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Supplementary Information

Near room-temperature formation of a skyrmion crystal in thin-films of the helimagnet FeGe

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Lorentz transmission electron microscopy (TEM) with the applied magnetic field

The supplementary Figure 1 is the schematic ray diagram in the transmission electron microscopy (TEM). The parallel electron beam emitted from the electron gun (not shown) irradiates the thin plate sample (thickness t < 100 nm) located in the gap of the objective lens (magnetic lens), and is focused by the objective lens to the back focal (diffraction) plane, and then magnified on the image plane. A magnetic field $B_z \approx \frac{B_0}{1+(z/a)^2}$ is induced by the objective magnetic lens and is applied on the thin plate sample in the TEM. B_0 is a maximum field at z = 0 and depends on the lens current, and a the half-width at half maximum[1]. In the conventional high-resolution TEM, B_0 (~2 T) is large enough to shorten the focal length f_0 (~ 1mm) and to obtain large magnification at the image plane. Under such a strong magnetic field, the single ferromagnetic domain along the z direction (parallel to the incident electron beam) should be realized in FeGe. In this case, no in-plane magnetic component can be seen. To get a weaker magnetic field normal to the thin plate, the objective lens current should be reduced. The decrease of objective lens current should provide the longer focal length f_1 to record easily deflected electron beams (the gray cones in supplementary Fig. 1(a)) in the imaging plane. As the normal field is weak enough, the in-plane magnetic components in the thin plate should be observed. In this case, the incident electron beam should be deflected by the Lorentz forces induced by the in-plane magnetic components of the specimen,

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resulting in converges (bright contrast)/diverges (dark contrast) on the defocused (under/over-focused) image planes (shown in supplementary Fig. 1(b)). The inversion of such magnetic contrast can be seen between the over- and under-focused images[2]. On the other hand, out-plane magnetic components can not affect the electron propagation. In this study, we changed the objective lens current to control the magnetic fields applied to the specimen along the z-axis.

Quantitative evaluation of magnetic component mapping in the magnetic domain:

Magnetic transport-of-intensity (MTIE) method

In addition to the high-resolution magnetic domain image, the quantitative evaluation of magnetic components in a thin plate can be achieved by using a package of software QPt[3, 4], where two Lorentz TEM micrographs (under-, and over-focused images) were analyzed using the transport-of-intensity equation (TIE Eq.)[5-6],

$$\frac{2\pi}{\lambda}\frac{\partial I(xyz)}{\partial z} = \nabla_{xy}\left[I(xyz)\nabla_{xy}\phi(xyz)\right], \qquad (1)$$

that was derived from Schrödinger equation under the small-angle approximation when the optical wave propagates through a phase object[5]. $I(xyz)$ and $\phi(xyz)$ stand for the intensity and phase of propagating optical wave, respectively. TIE Eq. was recently applied to the simulation for TEM images[3, 4], since the thin plate for TEM observations can be also viewed as a weak phase object[1]. Maxwell-Ampére equations lead to a relationship between $\phi(xyz)$ and the magnetization $\overrightarrow{M}$ as

$$\nabla_{xy}\phi(xyz) = -\frac{\mathrm{e}}{\hbar}(\overrightarrow{M}\times\vec{n})t, \qquad (2)$$

where t is the thin plate thickness, $\vec{n}$ the unit vector perpendicular to the thin plate. We can obtain the in-plane magnetic component distribution M_{xy} as $\phi(xyz)$ is known.

$\phi(xyz)$ can be obtained by the Eq. (1) if $\frac{\partial I}{\partial z}$ is known. The change of electron intensity in the two (over/under) defocused planes $\frac{\partial I}{\partial z}$ can be recorded by Lorentz TEM observations with an approximation that

$$\frac{\partial I}{\partial z} \approx \frac{I(x, y, f_1 + \Delta z) - I(x, y, f_1 - \Delta z)}{2\Delta z}, \qquad (3)$$

where $2\Delta z$ is the distance between the over-focused and under-focused planes and f_1 ($>>\Delta z$) the focal length of the objective lens.

Magnetic component map in the helical domain

Supplemental Figure 2(a) shows the schematic three-dimensional (3D) helical spin structure in FeGe. When the thickness of thin plate is smaller than the helical period, the thin plate can be viewed as a 2D helimagnet. The Monte Carlo simulation shows the stripe pattern (see Fig. 2(b)) for magnetic structure in such 2D helimagnet[4]. In our Lorentz TEM observations, the thin plate can be prepared thin enough as the 2D helimagnet. The stripe pattern in the under (Fig. 2(c))/over (Fig. 2(d)) -focused Lorentz image is seen in a thin plate FeGe with the thickness of ~50 nm. By analyzing the under- and over-focused Lorentz TEM images with Qpt, the in-plane magnetic component was mapped (Fig. 2(e)). Such a distribution map of magnetic components agrees closely with the Monte Carlo simulation result (Fig. 2(b)).

Fluctuating skyrmions

Supplemental Figure 3 represents the formation of skyrmion crystal and the motion of skyrmions under a perpendicular magnetic field *B*. It was observed that the skyrmions start to emerge around the thin-plate edge when the magnetic field is up to 700 Oe. The coexistence of helical structure (the stripe pattern at the region far from the

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sample edge in Fig. 3(c)) and skyrmion lattice (around the sample edge in Fig. 3(c)) was observed in the range of magnetic field between 700 Oe and 800 Oe. As the magnetic field is over 900 Oe, the skyrmion crystal with the hexagonal symmetry (Fig. 3(d)) is generated and eventually fills the whole space of the thin plate. With further increasing *B* above 1600 Oe, the ferromagnetic (FM) domains composed of the out-plane component along the external field occur. Through the mixed state of skyrmion lattice and the FM domains (no magnetic contrast in Fig. 3(e)), the skyrmions are persistently discened even when *B* is as high as 1800 Oe.

Magnetic component map in the skyrmion lattice

As we applied a magnetic field of 1000 Oe normal to the thin plate FeGe, the under (Fig. 4(a))/over (Fig. 4(b)) focused Lorentz TEM image show the vortex-like magnetic structure, respectively. The magnetic component map obtained by the afore-mentioned MTIE method is shown in Fig. 4(c). Such a MTIE image demonstrates that the magnetization in each skyrmion has the lateral components perpendicular to the external magnetic field other than the core and the peripheral regions of skyrmion (the dark color in Fig. 4(c)), where the magnetic components are antiparallel or parallel to the field. According to the distinguishable magnetic contrast (see Figs. 4(a)-4(b)) between the center and periphery of skyrmions as well as to the different size of dark regions between the outside and core of skyrmions, we can conclude that the outside spins are parallel to the field, while the core spins are antiparallel to the field, although the Lorentz TEM image can record only the in-plane magnetic components.

References

1. L. Reimer and H. Kohl, *Transmission electron microscopy.* Fifth Edition (Springer, 2008) 146-152; 262-271.

2. P. J. Grundy and R. S. Tebble, *Adv. Phys.* **17** (1968) 153.

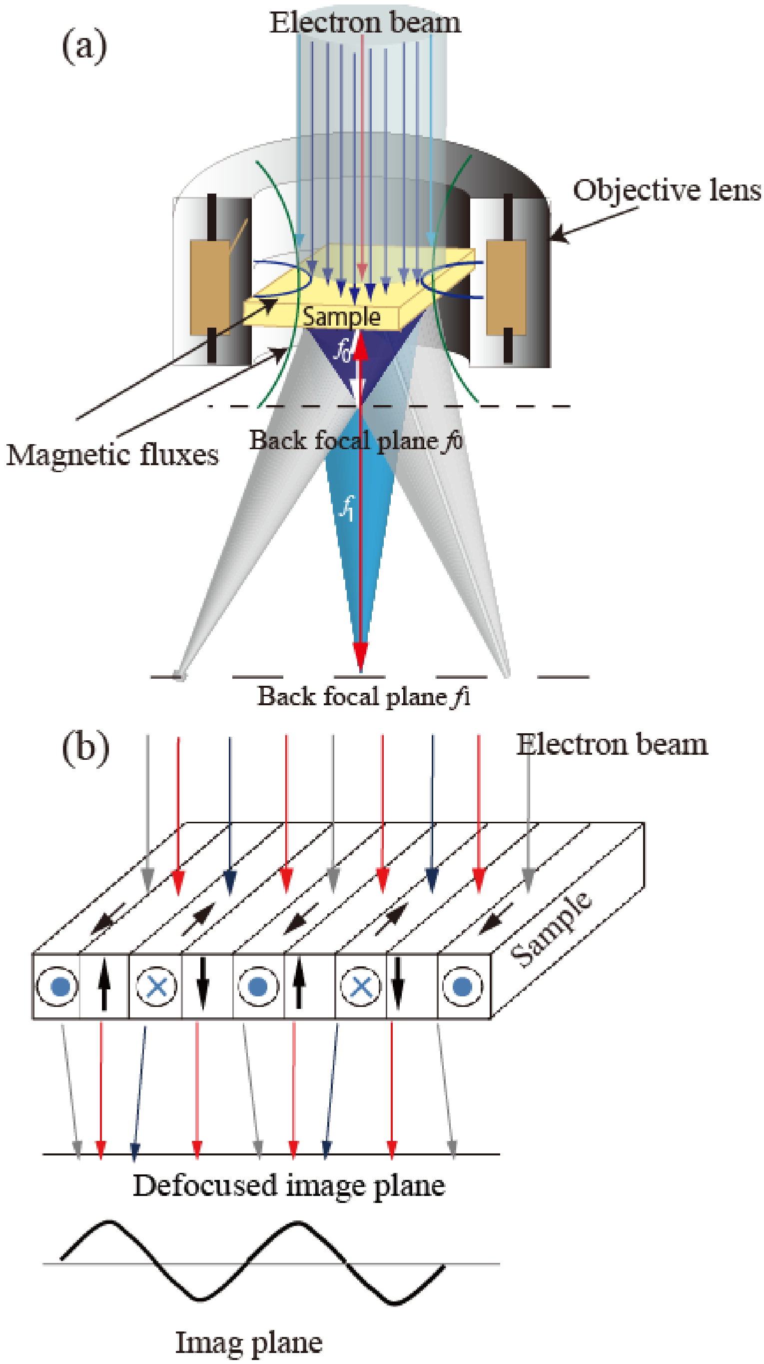
3. K. Ishizuka and B. Allman, *J. Electron Microsc.* **54** (2005) 191.

4. X.Z. Yu, et al., Nature **465** (2010) 901.

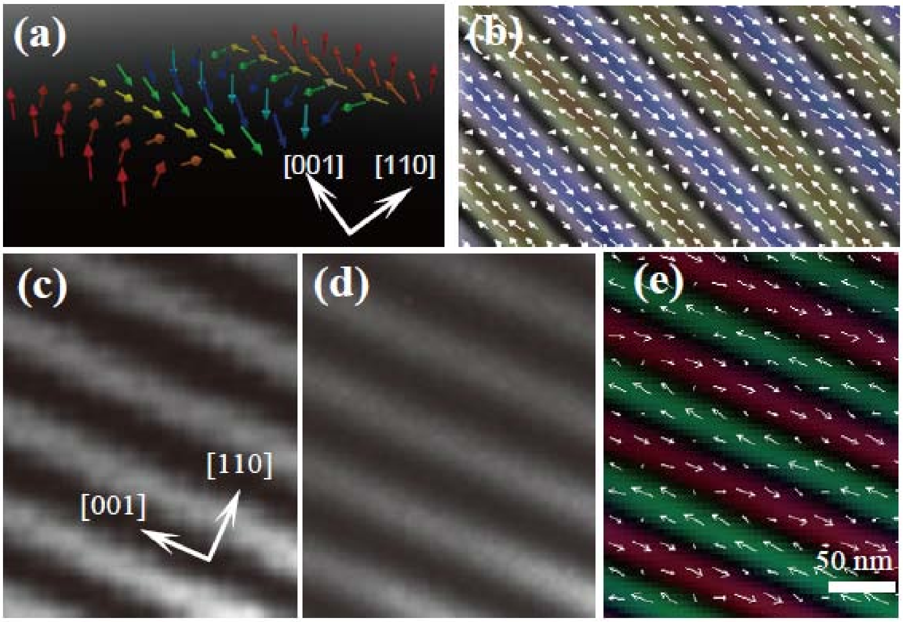
5. M.R. Teague, *J. Opt. Soc. Am*. **73** (1983) 1434.

6. S. Bajt, et al., *Uitramicroscopy* **83** (2000) 67.

 DOI: 10.1038/NMAT2916



Supplementary Fig. 1: Schematics of ray diagram for transmission electron microscopy (TEM) (upper panel) and Lorentz TEM (down panel).



Supplementary Fig. 2: Helical spin structure in FeGe. (a) Schematic of 3D helical spin structure. (b) Monte Carlo simulation image of 2D helical spin structure. (c-d) Under/over –focaused Lorentz TEM images. (e) Magnetic component map (MCM) of 2D helical spin structure. White arrows indicate the size and direction of magnetic components at every point.

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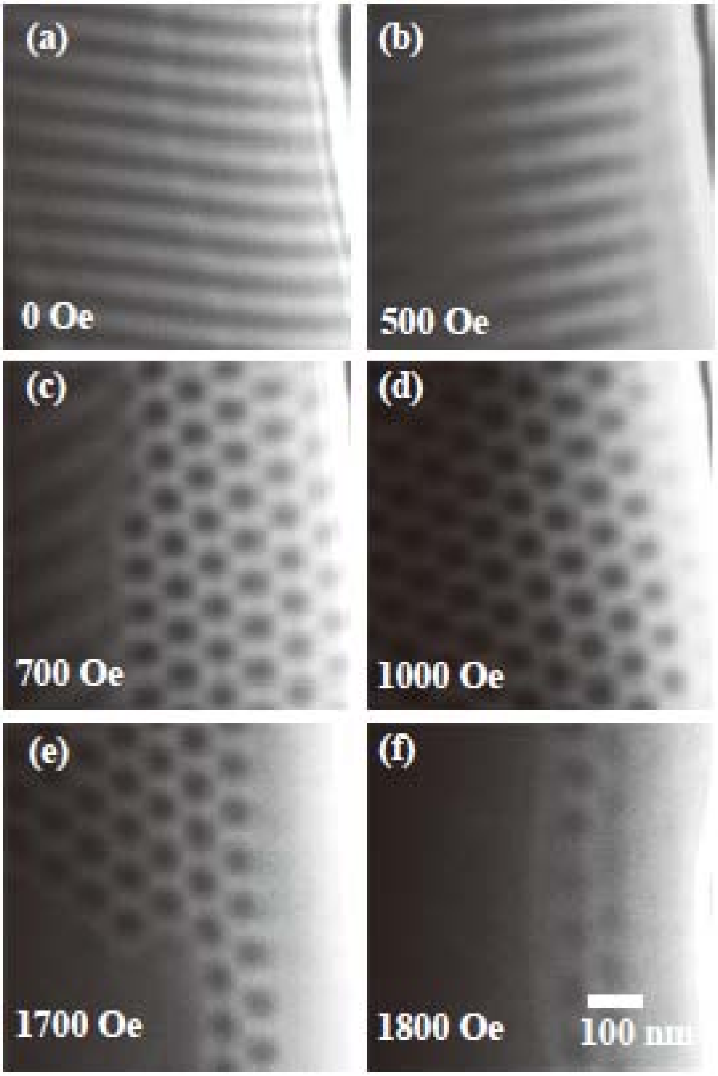
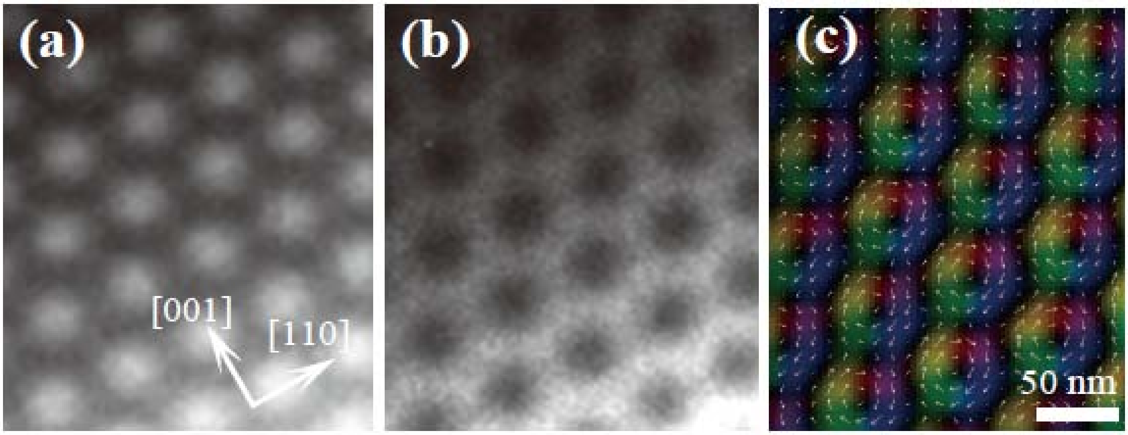


Supplementary Fig. 3: Over-focused Lorentz images of (110) FeGe at 250 K under various magnetic fields. The stripe pattern and the dark “particle” show the helical magnetic domain and the skyrmion, respectively.



Supplementary Fig. 4: Lorentz images (a-b) and (c) MCM of skyrmion lattice. (a) Under-focused image. (b) Over-focused image.