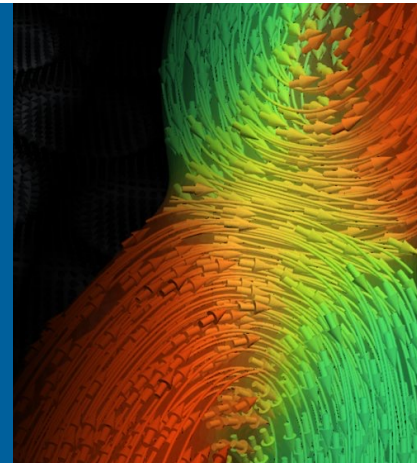


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WORKSHOP: NANOMAGNETISM IN 3D



# FRONTIERS OF IMAGING AND UNDERSTANDING 3D NANOSCALE MAGNETIC STRUCTURES



***C. PHATAK***

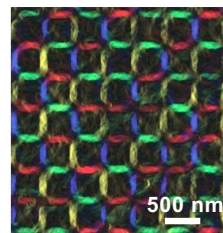
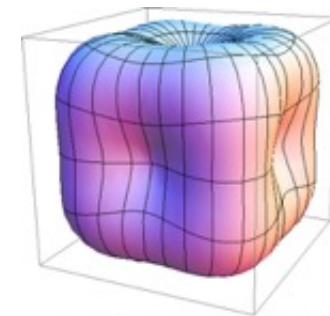
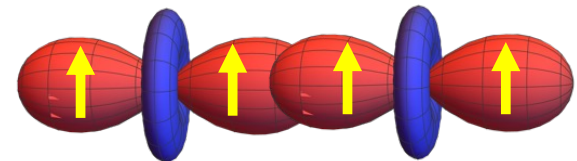
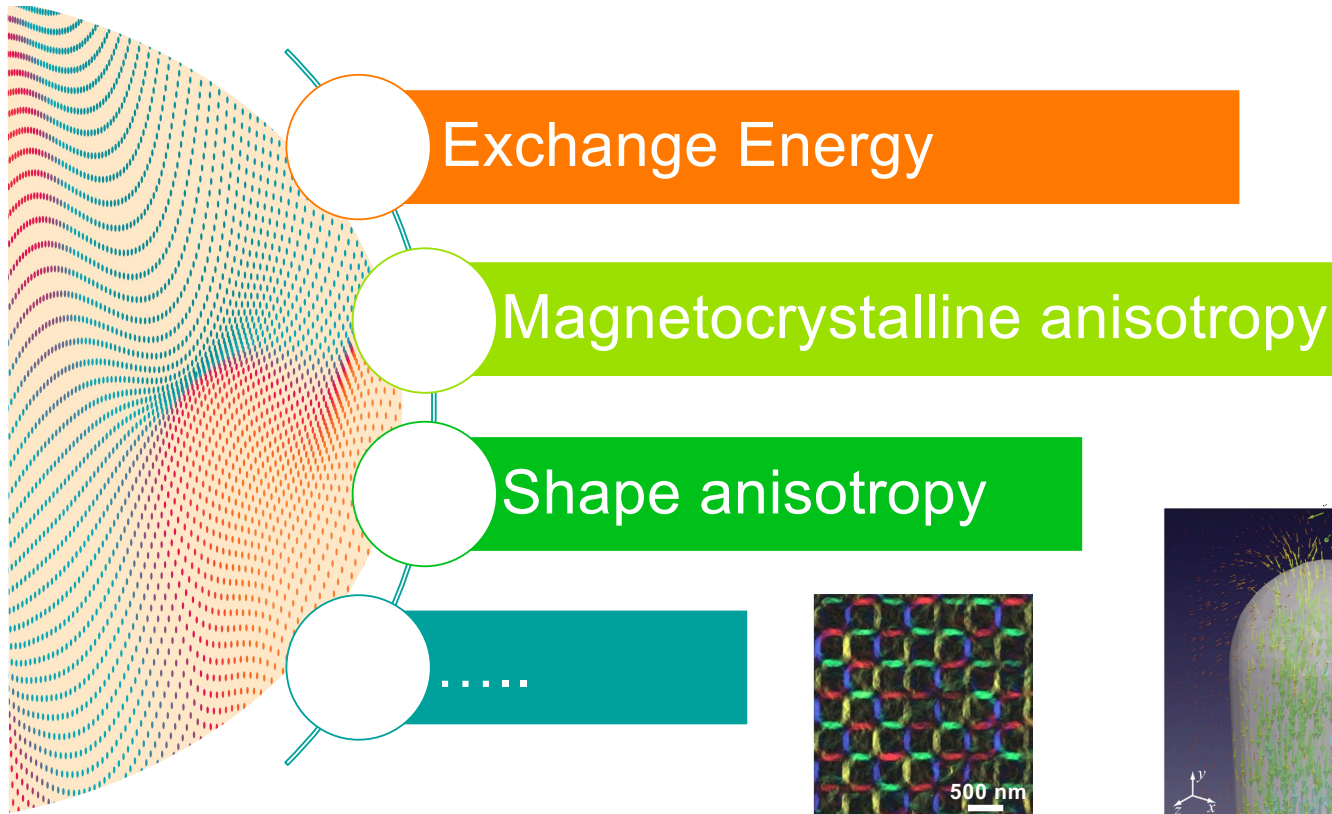
Deputy Division Director,  
Materials Science Division, Argonne National Laboratory

Department of Materials Science and Engineering,  
Northwestern University.

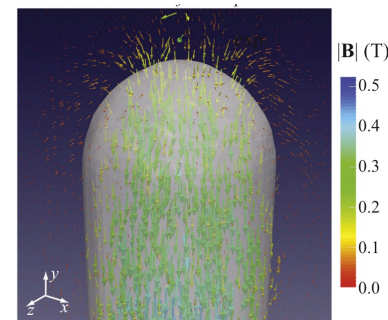
May 1, 2024  
Ingelheim, Germany

# MAGNETIC NANOSTRUCTURES

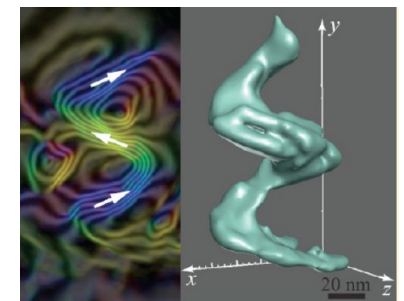
Exploring the energy landscape



C. Phatak, et al. Phys. Rev. B **83**(17), 174431 (2011).



C. Phatak, et al. Ultramicroscopy **164**, 24–30 (2016).

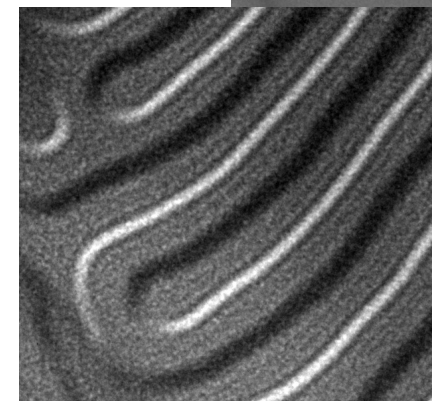
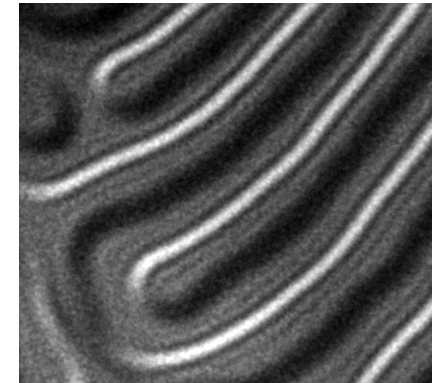
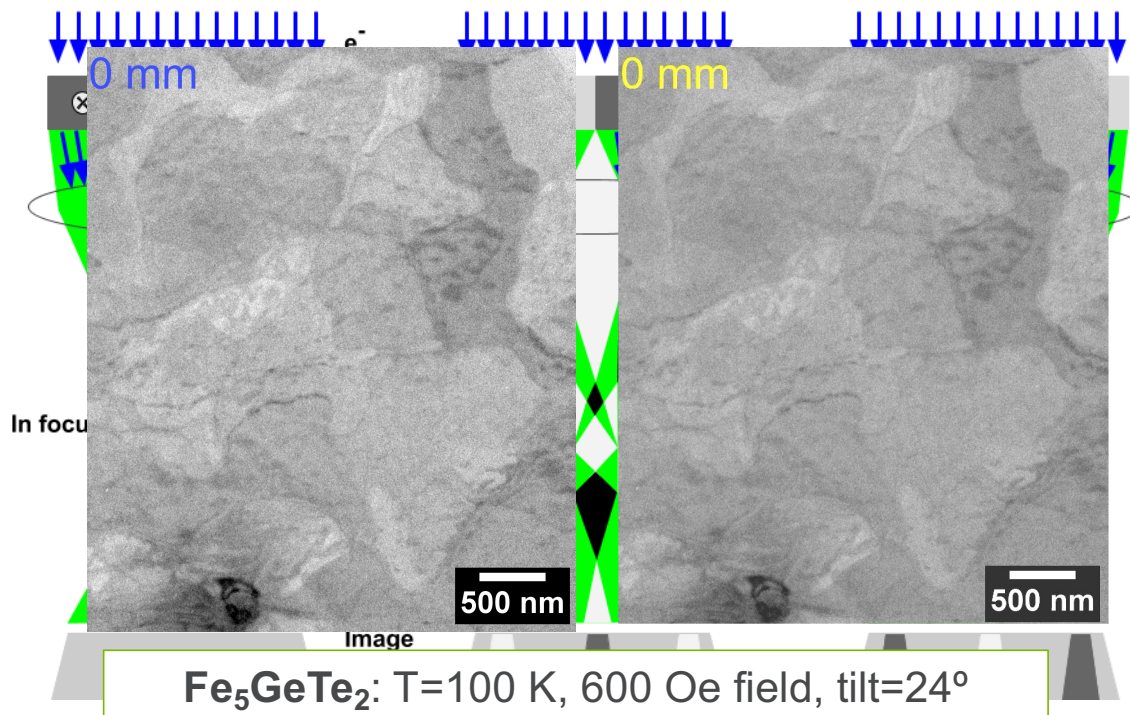


C. Phatak, et al. Nano Letters **14**(2), 759–764 (2014).

# LORENTZ TEM

# LORENTZ TEM – IMAGING

Fresnel Mode:



- Out-of-focus images highlight regions of change in magnetization
  - Domain walls, Vortices, Skyrmions

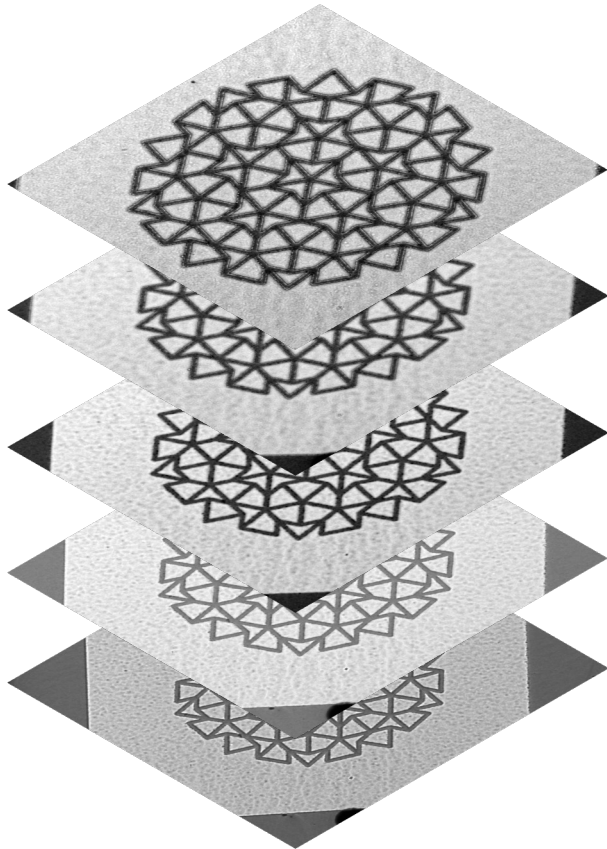
# PHASE RETRIEVAL USING TIE

## Transport of Intensity Equations (TIE)

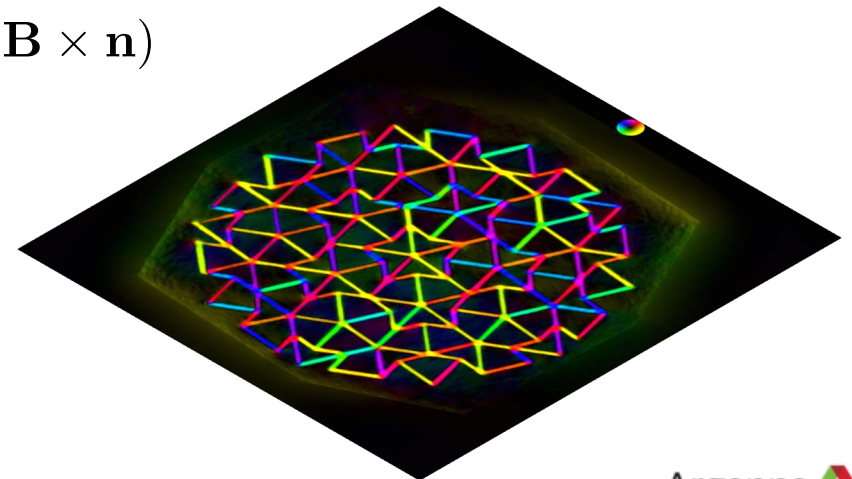
$$\varphi(\mathbf{r}_\perp, z) = -\frac{2\pi}{\lambda} \nabla_\perp^{-2} \left[ \nabla_\perp \cdot \left( \frac{1}{I(\mathbf{r}_\perp, 0)} \nabla_\perp \left\{ \nabla_\perp^{-2} \left[ \frac{\partial I(\mathbf{r}, 0)}{\partial z} \right] \right\} \right) \right], \quad I(\mathbf{r}_\perp, 0) \neq 0$$

In-focus image ↑ Approximated by difference between the over- and under-focus images

$$\nabla\varphi = \frac{2\pi et}{h} (\mathbf{B} \times \mathbf{n})$$

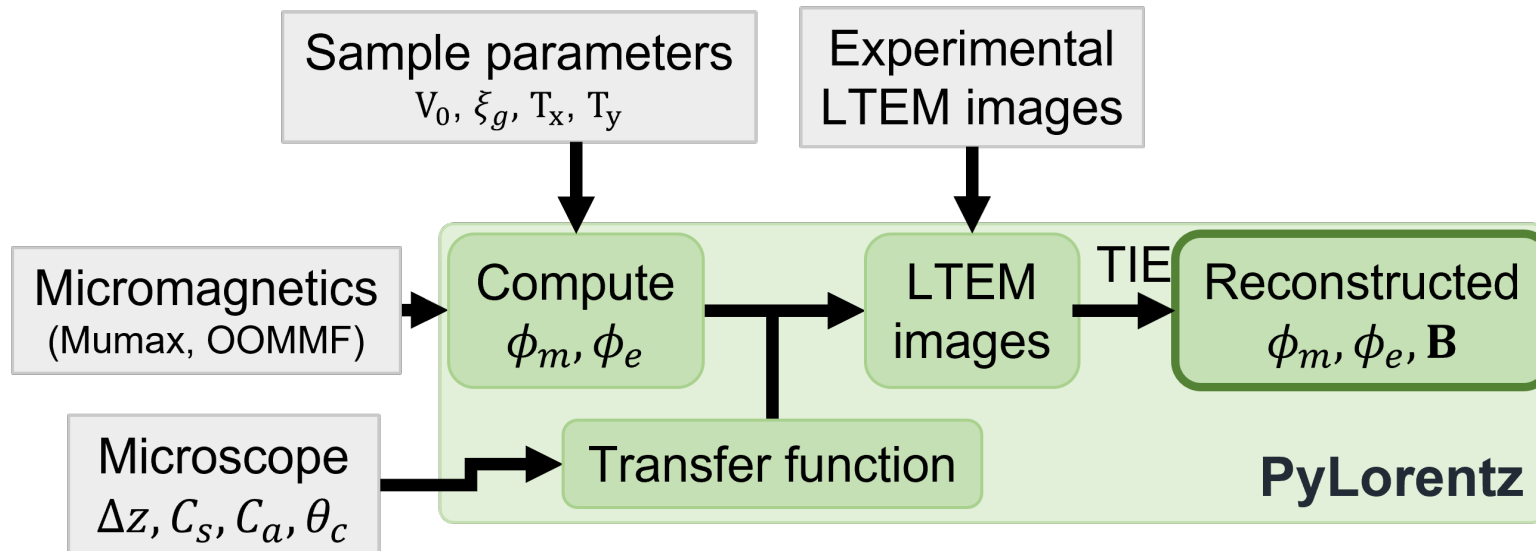


Through-Focus Series of TEM images



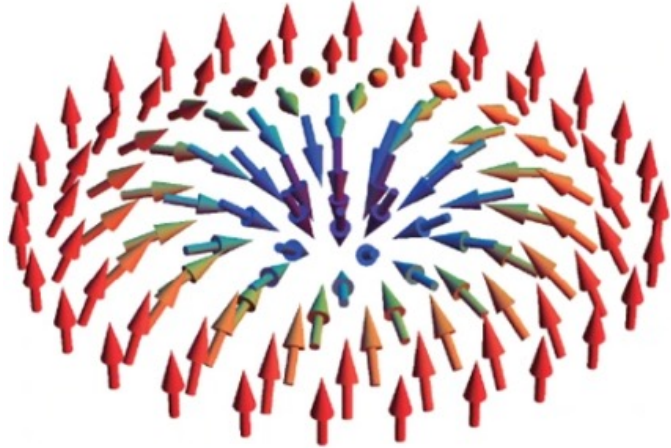
# IMAGE ANALYSIS AND SIMULATION

## Simulating LTEM images from micromagnetics output

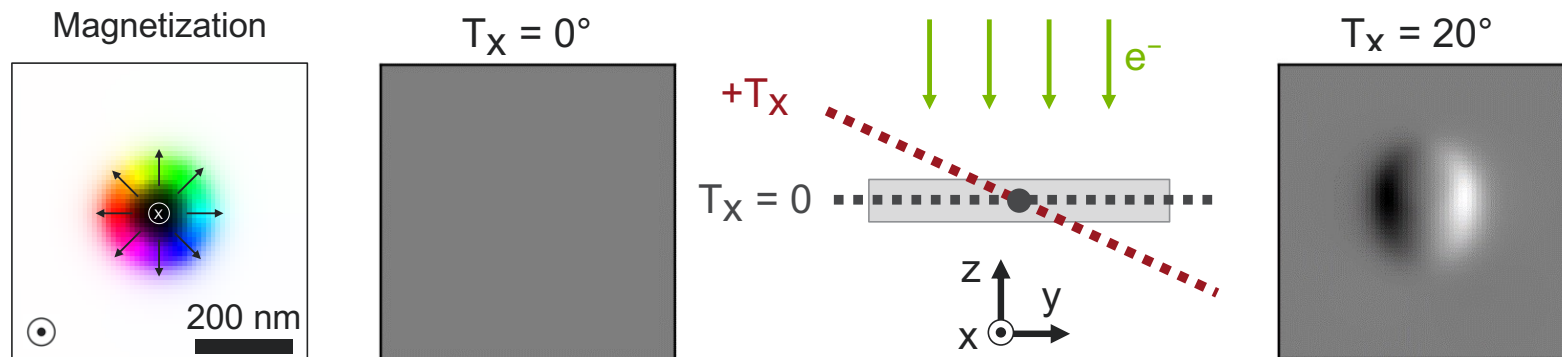


- We developed a new method for calculating electron phase shift through a magnetic material, using the linear superposition method
  - This works for 3D magnetization at any image tilt

# NÉEL SKYRMION: 3D MAGNETIC STRUCTURE



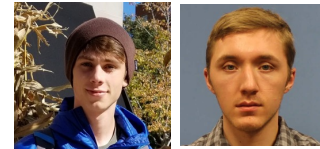
- Néel skyrmions must be tilted to create contrast



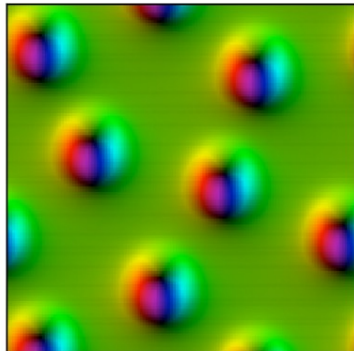
Simulated LTEM images at  $\Delta z = -25 \mu\text{m}$ , for  $0^\circ$  tilt and rotated  $20^\circ$  about the x axis



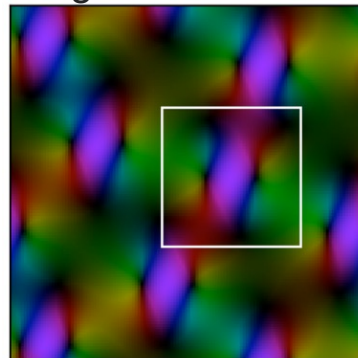
# NÉEL SKYRMIONS



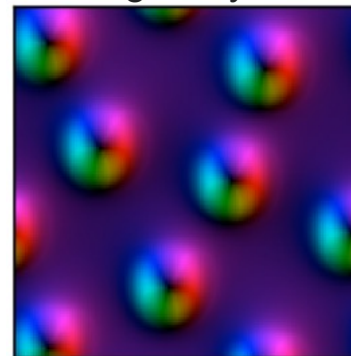
Magnetization



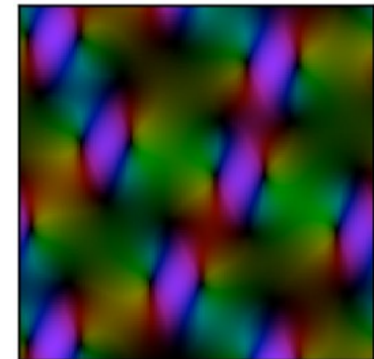
Reconstructed  
Magnetic Induction



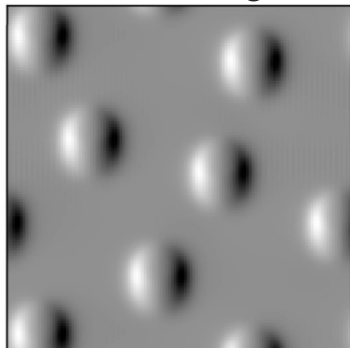
Demag/Stray Field



$B = \mu_0(M + H)$



LTEM Image



These do not match!!

**In LTEM, we measure the total projected magnetic induction which includes the stray fields**

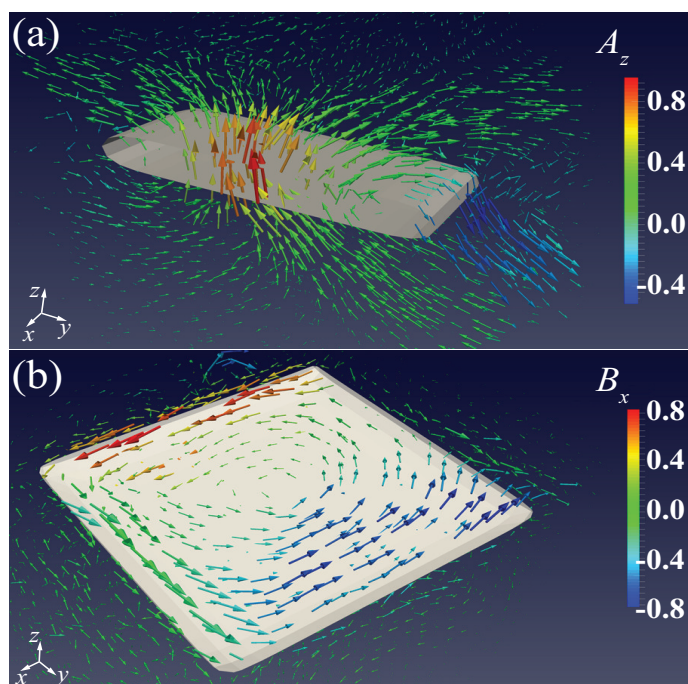
**Simulation is critical to account for tilt and stray fields**

**It allows us to interpret the magnetization from the magnetic induction**

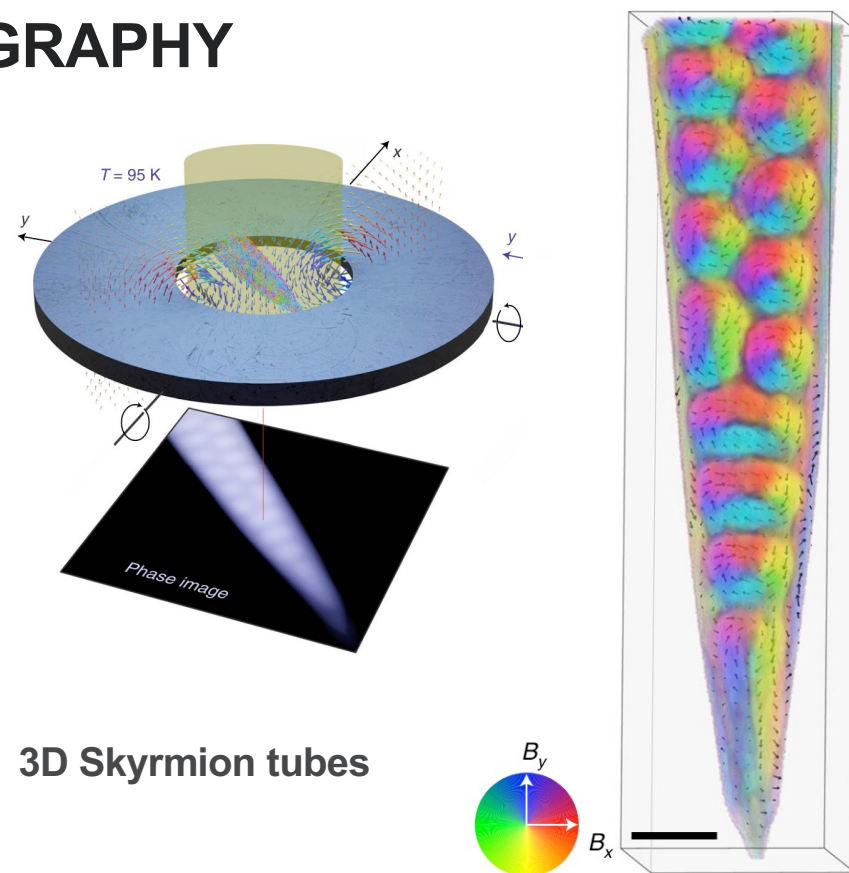
# VECTOR FIELD ELECTRON TOMOGRAPHY

## Reconstructing 3D Magnetic Fields

### Patterned Permalloy



C. Phatak, et al., Phys. Rev. Lett. **104**, 253901 (2010).



### 3D Skyrmion tubes

D. Wolf, et al., Nat. Nanotechnol. **17**(3), 250–255 (2022). © Creative Commons 4.0

# Can we reconstruct the sample magnetization from LTEM data?

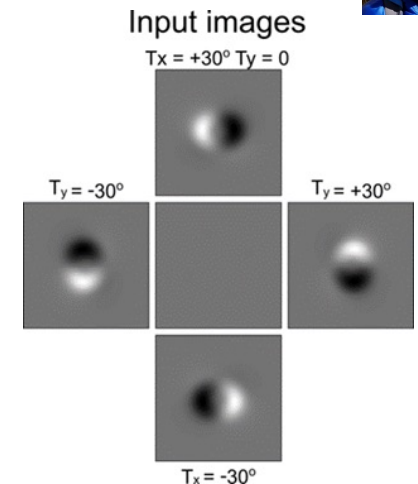
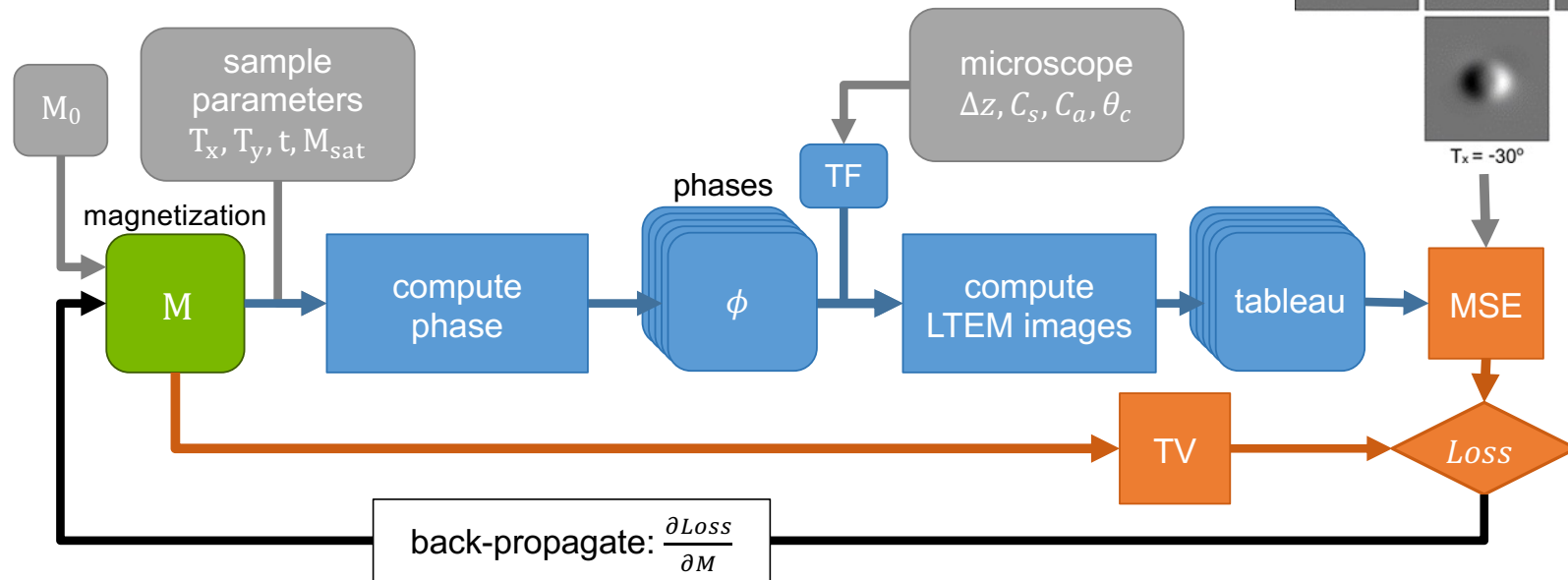


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# 3D MAGNETIZATION RECONSTRUCTION

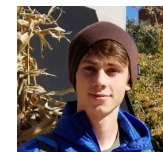
## Automatic differentiation (AD) of a forward model

- The forward model is composed of two steps:  $\mathbf{M} \rightarrow \phi \rightarrow \text{images}$
- Input is a tilt tableau of defocused LTEM images
- Iteratively learns the magnetization that creates those images

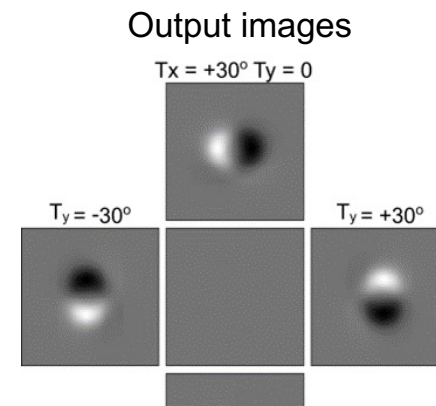
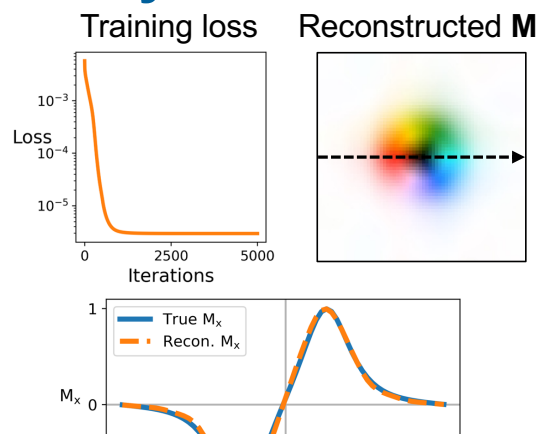
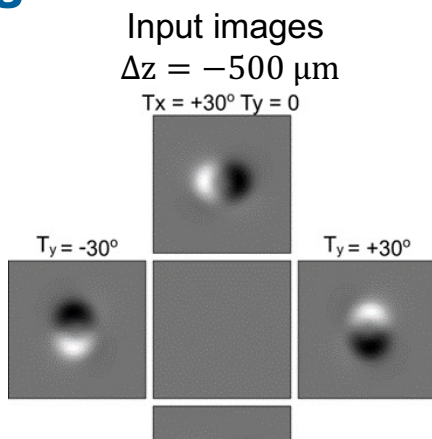
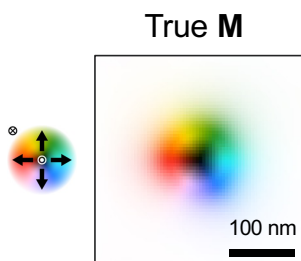


# RECONSTRUCTING A NÉEL SKYRMION

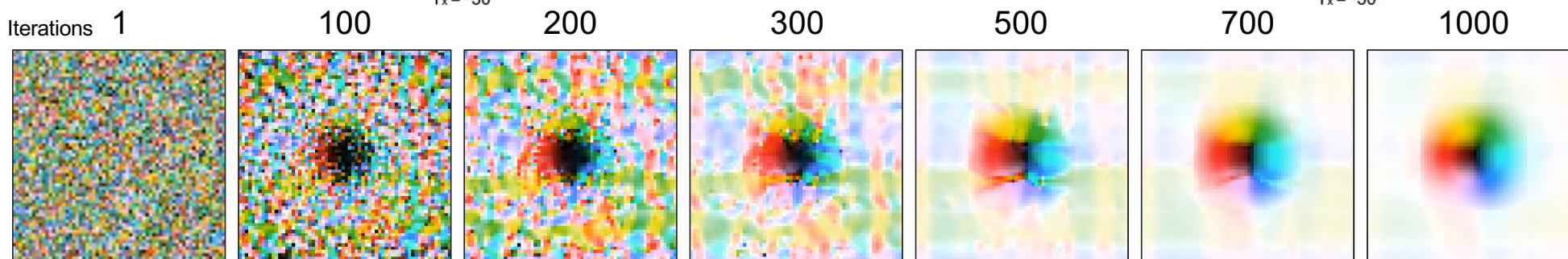
## 5 input images from a simulated skyrmion



64 x 64 x 1 voxels  
320 nm field-of-view

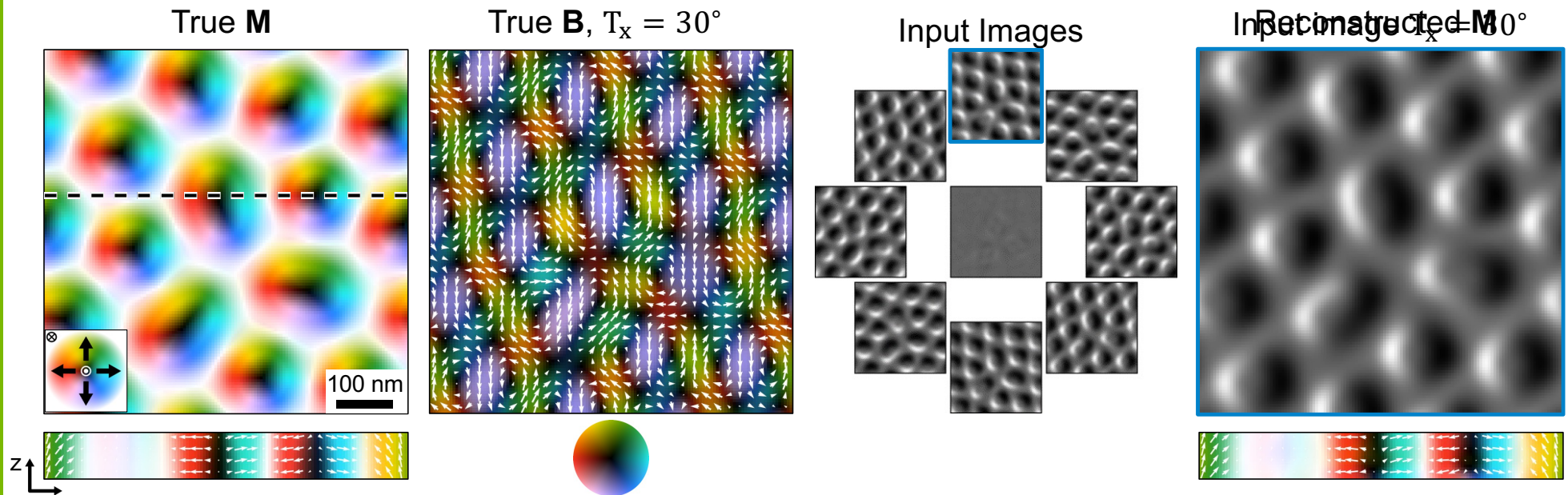


**We accurately reconstruct the skyrmion chirality!**



# NÉEL SKYRMION LATTICE RECONSTRUCTION

- Magnetization reconstruction is more important in cases with many spin textures
  - Skyrmions are often distorted and have complex integrated induction maps
- Skyrmion lattice region:  $(128 \times 128 \times 16)$ ,  $5 \text{ nm}^3$  cell size
  - 9 input images (conical tilt series,  $\theta = 30^\circ$ ) for reconstructing 3D samples



# COBALT NANOHELICES



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## Maren Hassinger: This Is How We Grow

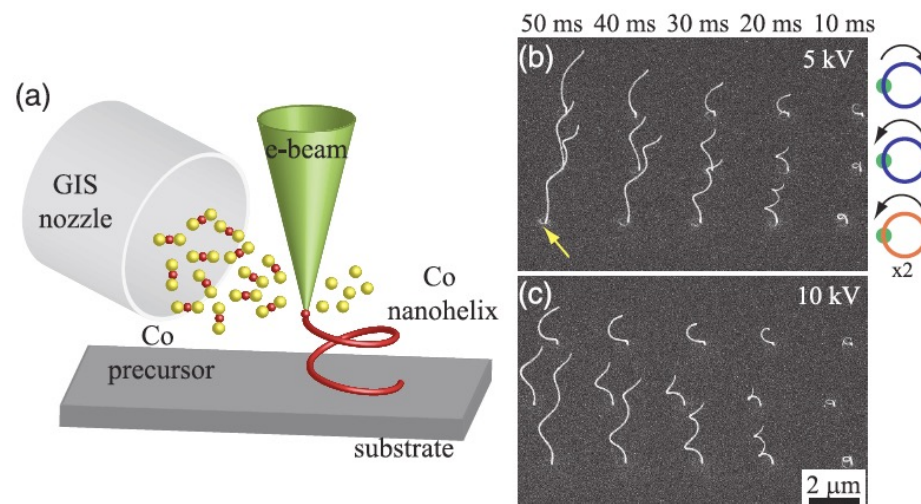
Art installation at Art Institute of Chicago titled “Paradise Regained (2020)” shows industrial rope curved towards Lake Michigan



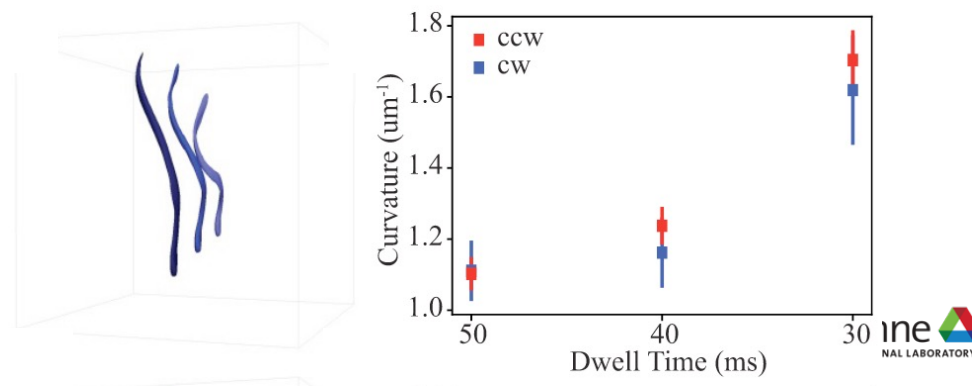
# CREATING MAGNETIC NANOHELICES

## Controlling curvature and torsion

- Creating magnetic nanostructures with controlled curvature can be challenging
- We use direct-write technique via focused electron beam-induced deposition (FEBID)
- This enables growth of helices with different curvature and torsion



Schematic of FEBID process for fabrication of Co nanohelices with different curvature and torsion



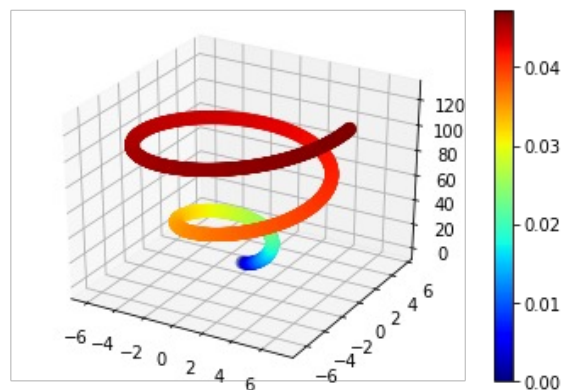
C. Phatak et al., *ACS Appl. Nano Mater.* **3**, 6009 (2020). © American Chemical Society 2020

# NANOHELICES

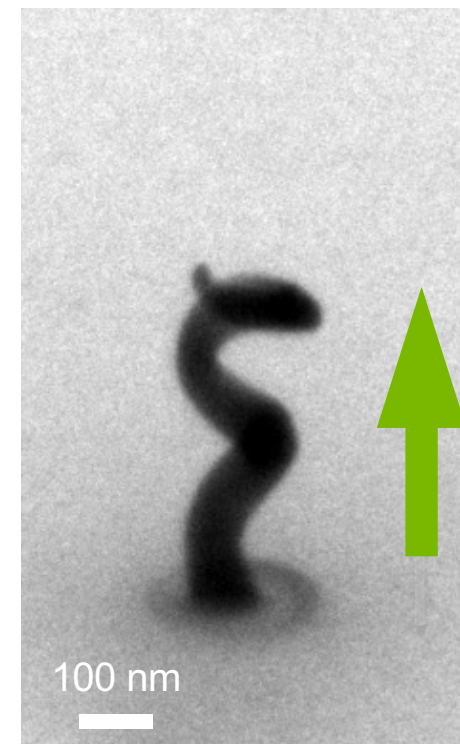
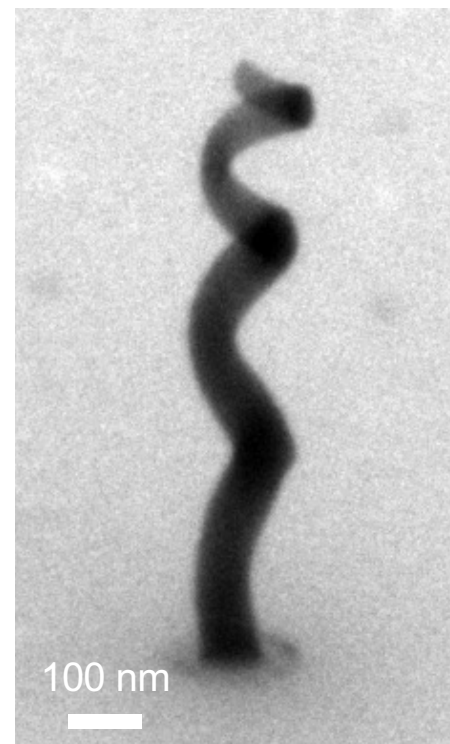
## Changing curvature in a single nanohelix

- The radius of the nanohelix changes with the height.

$$\kappa = \frac{|r|}{r^2 + c^2}, c = \frac{\text{Pitch}}{2\pi}$$



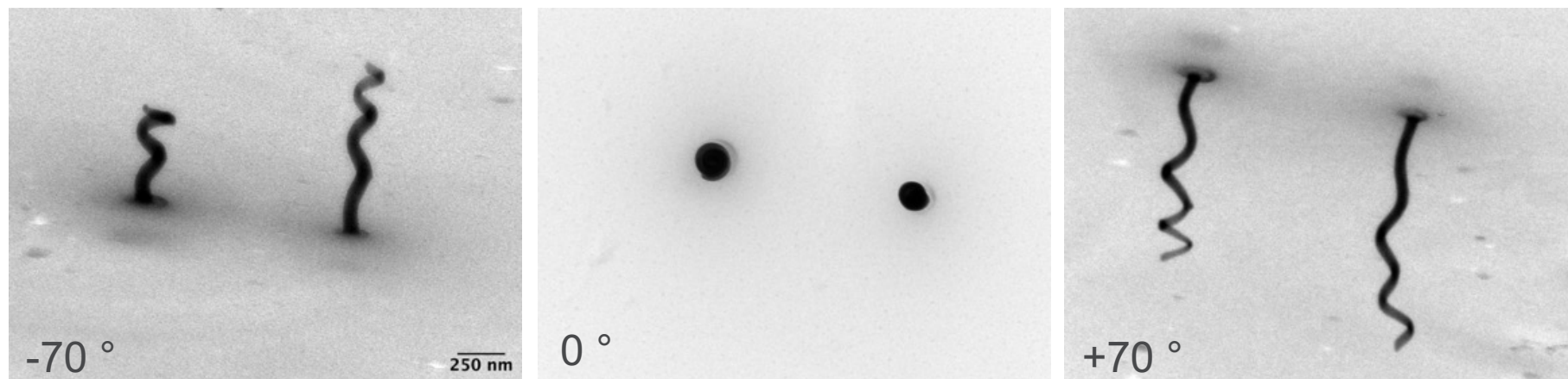
- Varying curvature and torsion with height of the nanostructure.



J. Fullerton, et al., Nano Lett., **24**, 2481-2487 (2024). © American Chemical Society 2024

# ELECTRON TOMOGRAPHY OF MAGNETIC NANOHELICES

- Depositing nanohelices directly on C-coated  $\text{SiO}_x$  TEM membranes allows for electron tomography and LTEM experiments

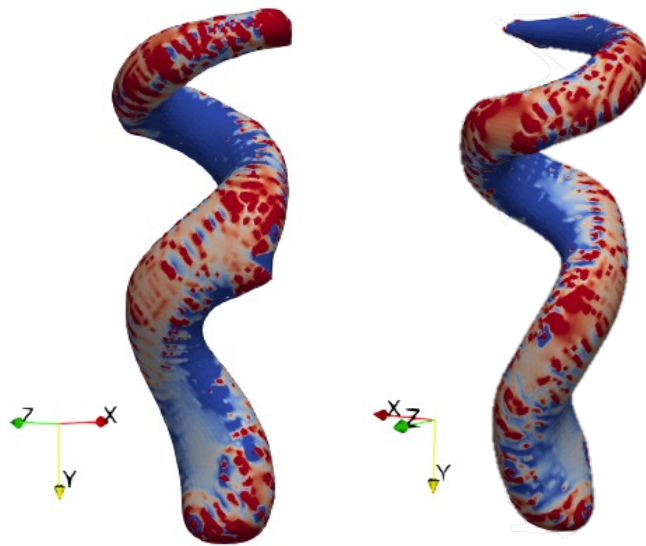


Tilt series of images showing the orientation of two different Co nanohelices grown using FEBID.

# GAUSSIAN CURVATURE OF 3D NANOHELICES



Helix 1:



Curvature ( $\mu\text{m}^{-2}$ )

-13      0      13



Helix 2:

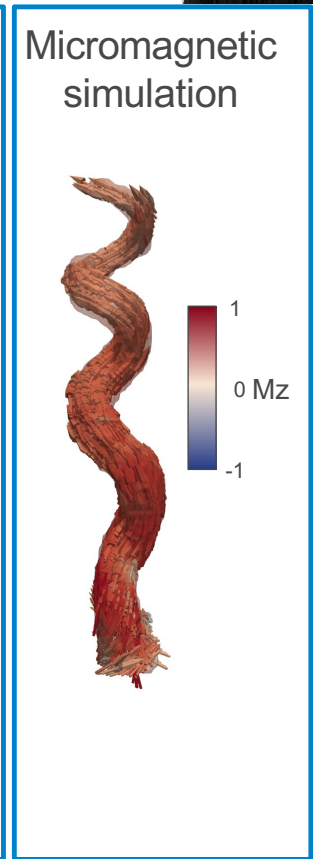
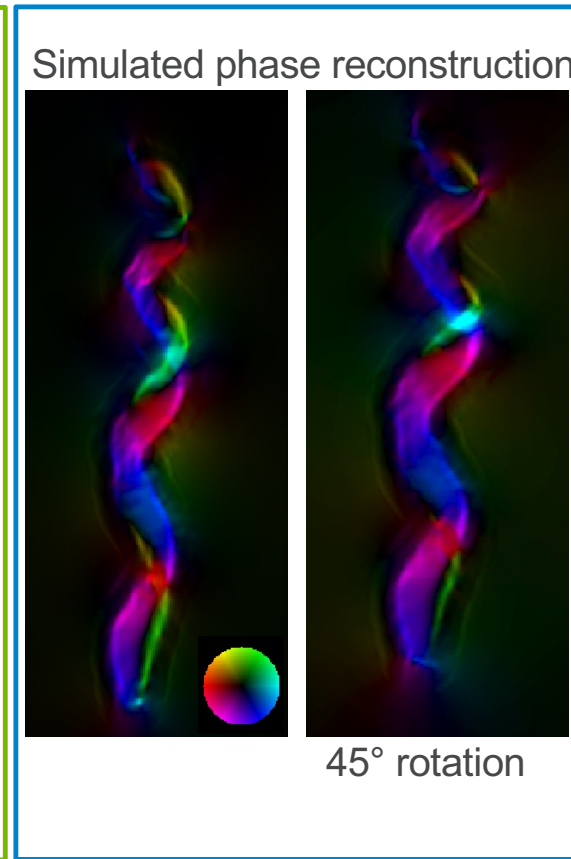
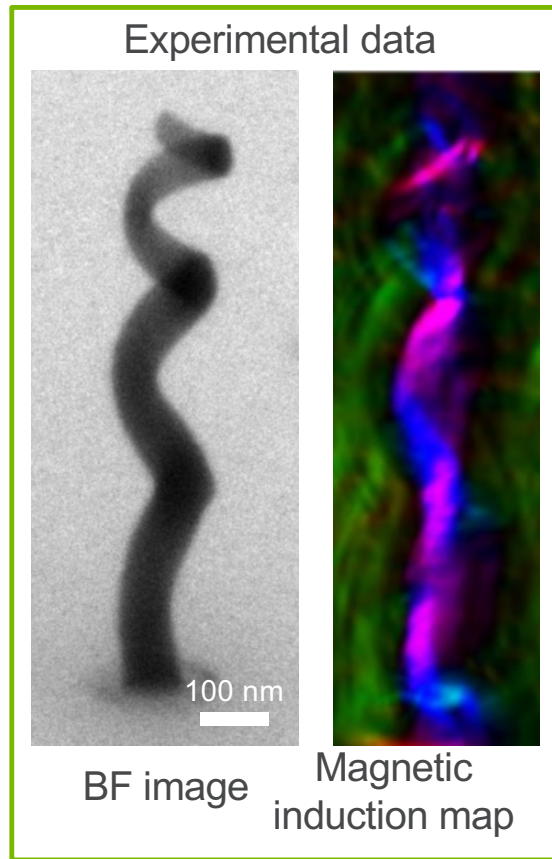


Increasing  
magnitude of  
curvature with  
height



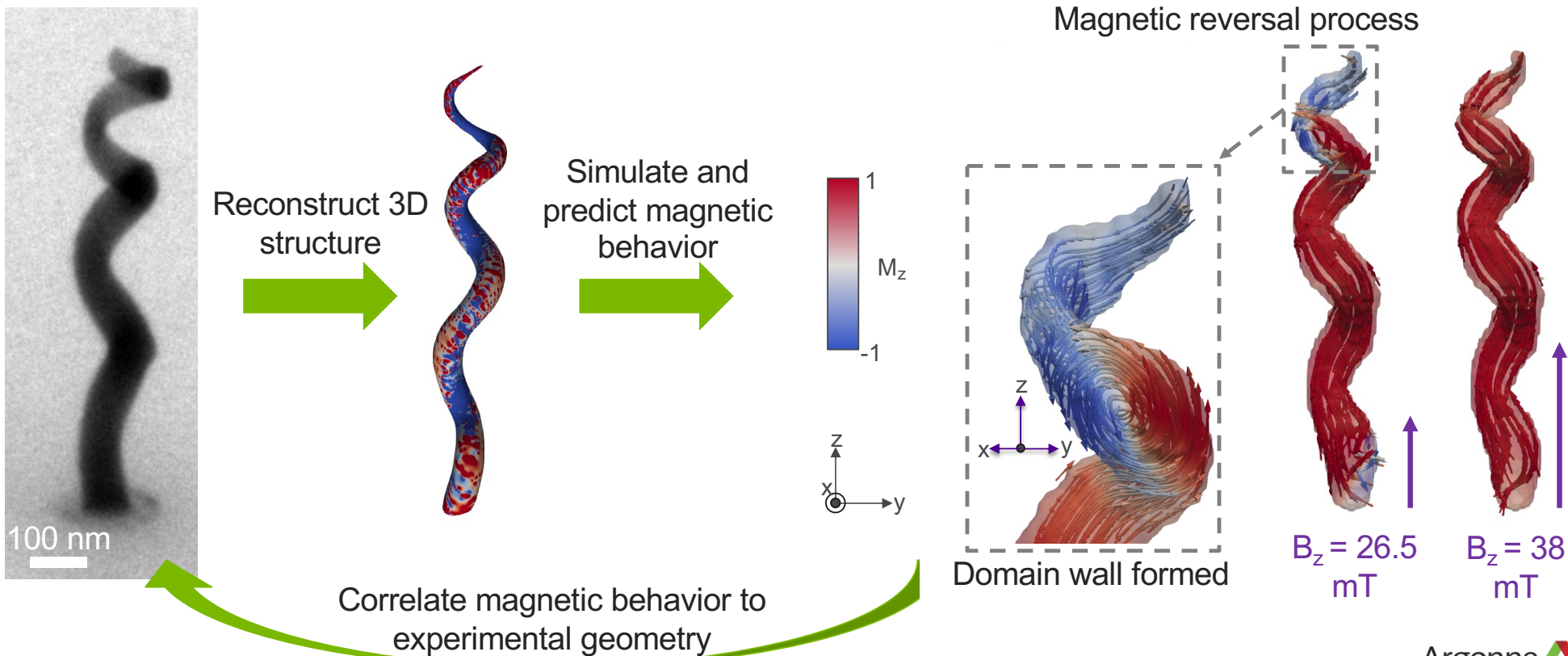
# Co FEBID NANOHELIX 1: RECONSTRUCTED PHASE

- Experimental LTEM data used to reconstruct magnetic induction map
- Experimental tomography data were used as the input for the helix shape in micromagnetic simulation (first time we have done this)
- Micromagnetic simulation used to reconstruct simulated magnetic induction map



**Magnetic induction maps for this nanohelix match a single domain state with magnetization following the length of the helix**

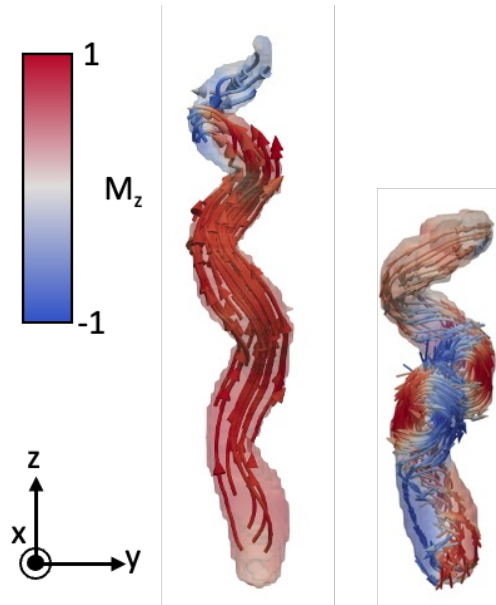
# SIMULATED MAGNETIC REVERSAL



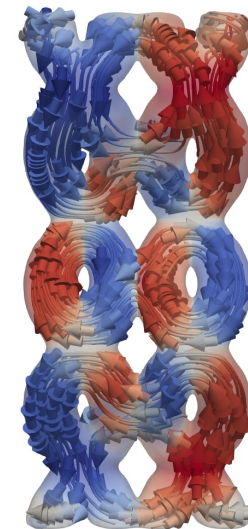
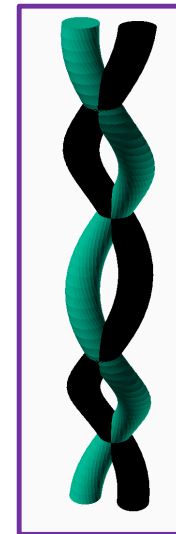
# NANOHELICES

## Beyond the single nanohelix

Domain walls appear with higher curvature and under applied field



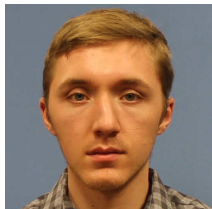
Designing interconnected nanowires  
– curvature, overlap



But single domain states are favored at remanence for single, non-interacting helices

# ACKNOWLEDGEMENTS

## Argonne National Laboratory



Tim Cote



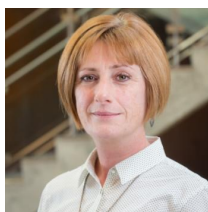
Arthur McCray  
(now at LBNL)



John Fullerton



Mathew Cherukara



Amanda Petford-Long

## Collaborators:

### Argonne National Lab

Center for Nanoscale Materials

- C. S. Miller
- D. Czaplewski

### Northwestern University

Materials Science & Engineering Dept

- E. B. Gulsoy
- Z. Thompson

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**THANK YOU FOR LISTENING !!**

<https://www.anl.gov/msd/functional-nanoscale-heterostructures>