Direct observation of topological magnetic monopoles using soft x-ray vector ptychography at 10 nm resolution

Chen-Ting Liao^{1,2*}, Arjun Rana^{2,3*}, Ezio Iacocca^{4,5}, Ji Zou¹, Minh Pham^{2,6}, Xingyuan Lu^{3,7}, Emma-Elizabeth Cating Subramanian^{1,2}, Yuan Hung Lo^{2,3}, Sinéad A. Ryan^{1,2}, Charles S. Bevis^{1,2}, Robert M. Karl Jr^{1,2}, Andrew J. Glaid⁸, Jeffrey Rable⁸, Pratibha Mahale^{8,9}, Joel Hirst¹⁰, Thomas Ostler^{10,11}, William Liu^{2,3}, Colum M. O'Leary^{2,3}, Young-Sang Yu¹², Karen Bustillo¹³, Hendrik Ohldag¹², David A. Shapiro¹², Sadegh Yazdi¹⁴, Thomas E. Mallouk^{8,9}, Stanley J. Osher^{2,6}, Henry C. Kapteyn^{1,2}, Vincent H. Crespi⁸, John V. Badding⁸, Yaroslav Tserkovnyak¹, Jianwei (John) Miao^{2,3}, and Margaret M. Murnane^{1,2}

¹JILA and Department of Physics, University of Colorado and NIST, 440 UCB, Boulder, Colorado 80309, USA. ²STROBE Science and Technology Center. ³Department of Physics & Astronomy and California NanoSystems Institute, University of California, Los Angeles, CA 90095, USA. ⁴Department of Mathematics, Physics, and Electrical Engineering, Northumbria University, Newcastle upon Tyne, NE1 8ST, UK. ⁵Center for Magnetism and Magnetic Materials, University of Colorado, Colorado Springs, CO 80918, USA. ⁶Department of Mathematics, University of California, Los Angeles, CA 90095, USA. ⁷School of Physical Science and Technology, Soochow University, Suzhou 215006, China. ⁸Departments of Chemistry, Physics, Materials Science and Engineering and Materials Research Institute, Penn State University, University Park, PA 16802, USA. ⁹Department of Chemistry, University of Pennsylvania, Philadelphia PA 19104, USA. ¹⁰Materials and Engineering Research Institute, Sheffield Hallam University, Howard Street, Sheffield S1 1WB, UK. ¹¹Department of Engineering and Mathematics, Sheffield Hallam University, Howard Street, Sheffield S1 1WB, UK. ¹²Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA. ¹³National Center for Electron Microscopy, Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA, 94720, USA. ¹⁴Renewable and Sustainable Energy Institute, University of Colorado, Boulder, CO 80309, USA. *ChenTing.Liao@colorado.edu*

Abstract: We developed soft x-ray vector ptychography at 10 nm spatial resolution without requiring a priori knowledge, which is then used to quantitatively image 3D magnetization vector fields of topological magnetic monopoles and their interactions. © 2022 The Author(s)

Magnetic topological defects and textures are energetically stable spin configurations characterized by symmetry breaking such as the well-known example 2D skyrmions that promise future spintronics technologies [1-4]. However, experimental evidence of the 3D spin textures has been largely indirect or qualitative to date due to the lack of magnetic characterization technique within nanoscale volumes. Here, we developed soft x-ray vector ptychography (also called vector ptychographic tomography) to quantitatively image the 3D magnetization vector fields in a frustrated magnetic meta-lattice with 10 nm spatial resolution for the first time [5]. The high-resolution and quantitative nature of our new method enabled us to directly observe emergent topological magnetic monopoles and their interactions in a ferromagnetic, nickel meta-lattice sample. This spatial resolution is comparable to the magnetic exchange length of transition metals, enabling us to determine the magnetization vectors and emergent magnetic fields and to probe monopole-monopole interactions.



Fig. 1. Experimental schematic of 3D soft x-ray vector ptychography. Left- and right-circularly polarized x-rays (pink) were focused onto a ferromagnetic, nickel meta-lattice sample (center), on which the green circles indicate the partially overlapped scan positions. The sample was titled around the x- and z-axis and diffraction patterns were collected by a camera. **a**, The reconstructed 3D electron density (green) and magnetization vector field (arrows) of the sample. **b**, Spatial distribution of 68 and 70 topological monopoles with positive (red dots) and negative charges (blue dots) in the meta-lattice, where the surfaces of the magnetic voids in red and blue represent virtual topological monopoles with positive and negative charges, respectively. The solid and dashed squares mark the region of interest shown in Fig. 2. Scale bar, 60 nm in (**b**).

Our experiment was conducted by focusing circularly polarized soft x-rays onto the ferromagnetic meta-lattice (Fig. 1). The magnetic contrast of the sample was obtained by using x-ray magnetic circular dichroism and tuning the x-ray energy to the L₃-edge of nickel. In each measurement, three independent tilt series were acquired from the sample, corresponding to three in-plane rotation angles (0°, 120° and 240°) around the z-axis (Fig. 1). Each tilt series was collected by rotating the sample around the x-axis with a tilt range from -62° to +61°. At each tilt angle, a focused x-ray beam was scanned over the sample with partial overlap between adjacent scan positions and a far-field diffraction pattern was recorded by a camera at each scan position. The full data set consists of six tilt series with a total of 796,485 diffraction patterns, which had been reconstructed using a regularized ptychographic iterative engine. The scalar tomographic reconstruction was performed from the three tilt series of 91 projections using a real space iterative algorithm, which can optimize the reconstruction by iteratively refining the spatial and angular alignment of the projections. To determine the magnetization vector field, we took the difference of the left- and right-circularly polarized projections of the three tilt series. The 3D vector reconstruction was performed from 91 difference projections by least-squares optimization with gradient descent. More experimental details can be found in Ref. [5].

Our 3D ferromagnetic meta-lattice was synthesized by self-assembly of a face-centered cubic template using silica nanospheres of 60 nm in diameter. The interstitial spaces between the nanospheres of the template were infiltrated with nickel to create a meta-lattice, comprising octahedral and tetrahedral sites interconnected by thin necks [6-7]. After image reconstructions (Fig. 1(a)), we applied topological theory to quantitatively analyze the experimental 3D magnetization vector field and identified nonlocal spin textures in the meta-lattice sample (Fig. 2). We found that the topological monopole pairs with positive and negative charges are separated closer, while the positively and negatively charged pairs are stabilized at comparatively longer distances. We also observed virtual topological magnetic monopoles (positive, red surface; negative, blue surface, Fig. 1(b)) created by magnetic voids in the meta-lattice. See, Ref. [5] for detailed analysis and findings. This work demonstrates that magnetic meta-lattices could be used as a new platform to create and investigate the interactions and dynamics of topological magnetic monopoles.

In summary, with the rapid development of advanced synchrotron radiation, x-ray free electron lasers, laser-driven high harmonic generation sources, and other compact x-ray light sources worldwide, we expect that 3D soft x-ray vector ptychography can find broad applications in magnetic and anisotropic materials at the nanoscale.



Fig. 2. (Left 4 panels) Quantitative 3D characterization of topological magnetic monopoles in the 281 ferromagnetic meta-lattice. a, b, The location (from Fig. 1(b)) and 3D spin textures of a positively charged monopole within a tetrahedral site of the face-cantered cubic meta-lattice. c, d, the location and 3D spin textures of a negatively charged monopole within an octahedral site. Scale bars, 25 nm (a) and 10 nm (b). (**Right 6 panels) Interactions of the topological magnetic monopoles. e-g**, Three representative topological monopole pairs with a positive and negative charge (e), two positive (f), and two negative charges (g), where the white lines represent the emergent magnetic field lines. h-j, Histograms of the nearest-neighbor distance for the topological monopole pairs with a positive and negative charges (j). The three histograms were fit to a generalized extreme value distribution, producing three curves in (h-j), where µ represents the centre of each fit and the standard error was determined from the fit's confidence interval. Scale bar, 5 nm.

References

[1] P. Milde et al., "Unwinding of a skyrmion lattice by magnetic monopoles." Science 340, 1076–1080 (2013).

- [2] C. Donnelly et al., "Three-dimensional magnetization structures revealed with X-ray vector nanotomography." Nature 547, 328-331 (2017).
- [3] M. Ray *et al.*, "Observation of Dirac monopoles in a synthetic magnetic field." Nature 505, 657–660 (2014).
- [4] J. Zou et al., Topological transport of deconfined hedgehogs in magnets. Phys. Rev. Lett. 125, 267201 (2020).
- [5] A. Rana*, C.-T. Liao* et al., in review (2022). Preprint: arXiv2104.12933
- [6] J. Han et al., "Abrupt topological transitions in the hysteresis curves of ferromagnetic metalattices." Phys. Rev. Lett . 89, 197203 (2002).
- [7] Y. Liu et al., "Confined chemical fluid deposition of ferromagnetic metalattices." Nano Lett. 18, 546–552 (2018).