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# Room Temperature Néel-type Skyrmions in a van der Waals Ferromagnet Revealed by Lorentz 4D-STEM

Yu-Tsun Shao[1], Hongrui Zhang[2], Rui Chen[2,3], Xiang Chen[3,4], Sandhya Susarla[2,3], Jonathan T. Reichanadter[5,6], Lucas Caretta[2], Xiaoxi Huang[2], Nicholas S. Settineri[5], Zhen Chen[1], Jingcheng Zhou[2], Edith Bourret-Courchesne[3], Peter Ercius[7], Jie Yao[2,3], Jeffery B. Neaton[3,4,8], Robert J. Birgeneau[2,3], Ramamoorthy Ramesh[2,3,4] and David A. Muller[1,9]

[1.] Department of Applied and Engineering Physics, Cornell University, Ithaca, New York, USA.
[2.] Department of Materials Science and Engineering, University of California, Berkeley, CA, USA.
[3.] Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA, USA.
[4.] Department of Physics, University of California, Berkeley, CA, USA.
[5.] Department of Electrical Engineering, University of California, Berkeley, CA, USA.
[6.] Department of Chemistry, University of California, Berkeley, CA, USA.
[7.] The Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA, USA.
[8.] Kavli Energy Nanosciences Institute at Berkeley, Berkeley, CA, USA.
[9.] Kavli Institute at Cornell for Nanoscale Science, Ithaca, New York, USA.

Two-dimensional van der Waals (2D vdW) magnets offer an excellent platform for exploring magnetic and topological phases, owing to their unique layered structure and stacking-dependent crystal symmetries [1,2]. Magnetic skyrmions, which are topologically protected real-space swirling spin textures, are often stabilized by an antisymmetric Dzyaloshinksii-Moriya interaction (DMI) present in materials with broken inversion symmetry. Among the vdW materials for studying 2D magnetism, the $Fe_NGeTe_2$ (FGT, N=3-5) system is exceptional due to its tunability of magnetic properties with chemical doping and the existence of ferromagnetism above room temperature.

Here, we explore the chemically driven structural and magnetic phase transitions in $(Fe_{1-x}Co_x)_5GeTe_2$ (FCGT) using atomic resolution imaging and Lorentz four-dimensional scanning transmission electron microscopy (4D-STEM) along with an electron microscopy pixel array detector (EMPAD) [3]. We find that the FCGT changes both in structure and magnetic properties as a function of Co-doping from an antiferromagnetic, centrosymmetric AA-stacking (x=0.46) to a ferromagnetic, polar AA'-stacking (x=0.50). More interestingly, room temperature Néel-type skyrmions emerge in the AA' phase as revealed by Lorentz 4D-STEM [4].

Figure 1 gives atomic-resolution evidence of both AA- (space group $P\bar{3}m1$, No. 164) and AA'-stacked (space group $P6_3mc$, No. 186) phases in FCGT. The FCGT unit cell features layers of Te-Te planes along the c-axis separated by vdW spatial gaps. Within each Te-Te layer, there are three unique Fe Wyckoff positions, giving rise to Te-Fe1-Fe3-Fe2-Ge-Fe2-Fe3-Fe1-Te plane-like structure. The difference in stacking sequence between the two Te layers is captured by high-angle annular dark-field (HAADF-)STEM images. These images are false-color coded to illustrate the differences in stacking sequence, showing zig-FCG in green and zag-FCG in blue. More interestingly, atomic resolution energy dispersive x-ray spectroscopy (EDS) maps reveal the different Fe and Co ordering between the two phases.

Figure 2 a & b show the experimental LTEM images of labyrinthine textures in a 110nm-thick AA' FCGT nanoflake at 0º and 18º stage tilts, revealing the Néel character. An external field applied along the beam direction induces the magnetic phase evolution from stripes to a mixture of bubbles and short stripes (Fig. 2c), and finally to the bubbles showing dark/bright contrast (Fig. 2d). Figure 2e shows the magnetic induction field obtained by Lorentz 4D-STEM consisting of clockwise and counter-clockwise curls, which agrees well with the calculated induction field distribution for Néel-type skyrmions. In summary, our work paves the way for studying structural and magnetic phase transition in vdW magnetic materials [5].

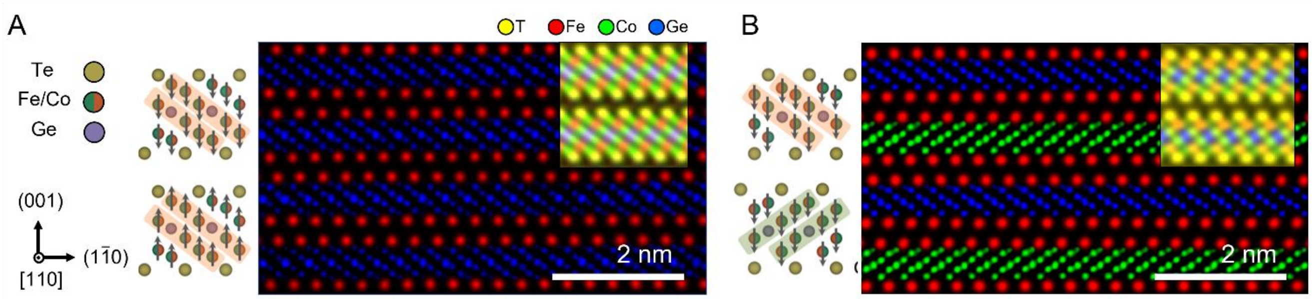


**Figure 1.** Structural phase transition of $(Fe_{1-x}Co_x)_5GeTe_2$. Structural model and corresponding HAADF-STEM cross-sectional images of FCGT for (A) AA stacking (x=0.45) and (B) AA' stacking (y=0.50) along the [110] direction. False colored sub-lattices of Te in red, zig-Fe-Co-Ge in green, and zag-Fe-Co-Ge in blue aided by 2D Gaussian fitting of atomic column positions. Inset, corresponding atomic resolution STEM-EDS images for both AA and AA' phases.

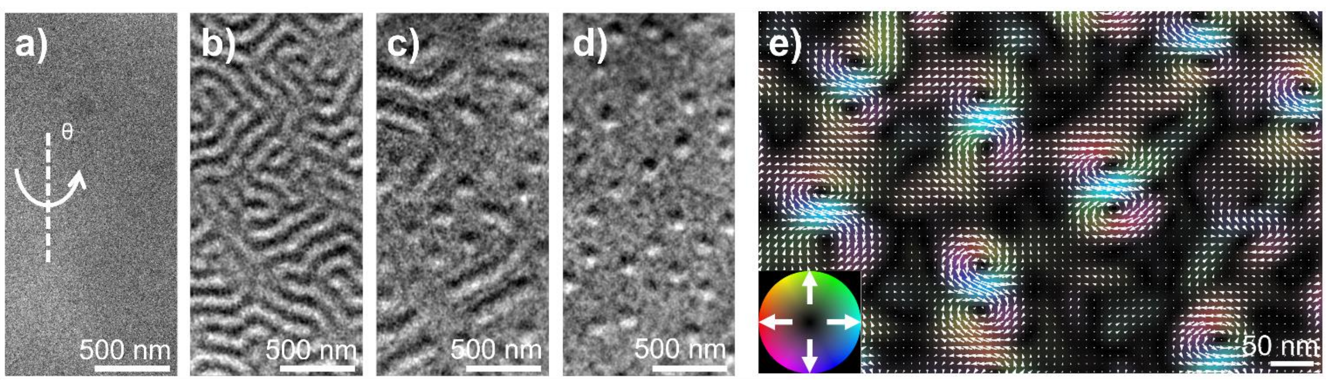


**Figure 2.** Room temperature magnetic textures probed by Lorentz (S)TEM. (a) Lorentz TEM images of the FCGT nanoflake acquired at the same region under zero-field and +4 mm defocus with 0º-tilt and (b) 18º-tilt reveal a labyrinth phase with Néel character. At the same region with 18º-tilt, Lorentz TEM images showing (c) a mixture of serpentine and skyrmions and (d) isolated skyrmions emerge at applied fields of 125 mT and 139 mT, respectively. (e) Magnetic induction map of Néel skyrmions obtained by Lorentz 4D-STEM recorded with an EMPAD. The color and arrows indicate induction field components perpendicular to the beam propagation direction for a 18º sample tilt.

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