

Tailoring Magnetic Spin Textures in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ -based Micromagnets

Prof. Yayoi Takamura

Department of Materials Science and Engineering
University of California, Davis

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Min-Fu Hsieh, Guenter
Reiss, Yayoi Takamura

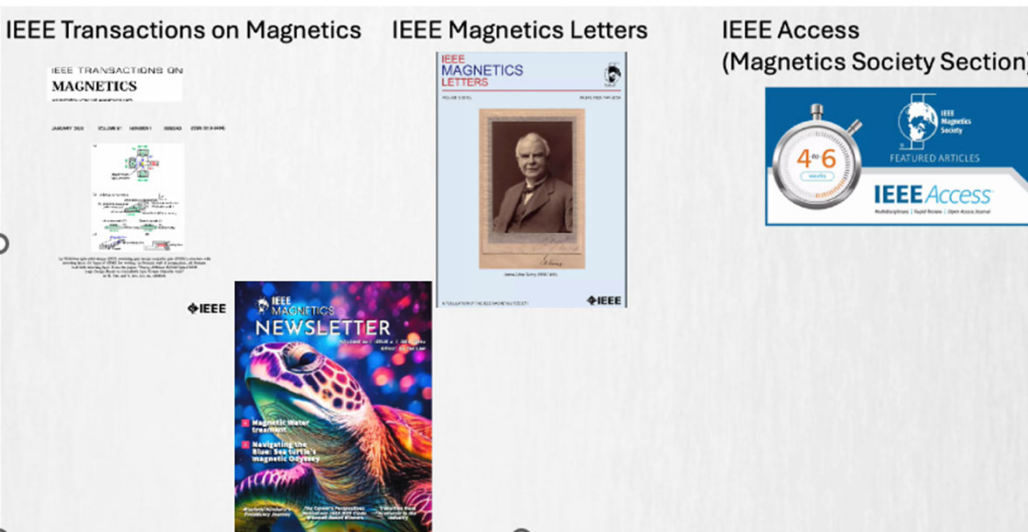
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Acknowledgements

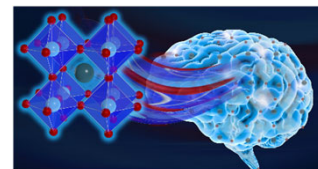


Takamura Research Group: pictured from left to right: Varun Rao, Nathan Tsui, **Dr. Dayne Sasaki** (now at ALS), Yayoi Takamura, Matthew Frame, Hudson Shih, Izoah Snowden, Ishmam Nihal (not pictured Tanaya Sahoo, **Dr. Michael Lee** (now at Niron), **Dr. Rajesh Chopdekar** (now at WD), Ethan Teanu, Steve Ho)

UC Davis: Prof. Jeremy Mason

ALS: Andreas Scholl, Barat Achinuq

ORNL: Scott Retterer



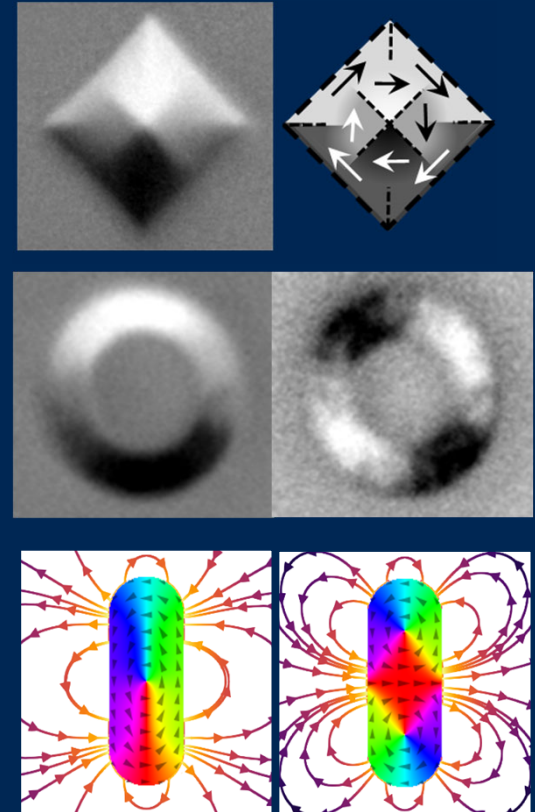
DE-SC0019273



National Laboratory Research
Program

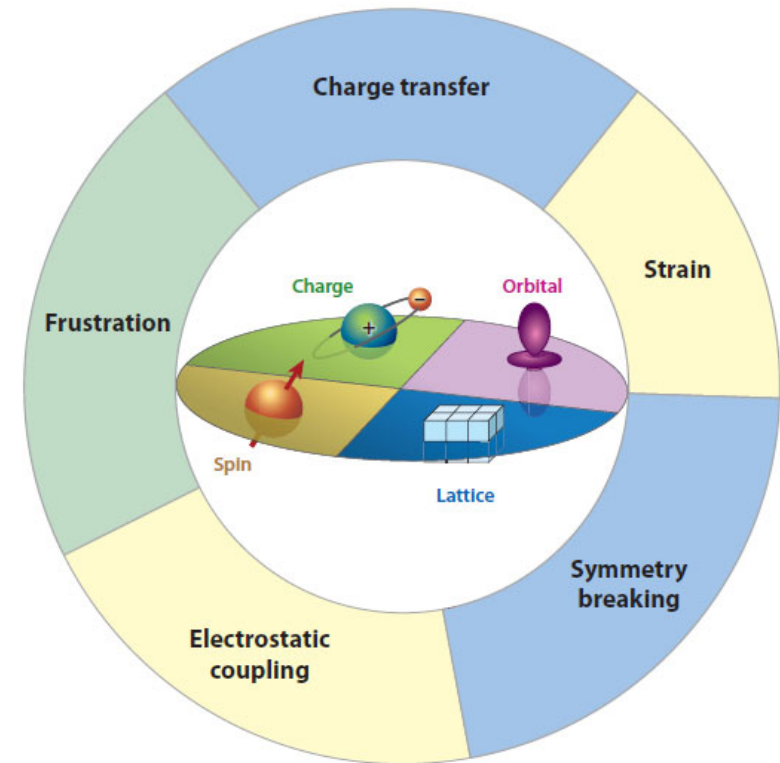
Outline

- Motivation
- Imaging magnetic spin textures
- Two short stories on tailoring magnetic spin textures
 1. $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ micromagnets
 2. $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ artificial spin ice structures
- Summary



Complex Oxides

- Strong interaction among the lattice, charge, spin, orbital degrees of freedom
 - Stimulus-sensitive functional properties
- Wide range of functional properties
 - Ferromagnetism, ferroelectricity, superconductivity, etc...
- Applications in spintronics, neuromorphic computer, magneto-ionics, and solid oxide fuel cells



Zubco *et al.*, *Annu. Rev. Condens. Matter Phys.*, **2**, 141 (2011)

ABO₃ Perovskite Oxides

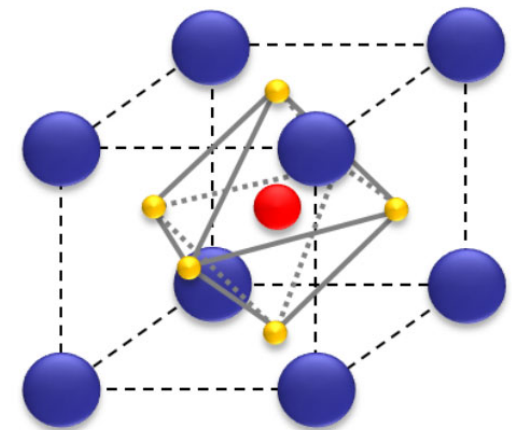
IA																	VIIIA				
H																	He				
	IIA																				
Li	Be															B	C	N	O	F	Ne
Na	Mg												Al	Si	P	S	Cl	Ar			
		IIIB	IVB	VB	VIB	VIIIB	VIII		IB	IIB											
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr				
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe				
Cs	Ba	La-Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn				
Fr	Ra	Ac-Lr	Rf	Db	Sg	Bh	Hs	Mt													

La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
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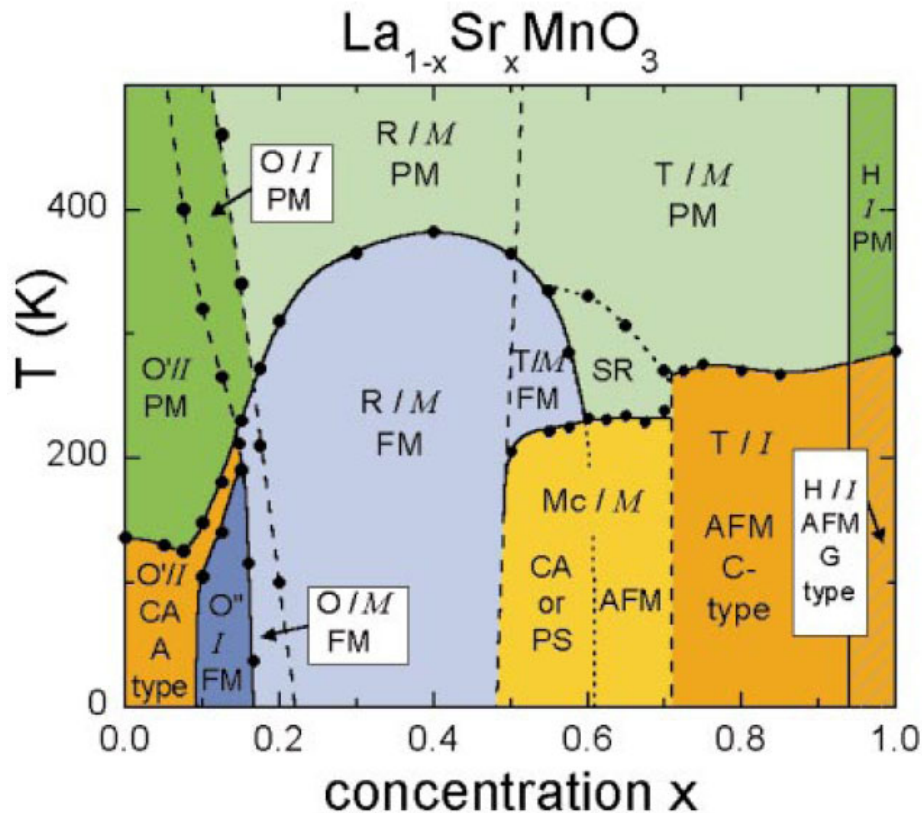
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
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Rondinelli and Fennie, *Adv. Mater.*, **24**, 1962 (2012)

- Compatible with many different elements leading to a wide range of functional properties

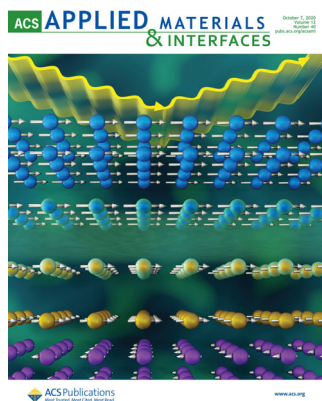


Magnetic Perovskite Oxides

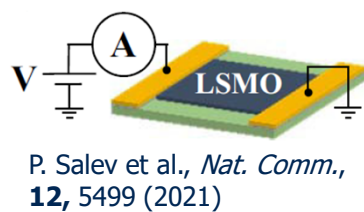
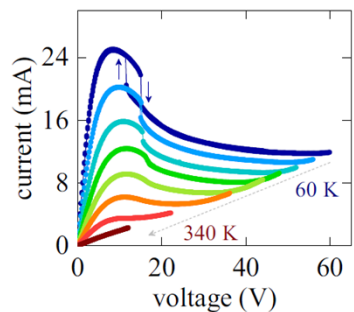


- $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO):
 - Ferromagnetic (FM) metal $< T_C = 360$ K, half metal, colossal magnetoresistance

Research Highlights

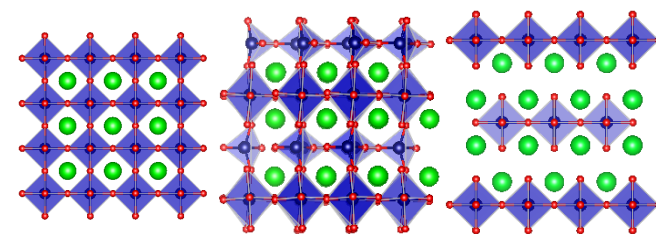
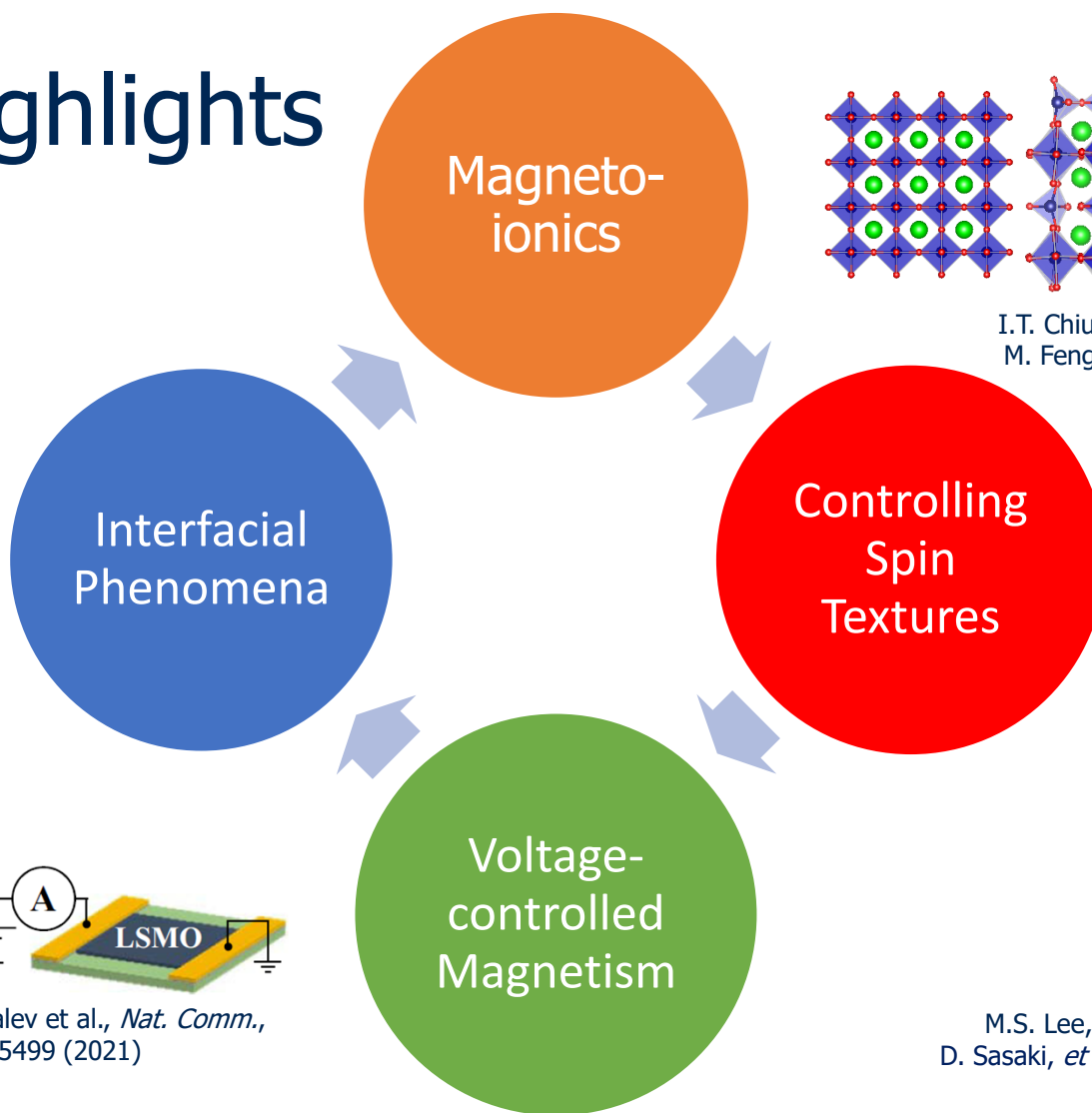


A.M. Kane *et al.*, *ACS Appl. Mater. & Int.*, **12**, 45437 (2020)
I. Nihal *et al.*, *PRB*, **110**, 014411 (2024)

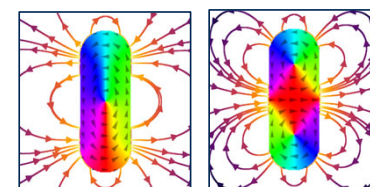
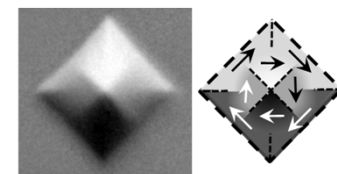


P. Salev *et al.*, *Nat. Comm.*, **12**, 5499 (2021)

11/23/2025



I.T. Chiu, *et al.*, *PRM*, **5**, 064416 (2021)
M. Feng, *et al.*, *J. Phys. Chem. C*, **128**, 17124 (2024)



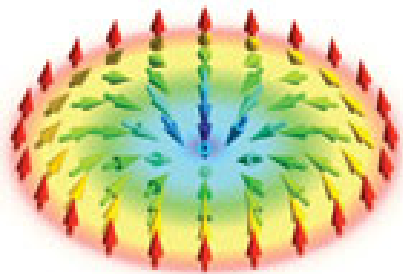
M.S. Lee, *et al.*, *ACS Nano*, **10**, 8545 (2016)
D. Sasaki, *et al.*, *PR Applied*, **17**, 064057 (2022)

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Spin Textures

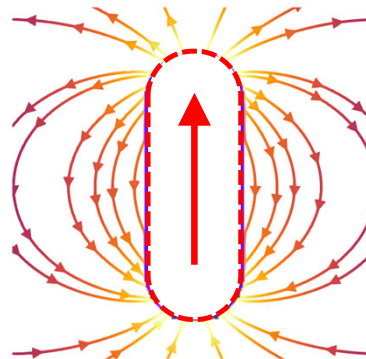
- Spatial variation of the local magnetization in a material/object

Skyrmions

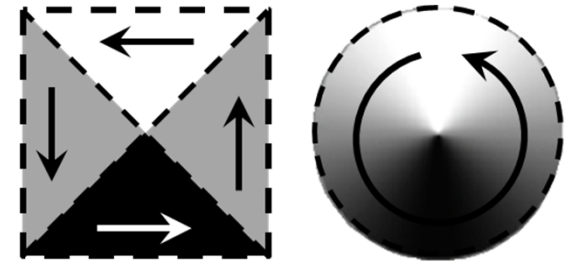


www.christophschuette.com

Single Domain



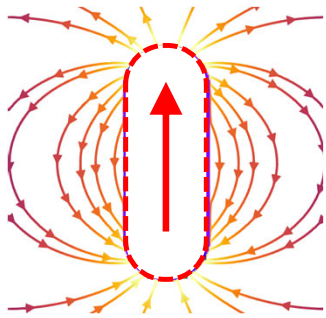
Landau/Vortex States



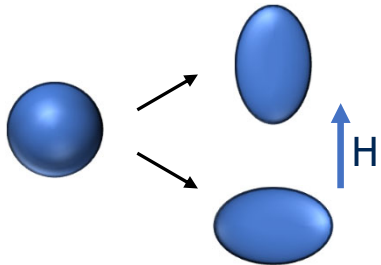
Increasing dimensions

Spin Texture Energy Terms

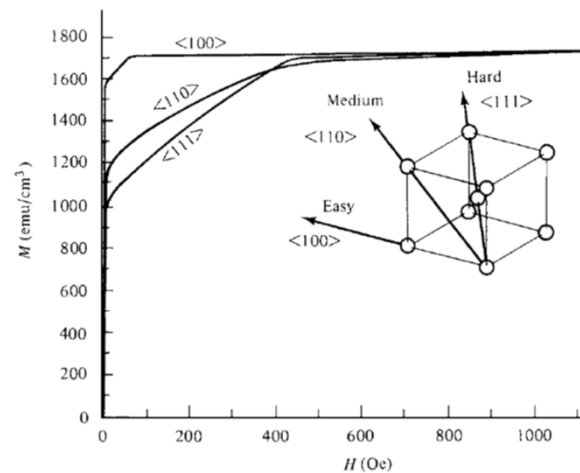
Magnetostatic Energy



Magnetoelastic Energy

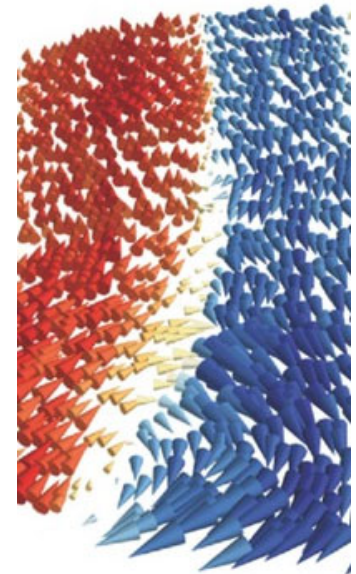


Magnetocrystalline Anisotropy Energy



Cullity and Graham, *Introduction to Magnetic Materials*, 2nd edition

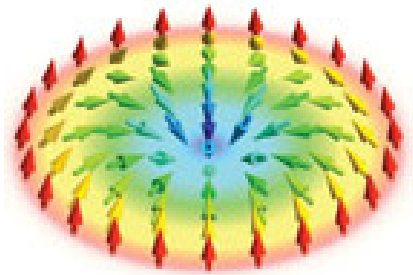
Domain Wall Energy



G. Chen & A.K. Schmid, *Adv. Mater.*, 27, 5738

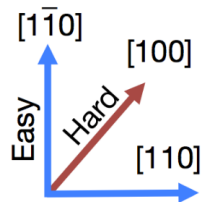
Spin Texture Energy Terms

Skyrmions



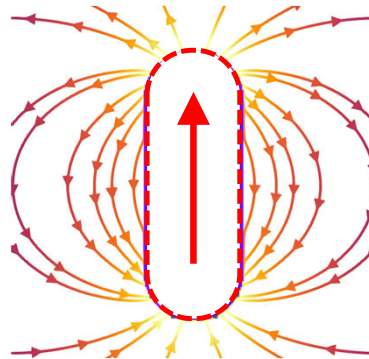
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Magnetocrystalline
Anisotropy Energy



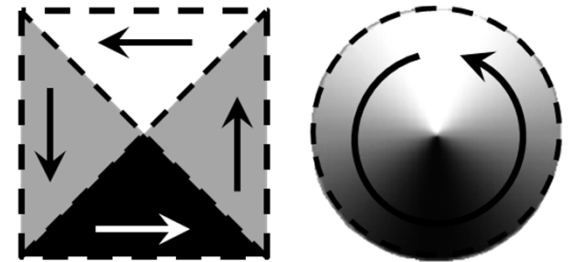
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Single Domain



Magnetostatic Energy
Magnetoelastic Energy

Landau/Vortex States



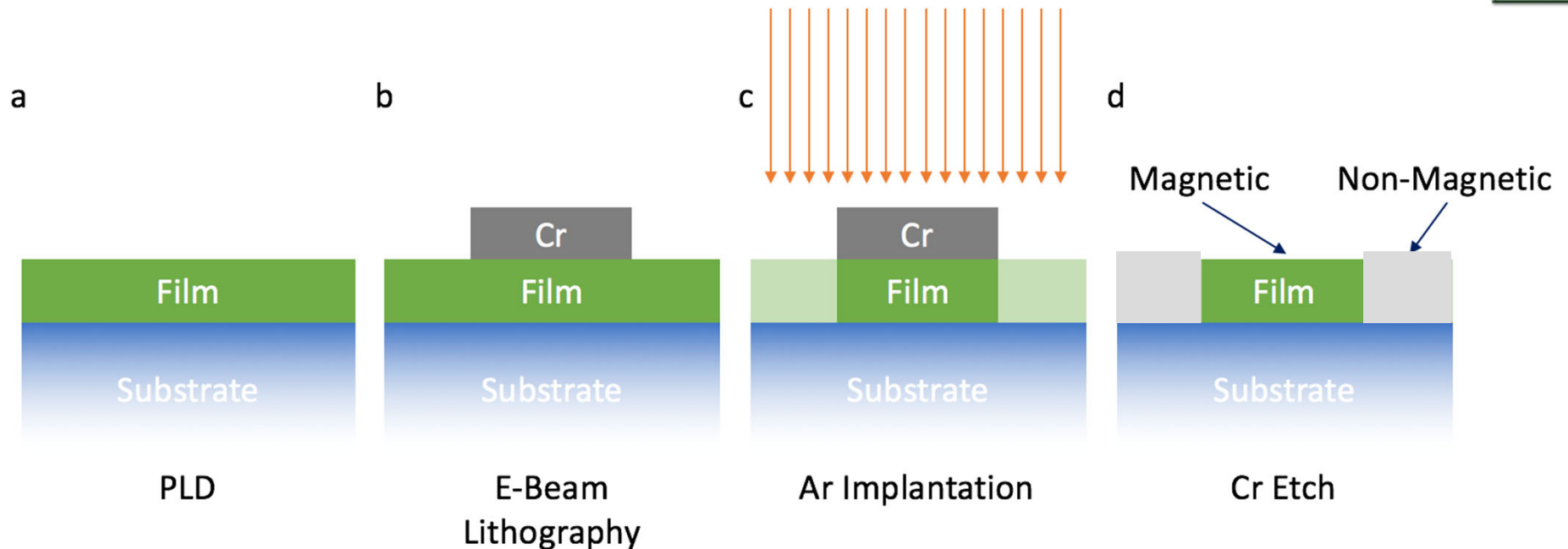
Domain Wall Energy (squares)
Magnetoelastic Energy (squares)

Magnetocrystalline Anisotropy
Energy (circles)

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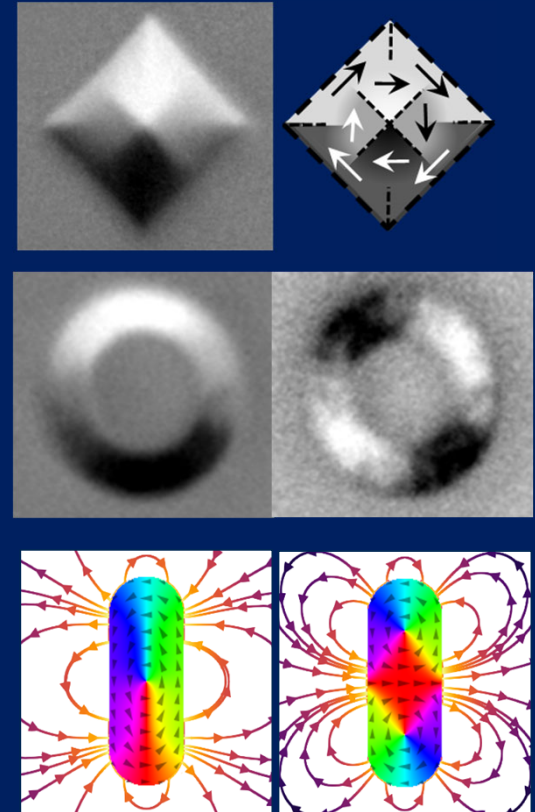
Ion Implantation-based Patterning



- Magnetic islands embedded in a non-magnetic matrix

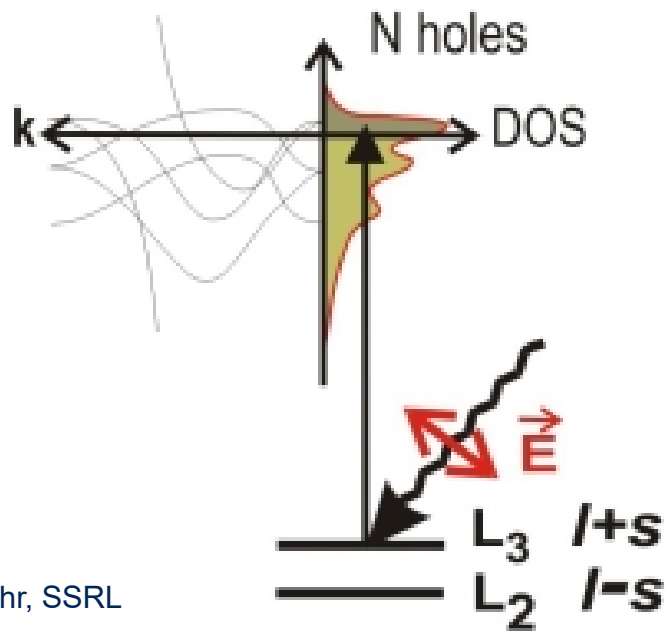
Outline

- Motivation
- Imaging magnetic spin textures
 - X-ray photoemission electron microscopy
- Two short stories on tailoring magnetic spin textures
 1. $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ micromagnets
 2. $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ artificial spin ice structures
- Summary

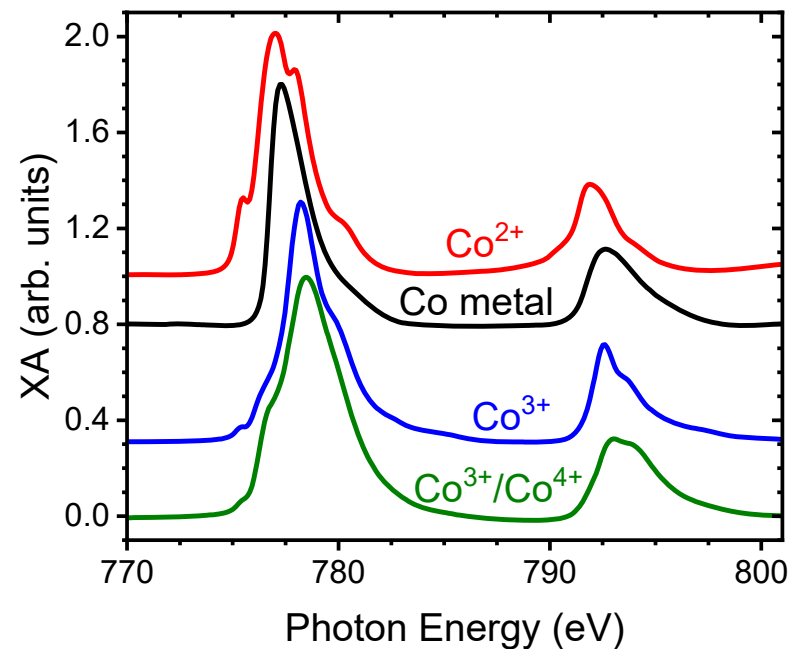


X-ray Absorption (XA) Spectroscopy

- Sensitive to valence states (e.g., Co^{2+} vs. Co^{3+} vs. Co^{4+} ions) and bonding configurations (e.g., octahedral vs. tetrahedral coordination)

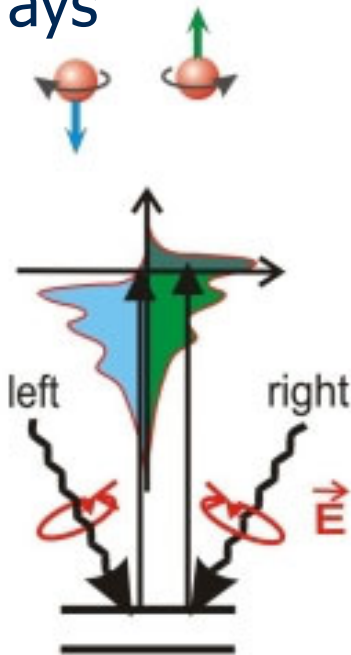


J. Stohr, SSRL

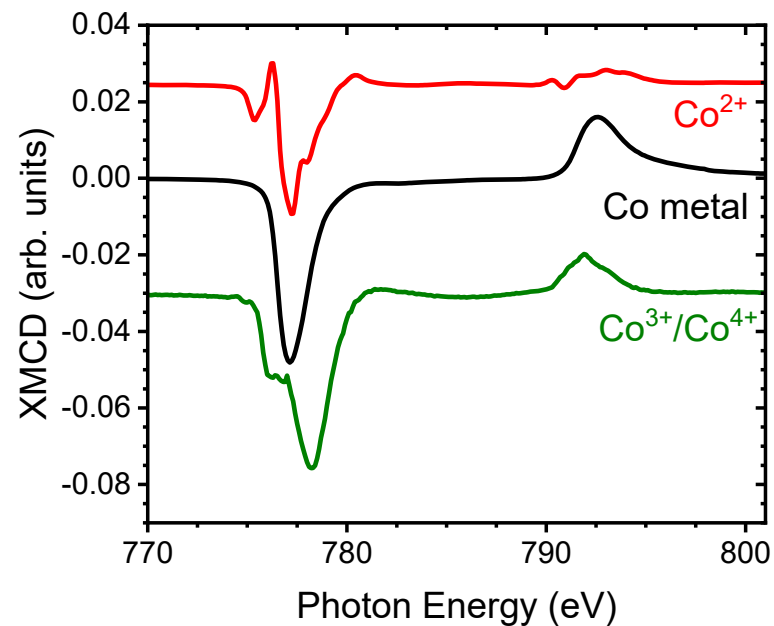


X-ray Magnetic Circular Dichroism (XMCD)

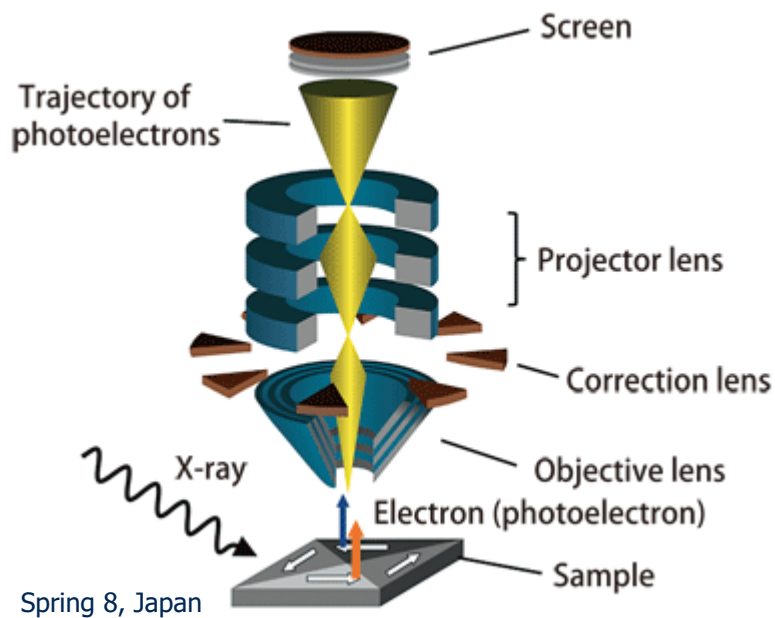
- Sensitive to ferromagnetically active ions
- Difference between XA spectra acquired with right/left circularly polarized x-rays



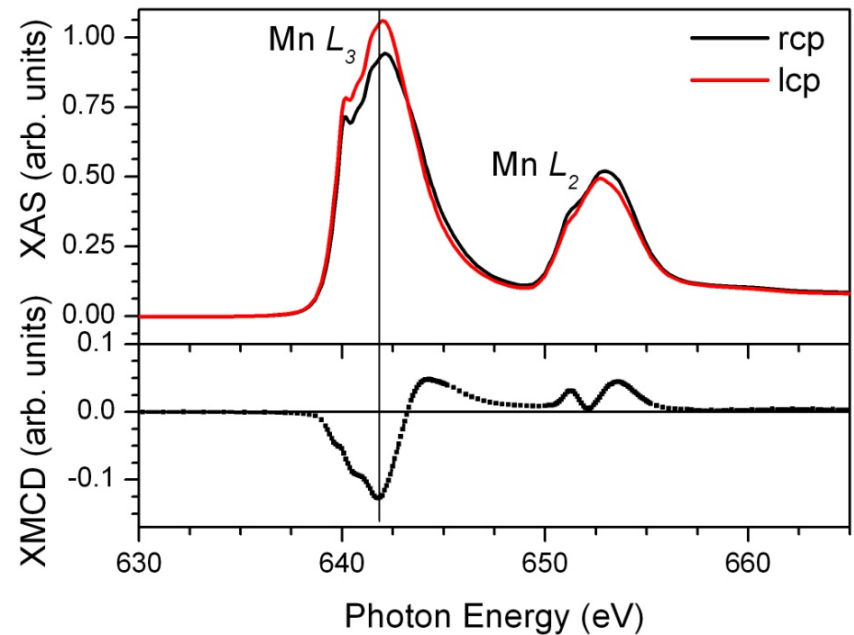
J. Stohr, SSRL



Photoemission Electron Microscopy (PEEM)

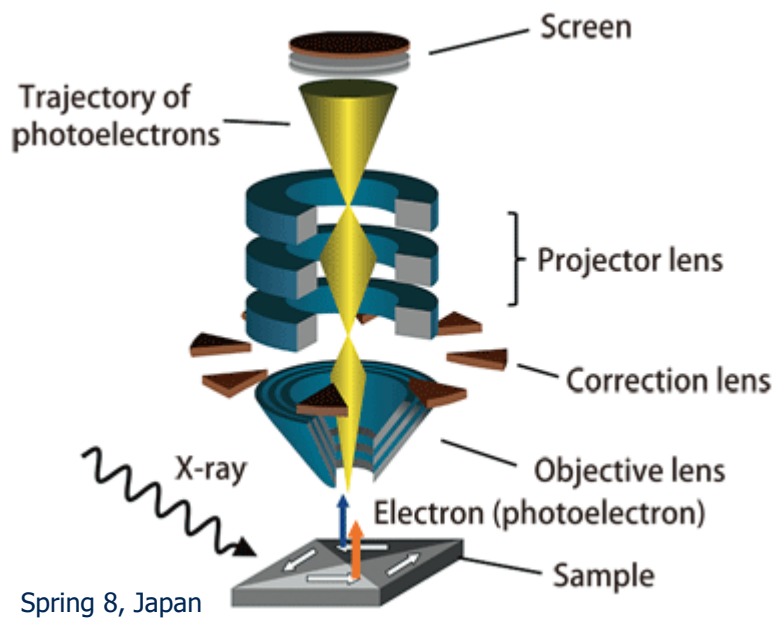


Performed using the PEEM-3 Microscope



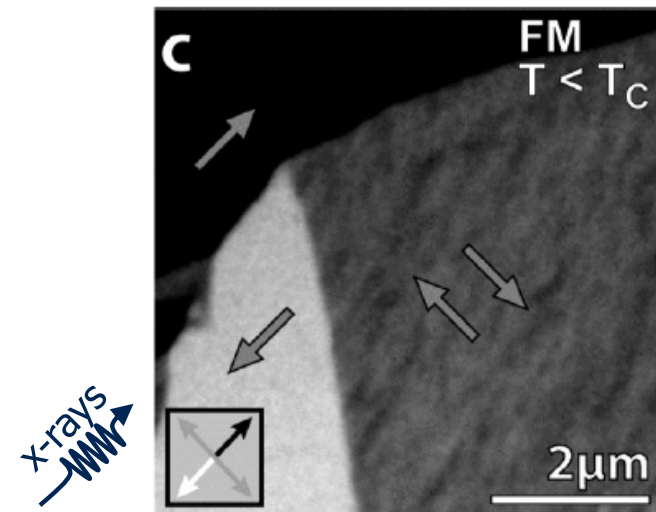
$$Asymmetry = \frac{rcp - lcp}{rcp + lcp}$$

Photoemission Electron Microscopy (PEEM)



Performed using the PEEM-3 Microscope

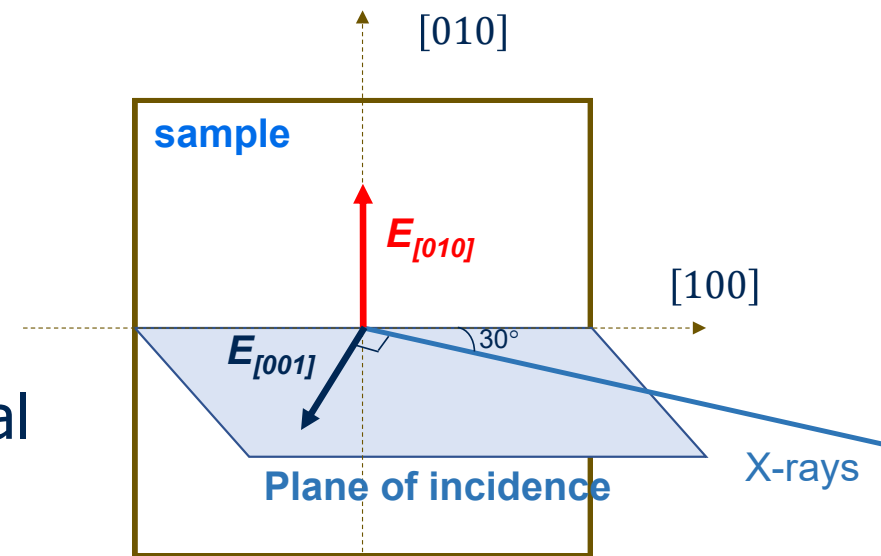
- XMCD $\sim \mathbf{M} \cos(\theta)$; θ = angle between x-ray helicity and magnetization, $\mathbf{M}$



White/black/grey = antiparallel/parallel/perpendicular alignment

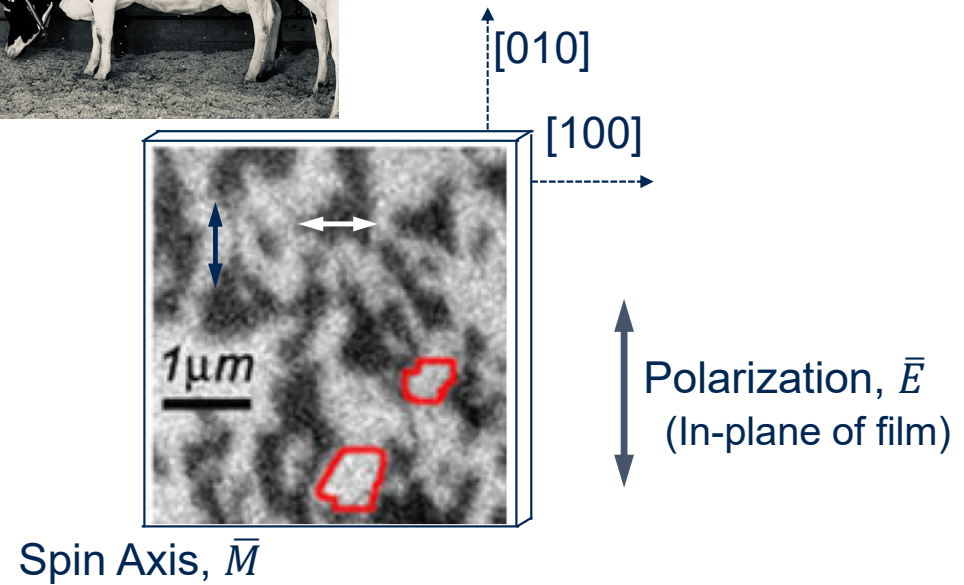
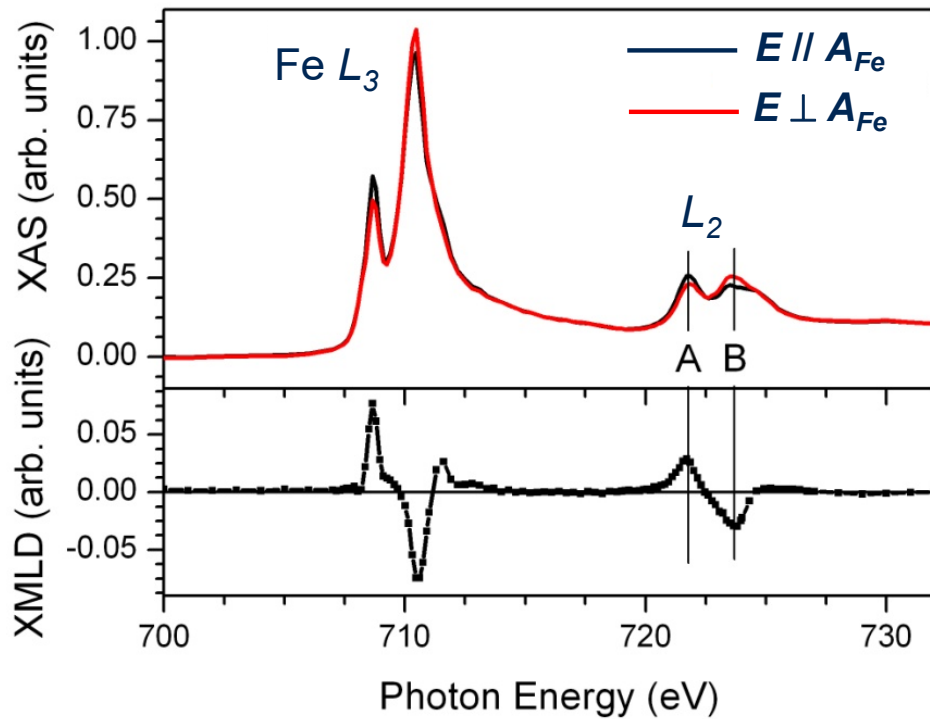
X-ray Magnetic Linear Dichroism (XMLD)

- Sensitive to *asymmetry* in the system
 - Orbital occupancy
 - Crystallographic directions/asymmetry
 - AFM spin axis
- Requires carefully designed experimental geometry



$$I_{XLD} = I_{XA}(E_{100}) - I_{XA}(E_{190})$$

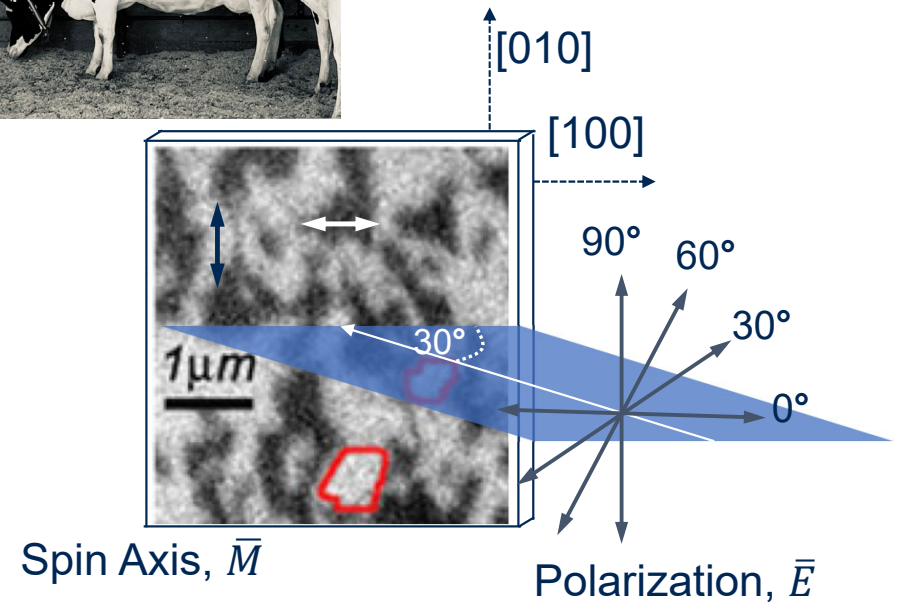
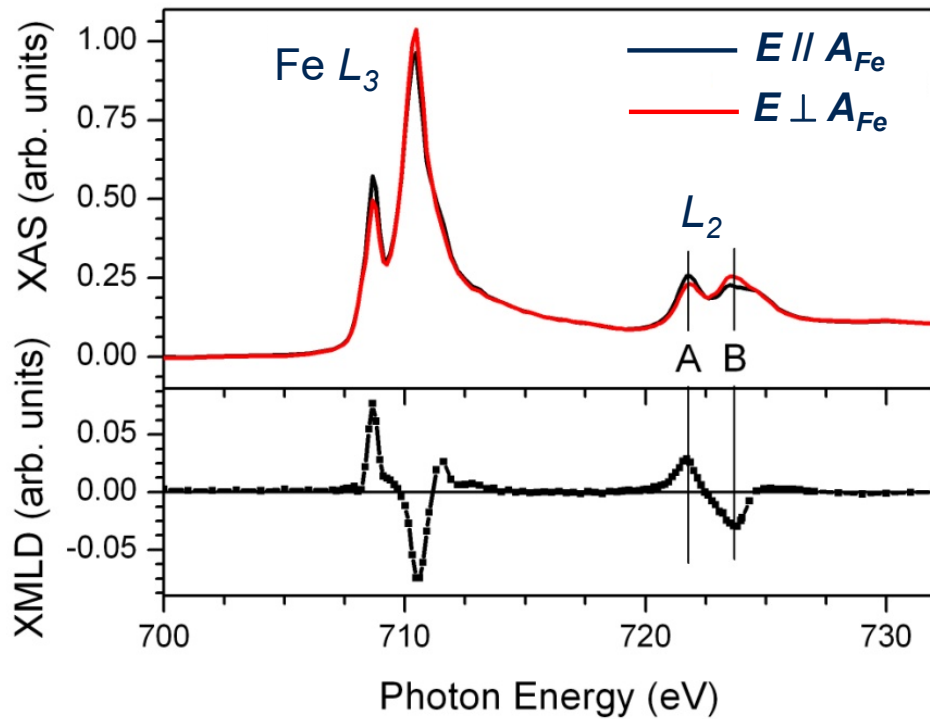
XMLD-PEEM



$$I(\omega, T) = a + b(3\cos^2\omega - 1)\langle A^2 \rangle_T$$

ω = angle between $\bar{E}$ and magnetic spin axis, $\bar{A}$

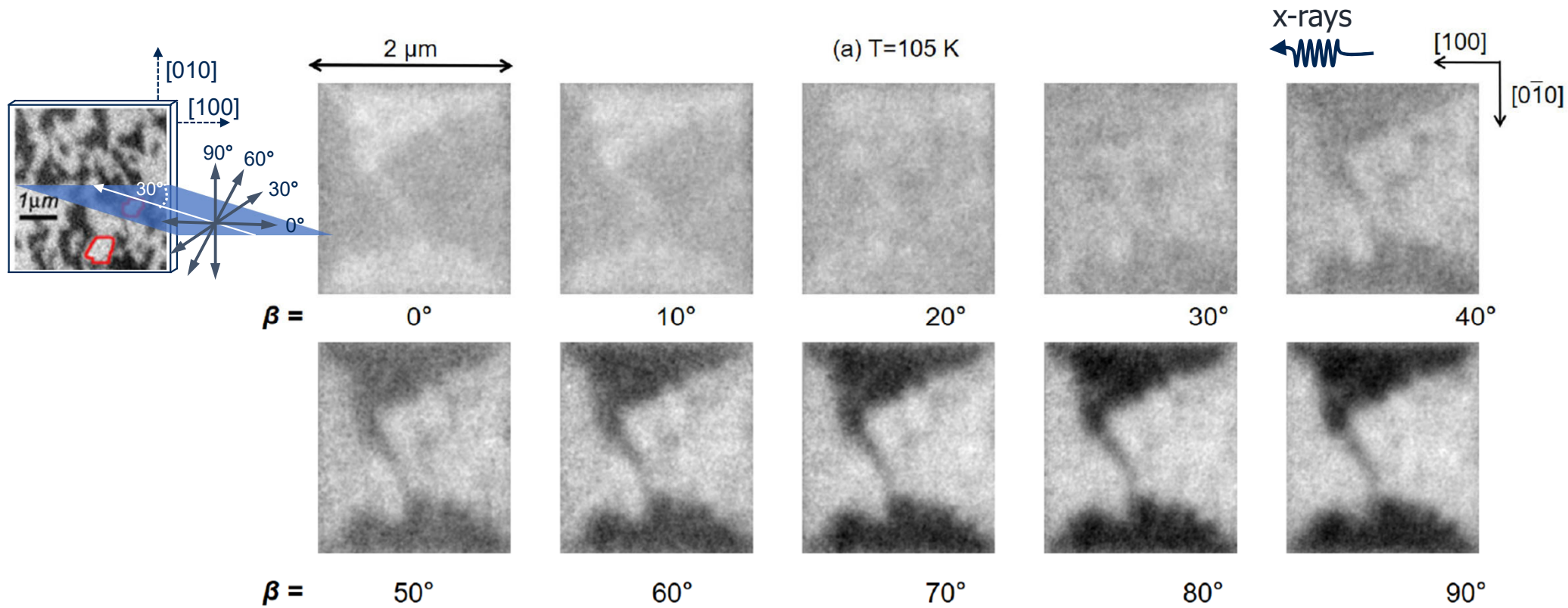
XMLD-PEEM



$$I(\omega, T) = a + b(3\cos^2\omega - 1)\langle A^2 \rangle_T$$

ω = angle between $\vec{E}$ and magnetic spin axis, $\vec{A}$

XMLD-PEEM

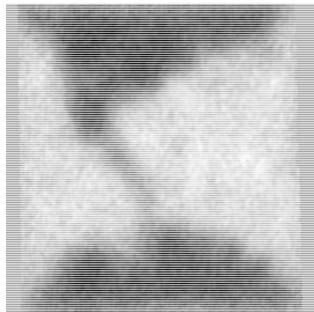


M.S. Lee, Y. Takamura *et al.*, *JAP*, **127**, 203901 (2020)

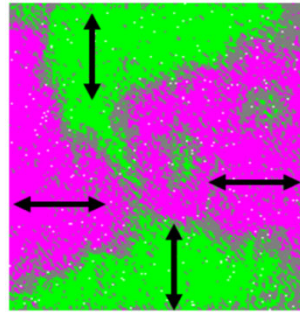
XMLD-PEEM

Square $\langle 100 \rangle$ edges

2 μm

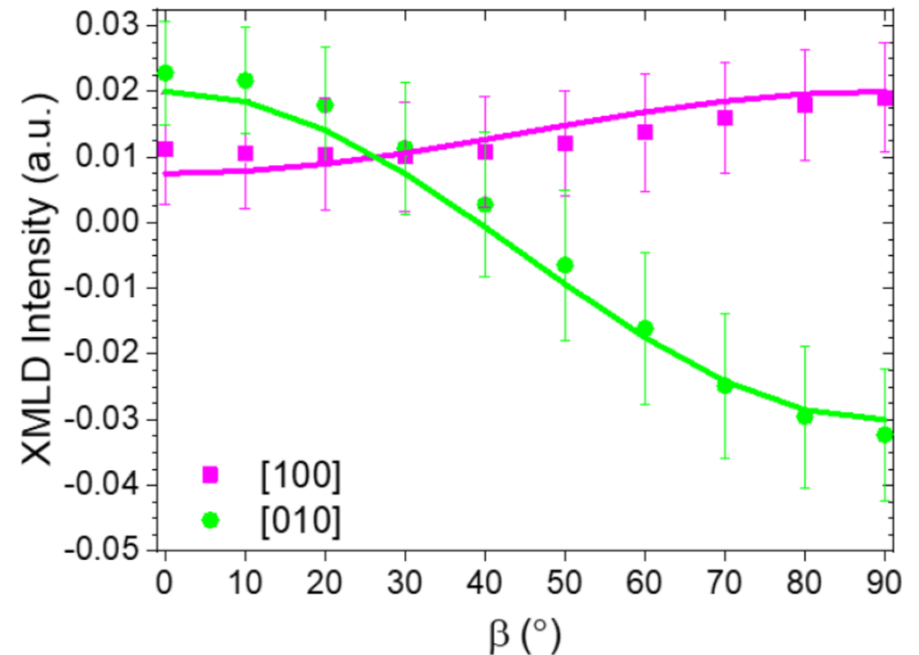


Fe XMLD, $\beta = 90^\circ$



6 u.c. $\text{La}_{0.7}\text{Sr}_{0.3}\text{FeO}_3$

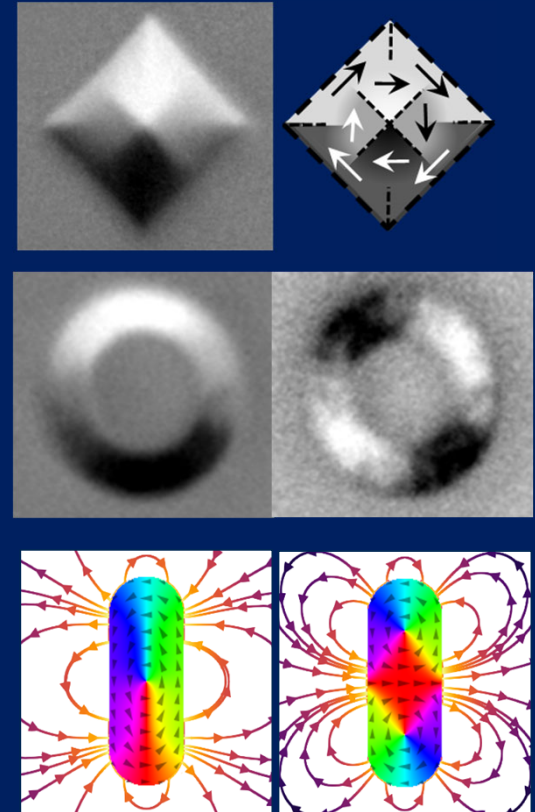
➤ AFM spins lie *in-plane* along the $\langle 100 \rangle$ directions



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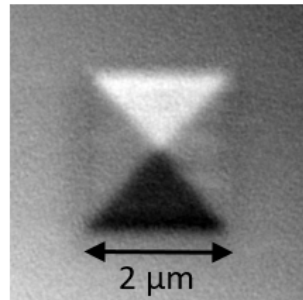
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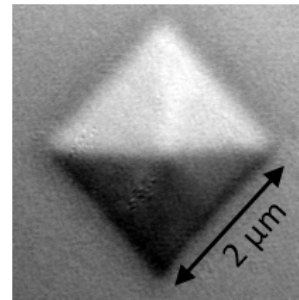


LSMO Micromagnets

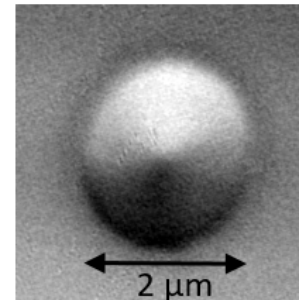
Square



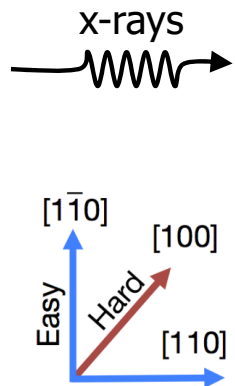
Diamond



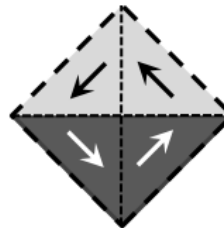
Circle



$T = 230 \text{ K}$
100 u.c. LSMO



Landau



Landau



Vortex

M.S. Lee, Y. Takamura *et al.*,
ACS Nano, **10**, 8545 (2016)

➤ Domain patterns determined by *shape anisotropy*

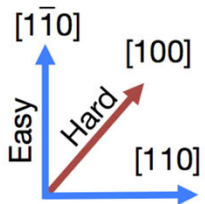
Temperature Evolution

100 u.c. LSMO



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ACS Nano, **10**, 8545 (2016)

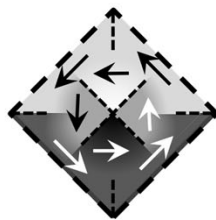
Magnetic
easy axes



Square



Diamond



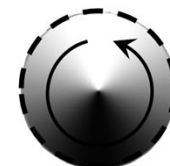
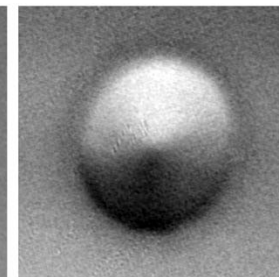
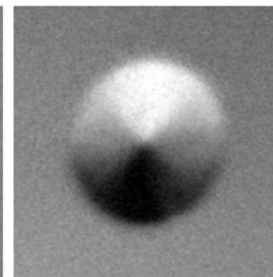
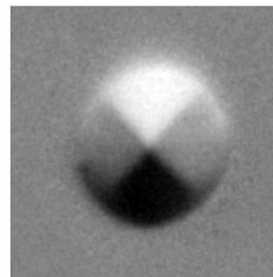
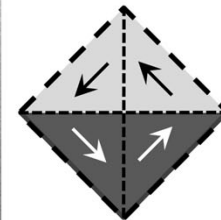
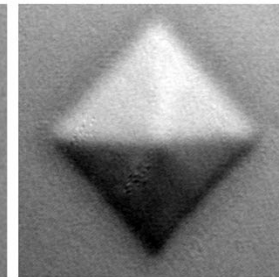
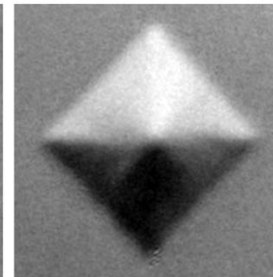
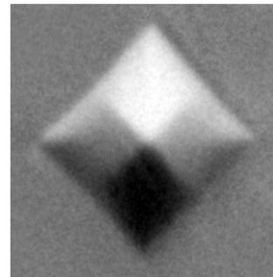
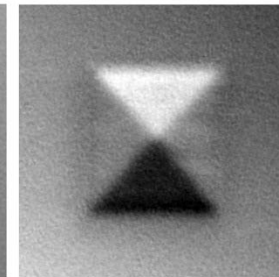
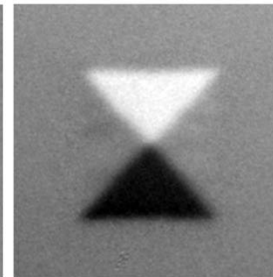
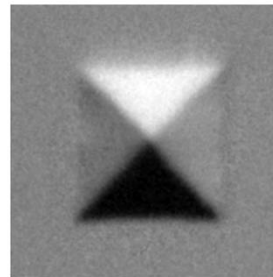
Circle



$T = 32\text{ K}$

150 K

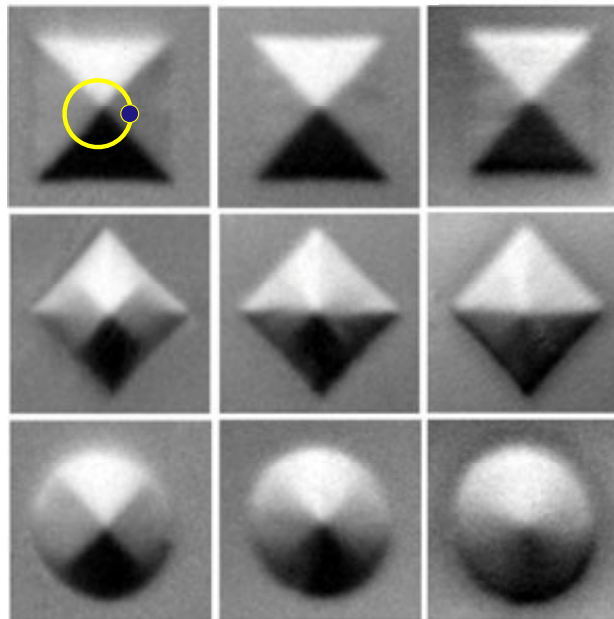
230 K



➤ Magnetocrystalline anisotropy dominates at low temperatures

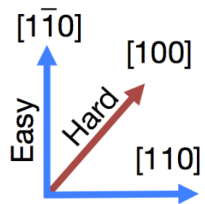
Contrast Analysis

T=32 K 150 K 230 K

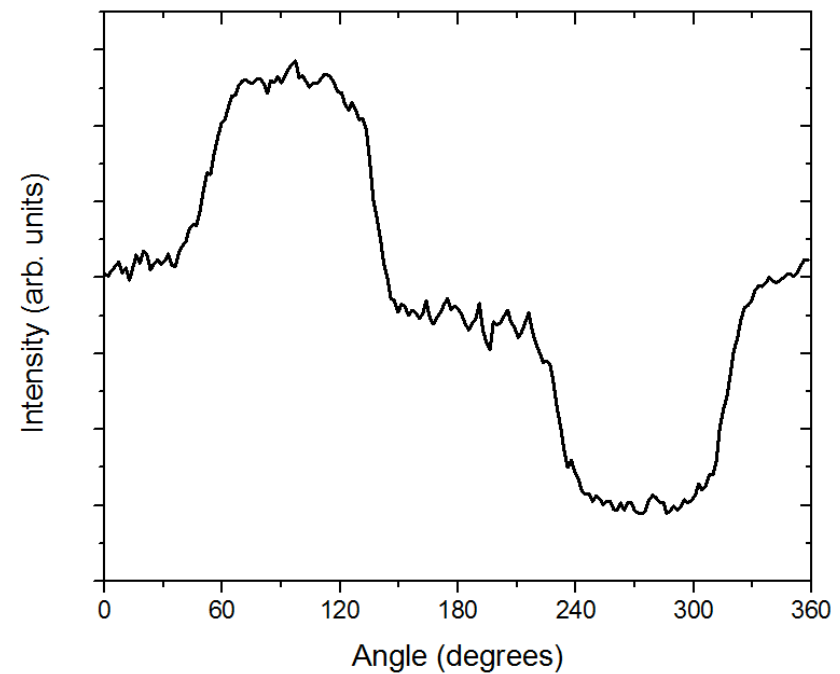


M.S. Lee, Y. Takamura *et al.*, *ACS Nano*, **10**, 8545 (2016)

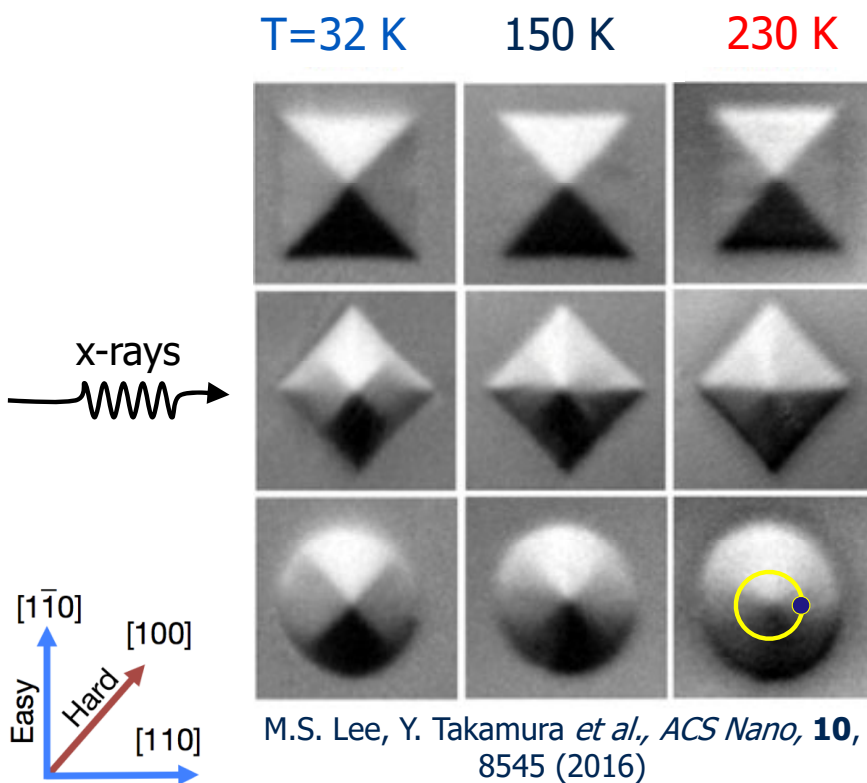
x-rays
→



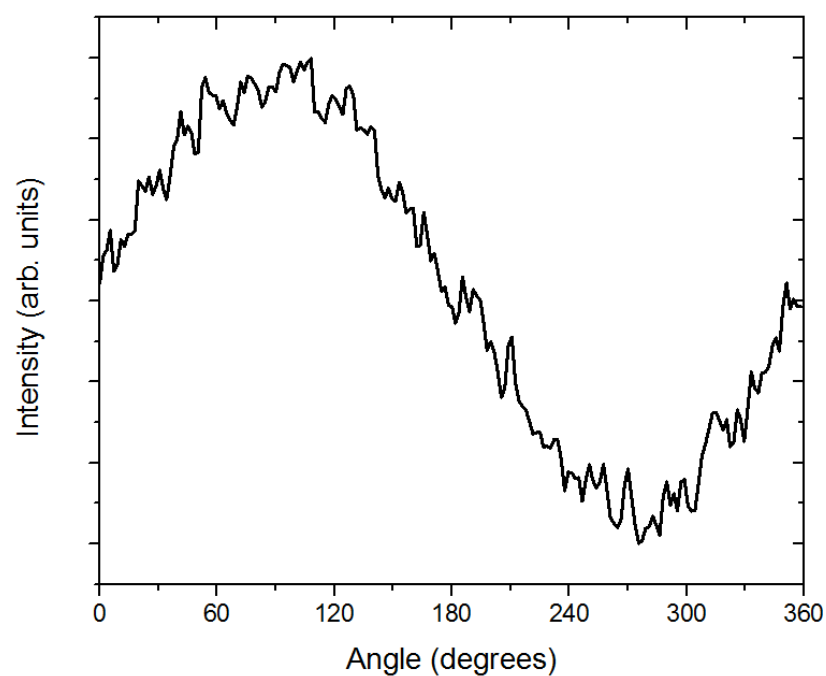
Square at 32 K



Contrast Analysis

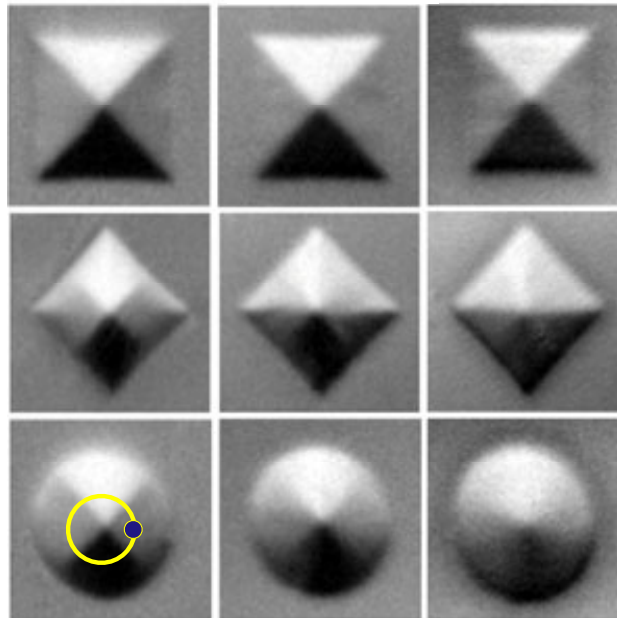


Circle at 230 K

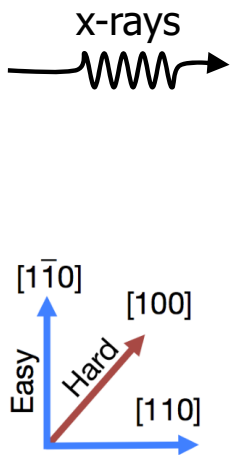


Contrast Analysis

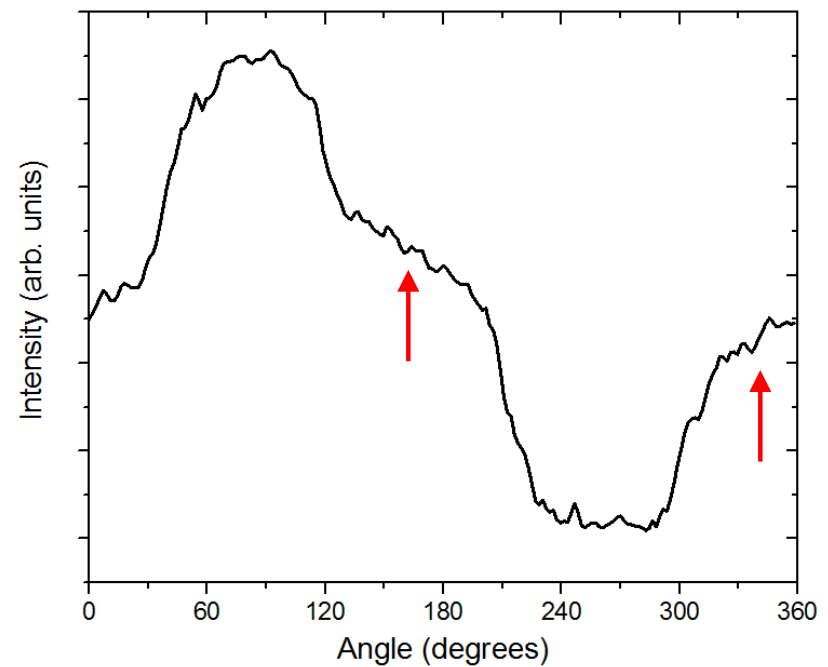
T=32 K 150 K 230 K



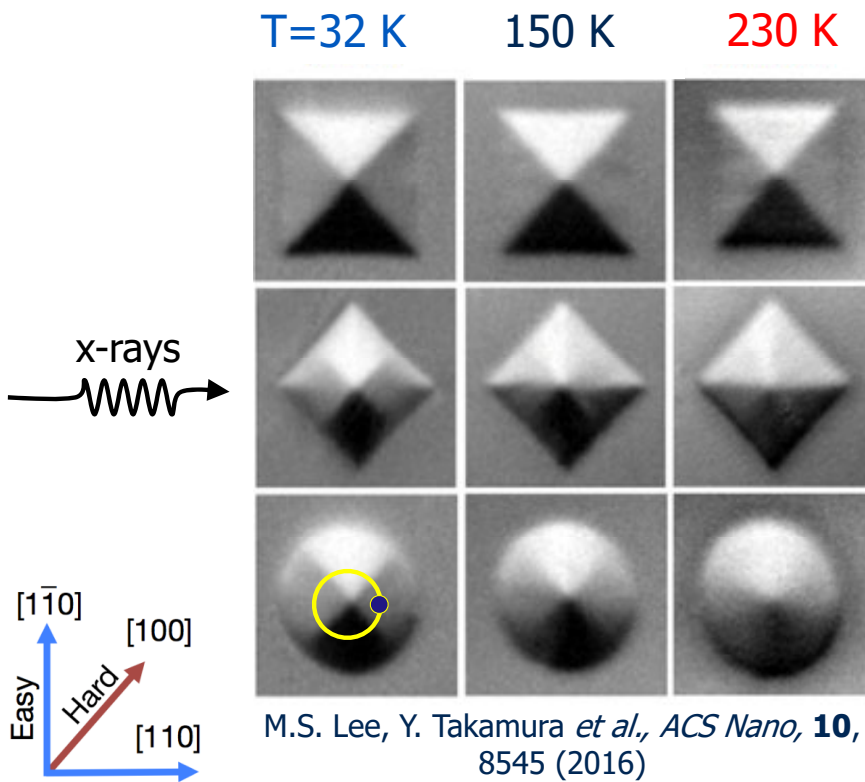
M.S. Lee, Y. Takamura *et al.*, *ACS Nano*, **10**, 8545 (2016)



Circle at 32 K



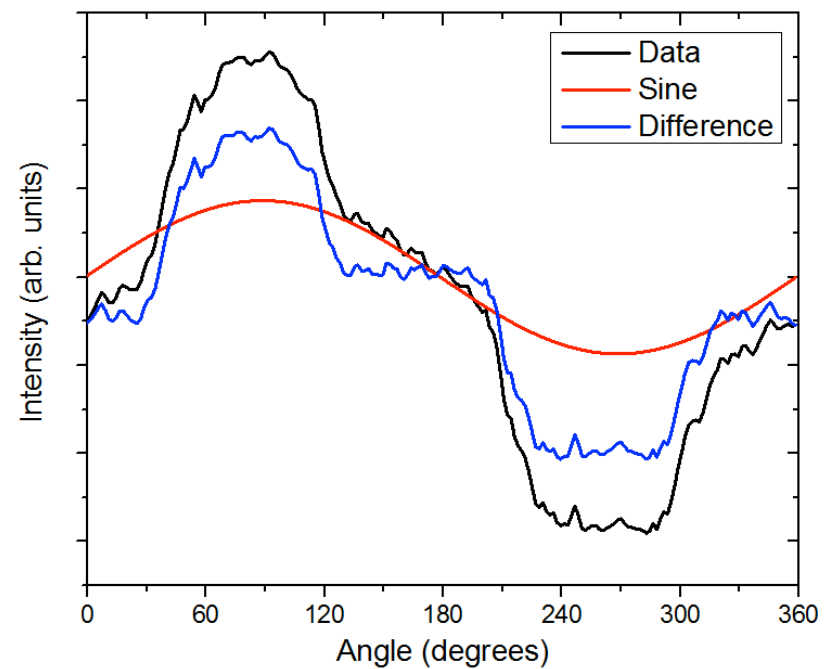
Contrast Analysis



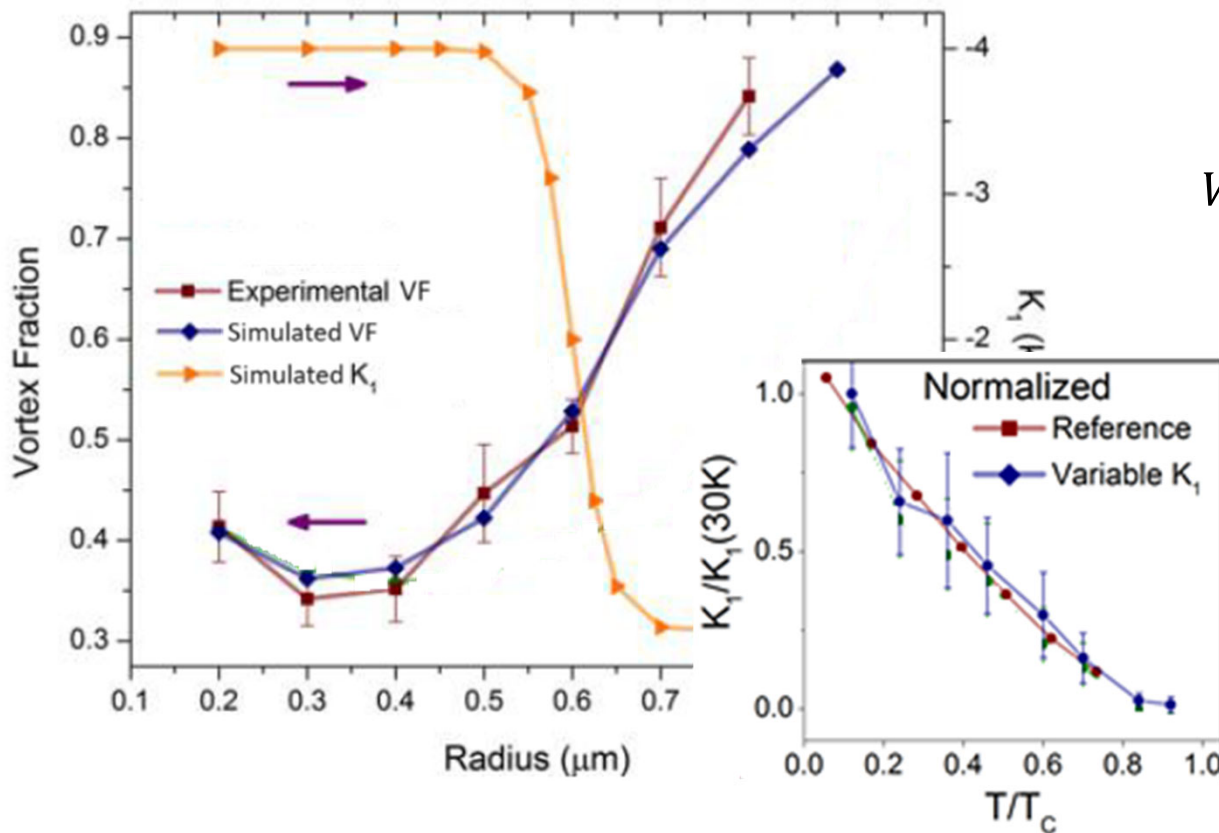
Vortex Fraction (VF)

$$VF = \frac{A_{Sine\ Func}}{A_{Sine\ Func} + A_{Step\ Func}}$$

Circles at 32 K



Magnetocrystalline Anisotropy



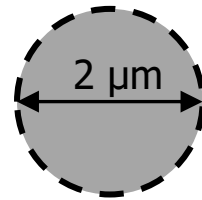
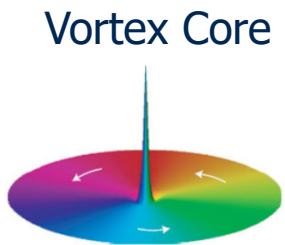
Vortex Fraction (VF)

$$VF = \frac{A_{Sine Func}}{A_{Sine Func} + A_{Step Func}}$$

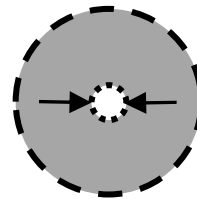
- Comparison to MuMax³ micromagnetic simulations to extract *local* magnetocrystalline anisotropy constant (K_1) values as a function of temperature

M.S. Lee, Y. Takamura *et al.*, *ACS Nano*, **10**, 8545 (2016)

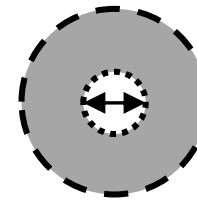
LSMO Micromagnetic Donuts



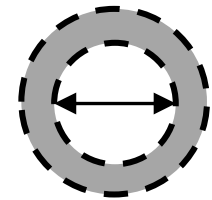
No hole



hole = 0.4 μm



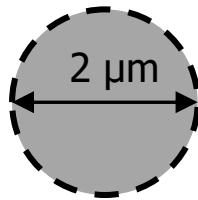
0.8 μm



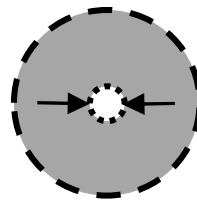
1.2 μm

LSMO Micromagnetic Donuts

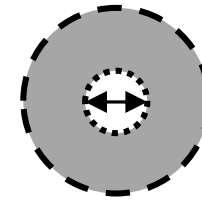
40 K



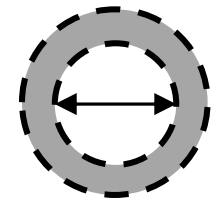
No hole



hole = 0.4 μm



0.8 μm

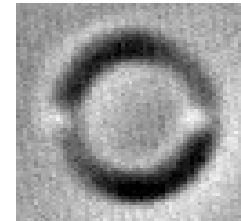
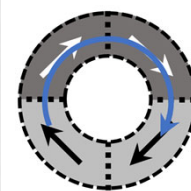
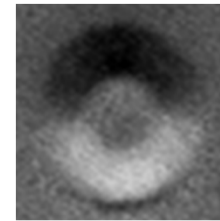
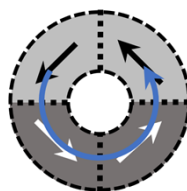
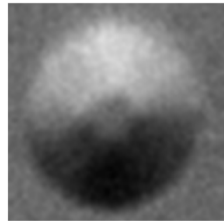
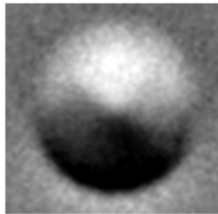
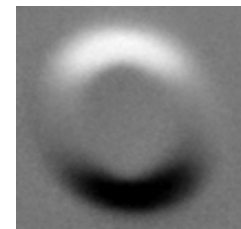
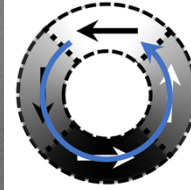
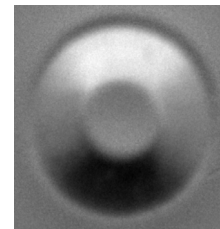
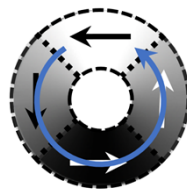
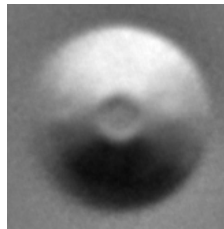
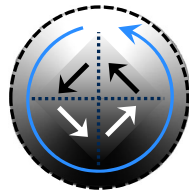
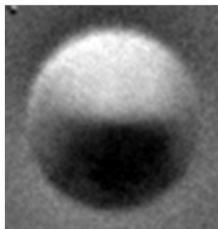


1.2 μm

[010]
↑
Hard
[100]

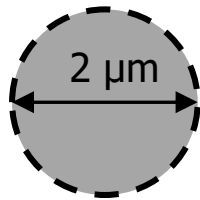
[1 $\bar{1}$ 0]
↑
Easy
[110]

x-rays

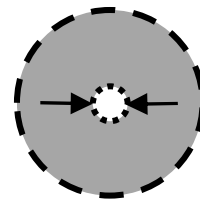


LSMO Micromagnetic Donuts

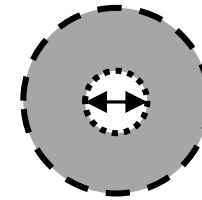
40 K



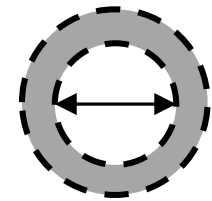
No hole



hole = 0.4 μm

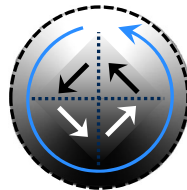
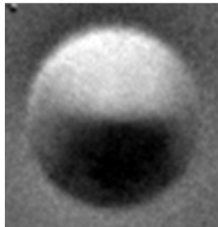


0.8 μm

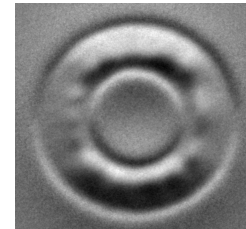


1.2 μm

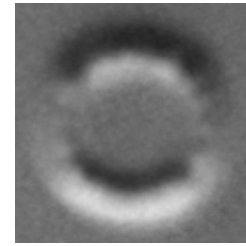
[010]
Hard
[100]



?



?



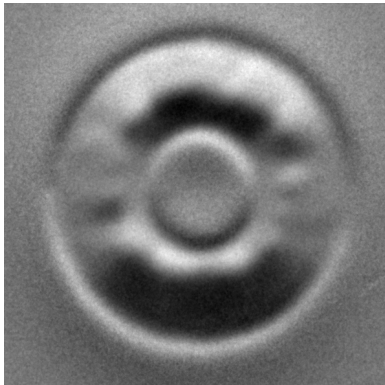
x-rays
←

11/23/2025

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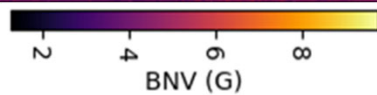
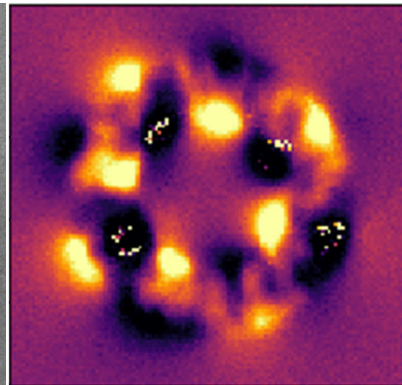
LSMO Micromagnetic Donuts

PEEM image



SNVM image

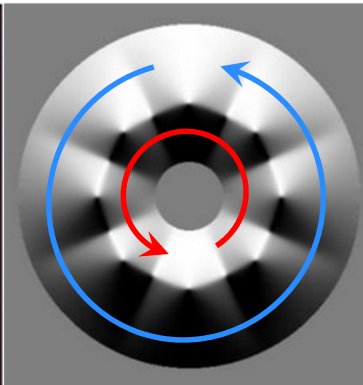
Sensitive to domain walls



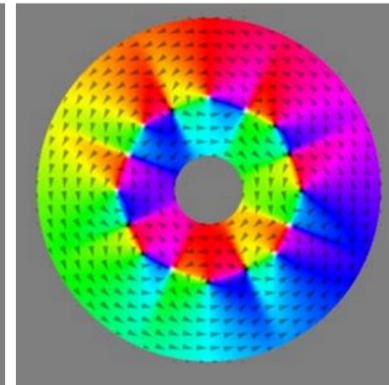
nami

MuMax³ simulations

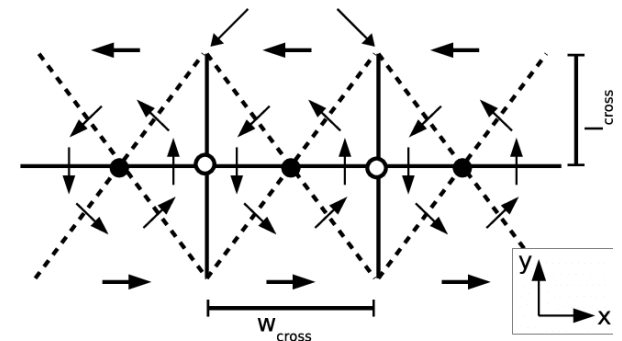
XMCD



m vector field



Cross-tie domains

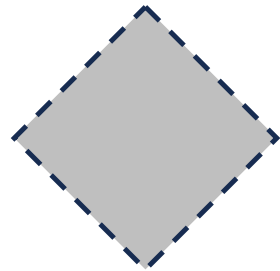


100 u.c. LSMO
[010] Hard [100] x-rays

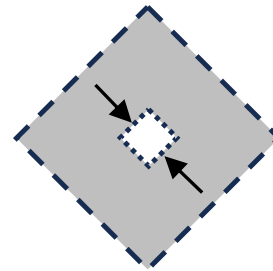
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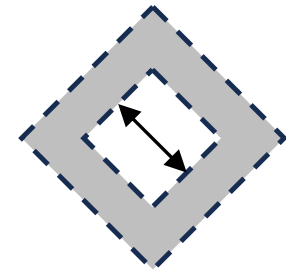
LSMO Micromagnetic Donuts



No hole

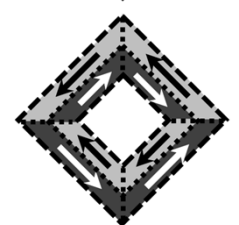
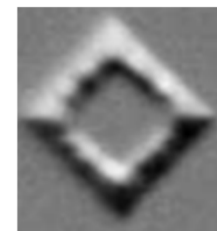
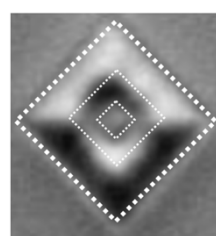
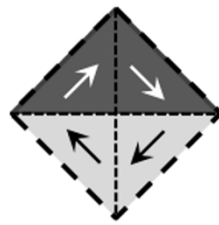
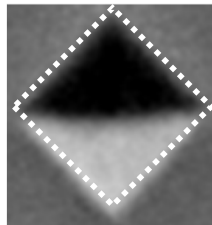
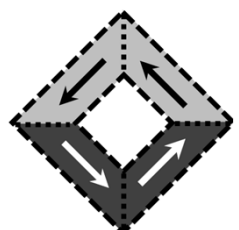
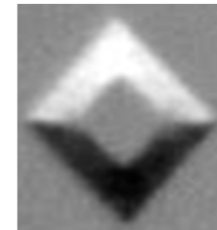
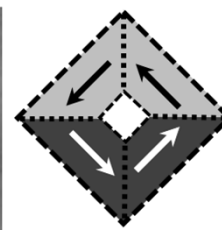
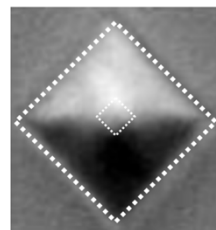
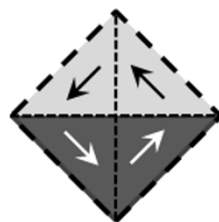
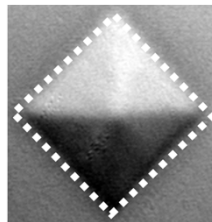


hole = 0.4 μm



hole = 1.0 μm

Edges
along easy
axes



$T=110\text{ K}$
100 u.c. LSMO

[010] ↑ Hard
[100] ← x-rays

11/23/2025

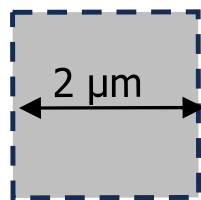
Autonomous University of Madrid

LSMO Micromagnetic Donuts

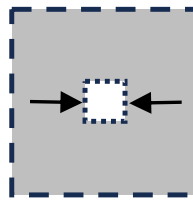
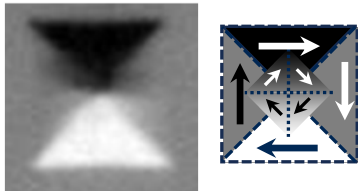
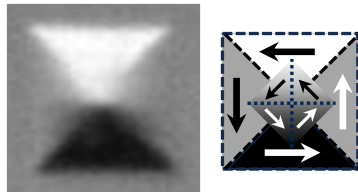
Edges
along hard
axes

$T=110\text{ K}$
100 u.c. LSMO

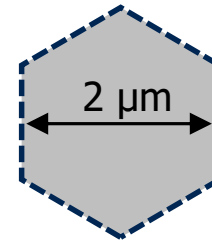
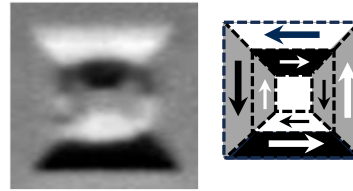
[010] ↑ Hard
[100] ← x-rays



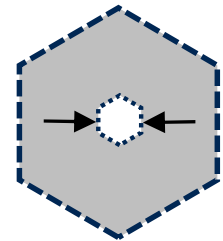
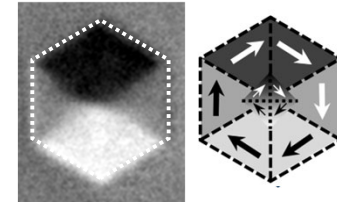
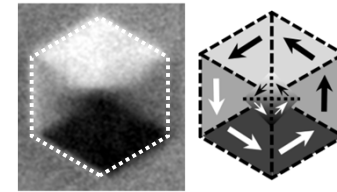
No hole



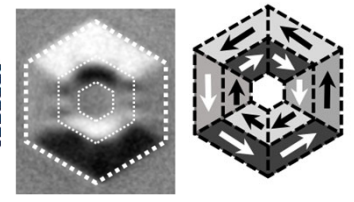
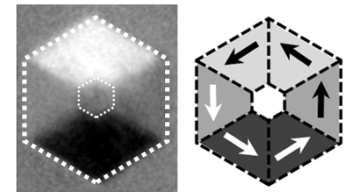
hole = $0.4\text{ }\mu\text{m}$



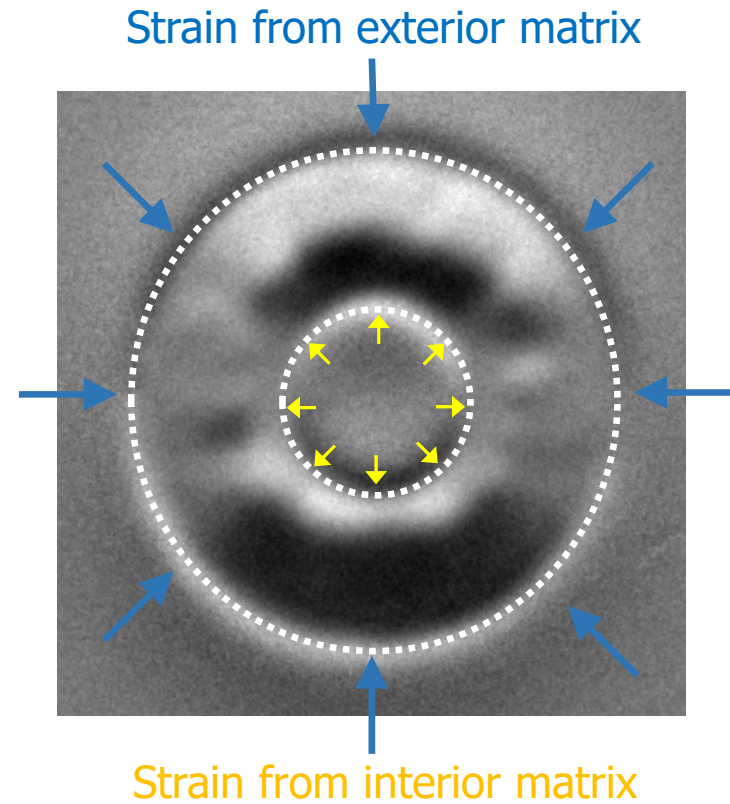
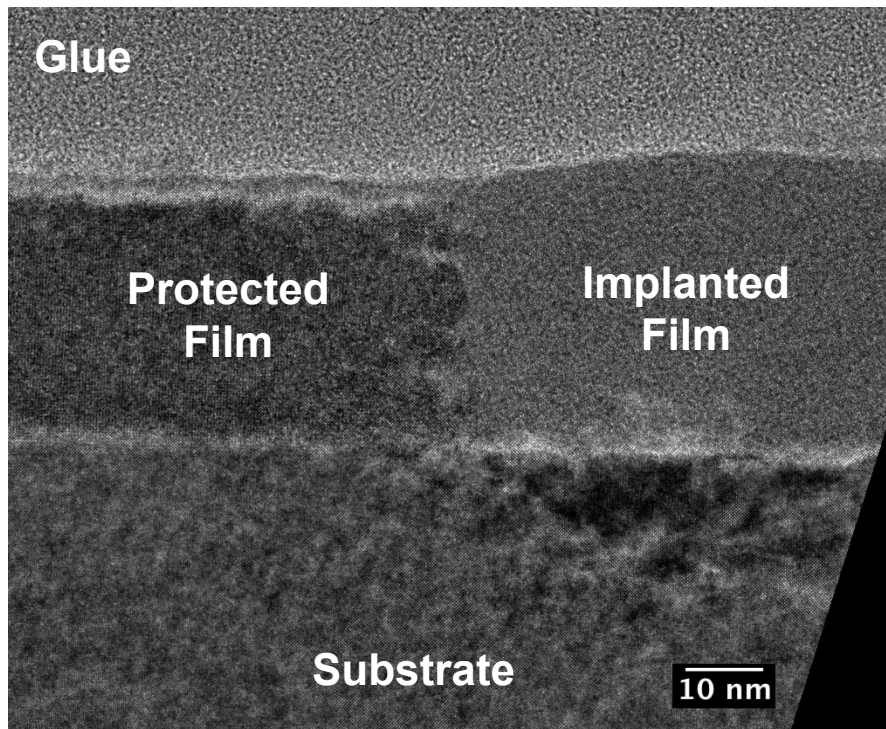
No hole

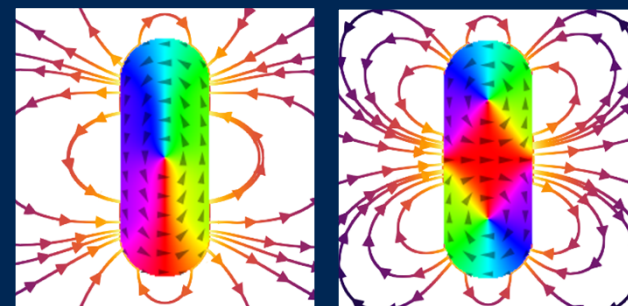


hole = $0.4\text{ }\mu\text{m}$



Matrix Strain Effects

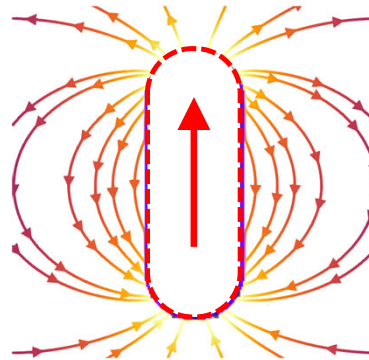




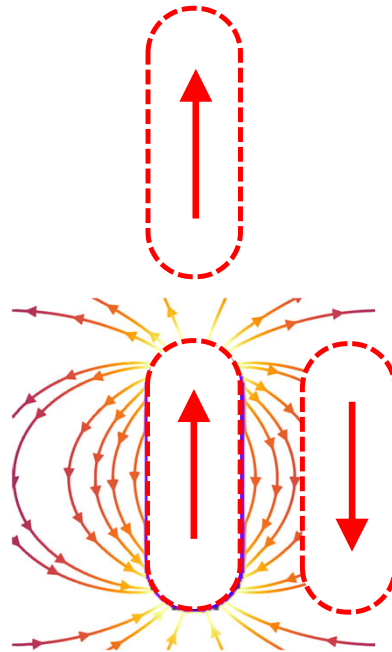
2. $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ Artificial Spin Ice Structures

R.V. Chopdekar, Y. Takamura *et al.*, *PR Materials*, **1**, 024401 (2017)
D.Y. Sasaki, Y. Takamura *et al.*, *PR Applied*, **17**, 064057 (2022)
D.Y. Sasaki, Y. Takamura *et al.*, *under review*

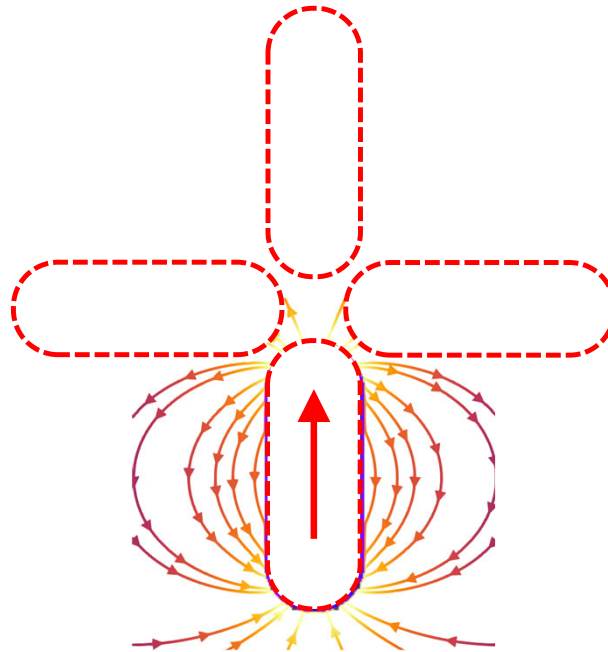
Artificial Spin Ice Structures



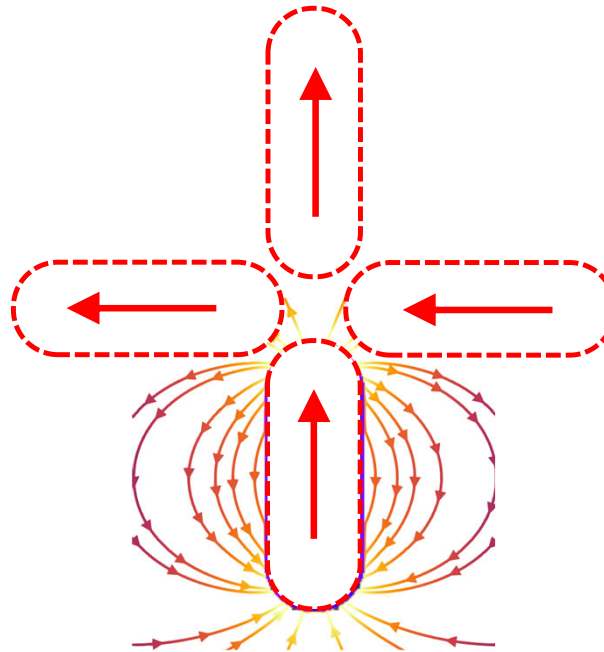
Artificial Spin Ice Structures



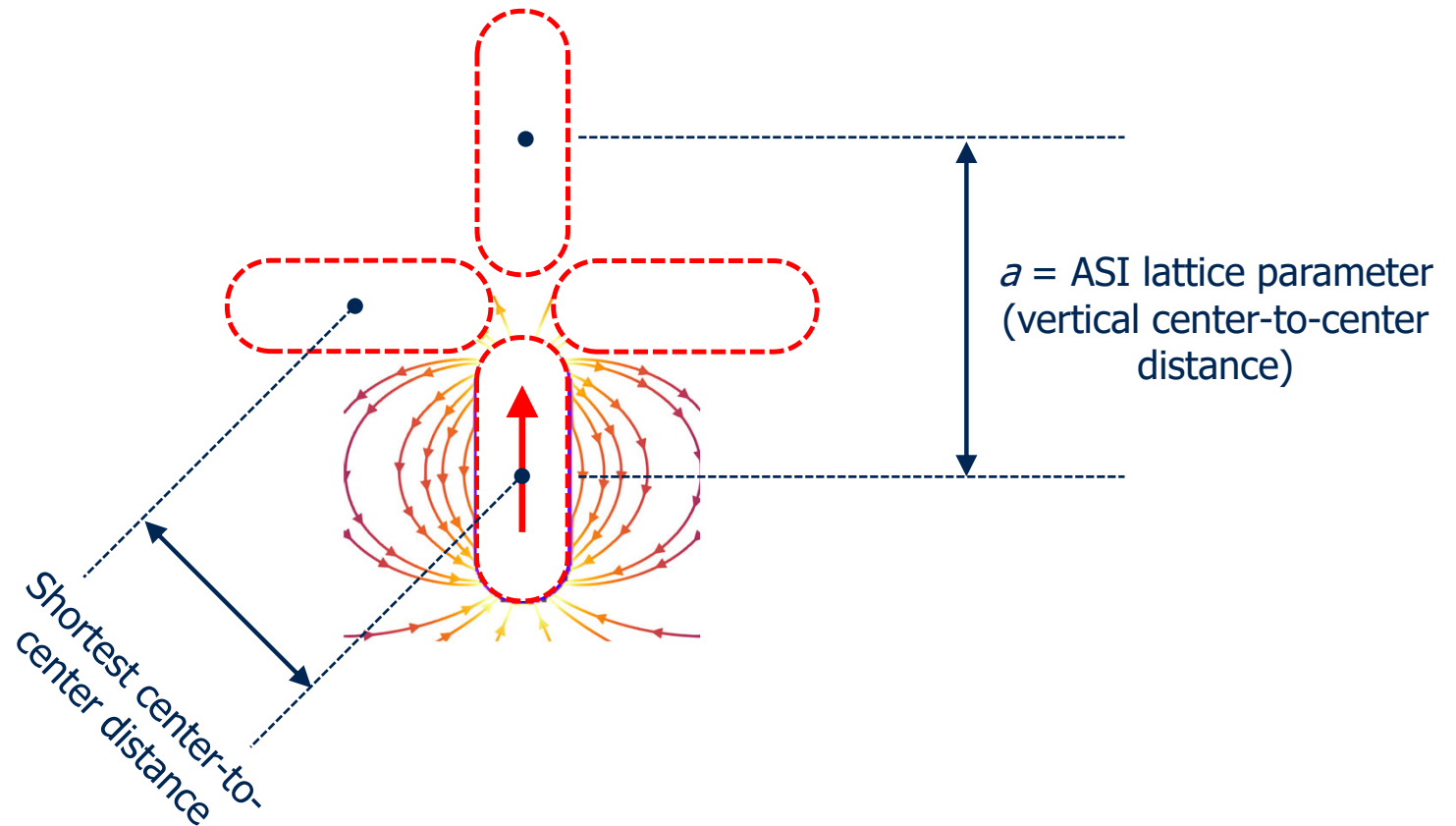
Artificial Spin Ice Structures



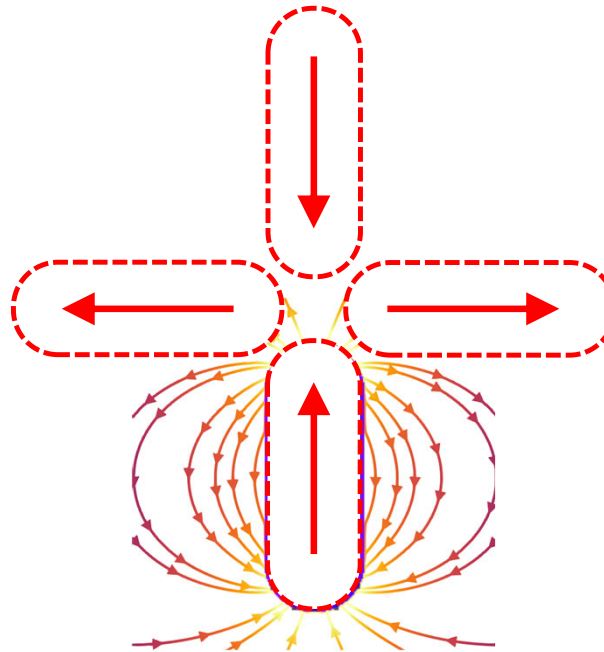
Artificial Spin Ice Structures



Artificial Spin Ice Structures

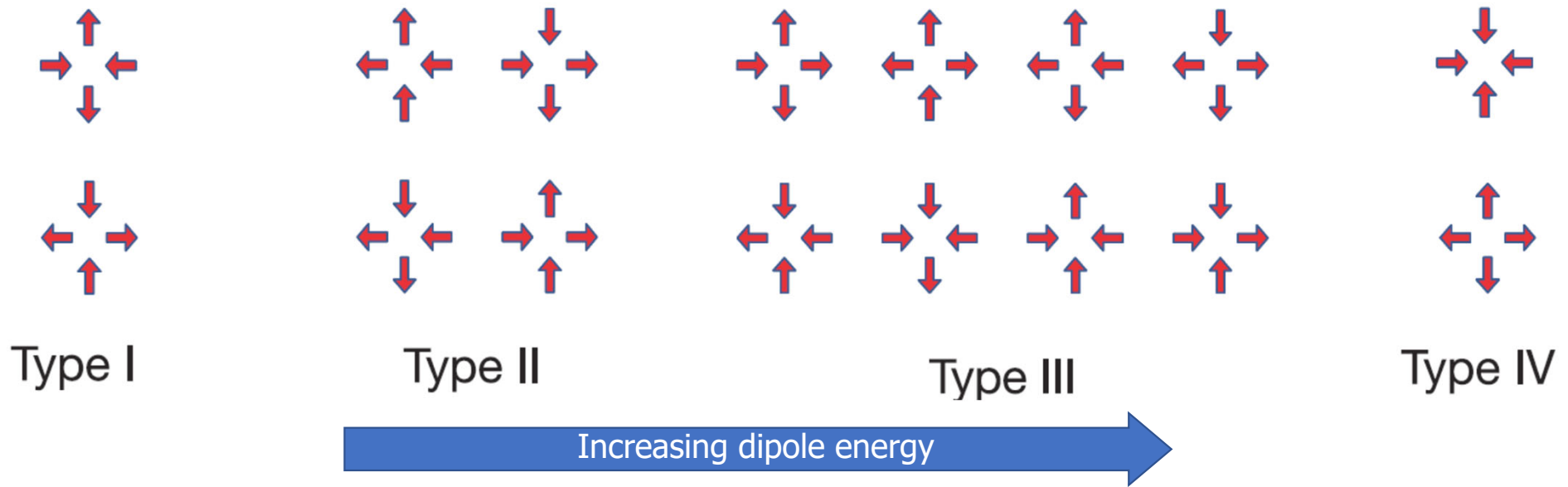


Artificial Spin Ice Structures



Lowest energy
configuration

Artificial Spin Ice Structures

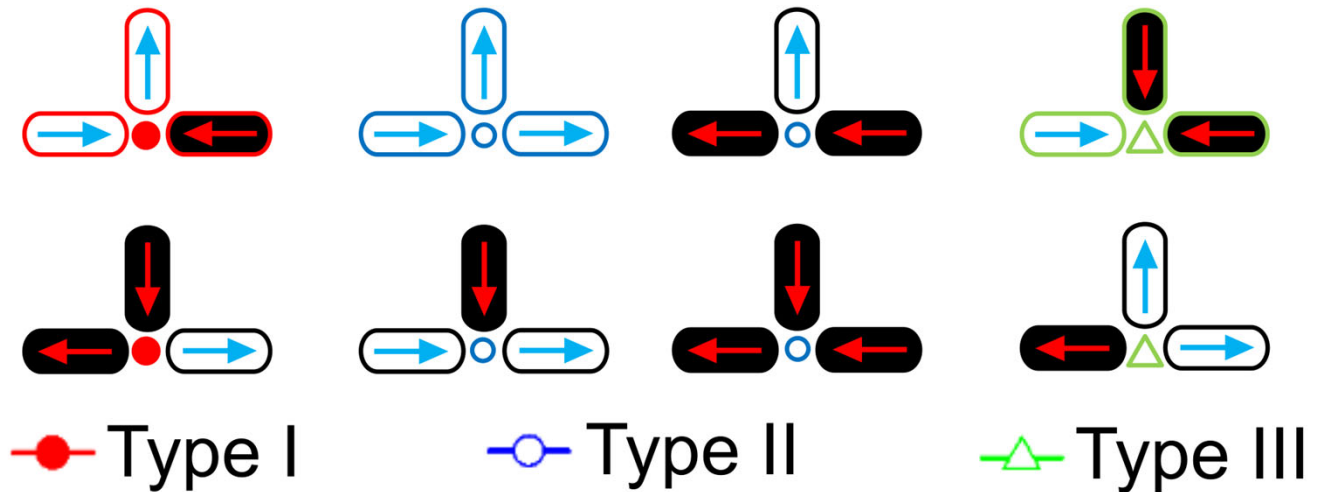
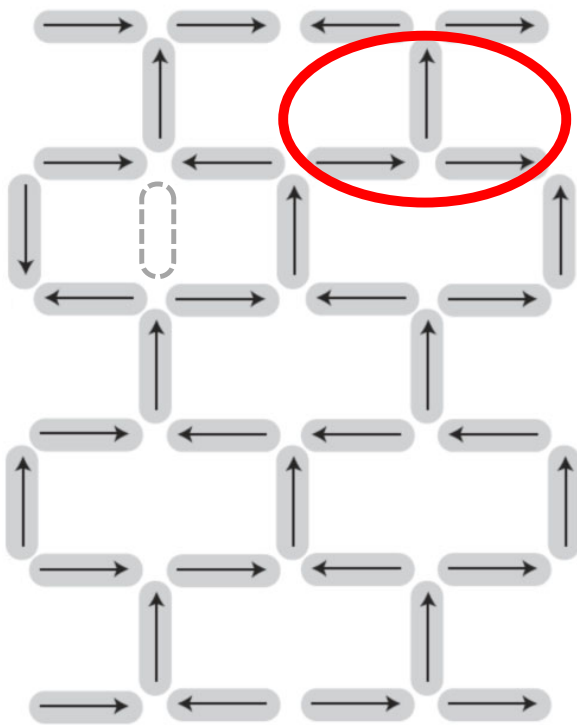


- Magnetic nanoisland arrays with configurations where all magnetic interactions cannot be satisfied simultaneously

Wang *et al.*, *Nature*, **439**, 303 (2006)

Brickwork Arrays

3-island vertices



Increasing dipole energy

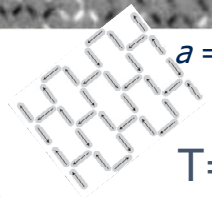
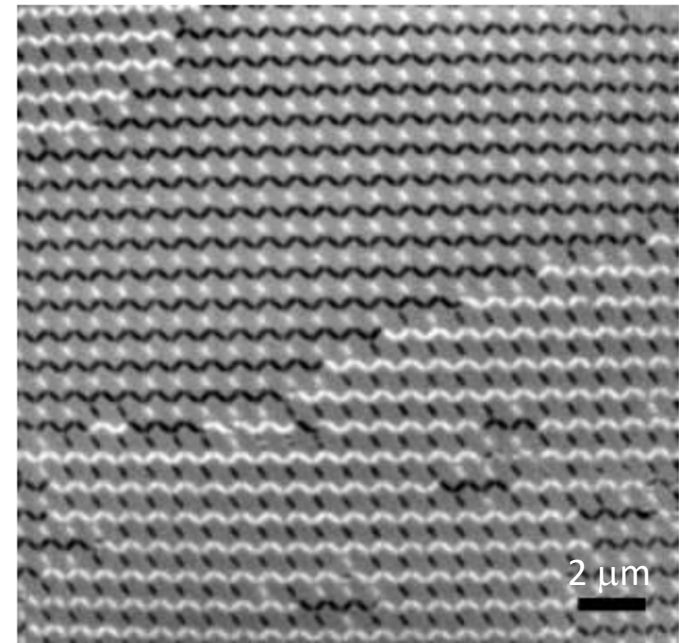
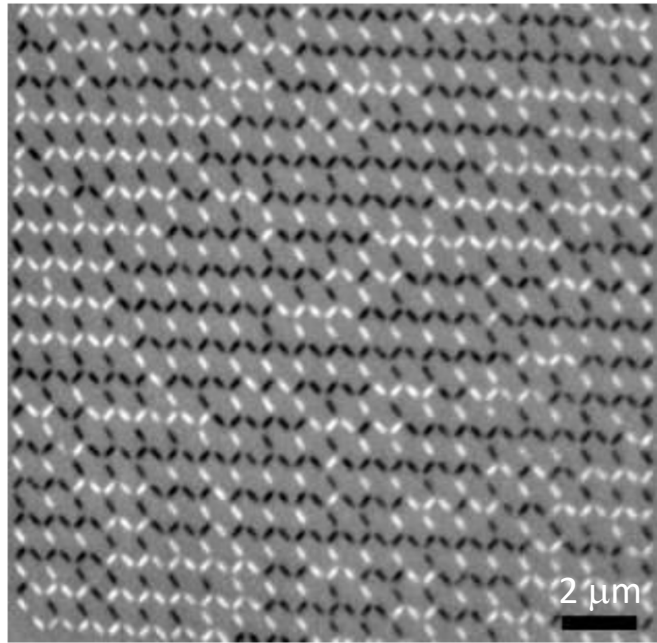
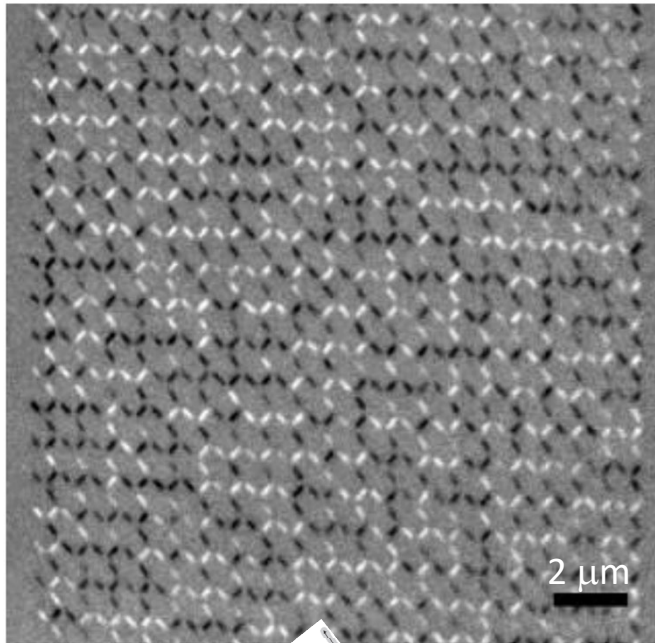
$$\text{Coupling strength, } J = \frac{m^2}{a^3}$$

m = nanoisland moment
 a = ASI lattice parameter

Effect of Coupling Strength

Random

Ordered patches



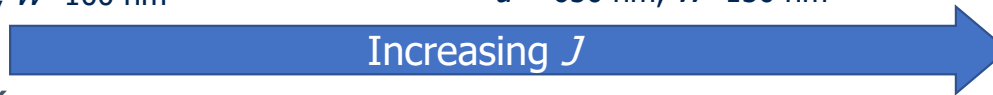
$a = 650 \text{ nm}$, $W = 100 \text{ nm}$

$a = 650 \text{ nm}$, $W = 150 \text{ nm}$

$a = 600 \text{ nm}$, $W = 175 \text{ nm}$
 $L = 470 \text{ nm}$

x-rays
←

$T = 110 \text{ K}$

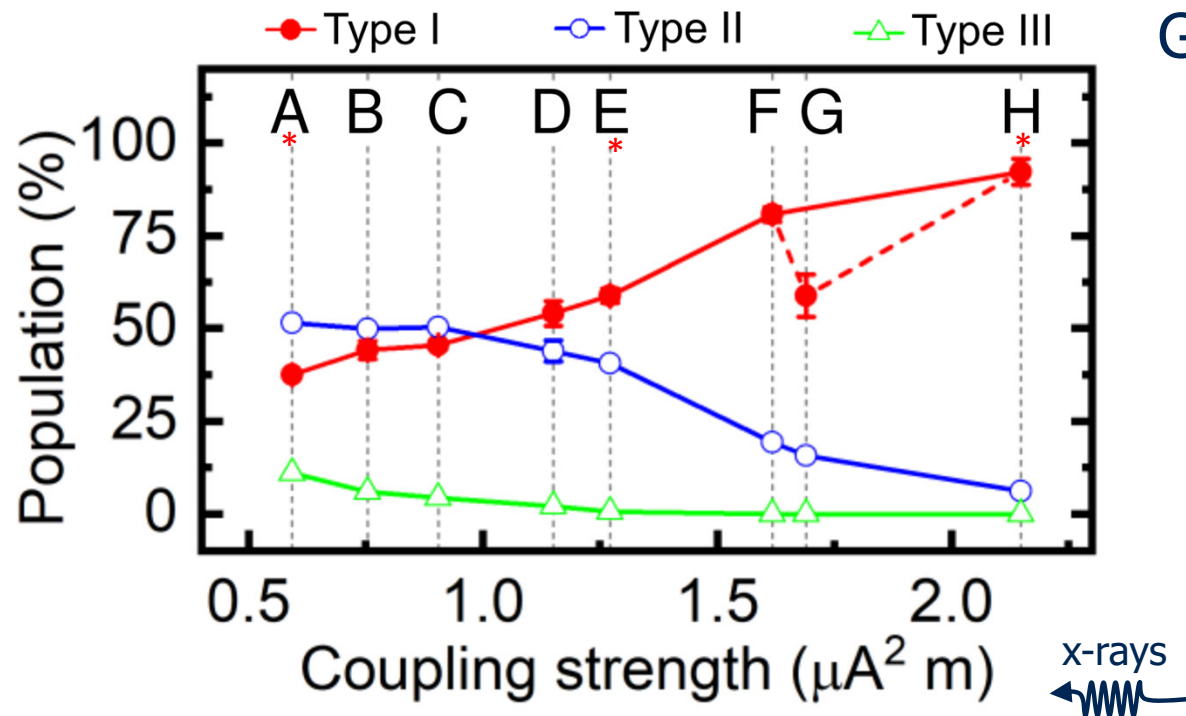


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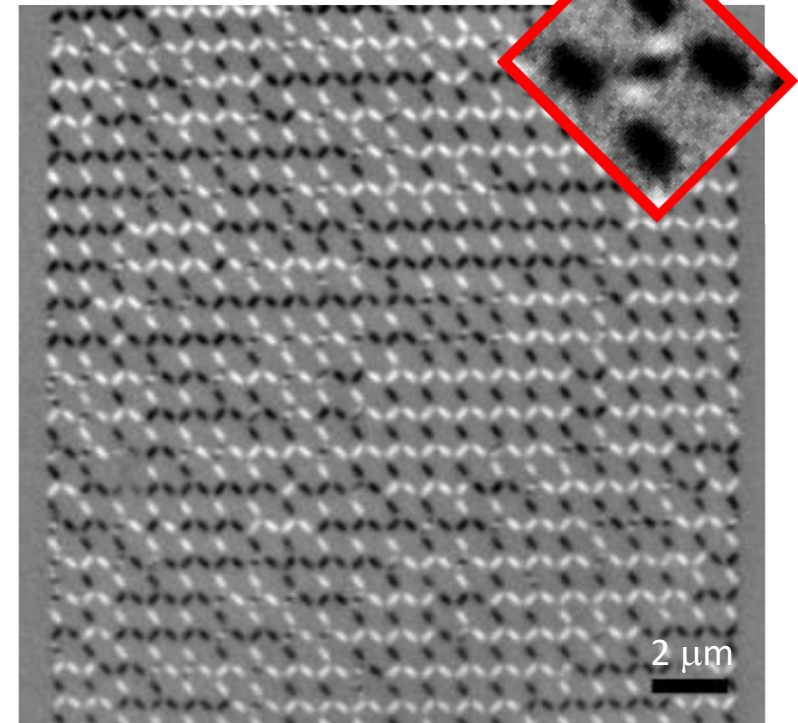
48

Effect of Coupling Strength



G

$a = 650 \text{ nm}$, $W = 175 \text{ nm}$



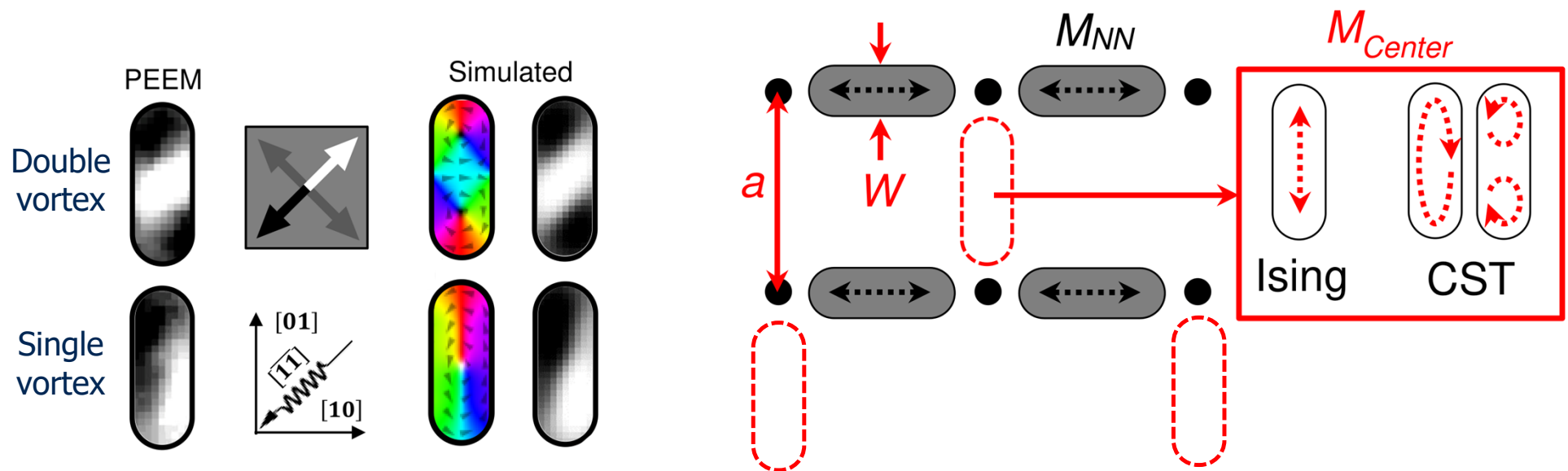
$T = 110 \text{ K}$

D. Sasaki, Y. Takamura *et al.*, *PR Applied*, **17**, 064057 (2022)

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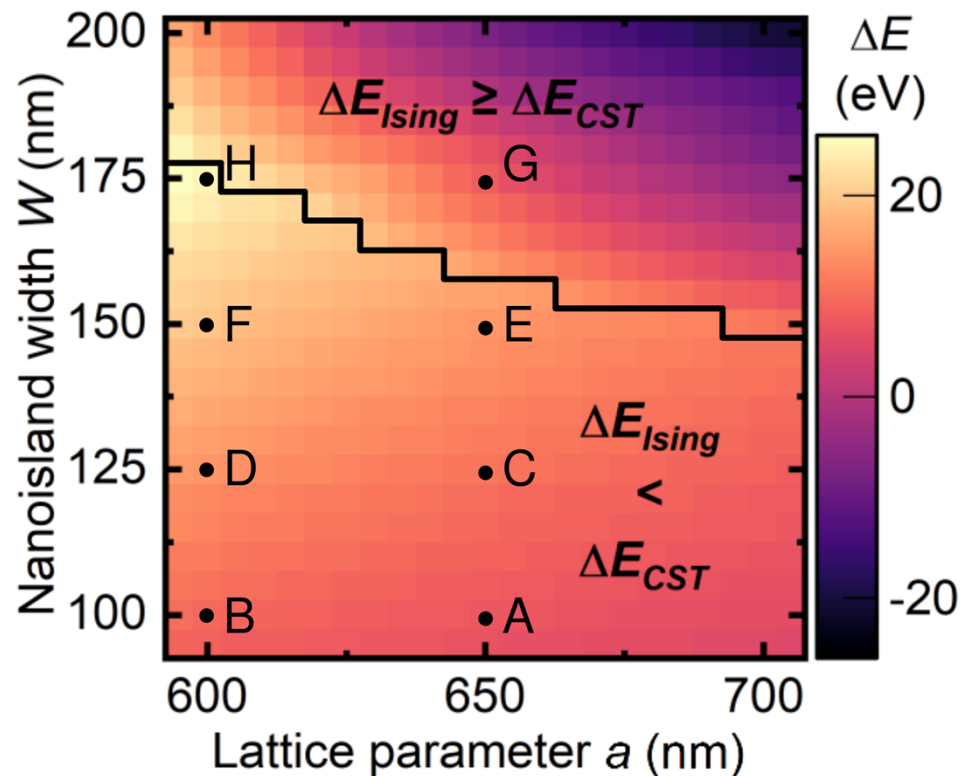
Complex Spin Textures (CSTs)



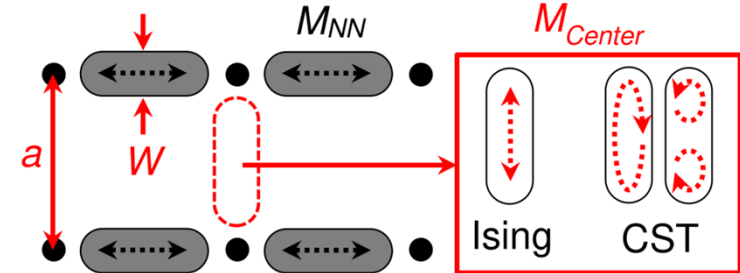
- 95% of observed CSTs occur in vertical nanoislands

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Energetics of CSTs



MuMax³ simulations

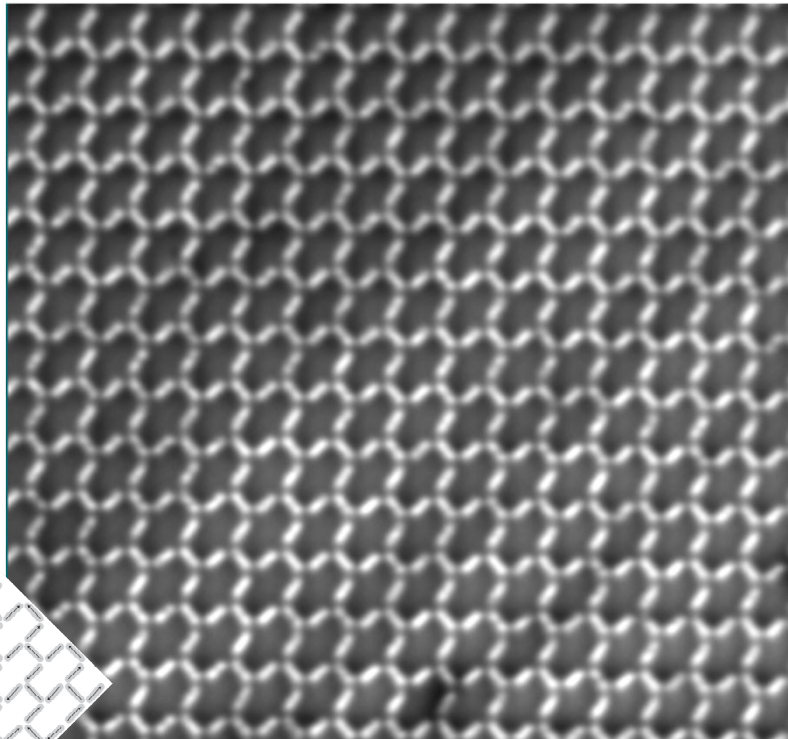


- Prevalence of CSTs controlled by ASI geometric parameters (W and a) and LSMO magnetic parameters

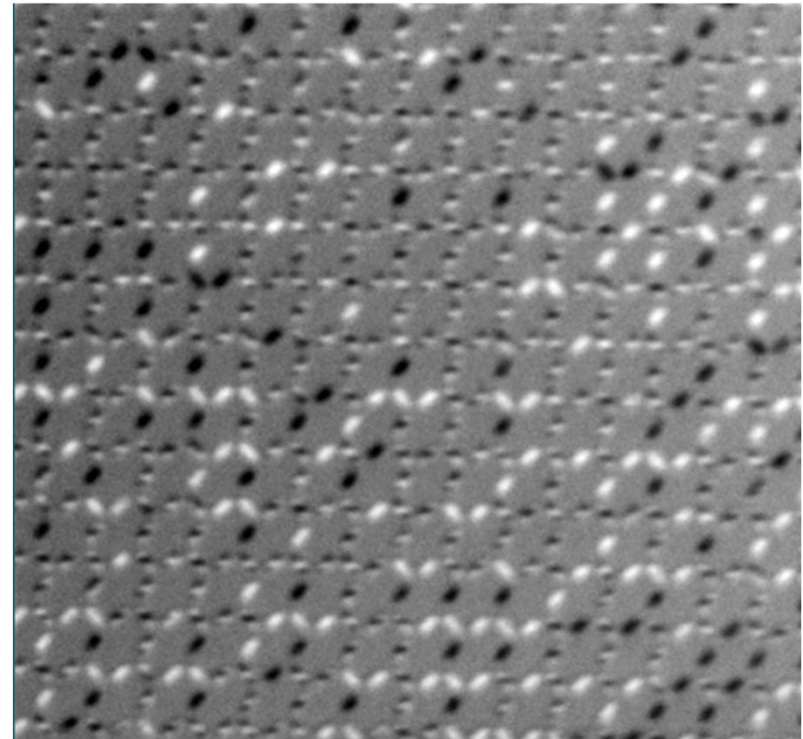
D. Sasaki, Y. Takamura *et al.*, *PR Applied*, **17**, 064057 (2022)

Prevalence of CSTs

XA image



XMCD image



T=110 K

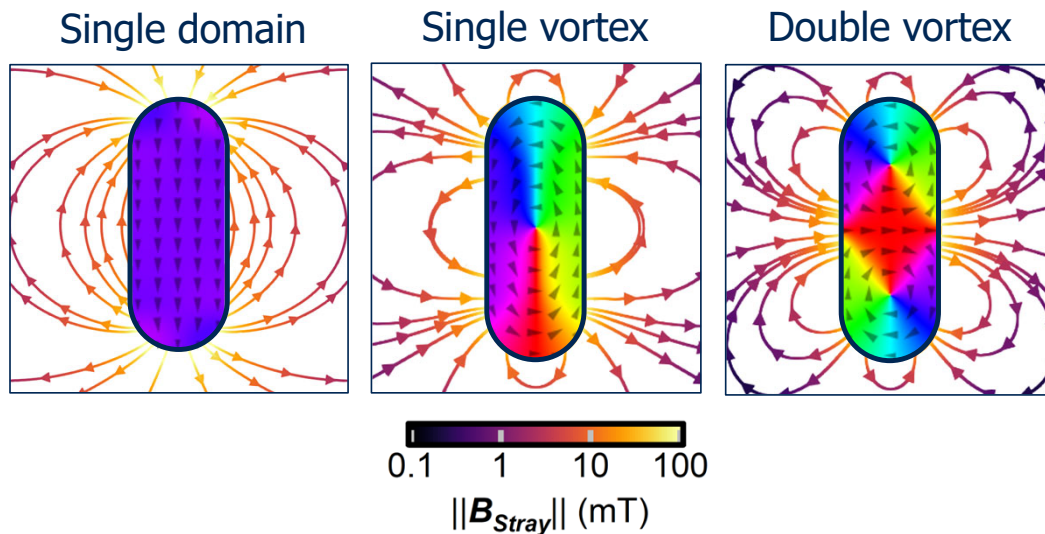
x-rays



$a = 675 \text{ nm}$, $W = 175 \text{ nm}$

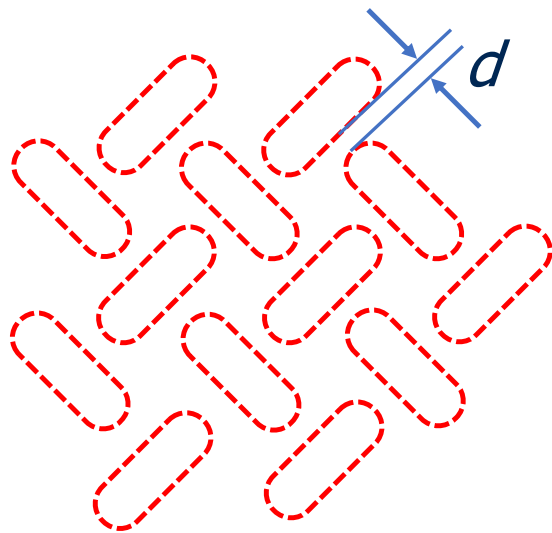
$L = 470 \text{ nm}$

ASI Structures Designed for CSTs



- Balance between *intra*-island shape anisotropy and *inter*-island dipolar interactions
- Modified stray field patterns provide opportunities for new ASI structures or exotic phase transitions in existing ASI structures

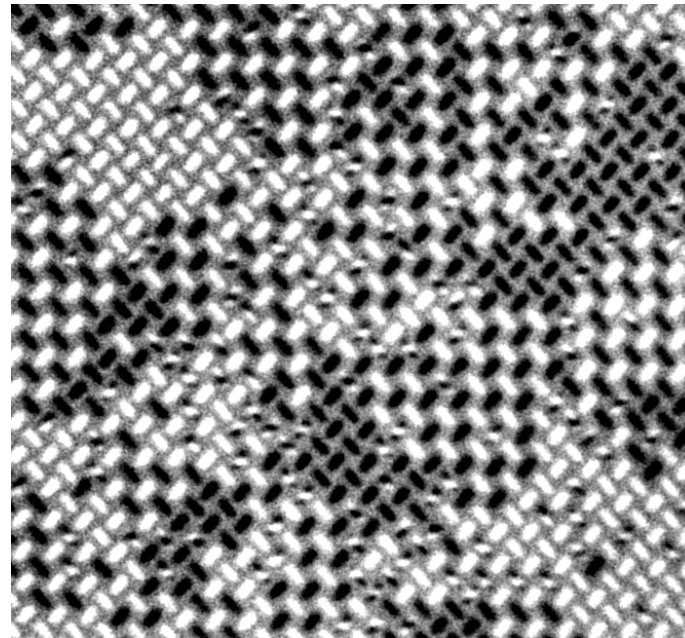
Pinwheel Structures



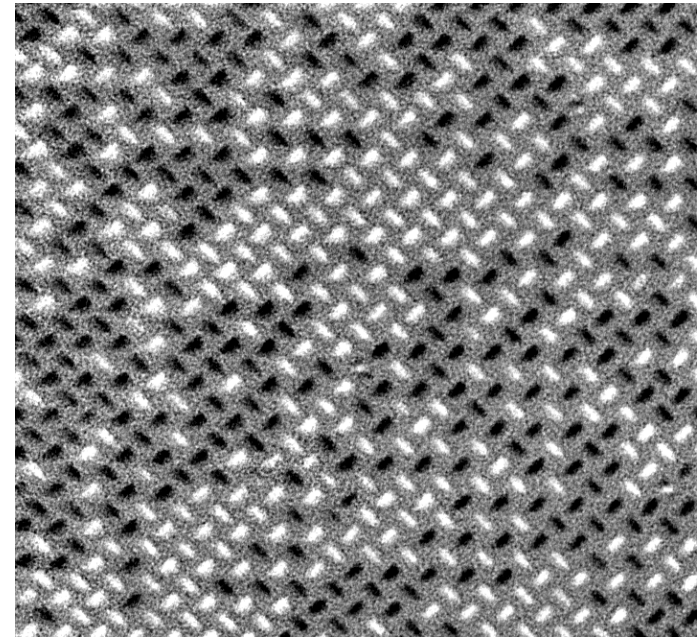
$W = 175 \text{ nm}$
 $L = 470 \text{ nm}$



$d = 25 \text{ nm}$

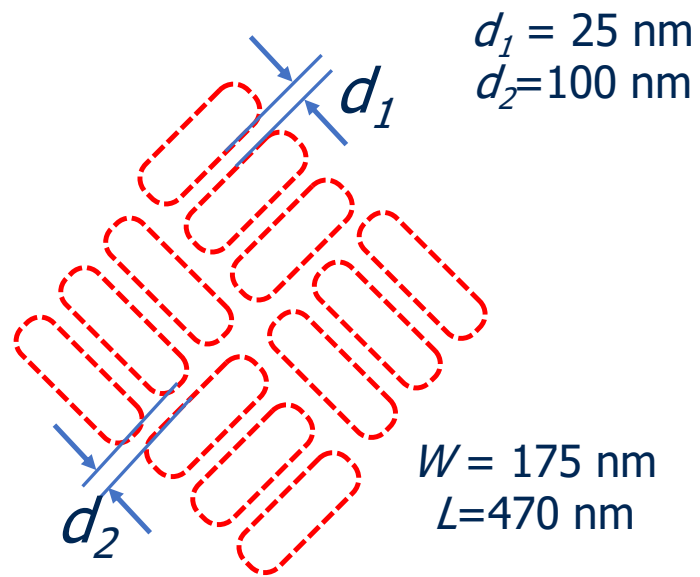


$d = 150 \text{ nm}$



- Decreasing d results in increased CST formation

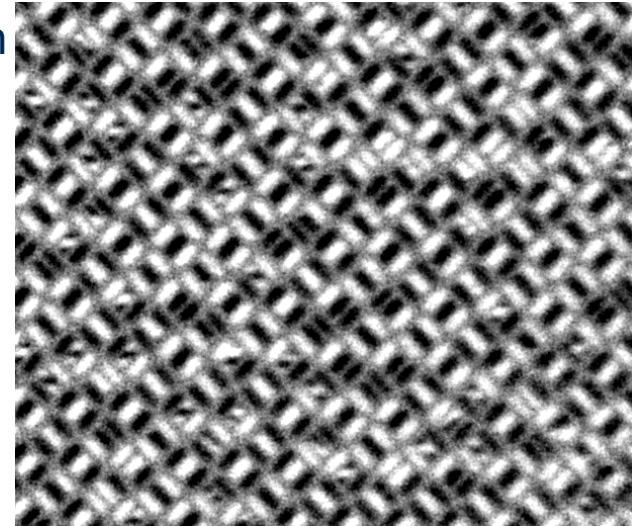
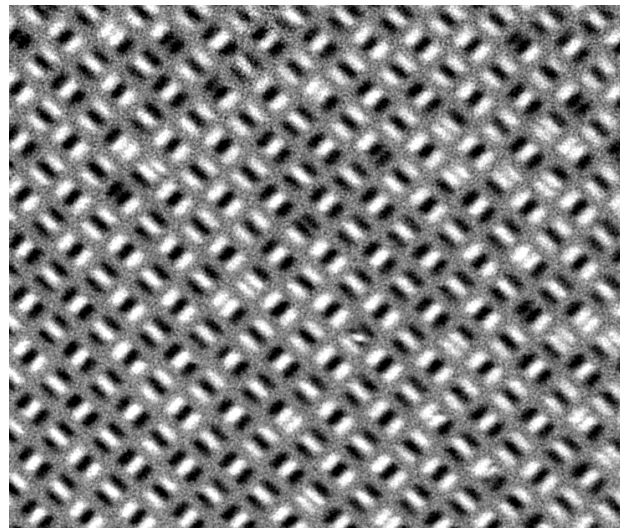
Trident Structures



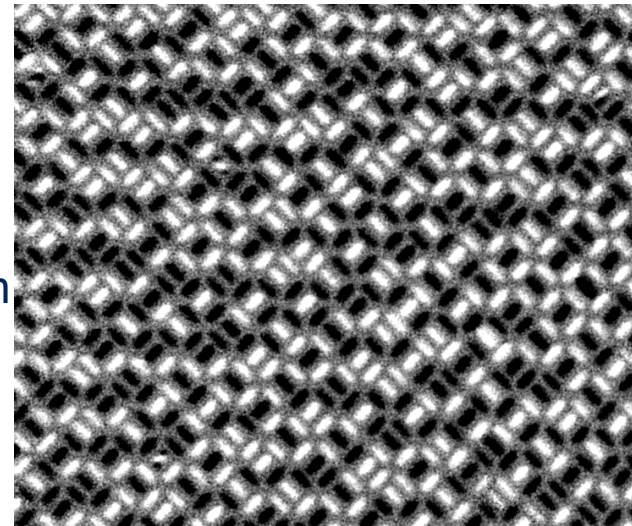
- CST formation depends on d_1 and d_2 and predominantly in center nanoisland

$d_1 = 25 \text{ nm}$
 $d_2 = 25 \text{ nm}$

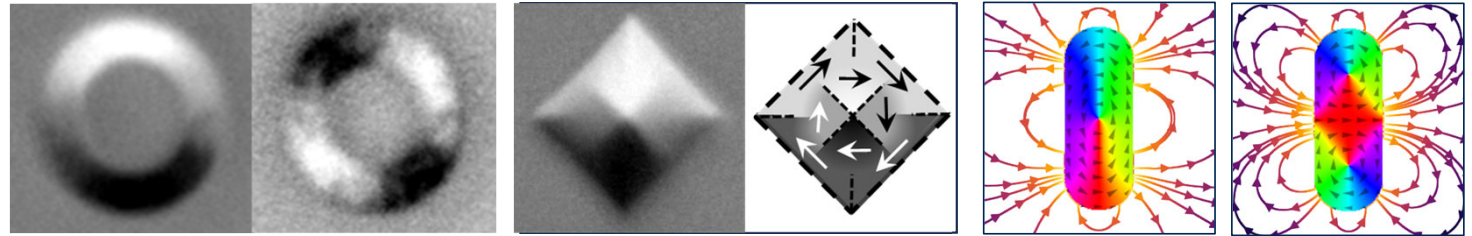
x-rays
←



$d_1 = 100 \text{ nm}$
 $d_2 = 25 \text{ nm}$



Summary



- Complex oxide heterostructures and micro-/nanostructures offer a rich materials playground where structural, chemical, magnetic, and electronic interactions offer fine control of functionality.
- Patterning of LSMO microstructures allows for:
 - Manipulation of a rich variety of FM (and AFM) spin textures
 - Complex competition between shape anisotropy, magnetocrystalline anisotropy, exchange coupling, and magnetoelastic energy.
- LSMO magnetic parameters enable formation of CSTs under specific ASI geometries.
 - Stray field patterns modify magnetic interactions between magnetic nanoislands