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Correlative Lorentz and Dark Field TEM for Studying Skyrmion-Defect Interactions in Van-der-Waals Ferromagnet Co-doped Fe₅GeTe₂

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Magnetic spin textures are an exciting platform for next-generation low-energy devices. Electricfield control of topologically-protected magnetic skyrmions allows low-current reading and writing of stable, switchable bits. The stability granted skyrmions by their non-trivial topology is guaranteed in pristine substrates, but a skyrmion in any real device will interact with crystalline defects and interfaces. Skyrmion ordering behavior [1,2] and skyrmion behavior in a finite-sized substrate [3] have been studied in-depth, but skyrmion interactions with crystalline defects and interfaces remain uncharacterized.

Van-der-Waals layered Fe_5GeTe_2 becomes an inversion-symmetry-breaking ferromagnet with nonzero Dzialoshinskii-Moriya interaction (DMI) when doped to exactly 50% Co substitution of Fe sites, allowing for the formation of magnetic skyrmions [4,5]. This quasi-2D ferromagnet provides an opportunity to investigate the relationship between magnetic textures and material structure. The transmission electron microscope (TEM) allows for multi-length-scale correlational imaging of structure, defects, and magnetic field distribution under various temperature and magnetic field conditions. By imaging magnetic skyrmions with varying temperature and magnetic field and correlating those measurements with images of the material's structure and defects, the relationship between skyrmions and defects can be investigated.

Here we use Lorentz transmission electron microscopy (LTEM) correlated with dark field TEM imaging to study skyrmion behavior and ordering at and around crystalline line defects in FCGT. In LTEM mode, the objective lens of the microscope is turned off and a ferromagnetic sample can be imaged under relatively field-free conditions. Under ambient conditions, FCGT displays helical domains. In combination with a heating or cooling holder, FCGT can be field-cooled *in situ* in the TEM by heating to just above its Curie temperature (~100°C depending on thickness) and then cooled to room temperature with the TEM objective lens slightly excited to place the sample in a small out-of-plane magnetic field. This field-cooling process reliably creates skyrmions that are stable at room temperature and zero magnetic field (Fig. 1a). Crystalline defects are visible as lines in L/TEM images and can be characterized with dark field TEM imaging (Fig. 1b,c). Automated skyrmion and defect identification allow statistical analysis of

skyrmion locations with respect to defects (Fig. 1d), which provides evidence for interaction between skyrmions and crystalline defects (Fig. 1e) [6].



Fig 1. Analysis of skyrmion-defect interaction. (a) LTEM image of skyrmions and crystalline defects in FCGT. (b) On-axis bright-field TEM image of defects with inset diffraction pattern and schematic diffraction pattern. (c) Two-beam condition bright field TEM image with the - 3030 diffraction peak excited, shown in inset schematic diffraction pattern. (d) Map of automatically detected skyrmions and defects showing skyrmion-defect distances. (e) Histogram of skyrmion-defect distances, scaled by the average skyrmion radius

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