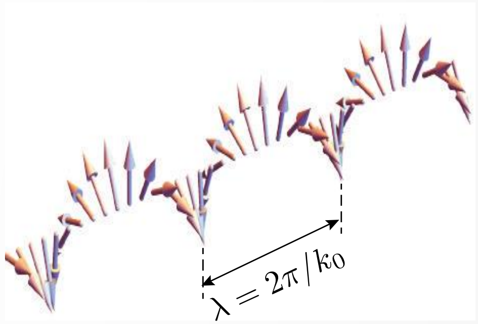
# Skrymionic Materials Magnetic Phase Diagrams

## In crystals with broken inversion symmetry and in the presence of spin-orbit interaction

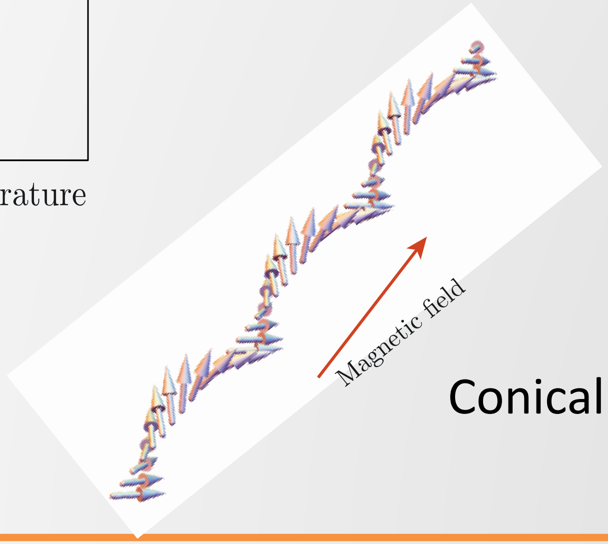



Skyrmion lattice

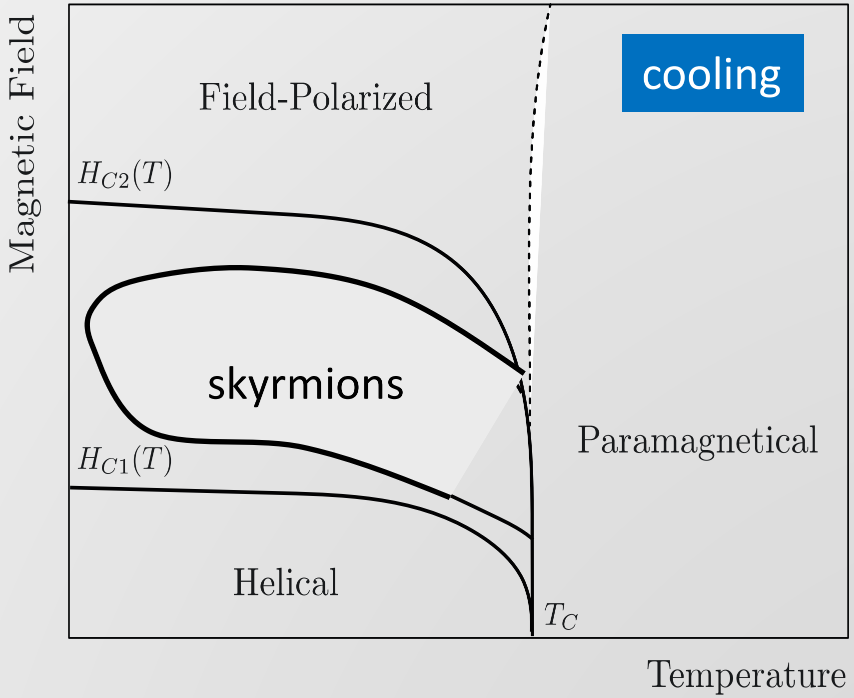





Helical




Conical




Hysteretic phase diagram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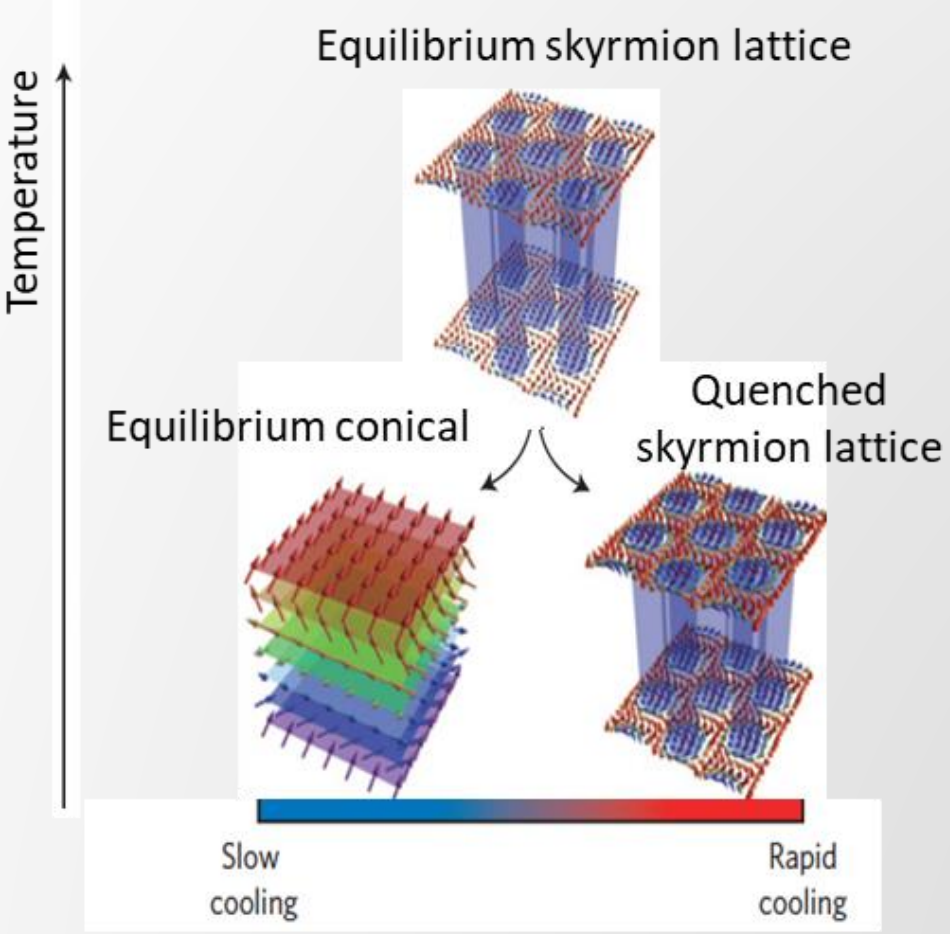
# Topology vs. Thermodynamics

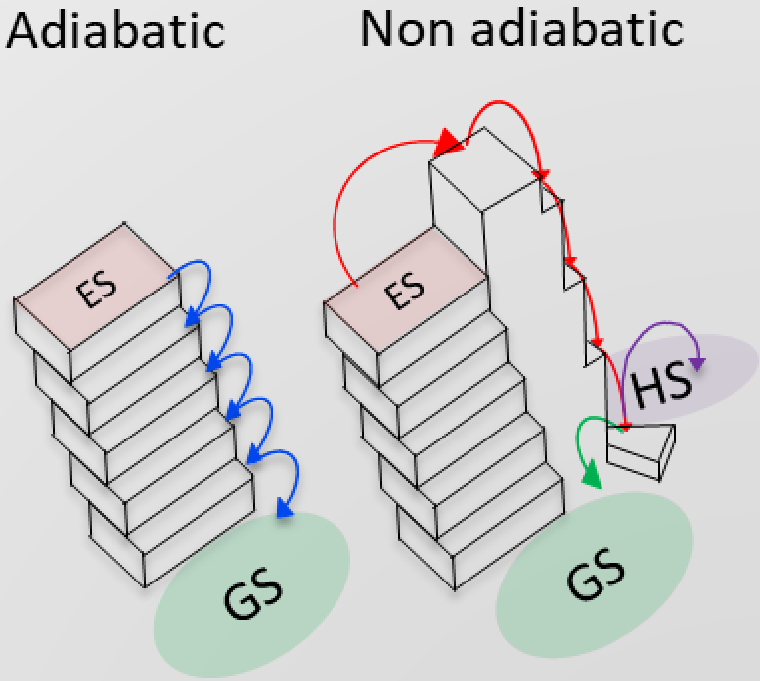
Topological magnetization patterns determined by dynamical interplay between

- Topological protection
- Thermodynamical fluctuations
- Out of equilibrium: The Mpemba effect

*"The Mpemba effect runs in reverse"*
A. Piccone, **Physics Today** 2022




Nat. Phys. 12, 62 (2015)



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# Skyrmions in Multiferroic $Cu_2OSeO_3$ (COSO)

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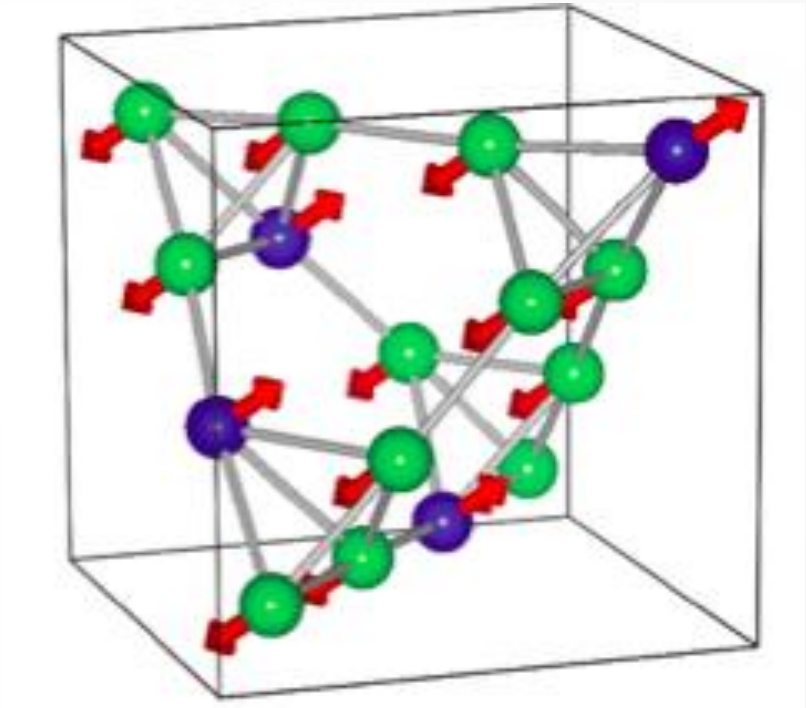
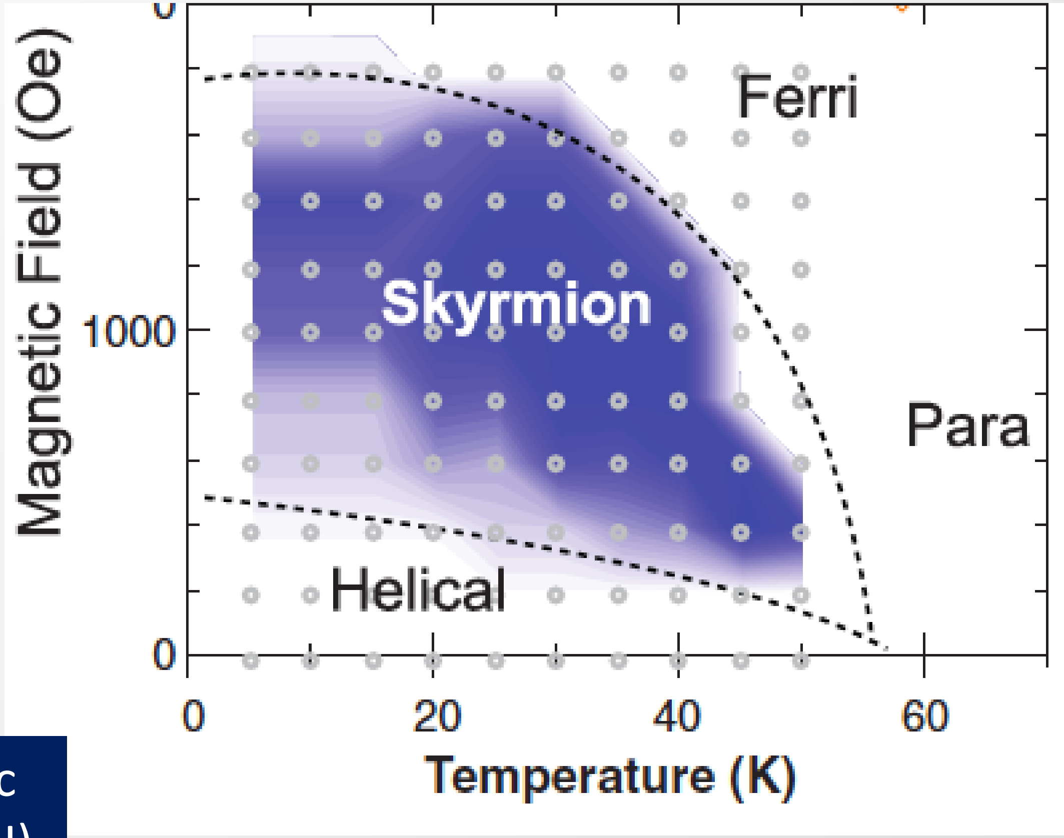
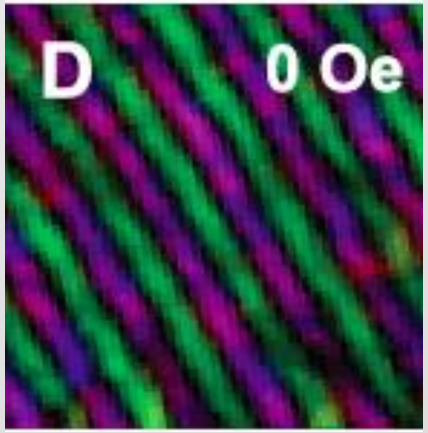


- COSO unit cell is noncentrosymmetric with broken inversion symmetry (DMI)
- There are two chirality, giving different Skrymion polarities

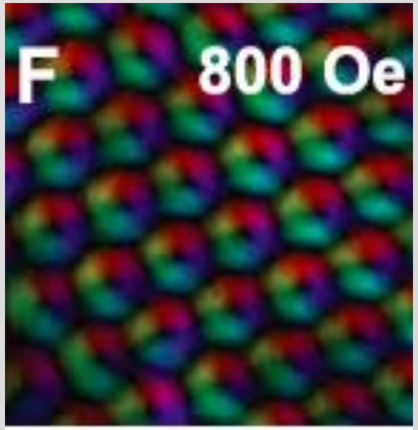



**Observation of Skyrmions in a Multiferroic Material**
S. Seki *et al.*
*Science* **336**, 198 (2012);

Helical Phase




SkL Phase



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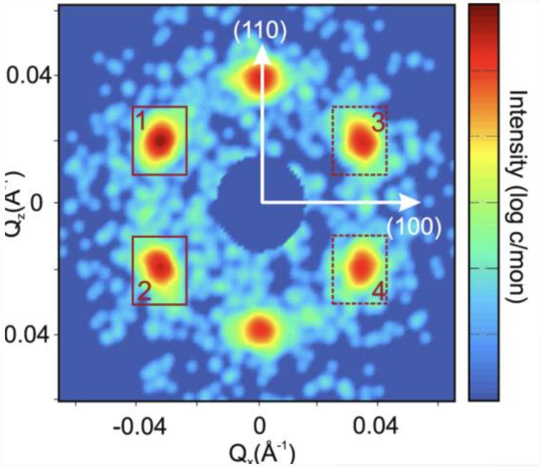
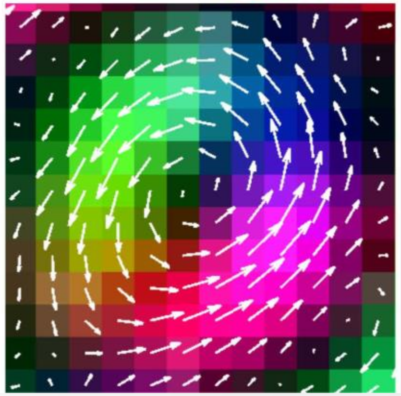
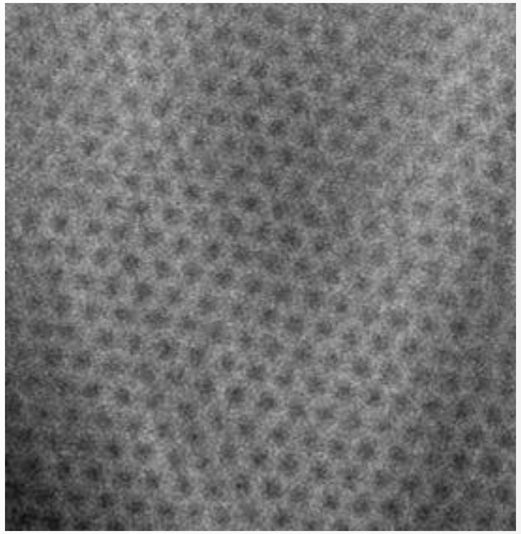
# Lorentz TEM images of Skrymions

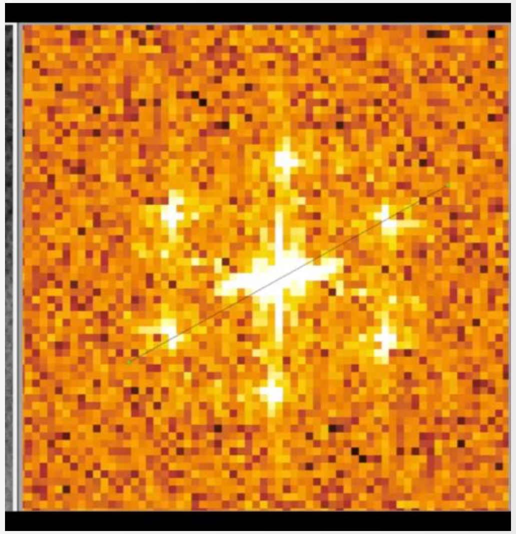
Reciprocal space:
neutron scattering

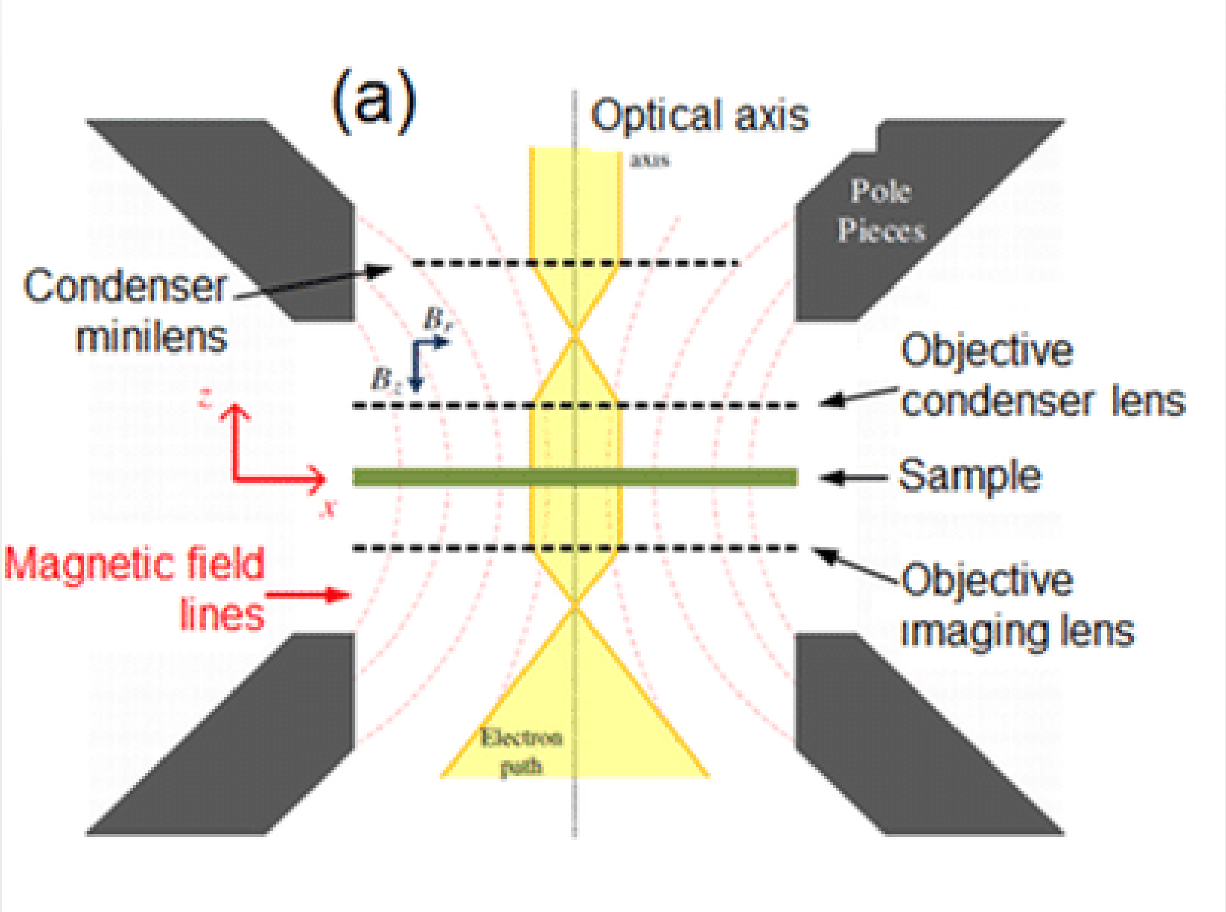



Real Space whirling $\vec{B}$










Out-of-focus observation
Interference contrast that distinguishes the intersection between domains or changing spin states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# Coherent Control of COSO skrymion lattice

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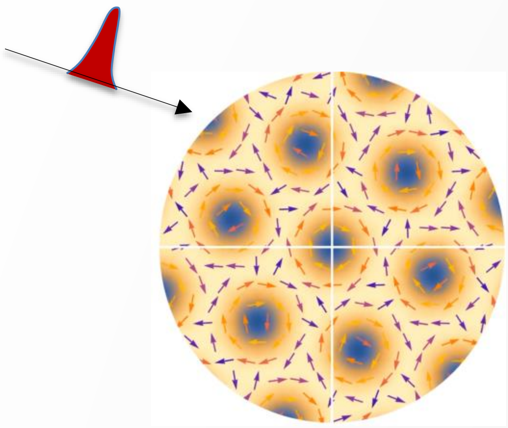
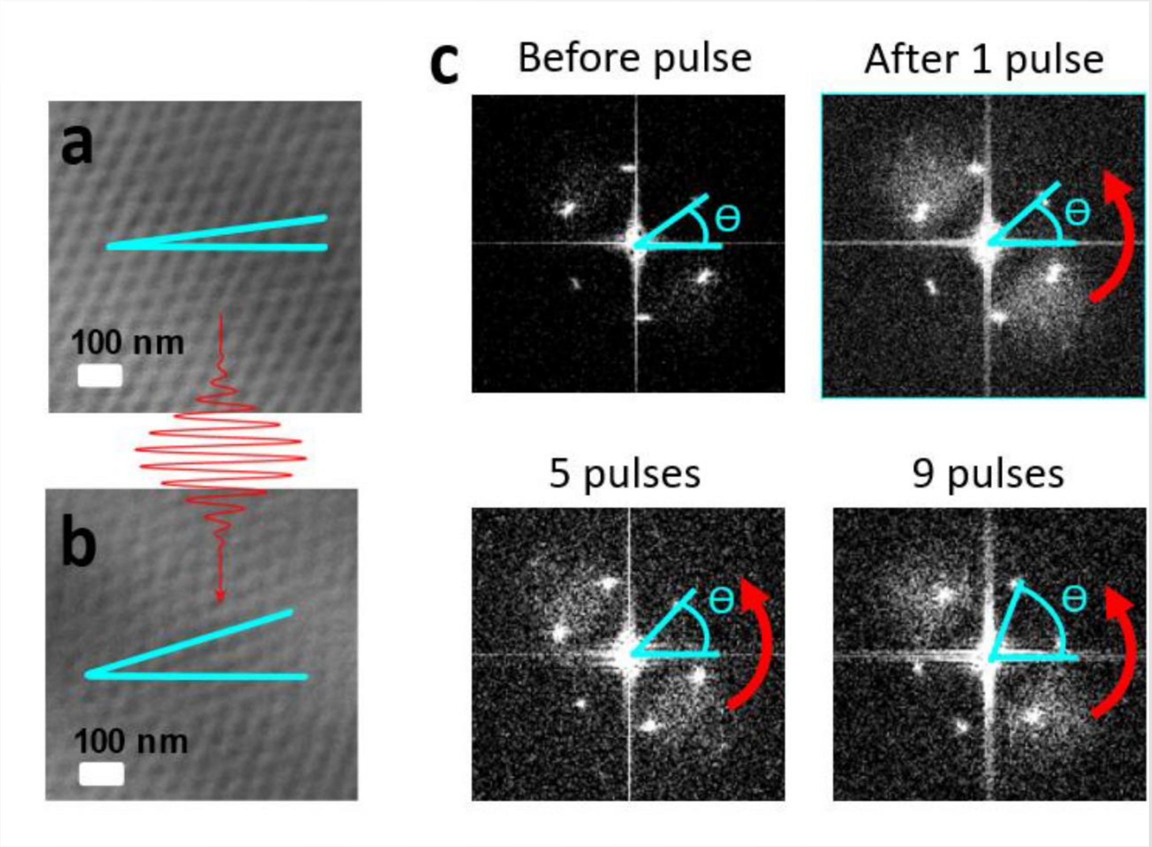
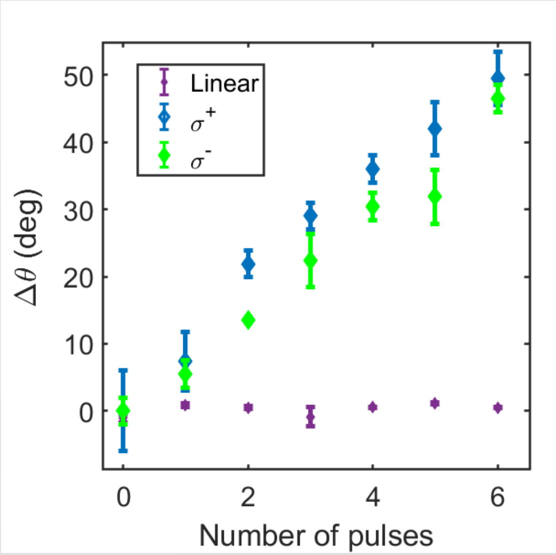
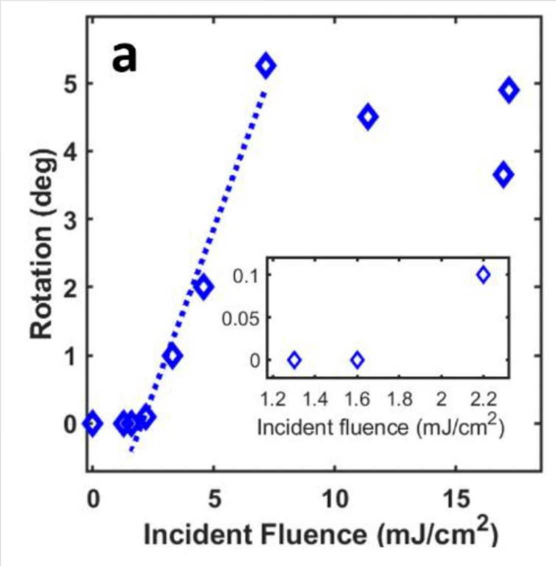


35 fs circularly polarized IR (1200nm) laser pulse induces a rotation




How fast does it rotate? What causes it?

- First hint: rotation has a threshold 2mJ/$cm^2$



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# Coherent Control of COSO skrymion lattice

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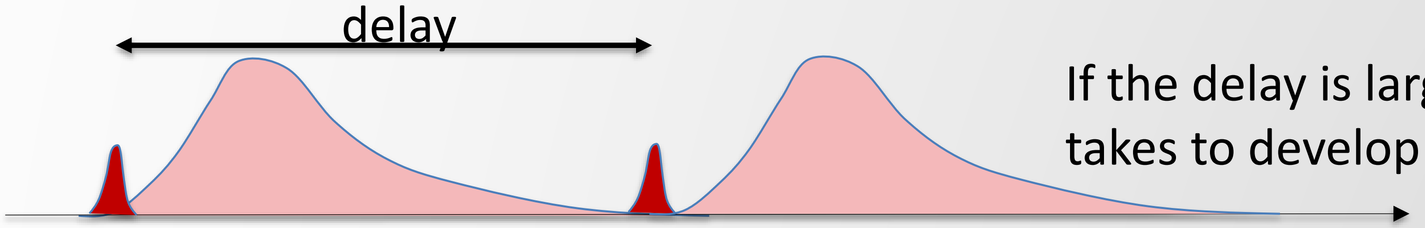
- Is it a thermal effect? Above a certain temperature jump the lattice rotates?
  - Happens on a slow time-scale (ns to $\mu$s) ?
  - Should not depend on polarization (CuOSeO is optically isotropic)
  - Threshold fluence should be higher for wavelengths corresponding to lower absorption coefficient

Rotation is an **IRREVERSIBLE** effect (can't use Ultrafast LTEM imaging approaches)

Two pump pulses, CW imaging with LTEM




If the delay is larger than the time heat takes to develop nothing happens

Each pulse is **half** what is needed to induce rotation

time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# Coherent control of skyrmions

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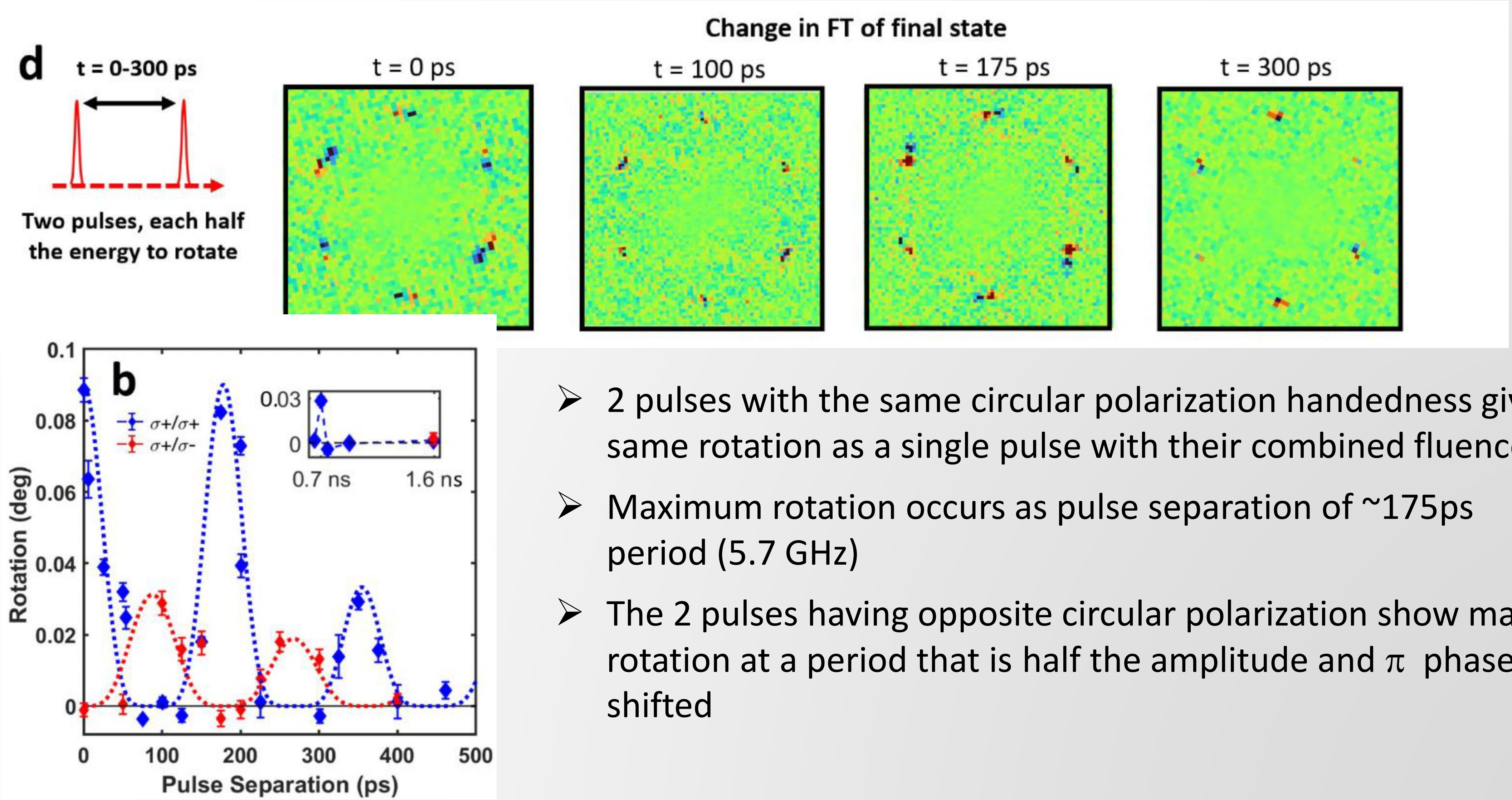


- 2 pulses with the same circular polarization handedness give same rotation as a single pulse with their combined fluence.
- Maximum rotation occurs as pulse separation of ~175ps period (5.7 GHz)
- The 2 pulses having opposite circular polarization show max rotation at a period that is half the amplitude and $\pi$ phase shifted


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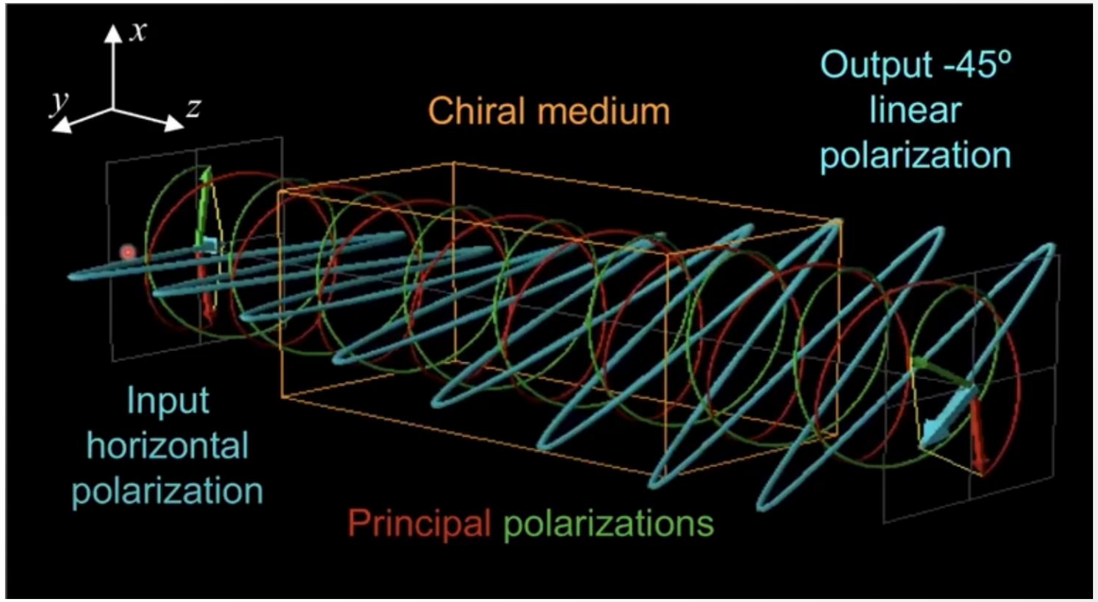
# Rotation is caused by inverse faraday effect




**Optical activity:**
In a chiral medium (structural):
Linearly polarized light can be decomposed in the two circular polarizations

Circular polarizations are dephased.
The result is a rotation of polarization

**Faraday effect:**
This effect can be induced in a material by applying a magnetic field to a magneto-optical material:

Electrons in a magnetic field follow looped paths. Circularly polarized light with the same handedness as the current loops experiences a different refractive index than light with the opposite handedness.

**Inverse Faraday effect**

Absorption of circular polarized light can induce current loops with a preferred hardiness resulting in a net magnetic field generated in the material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# Calculations indicated the IFE cannot "affectively" explain the obervastions

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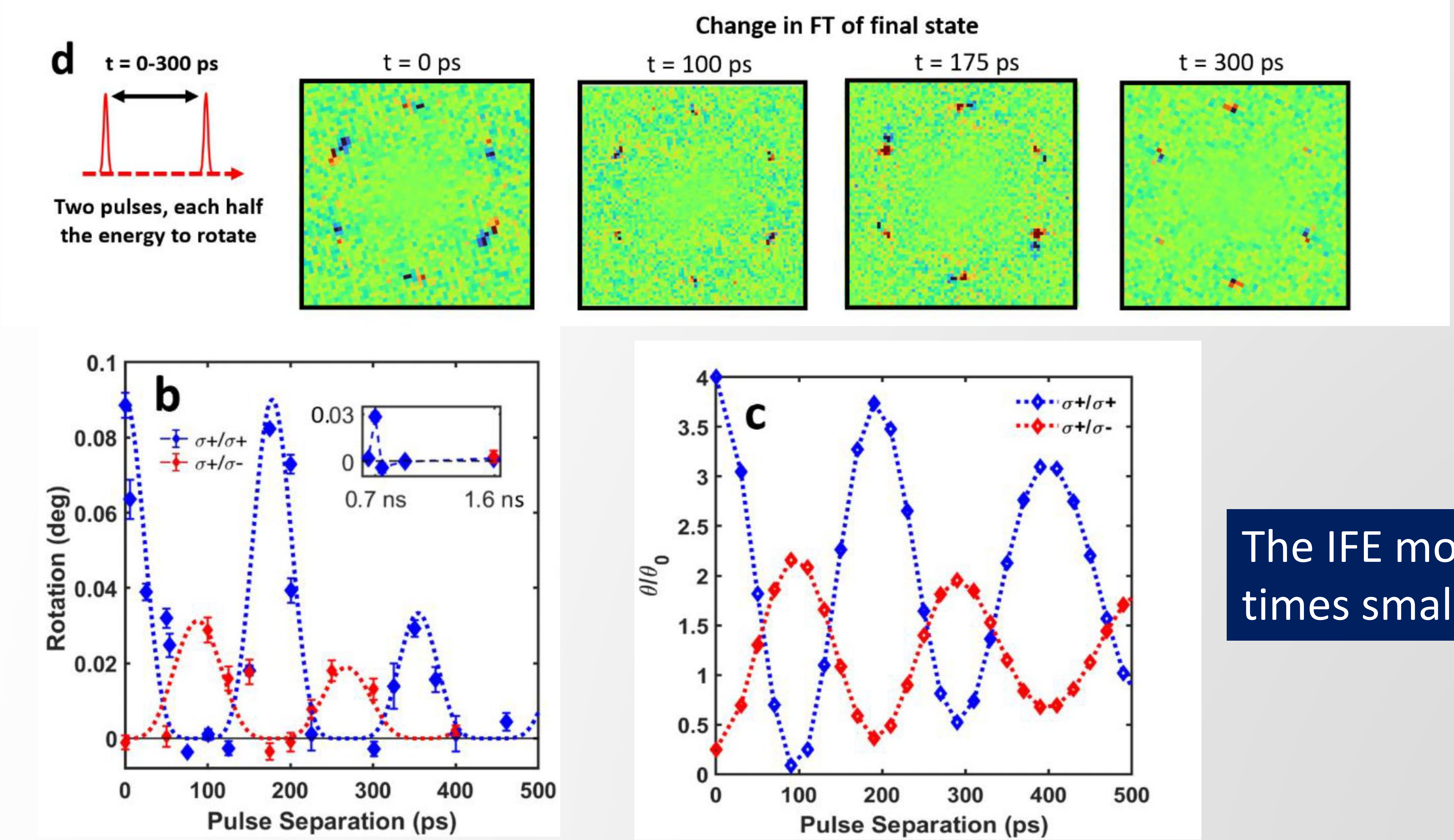


The IFE models suggest $10^4$ times smaller rotation…..


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# Inverse-Faraday induced coherent magnons

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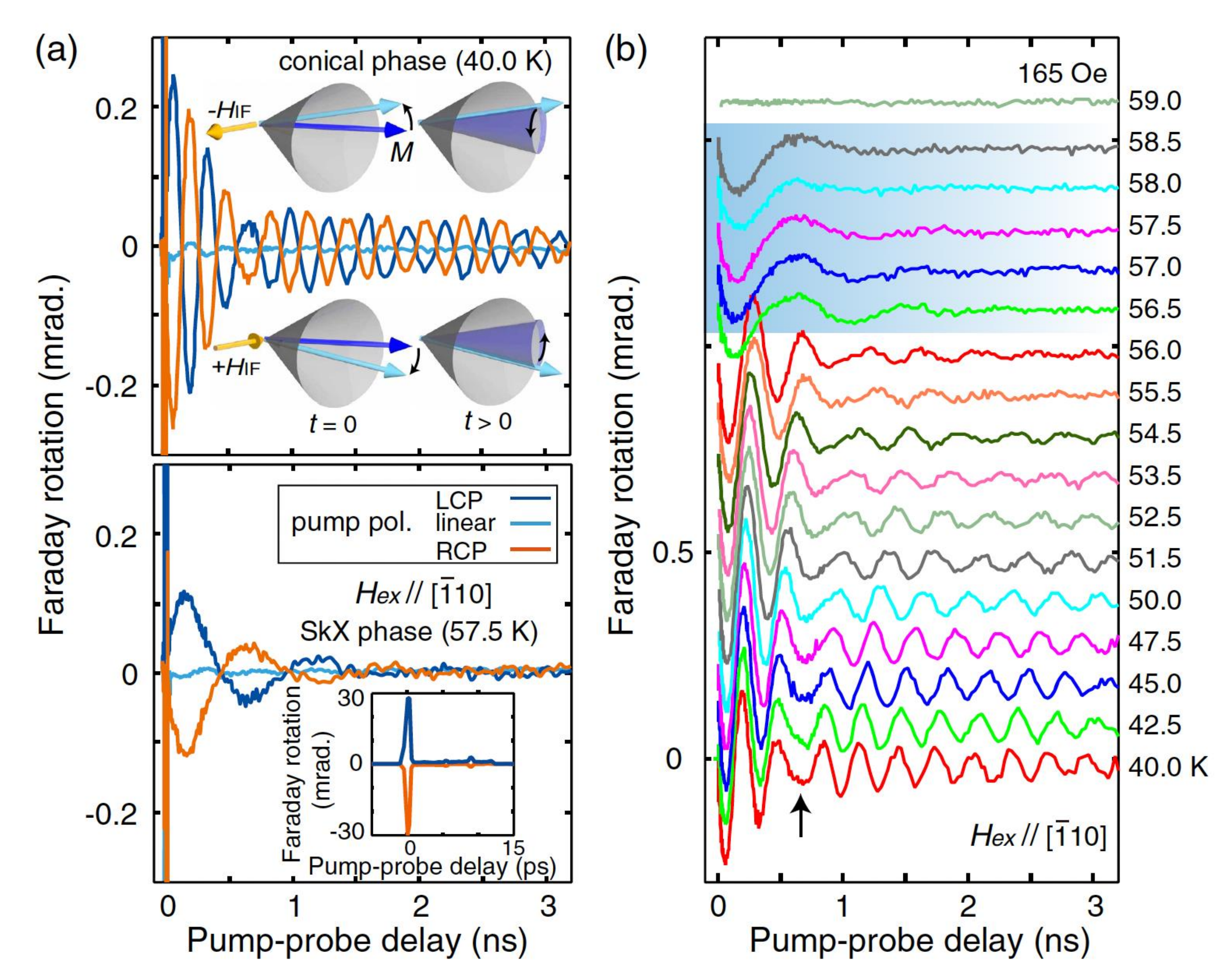


- IR pump, induces coherent oscillations of the magnetization via inverse Faraday effect.
- Oscillations are vibration modes of the SKL
- These Breathing modes have a frequencty of ~5.5 GHz

Ultrafast MOKE on CuOSeO

Ogawa, Seki, Tokura,
Sci. Rep. (2015)


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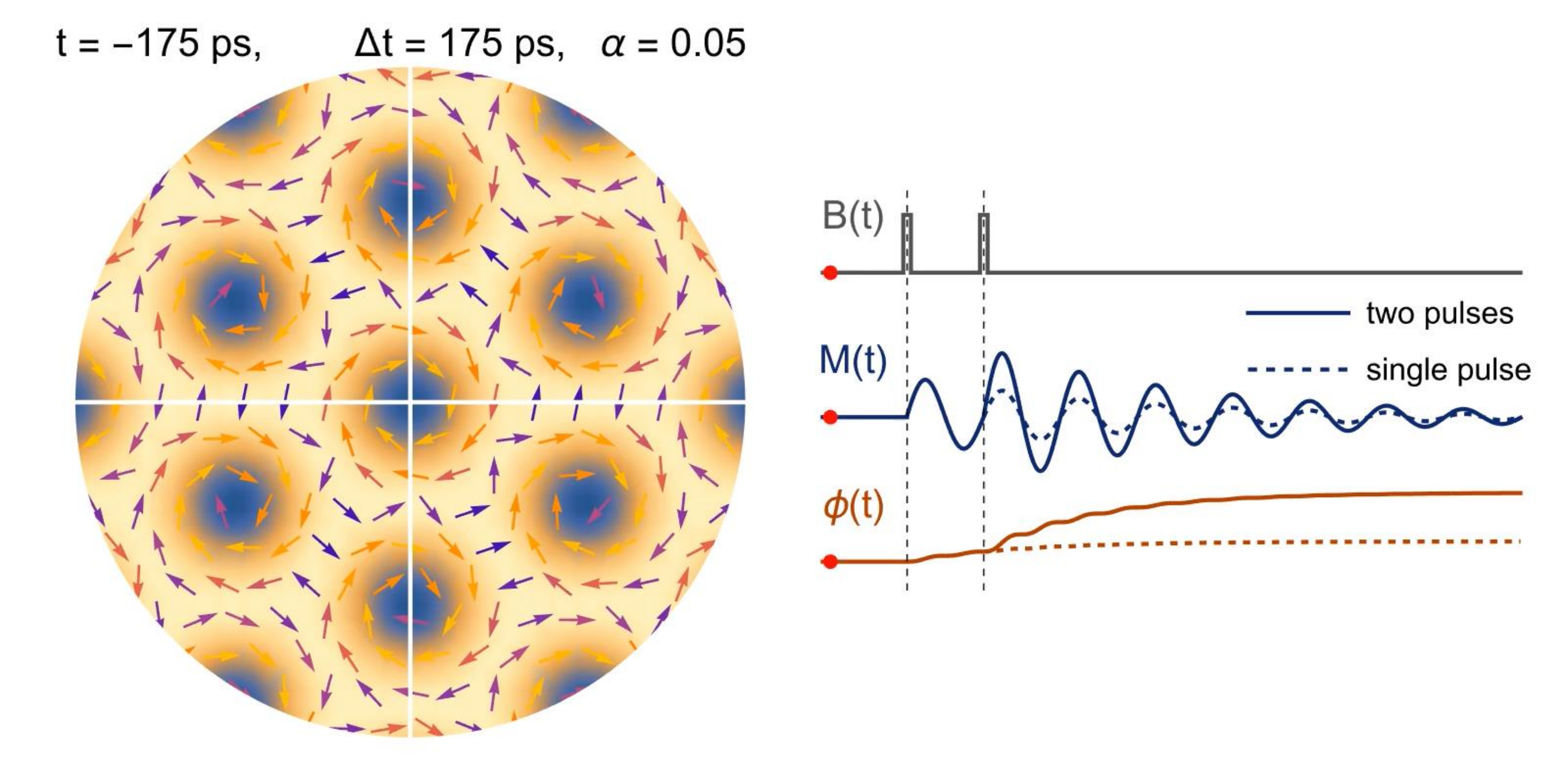
# Simulations of coherent control of skyrmions





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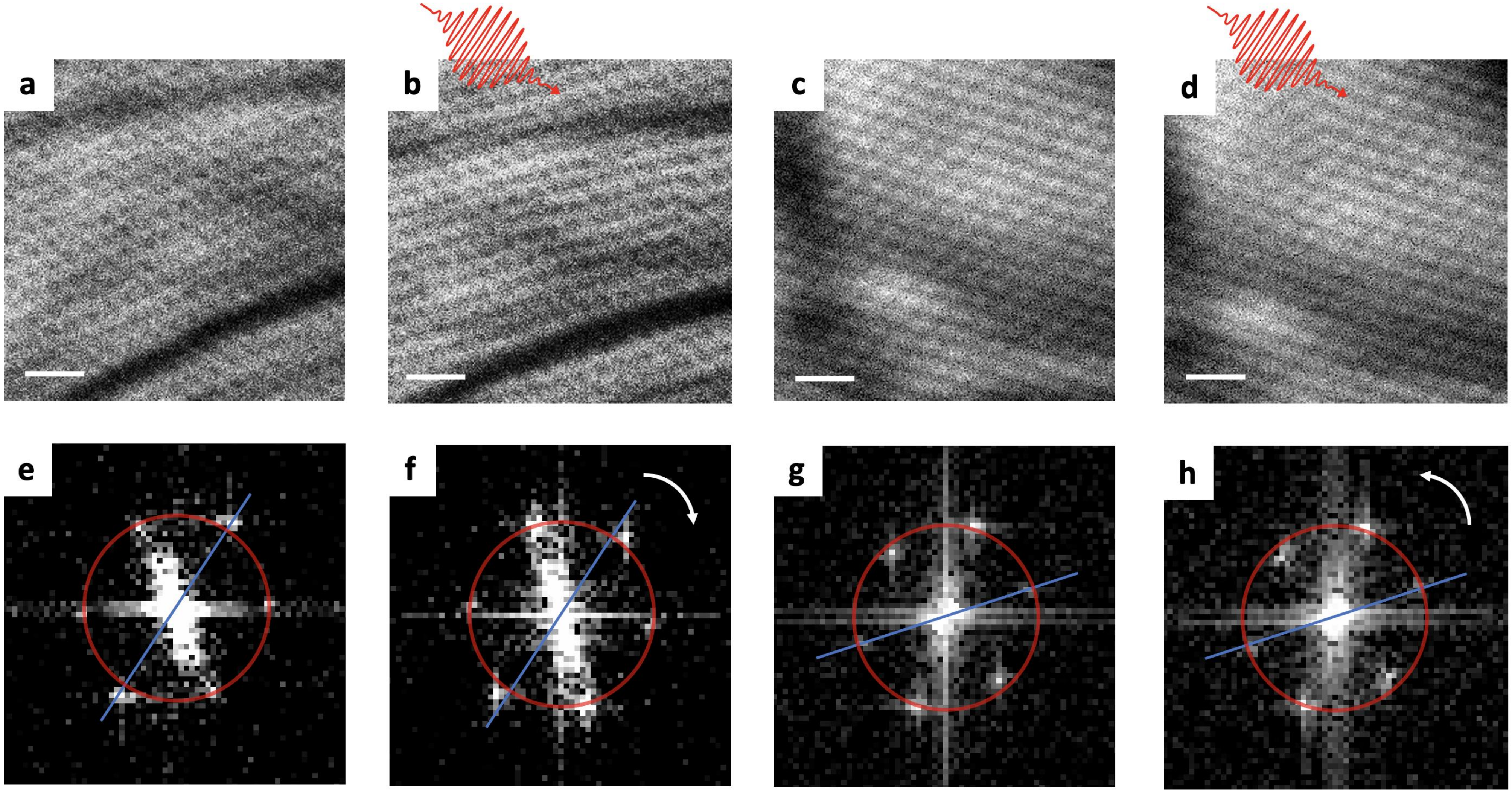
# What controls the sense of rotation?




a-b Applied field of the objective lens has positive polarity – gives clockwise rotation

c-d Applied field of the objective lens has negative polarity – gives counterclockwise rotation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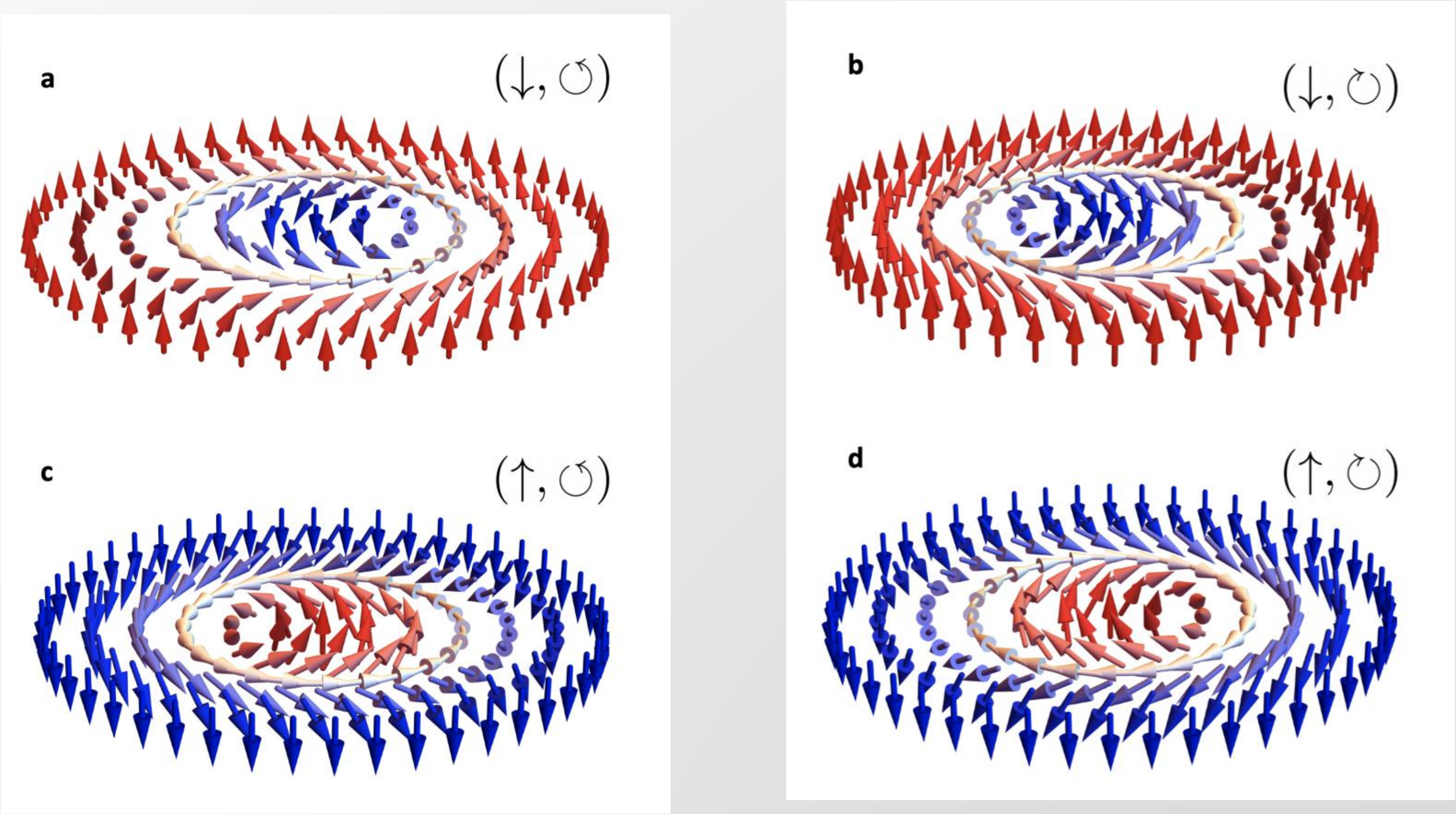
# Experiments using Vortex light for coherent control

But… What controls the sense of rotation?

- Polarity of the skyrmion?
- Chirality of COSO crystal?
- OAM topological charge?




Vortex photons CAN control the sense of rotation
Circularly polarized photons CANNOT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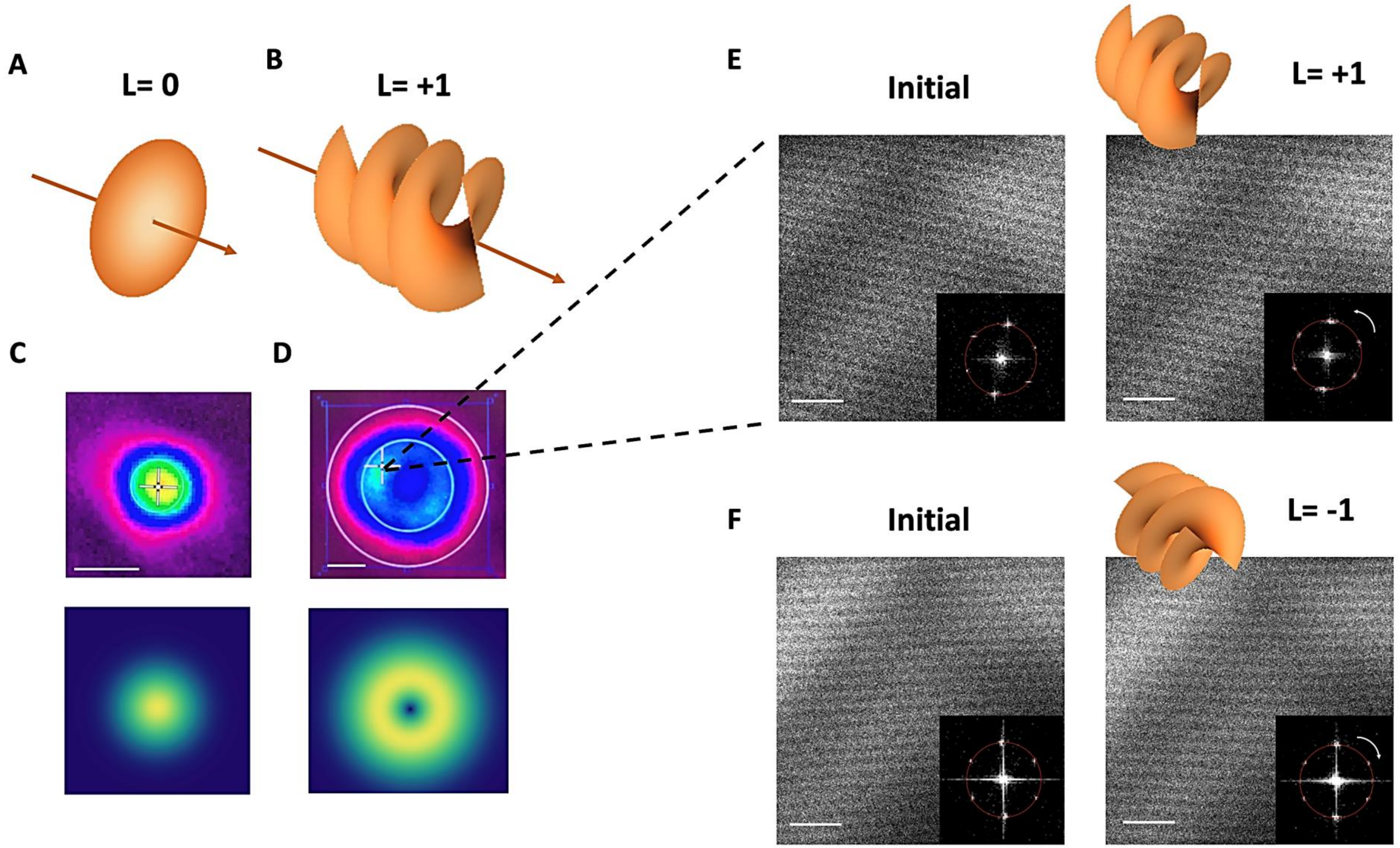
# Skyrmion Crystal Control using 35fs, 1200nm Vortex OAM beam




Counterclockwise Rotation

Clockwise Rotation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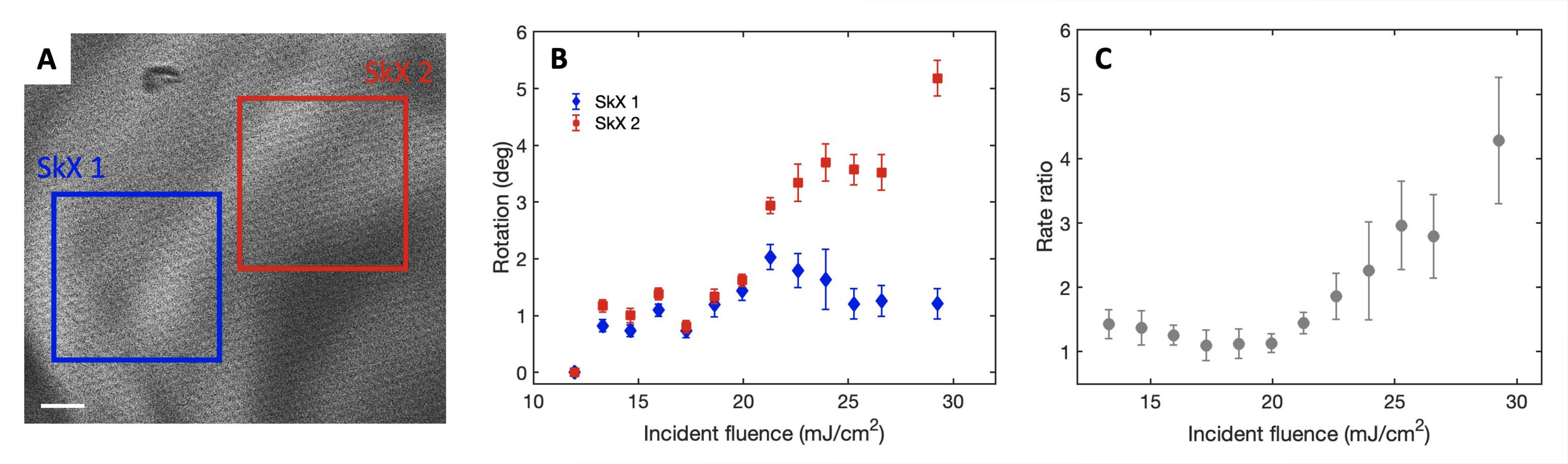
# Evidence for SkX Dynamics




- Threshold behavior + two rotating regimes
- Evidence for skyrmion crystal interactions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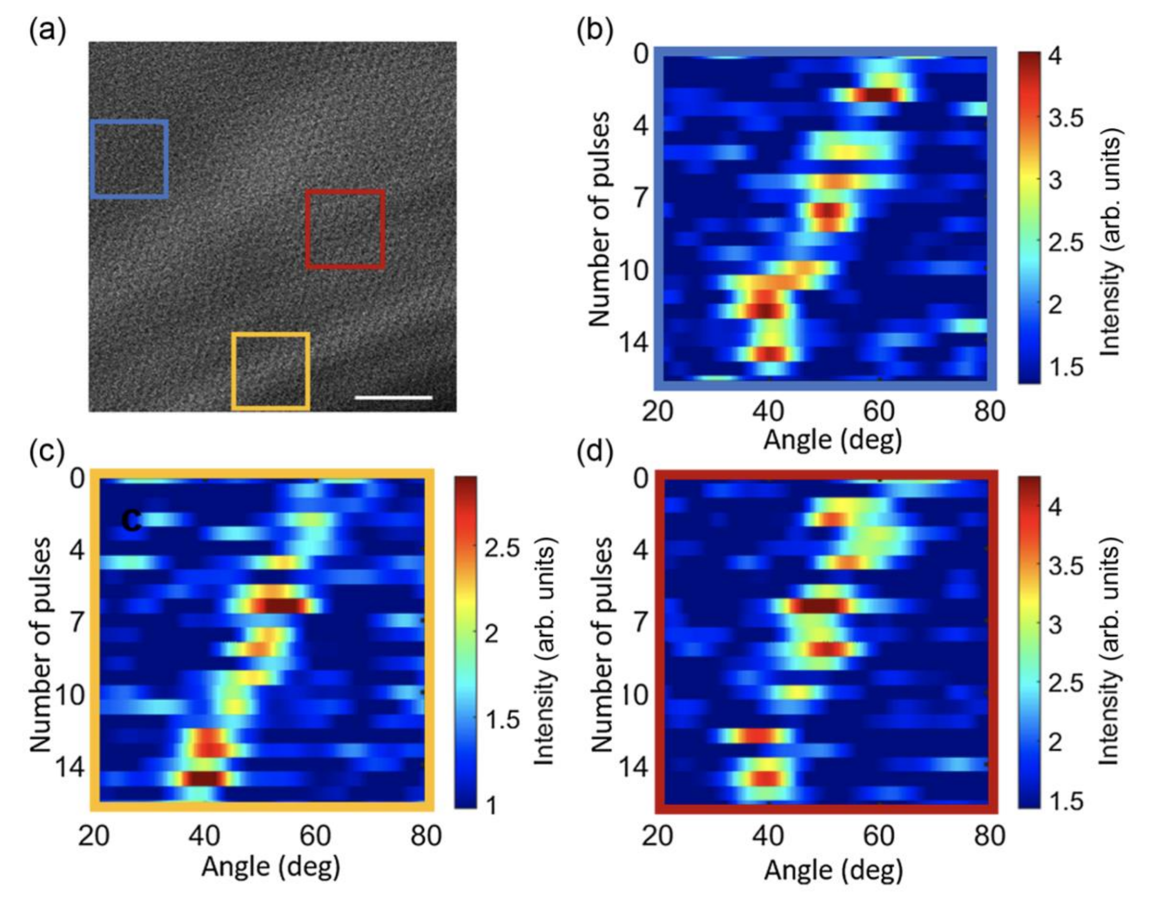
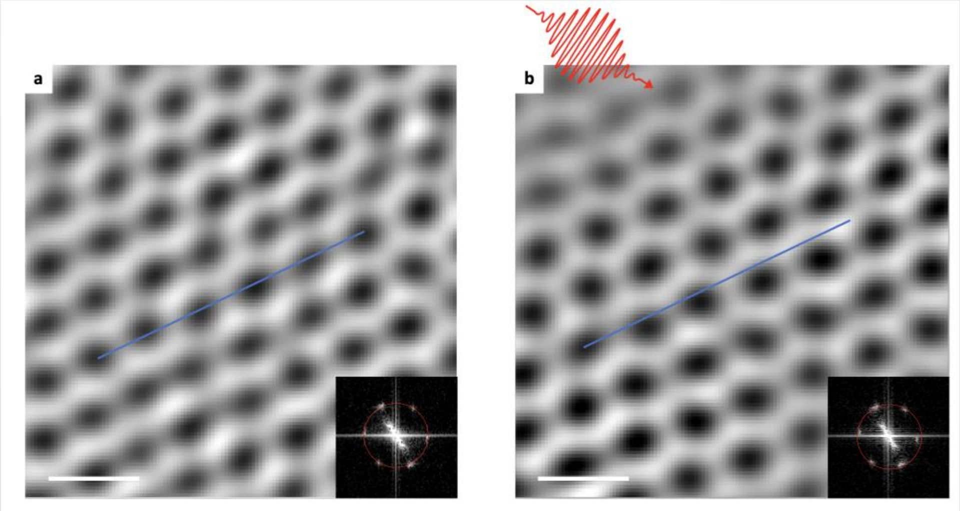
# Mapping in real-space the skyrmion clusters

- Fourier transform analysis of sub-domains
- Breakdown into smaller skyrmion crystallites with longer pitch length





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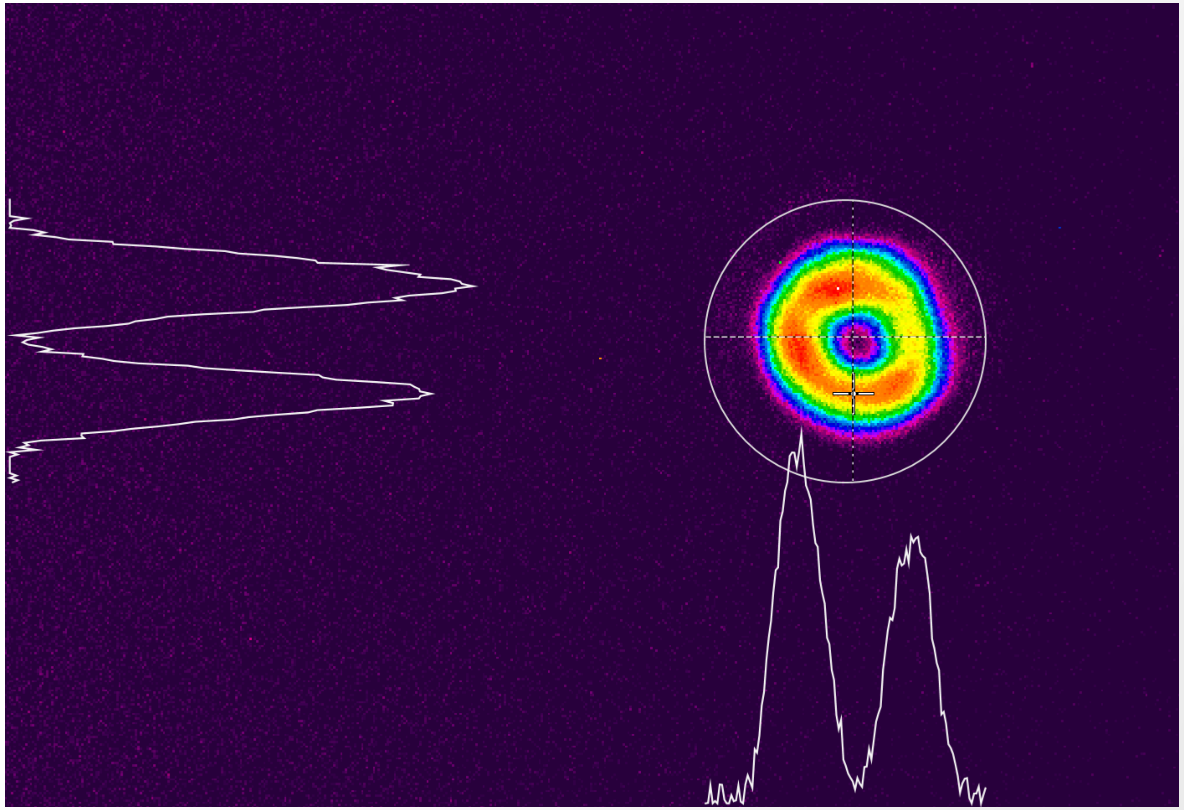
# Preliminary 1030nm OAM beam data



- 1030nm, 200fs, single-shot to 40MHz, 20µJ
- Better mode and smaller spot (65µm FWHM) than 1200nm laser
- More stable – can collect staticially relevant data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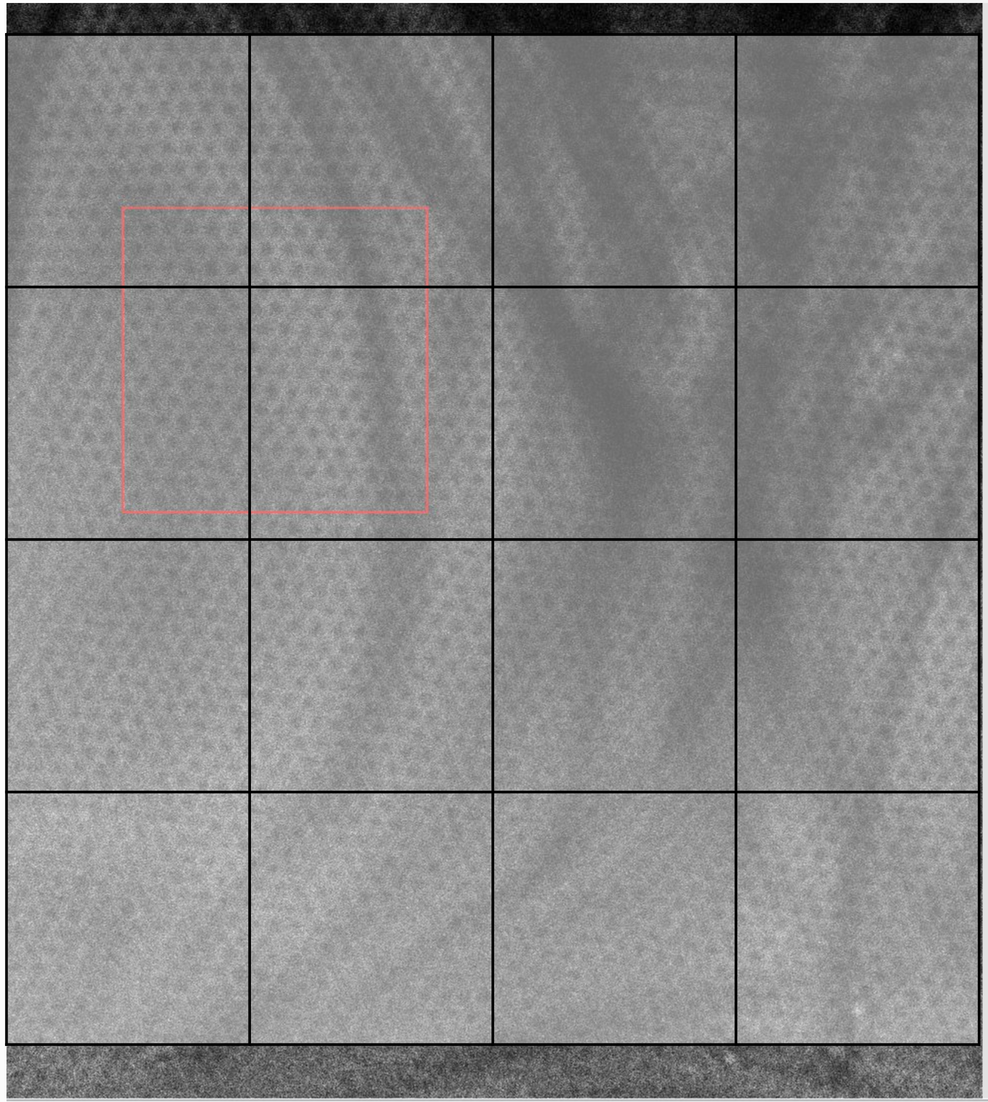
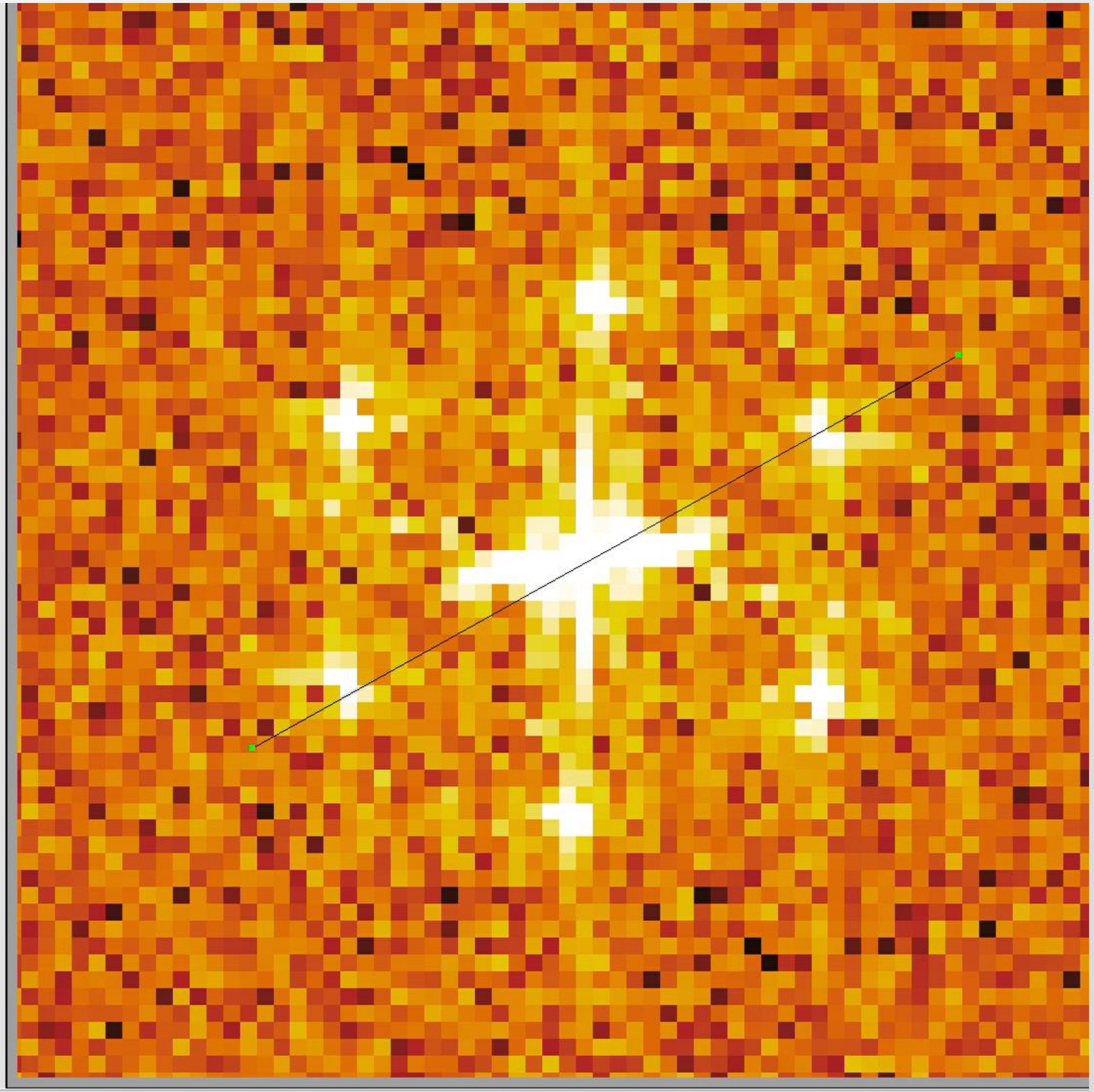
# We don't observe the same behavior (no clear rotations)



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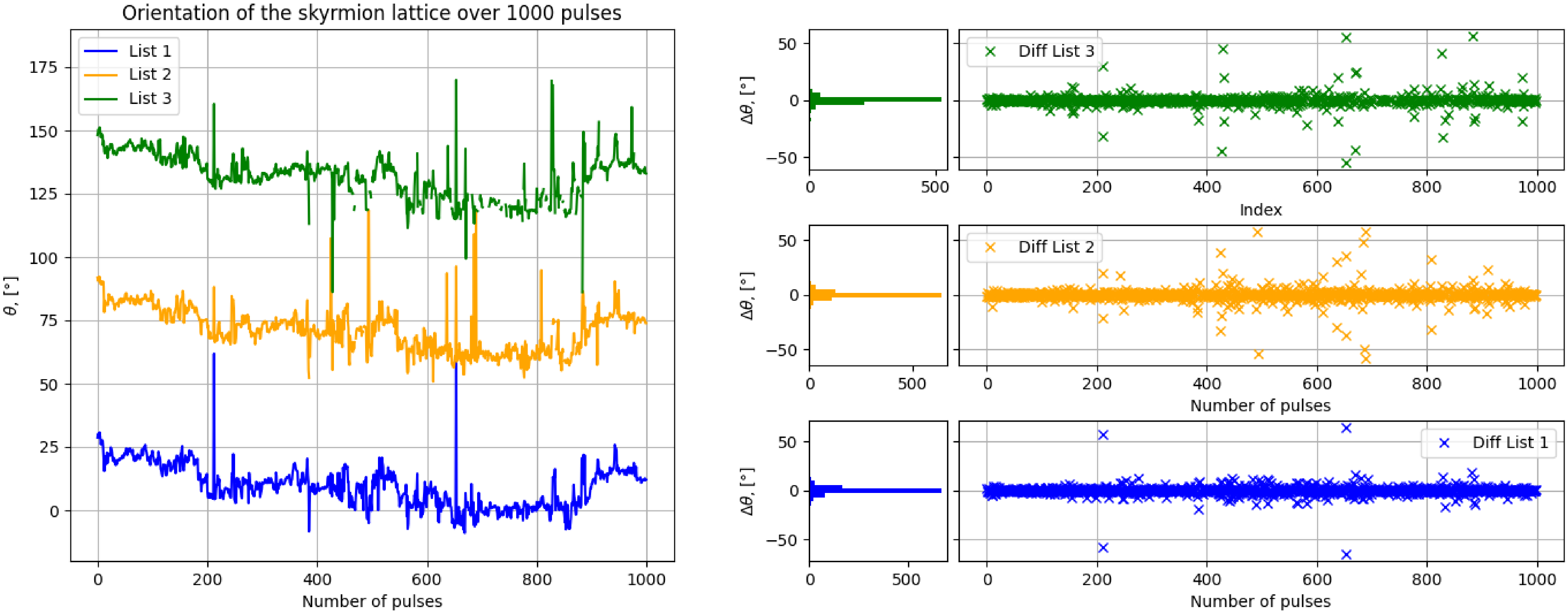
# Statistically, there is no rotation with 1030nm, and in both cases with single and 2-pulse pumping



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# 1030 nm wavelengths are not ideal for this study

communications physics

ARTICLE

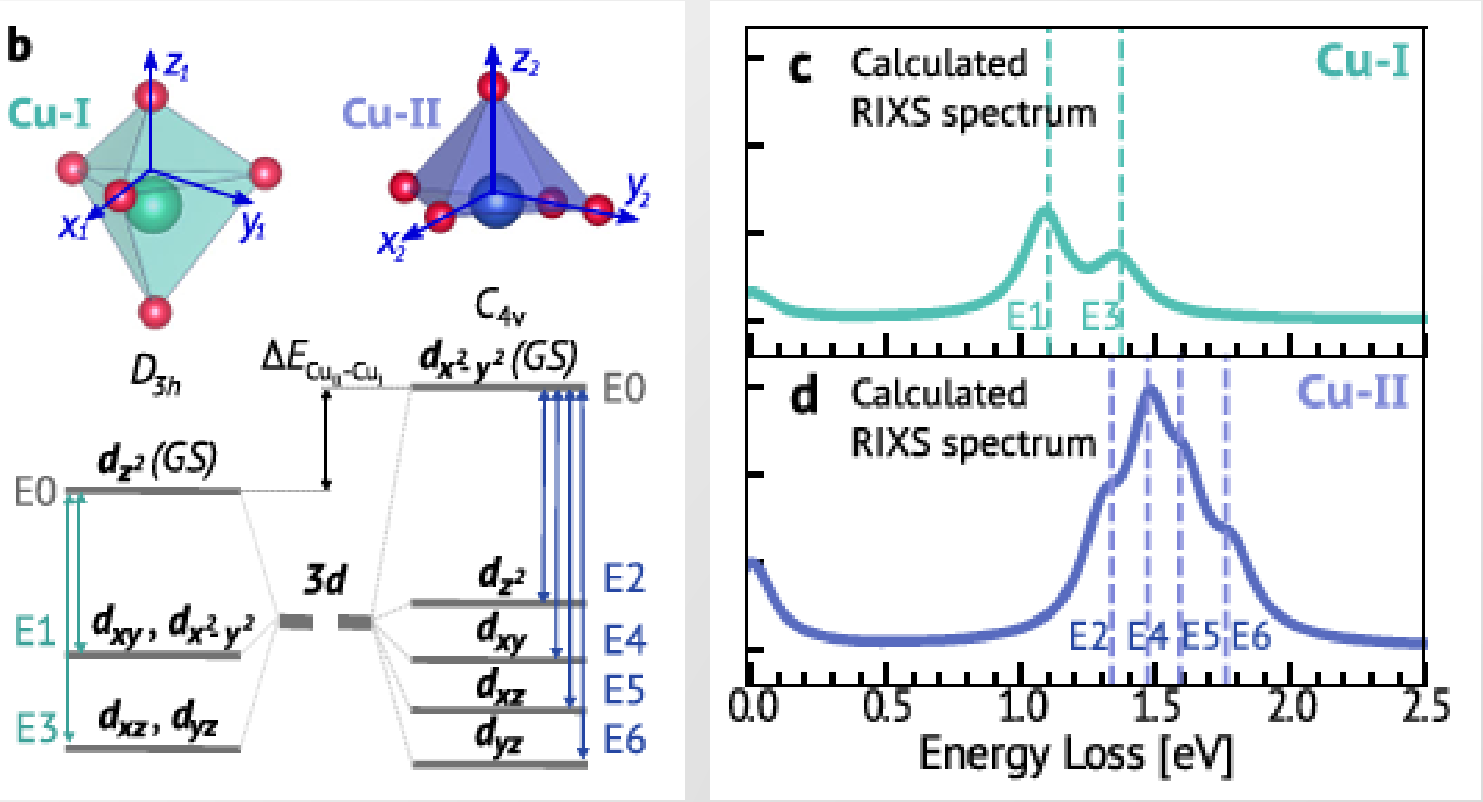
https://doi.org/10.1038/s42005-022-00934-y OPEN

Check for updates

## Site-specific electronic and magnetic excitations of the skyrmion material $Cu_2OSeO_3$

Yanhong Gu[1], Yilin Wang[2,5], Jiaqi Lin[2,6], Jonathan Pelliciari[1], Jiemin Li[1], Myung-Geun Han[2], Marcus Schmidt[3], Gabriel Kotliar[2,4], Claudio Mazzoli[1], Mark P. M. Dean[2] & Valentina Bisogni[1✉]

The manifestation of skyrmions in the Mott-insulator $Cu_2OSeO_3$ originates from a delicate balance between magnetic and electronic energy scales. As a result of these intertwined couplings, the two symmetry-inequivalent magnetic ions, Cu-I and Cu-II, bond into a spin S = 1 entangled tetrahedron. However, conceptualizing the unconventional properties of this material and the energy of the competing interactions is a challenging task due to the complexity of this system. Here we combine X-ray Absorption Spectroscopy and Resonant Inelastic X-ray Scattering to uncover the electronic and magnetic excitations of $Cu_2OSeO_3$ with site-specificity. We quantify the energies of the 3*d* crystal-field splitting for both Cu-I and Cu-II, fundamental for optimizing model Hamiltonians. Additionally, we unveil a site-specific magnetic mode, indicating that individual spin character is preserved within the entangled-tetrahedron picture. Our results thus provide experimental constraints for validating theories that describe the interactions of $Cu_2OSeO_3$, highlighting the site-selective capabilities of resonant spectroscopies.




**1030nm laser light can induce a crystal field excitation ($d_{xy}$, $d_{x^2+y^2}$) which can launch incoherent thermal processes that convolute with inverse faraday effect we want to study**

# Summary

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- Magnetic background influences skyrmions motion
- Light can induce new metastable skyrmion phases, though these light-induced states are **metastable**
- Inverse Faraday effect provides the possibility to control the skyrmions coherently
- OAM beams can provide an extra handle to manipulate skyrmions
- Avoid pumping crystal field excitations
- Perspective – 3D electron holography to observe twists in the skyrmions tubes


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# We want to directly observe coherent magnons induced by the magnetic fields of microwave antennae


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Editors' Suggestion | Featured in Physics

**Direct observation of coherent magnons with suboptical wavelengths in a single-crystalline ferrimagnetic insulator**

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Spin-wave dynamics were studied in an extended thin film of single-crystalline yttrium iron garnet using time-resolved scanning transmission x-ray microscopy. A combination of mechanical grinding and focused ion beam milling has been utilized to achieve a soft x-ray transparent thickness of the underlying bulk gadolinium gallium garnet substrate. Damon-Eshbach type spin waves down to about 100 nm wavelength have been directly imaged in real space for varying frequencies and external magnetic fields. The dispersion relation extracted from the experimental data agreed well with theoretical predictions. A significant influence of the ion milling process on the local magnetic properties was not detected.

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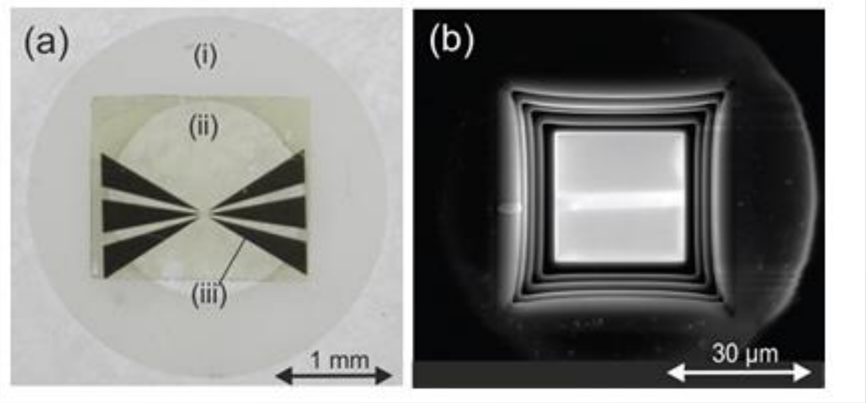
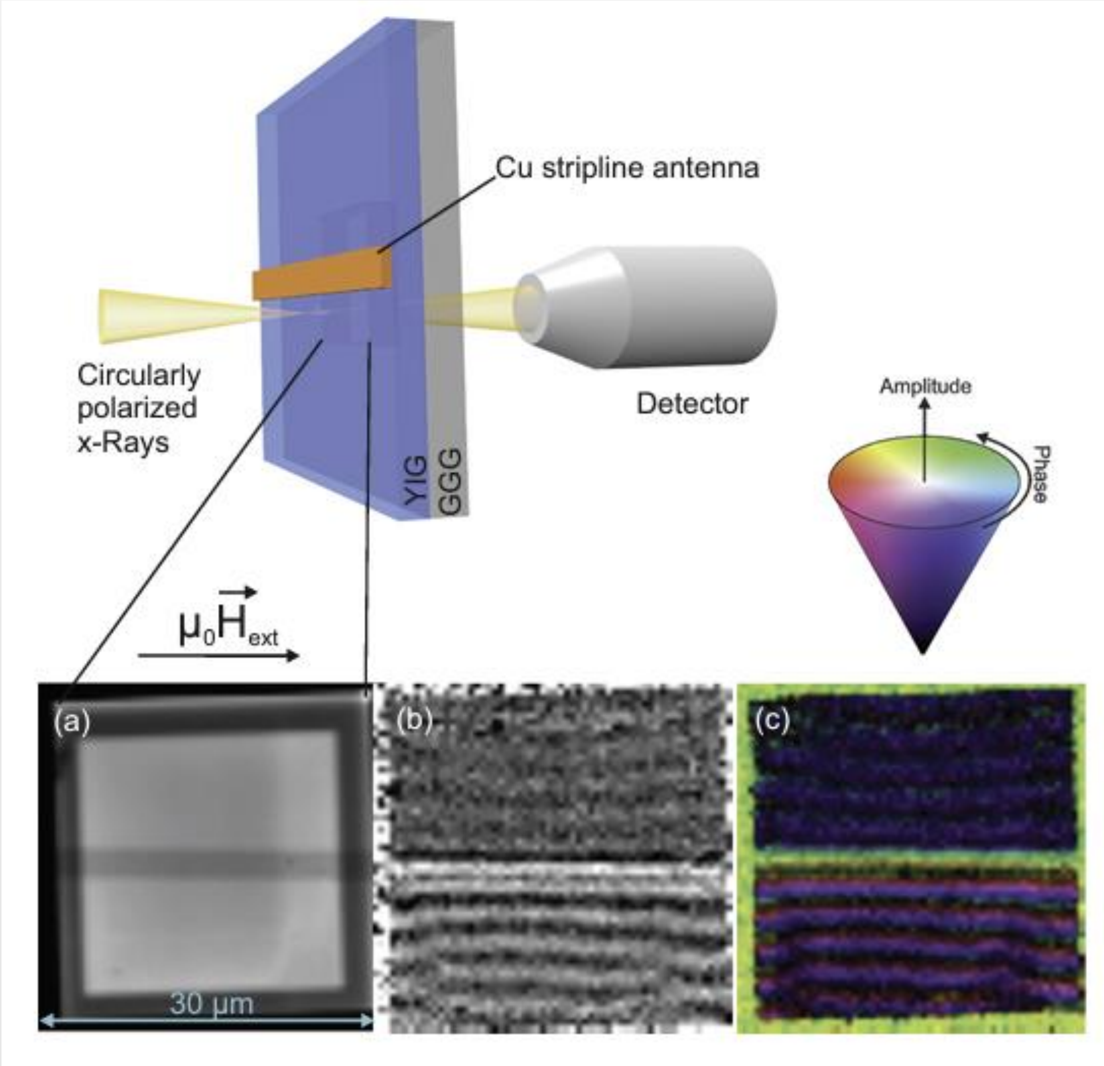



185nm thick single crystal YIG film deposited on Cu stripline antennae





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# We constructed a RF holder that can excite dynamics in TEM samples with microwave frequencies up to 15 GHz

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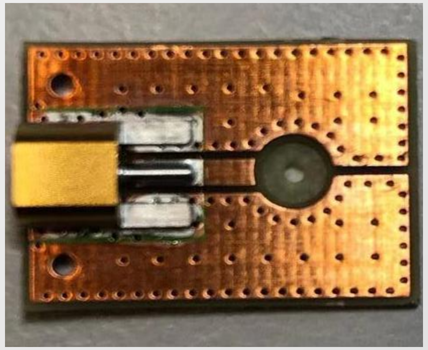
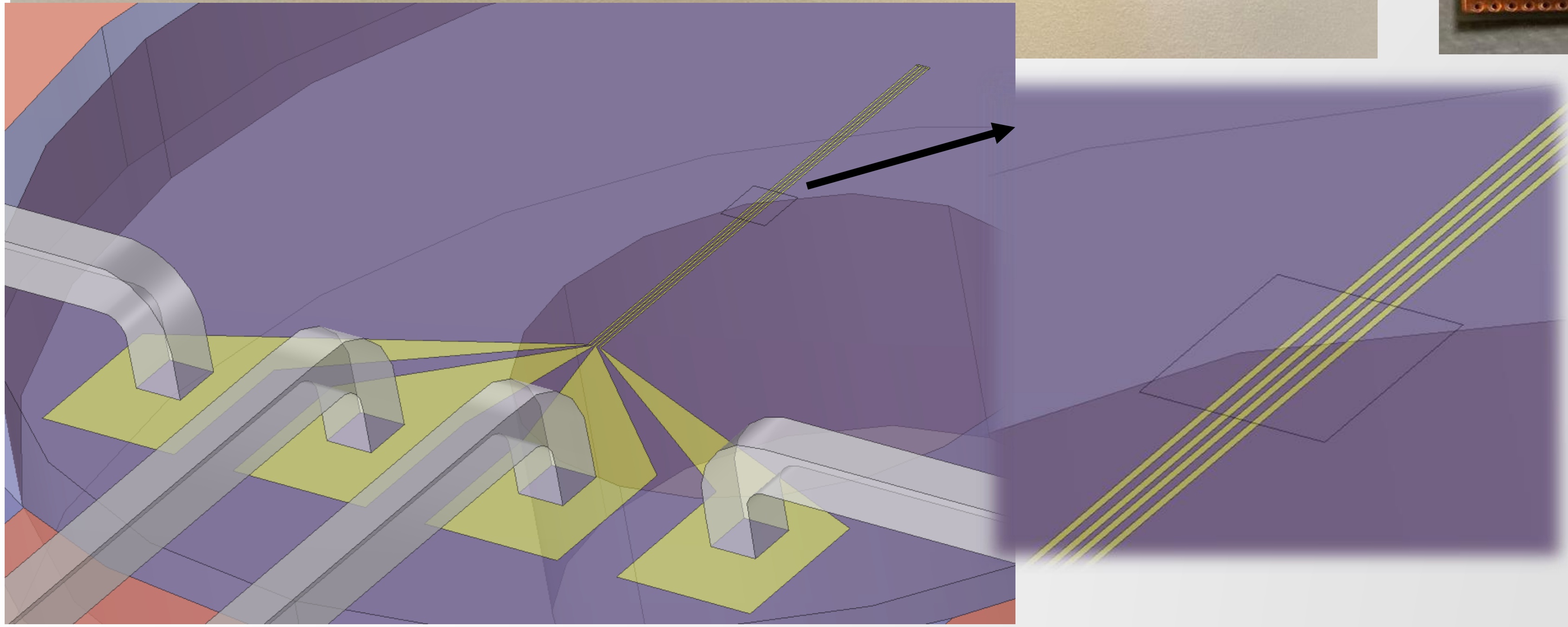
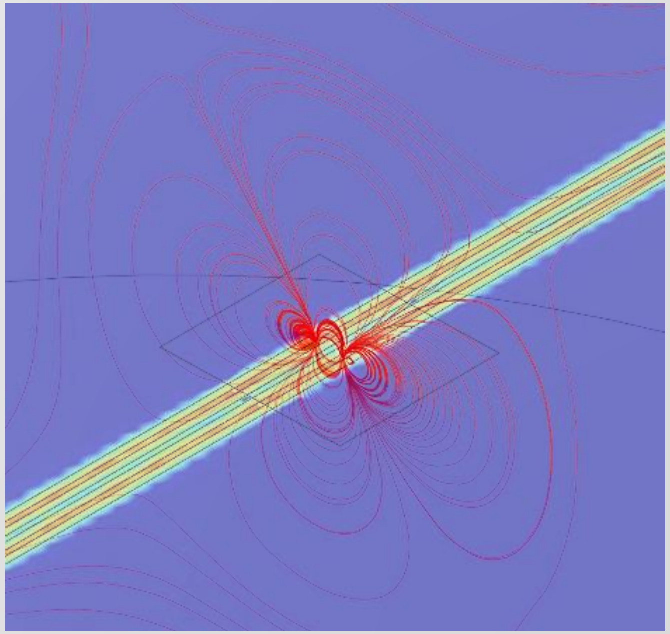
Wired bonded custom chips to single mode microwave PCB









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# Thank you!

# Near-term UTEM Experiments

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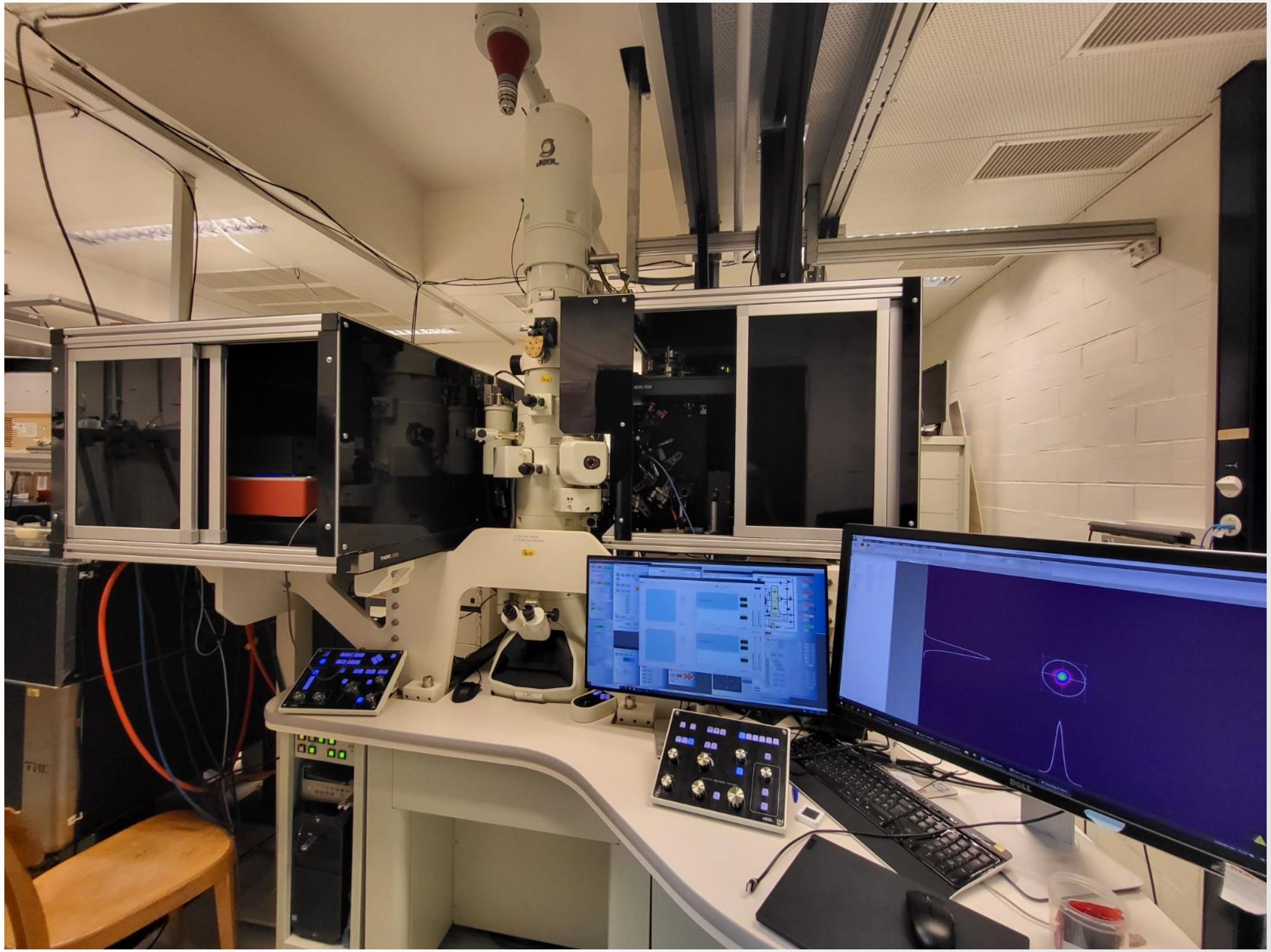

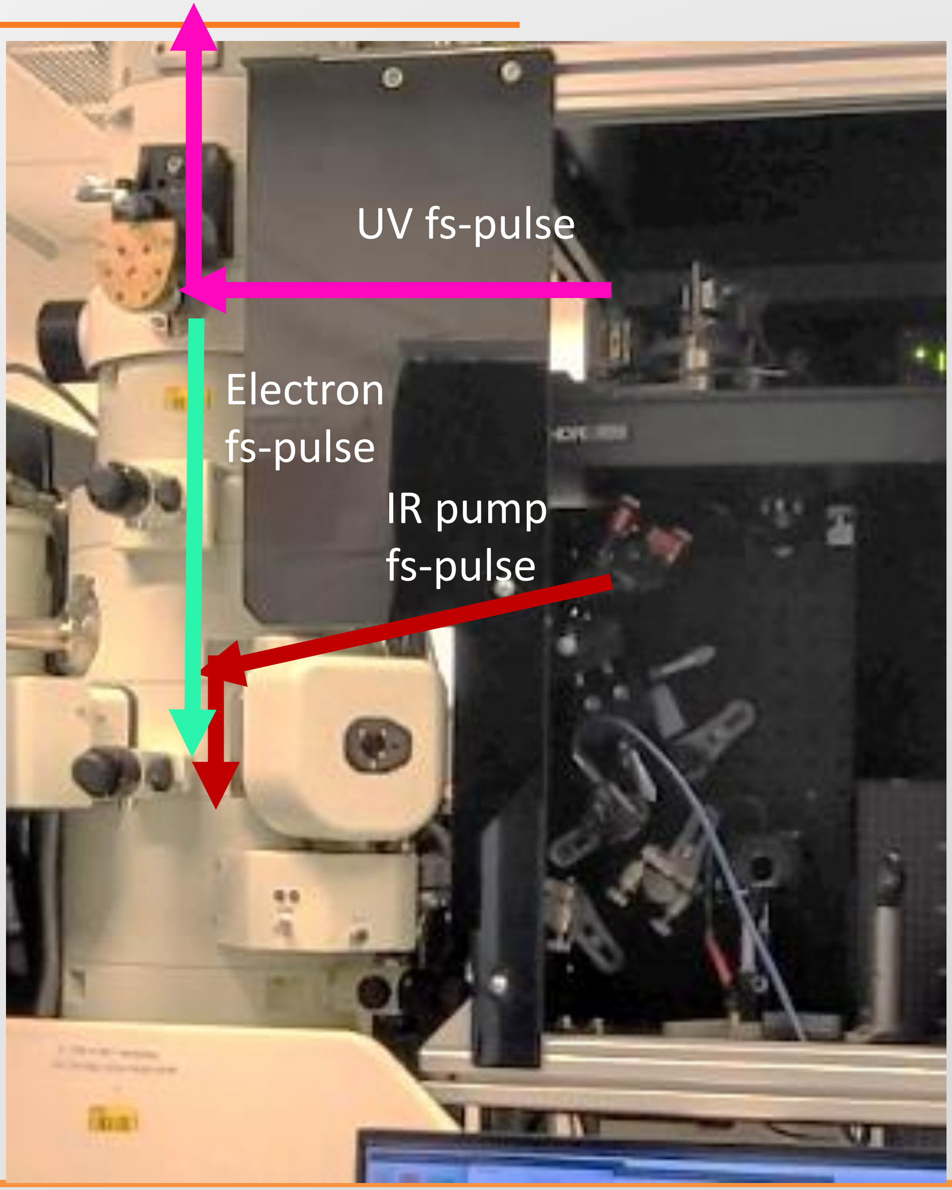



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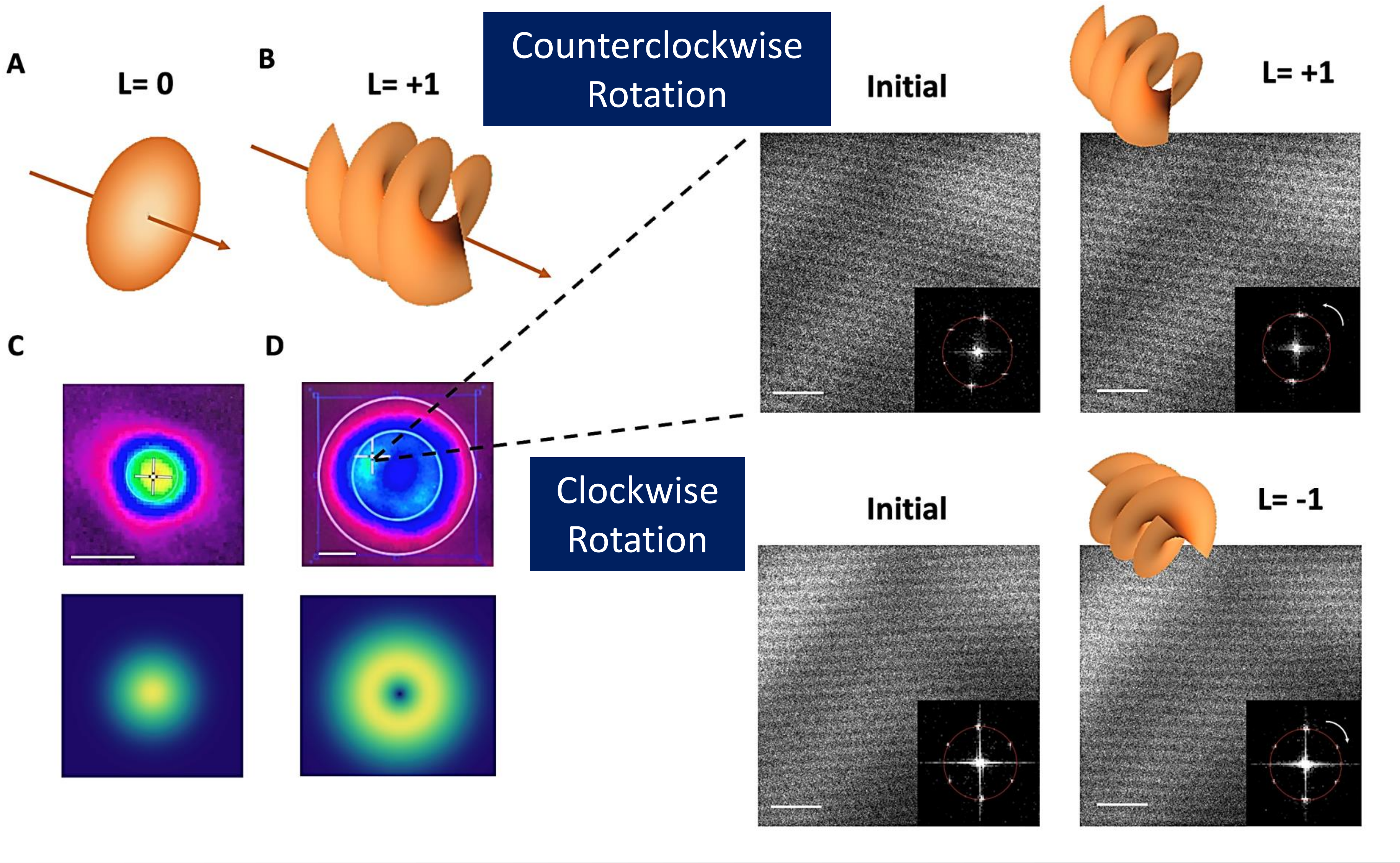
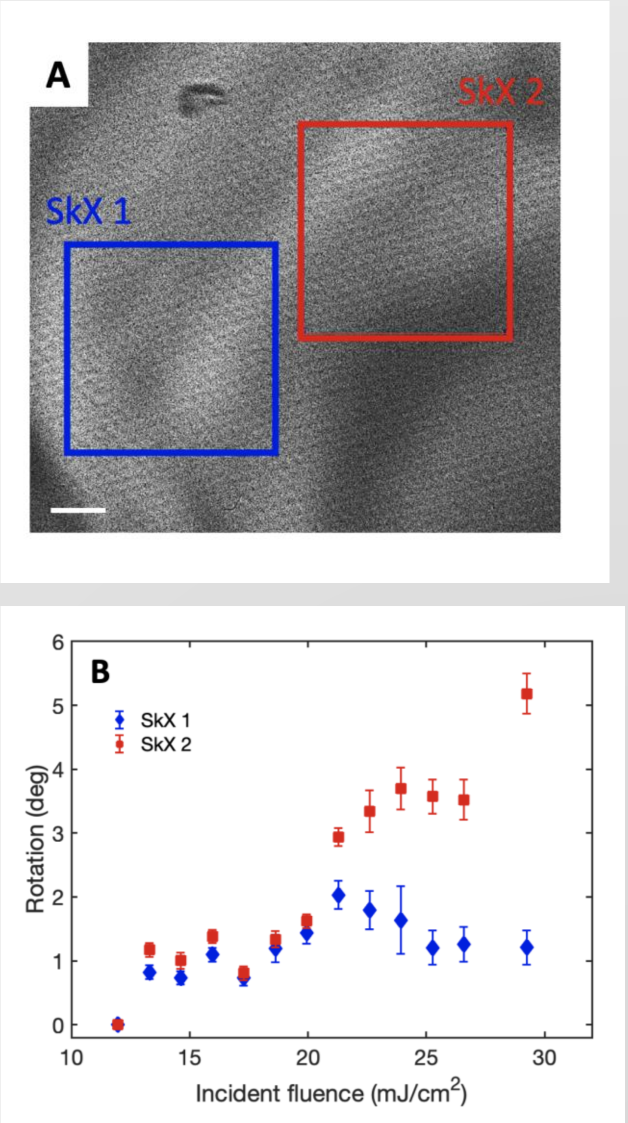
# Skrymion lattice rotation using 1200nm vortex light





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