

## Evolution of Skyrmion Lattice Order in the van der Waals Ferromagnet $\text{Fe}_3\text{GeTe}_2$

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

<sup>1</sup>. Materials Science Division, Argonne National Laboratory, Lemont, IL, USA

<sup>2</sup>. Applied Physics Program, Northwestern University, Evanston, IL, USA

<sup>3</sup>. Department of Physics, University of Arkansas, Fayetteville, AR, USA

<sup>4</sup>. Materials Science and Engineering Program, University of Arkansas, Fayetteville, AR, USA

<sup>5</sup>. Center for Nanoscale Materials, Argonne National Laboratory, Lemont, IL, USA

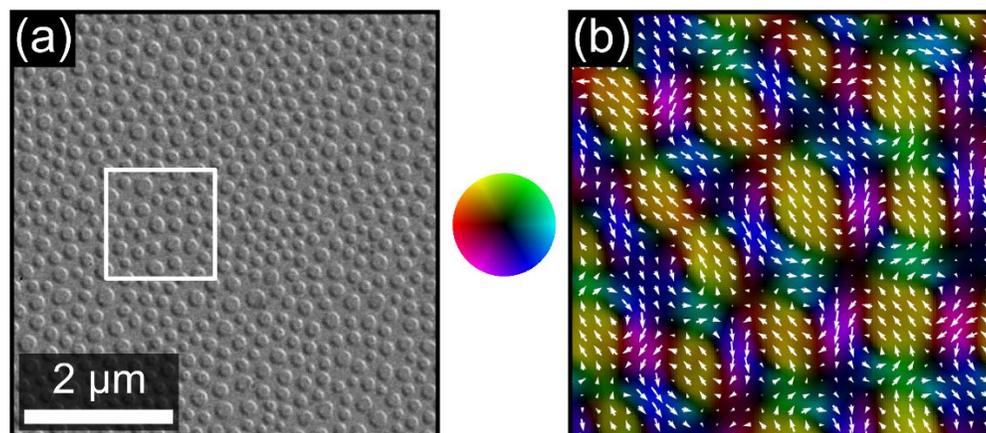
<sup>6</sup>. Department of Materials Science and Engineering, Northwestern University, Evanston, IL, USA

\* Corresponding author: amccray@anl.gov

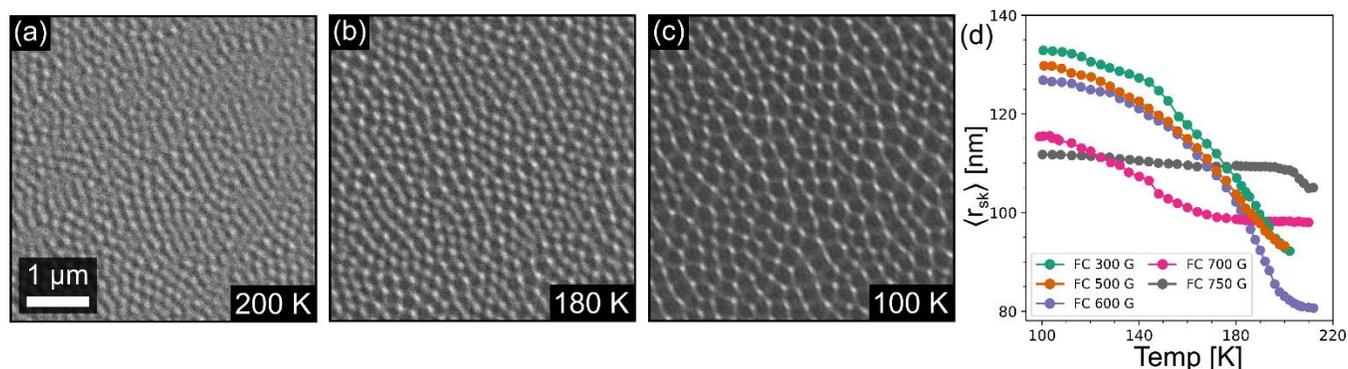
Magnetic skyrmions are topologically protected magnetic spin textures that can exist both as individuals and in dense quasiparticle lattices [1]. While individual skyrmion properties have been studied extensively, their collective behavior in skyrmion lattices is poorly understood, both on the micron-scale where lattice ordering is observed and the nanoscale where inter-skyrmion interactions occur. Skyrmion lattices have been proposed as a system in which to study 2D phase transitions. Bloch skyrmion lattices have been shown to display crystalline, hexatic, and skyrmion liquid phases [2], and Néel skyrmion lattices have been predicted to display both hexatic and skyrmion liquid phases [3]. Here we examine Néel skyrmion lattice ordering in the van der Waals (vdW) ferromagnet  $\text{Fe}_3\text{GeTe}_2$  using cryo-Lorentz transmission electron microscopy (LTEM). FGT is an itinerant ferromagnet that displays magnetism down to monolayer thickness [4], and it is of particular interest for this study as its magnetic parameters change greatly between the observed Curie temperature of 216 K and our minimum achievable temperature of 100 K. Due to these changing magnetic parameters, the average skyrmion size increases with decreasing temperature. By studying how the skyrmion lattice order responds to this size change, we gain insight into how skyrmions interact with each other, are created and destroyed, and how the lattice itself evolves as a collection of skyrmions.

Néel skyrmions, when imaged with LTEM, create an alternating bright/dark contrast pattern as seen in Fig. 1(a), with the reconstructed integrated magnetic induction shown in Fig. 1(b). Individual skyrmions are difficult to distinguish when imaged at the high defocus lengths needed to observe magnetism close to the Curie temperature. We therefore apply a machine-learning algorithm that employs a convolutional neural-network (CNN) to identify over 600,000 skyrmion centers across 383 images. With the skyrmions identified, we quantitatively analyze the skyrmion lattice structure for both translational and orientational order and determine that all observed skyrmion lattices exist in a skyrmion liquid phase. In this talk, we will present results of Néel skyrmion lattice evolution when field-cooling (FC) and field-heating (FH) under different applied field strengths. We observe a higher degree of skyrmion lattice order close to the Curie temperature; the order is lost when FC but is regained when FH and displays a thermal hysteresis effect. We will explain how this loss of order is due to the average skyrmion size increasing when cooling, as shown in Fig. 2(a-c), and how the order returns despite the skyrmion sizes remaining at their low-temperature values when heating. We find that the strength of the applied field affects skyrmion lattice ordering as well as average skyrmion size, as shown in Fig. 2(d) which plots average skyrmion radius as a function of temperature when FC with five different applied field values. Finally, we will show how the skyrmion size changes can be understood by examining the temperature dependent magnetic parameters of FGT and how the skyrmion magnetic domain energy scales as a

function of temperature [5].



**Figure 1.** Néel skyrmion lattice in  $\text{Fe}_3\text{GeTe}_2$ . **(a)** LTEM image of FGT taken at -1 mm defocus and a  $22^\circ$  tilt angle at 100 K. **(b)** The reconstructed integrated magnetic induction shown for the white box in (a). Color wheel denotes the direction of the in-plane component of the magnetic induction.



**Figure 2.** Skyrmion size variation as a function of temperature. **(a-c)** LTEM images taken at -7 mm defocus taken during a field-cooling sequence with a 500 G out-of-plane magnetic field. Skyrmion sizes increase as temperature decreases, with images shown at (a) 200 K, (b) 180 K, and (c) 100 K. All images have the same scale. **(d)** Average skyrmion radius,  $\langle r_{\text{sk}} \rangle$ , plotted as a function temperature for five different applied field strengths.

#### References:

- [1] Fert, A., Reyren, N. and Cros, V., Nature Reviews Materials, **2** (2017), p. 17031. doi:10.1038/natrevmats.2017.31
- [2] Huang, P. et al., Nature Nanotechnology, **15** (2020), p. 761. doi:10.1038/s41565-020-0716-3
- [3] Zázvorka, J. et al., Advanced Functional Materials, **30** (2020), p. 2004037. doi:10.1002/adfm.202004037
- [4] Fei, Z. et al. Nature Materials, **17** (2018), p. 778. doi:10.1038/s41563-018-0149-7
- [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.