## In-situ Magnetic Domain Behavior in van der Waals Fe<sub>3</sub>GeTe<sub>2</sub>

Yue Li<sup>1</sup>, Rabindra Basnet<sup>2</sup>, Krishna Pandey<sup>3</sup>, Jin Hu<sup>2,3</sup>, Wei Wang<sup>4</sup>, Xuedan Ma<sup>4</sup>, Arthur R. C. McCray<sup>1,5</sup>, Amanda K. Petford-Long<sup>1,6</sup> and Charudatta Phatak<sup>1,6\*</sup>

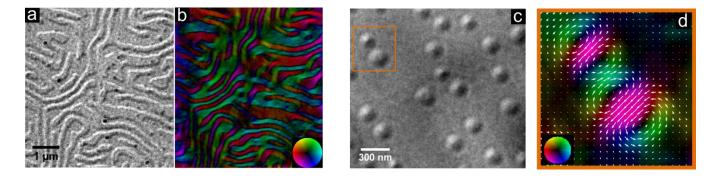
- <sup>1.</sup> Materials Science Division, Argonne National Laboratory, Lemont, IL, USA.
- <sup>2</sup> Department of Physics, University of Arkansas, Fayetteville, AR, USA.
- <sup>3</sup> Materials Science and Engineering Program, University of Arkansas, Fayetteville, AR, USA.
- <sup>4.</sup> Center for Nanoscale Materials, Argonne National Laboratory, Lemont, IL, USA.
- <sup>5.</sup> Applied Physics Program, Northwestern University, Evanston, IL, USA.
- <sup>6</sup> Department of Materials Science and Engineering, Northwestern University, Evanston, IL, USA.
- \* Corresponding author: cd@anl.gov

Two-dimensional (2D) magnetic van der Waals (vdW) materials exhibit intriguing magnetic physics [1] and are potential candidates for spintronic devices [2] owing to their atomically-layered heterostructures and strong intrinsic spin interaction. In particular, various nontrivial magnetic spin structures, such as the Bloch or the Néel-type stripes, skyrmions or bubble domains, have been reported in 2D magnetic vdW materials under certain external stimuli [3-4]. It is essential to understand the magnetic domain behavior in vdW materials in response to external stimuli such as electric field, magnetic fields and temperature in order to control them by tuning the governing energy terms. Fe<sub>3</sub>GeTe<sub>2</sub> is one of the promising magnetic vdW materials as it is an itinerant ferromagnet in the 2D-limit with a Curie temperature of 230 K. In this work we examine systematically the role of magnetic field on the formation and manipulation of magnetic domain structures in vdW Fe<sub>3</sub>GeTe<sub>2</sub> by in-situ cryo Lorentz transmission electron microscopy (LTEM). Quantitative retrieval of magnetic induction maps enables us to interpret the resulting magnetization configuration of Néel-type stripe domains or skyrmion lattices.

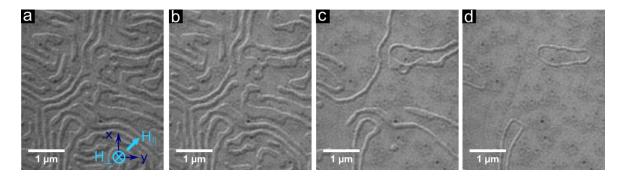
Two categories of distinct magnetic textures, Néel stripe domains and skyrmion lattices, are formed in Fe<sub>3</sub>GeTe<sub>2</sub> after following a zero field cooling (ZFC) and a field cooling (FC) protocol across the Curie temperature respectively. Figures 1(a) shows a LTEM image of magnetic stripe domains in Fe<sub>3</sub>GeTe<sub>2</sub> after following ZFC protocol at 100 K. Figure 1(b) shows the corresponding projected magnetic induction color map of the stripe domains, indicating the orientation of Néel magnetic domains. Figure 1(c) displays the skyrmions formed after a FC process in an externally applied perpendicular field of 400 Oe at 100 K. The projected stray field inside the skyrmion core (cyan) and outside the skyrmion are shown in Figure (d). The sample tilt makes it difficult to understand the projected magnetic induction map in the experiment. We will present an interpretation of experimental result by use of micromagnetic simulation and simulated LTEM approaches.

The response of magnetic domains to an external magnetic field applied in-situ at a fixed low temperature is of importance in order to understand their behavior and stability. Figure 2 shows the evolution of magnetic stripe domains as a function of the perpendicular magnetic field at 100 K, which also introduces an in-plane magnetic field with respect to the sample because of the sample tilt. We will present the implications of the in-plane component introduced which causes deformation of certain magnetic domains. The magnetic domains, where the magnetization orientation anti-aligns to the field direction, shrink and disappears as the field strength increases. We will also discuss the formation of magnetic domains with higher topological charge such as those in Figure 2(d) which remain stable until large applied fields [5].





**Figure 1.** (a) Lorentz TEM image and (b) corresponding reconstructed in-plane magnetic induction map of Néel stripe domains in Fe<sub>3</sub>GeTe<sub>2</sub> after a zero field cooled protocol with sample tilting angle 23° at 100 K. (c) Magnetic contrast of Néel skyrmions tilted by the angle of 15° after following a field cooled protocol in 400 Oe perpendicular magnetic field. (d) Projected magnetization map of two Néel skyrmions in (c) highlighted by the orange frame. The color code indicates the magnetization orientation.



**Figure 2.** (a-d) Response of stripe domains in Fe<sub>3</sub>GeTe<sub>2</sub> to an externally perpendicular magnetic field (H<sub> $\perp$ </sub>) of (a) 0 Oe, (b) 200 Oe, (c) 400 Oe and (d) 800 Oe at 100 K. An in-plane magnetic field (H<sub> $\parallel$ </sub>) is introduced with respect sample due to sample being tilted for LTEM imaging.

## References:

- [1] B Huang, G Clark and E Navarro-Moratalla et al., Nature **546** (2017), p. 270. doi: https://doi.org/10.1038/nature22391
- [2] W Xia, L Dai and P Yu et al., Nanoscale, 9 (2017), p. 4324. doi: 10.1039/C7NR00844A
- [3] T-E Park, L Peng, J Liang et al. Phys. Rev. B **103** (2021), p. 104410. doi:
- 10.1103/PhysRevB.103.104410
- [4] B Ding, Z Li, G X et al., Nano. Lett. **20** (2020), p. 868. doi: https://doi.org/10.1021/acs.nanolett.9b03453
- [5] This work was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Materials Sciences and Engineering Division. Use of the Center for Nanoscale Materials, an Office of Science user facility, was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.