Supporting Information

Real-Space Observations of Three-Dimensional Antiskyrmions and Skyrmion Strings

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1. Sample preparation

Single crystals of Fe1.9Ni0.9Pd0.2P (FNPP) and Co9Zn9Mn2 were grown by a self-flux method and the Bridgman method followed by water quenching, respectively. The crystal orientations were checked by X-ray Laue diffraction as well as electron diffraction. To perform tomographic Lorentz TEM, the micron-sized samples were sliced along the (001) plane from bulk crystals and shaped using a focused ion beam system (Hitachi NB5000).

2. Methods

Generation of zero-field antiskyrmions and skyrmions

To excite zero-field antiskyrmions in FNPP, as shown in Figs. 1 and 2, we performed field cooling (FC) the sample in a transmission electron microscope (JEM2800, JEOL). Firstly, we attached a micron-size sample to a heating holder (HC3500, Gatan), which was placed on the sample stage above
an objective lens coil. Next, a current of approximately 1 A was passed through the objective lens coil to generate a field of 50 mT antiparallel to the incident electron beam and normal to the (001) plane of the FNPP sample. Then, we cooled the FNPP sample from 400 K to a room temperature in the 50 mT field and then turned off the objective lens current to obtain the zero-field magnetic configuration (Fig. S1). Finally, we moved the FNPP sample to a high-tilt sample holder (JEOL), allowing the TEM sample to be tilted by ±80°.

Metastable skyrmion strings involving a chiral bobber were generated in a micron-size magnet of Co$_9$Zn$_9$Mn$_2$ using the aforementioned FC procedure.

**Figure S1.** 2D Lorentz TEM images observed in a thin FNPP at zero magnetic field and RT. a-b, The (001)-zone over-focus (a) and under-focus (b) Lorentz TEM images. c, The corresponding phase image obtained by calculating over- and under-focus images using the transport of intensity equation$^{32-33}$.

**Tomographic Lorentz TEM imaging and reconstruction of 3D vector field maps for antiskyrmions and skyrmion strings**

1) **Tomographic Lorentz TEM**
Tomographic Lorentz TEM was performed in a low-magnification mode of a commercial transmission electron microscope (JEM-2800, JEOL), where the objective lens current was turned off to realize the zero external magnetic field state in a micron-size sample.

Firstly, we used software “Recorder” (tomography.com) to acquire a series of projected 2D Lorentz TEM images automatically by varying the tilt angle $\alpha$ around the $y$-axis from $-50^\circ$ to $+50^\circ$ in a $2^\circ$ increment, as illustrated in Fig. S2a. These Lorentz TEM intensity images relate to the phase differential $(\nabla_{xy}^2 \varphi)$, which are fundamental to observing magnetic configurations in magnets by Lorentz TEM: the magnetic components in materials interact with the incident electron beam and result in the Lorentz force, which is normal to both magnetization and the electron beam; the Lorentz force deflects the incident electron beam and gives rise to the intensity change corresponding to beam convergence (bright) and divergence (dark) in the defocus Lorentz TEM image plane when antiparallel magnetic domains, like a 180-degree domain structure, exist in the FM sample (this mode is called Fresnel Lorentz TEM)\textsuperscript{S1}. Accordingly, Lorentz TEM intensity images can visualize magnetic configurations in a wide range of magnets.

A Oneview camera from Gatan was used to collect data with an exposure time of 20 ms. The correlation for each image at different tilt angles was taken by referring to markers in the sample, and the focus was corrected before acquiring each image. The aforementioned procedure was carried out by varying the tilt angle $\beta$ around the $x$-axis. Then, the obtained images were stacked by the software package “Composer,” which enables 3D reconstruction from the previously recorded 2D Lorentz TEM images. The procedure for 3D reconstruction is as follows:

1) Align the images for the dual-axis tilt series using the markers; this reduces artifacts, such as diffraction contrast, during the sample tilting;

2) Reconstruct the 3D image by using the simultaneous iterative reconstruction technique;
3) Check the accuracy of the cross-correlations of the stacked images by using the line alignment cross-correlation.

Finally, the 3D viewer “Visualizer” (tomography.com) was employed to determine the volume function of the 3D magnetic configuration according to the real sample geometry. The color and transparency maps, as well as multislice images and a movie of the 3D object, were obtained at selected angles and positions, as shown in the main text and Supporting movies (Movie S1-S2).

2) Reconstruction of 3D vector field maps of an antiskyrmion and a skyrmion string

3D magnetic field reconstruction was performed with a custom program written in Python, which utilizes two open-source Python packages, PyLorentz and TomoPy (Fig. S2b for the reconstruction algorithm). Both packages have been described in detail by their creators, and here we shall only mention some pertinent features of TomoPy, which is the most essential part of our program. Besides traditional 3D reconstruction, TomoPy can generate a 3D vector field from a stack of 2D vector maps obtained at different tilt angles. TomoPy uses all tilt data for each iteration, which increases the convergence speed and allows reconstructing more than one vector component in one run. In contrast, most alternative programs use only one tilt axis set for each iteration, and hence can yield only one vector component at a time.
Figure S2. Schematic of tomographic Lorentz TEM imaging of 3D magnetic configurations of a single skyrmion string (a) and the flowchart of an algorithm for 3D vector field reconstruction (b).

Firstly, we aligned, cropped, and noise-filtered 2D projections of Lorentz TEM images acquired by tilting the sample along the x and y axes. Then, we processed the images with PyLorentz, which yielded 2D field maps of lateral magnetic field components $B_x$ and $B_y$ via the transport of intensity equation (TIE):

$$
\frac{2\pi}{\lambda} \frac{\partial I(xyz)}{\partial z} = -\nabla_{xy} \left[ I(xyz) \nabla_{xy} \varphi(xyz) \right].
$$

(1)
Here $\nabla_{xy}$ is the 2D Laplacian operator and $I(\text{xyz})$ is the electron intensity at the position with coordinates $(x,y,z)$. TIE relates the quantitative phase $\varphi$ and the difference of intensities $\frac{\partial I(\text{xyz})}{\partial z}$ between the over-focused and under-focused Lorentz TEM images. Using the Maxwell-Ampère equation\textsuperscript{S3}

$$\nabla_{xy} \varphi (\text{xyz}) = -\frac{e}{\hbar} \left( \vec{B} \times \vec{n} \right) t,$$

we can obtain $B_x$ and $B_y$ accumulated through the sample thickness from the phase $\varphi$. In equation (2), $t$ is the sample thickness and $\vec{n}$ is the unit vector parallel to the incident electron beam. To reduce artifacts in the retrieved phase image, we have adjusted the regularization parameter to $q_c=0.001$ using test images with known spin textures\textsuperscript{7}.

The projected 2D field maps were fed to TomoPy to generate 3D maps of $B_x$ and $B_y$ using the iterative algebraic reconstruction technique\textsuperscript{S4}. The third magnetic field component, $B_z$, was calculated from the zero-divergence condition $\partial B_x/\partial x + \partial B_y/\partial y + \partial B_z/\partial z = 0$ with the boundary condition of $B_z = 0$ at the sample surfaces. All $B$ values are presented in arbitrary units, as it was not possible to extract the absolute values using the current reconstruction method.

Our Lorentz TEM experiments were performed in the tilt angle range from $+50^\circ$ to $-50^\circ$. The main reasons for the limitation of the tilt angle were 1) the isolated zero-field antiSks can not be stabilized in samples thinner than 130 nm. Hence thicker samples were used but their imaging at 200KV-Lorentz TEM would become difficult at large tilt angle; 2) the mixture of antiSks and helices is hard to disentangle at high tilt angle. The relatively small range of experimentally accessible tilt angles (known as the "missing-wedge" problem\textsuperscript{S5}) reduces the accuracy of 3D reconstruction for all tomography techniques.

3) Acquisition of tilt series of Lorentz TEM images for an antiskyrmion

Figures S3a, c–j display a series of projected 2D Lorentz TEM images in the under-focused mode by sequentially varying the tilt angles $\beta$ around the x-axis (c-f) and $\alpha$ around the y-axis (g-j), while
Fig. S3b presents the 2D magnetic induction field map at zero tilt angle obtained by TIE analyses. Figs. S4a, c–j presents the simulated Lorentz TEM images at the same tilt angles as those in Figs. S3a, c–j, while Fig. S4b shows the simulated magnetic field map at zero tilt angle. The color wheel with white arrows encodes the direction of the in-plane field at every point in the antiskyrmion. Red, blue-green, yellow-green, and blue-magenta represent magnetic fields pointing to the right, to the left, up, and down, respectively; the relatively dark color indicates the dominance of out-of-plane fields, which are prominent on the core and periphery. The simulation results qualitatively agree well with the experimental observations.

Figure S3. A series of 2D projected Lorentz TEM images at tilt angles of (α, β) and an in-plane magnetic induction field (B) map (color image) at zero tilt of an antiskyrmion in the Fe$_{1.9}$Ni$_{0.9}$Pd$_{0.2}$P (FNPP) thin plate (thickness $t \approx 200$ nm). The tilt angles (α, β) and a color wheel for encoding the in-plane field direction are indicated in insets of the figure, respectively. The yellow
arrow shows the incident electron beam antiparallel to the z-axis. The coordinate systems (x, y, z) and (x’, y’, z’) correspond to a non-tilt sample and a tilted sample, respectively.

4) Lorentz TEM image simulations

Lorentz TEM images were simulated with a home-built program using a multislice Fourier transform approach with added Poisson noise. The images in Fig. S4 a, c-j were simulated at a defocus distance Δf of −50 µm. The simulated field map is represented in Figs. S4 b. All images were simulated via micromagnetic simulations using material parameters from Ref. [7] with a mesh size of 128 × 128 × 40 pixels³ corresponding to a sample size of 640 × 640 × 200 nm³.

Figure S4. A series of simulated Lorentz TEM images at tilt angles of (α, β) and an in-plane magnetic induction field (B) map (color image) at zero tilt for an antiskyrmion. The tilt angles (α, β) and a color wheel for encoding the in-plane field direction are indicated in insets of the figure.

5) Demagnetization effect on the surface magnetic structure of the antiskyrmion
The demagnetization field should affect the antiskyrmion spin texture in a thin TEM sample, but the expected effect is significant only at the sample surfaces, as shown in Fig. S5. The figure shows results of micromagnetic simulations using an open-source package Mumax3\textsuperscript{88} and the following standard energy functional:

$$E = \int \left\{ A (\nabla m)^2 + D m \cdot (\hat{x} \times \partial_x m) \times (\hat{y} \times \partial_y m) - M_s \cdot H - K_u (m \cdot \hat{z})^2 - \frac{1}{2} M_s m \cdot B_d \right\} dV. \quad (3)$$

Here, $A$ is exchange stiffness, $m$ is normalized magnetization, $M_s$ is saturation magnetization, $D$ is anisotropic bulk DMI constant, $H$ is external magnetic field, $K_u$ is the uniaxial anisotropy constant and $B_d$ is demagnetizing field. The simulation was performed in a volume of 368×368×192 nm$^3$ (the thickness is close to the experimental value) with Neumann boundary conditions (open boundary conditions). We started with a polarized state and then relaxed the magnetic state at zero field, creating one stable antiskyrmion. The parameters were taken from the literature\textsuperscript{7} were as follows: $M_s = 4.17 \times 10^5$ A m$^{-1}$, $K_u = 3.1 \times 10^4$ J m$^{-3}$, $D = 4 \times 10^{-4}$ J m$^{-2}$ (the same order as the value shown in the literature), $A = 8.1 \times 10^{-12}$ J m$^{-1}$, $H = 0$, and $T = 0$ K. The total magnetic induction $B$ (what we measure in the experiment) in a 208×208×192 nm$^3$ slice of the simulated array in which the antiskyrmion stabilized is shown in Fig. S5a. This figure shows the effect of the demagnetization field $B_d$ (Fig. S5b) on the magnetization $M$ (Fig. S5c). Please note that for each image stack we selected three images with the largest difference along the sample thickness ($z$ axis); the spin texture was uniform in the $z$ direction, and changed only at the sample surfaces.

The main features of the antiskyrmion affected by the demagnetization field are 1) expansion of the domain walls on the top and bottom surfaces, 2) a diagonal deformation on the top and bottom surfaces, and 3) the decrease in out-of-plane magnetic induction at the surfaces.
Figure S5. Simulated B-field (a), demagnetizing field $B_d$ (b), and magnetization $M$ (c) for the antiskyrmion at zero magnetic field. The dark/white arrows indicate the down/up fields or magnetizations, while colored arrows show the twisted fields or magnetizations in the antiskyrmion. In each stack, the top and bottom images correspond to the sample surfaces, and the middle image to the middle of the sample.

6) Acquisition of phase image series by sequentially tilting the needle-like FNPP sample

Figure S6 shows a tilt series of 2D phase images of needle-like FNPP that were used to reconstruct the 3D vector field map. At zero tilt, a mixture of antiskyrmions and elongated spin textures parallel to the sample edges in the FNPP can be discerned. When the sample is tilted around the x-axis, both the projected 2D antiskyrmions and elongated spin textures extend or shorten along the y-axis, whereas they expand or shorten along the x-axis as the sample is tilted around the y-axis. Some projected phase images (see the images at tilt angles of $-14^\circ$ and $-18^\circ$ around the x-axis and of $28^\circ$ and $-22^\circ$ around the y-axis) show extremely weak signals due to the local strain, diffraction effects
or interference between the incident electrons and crystal lattice during sample holder tilting (Fig. S6).

Figure S6. Projected phase images of a mixture of antiskyrmions and elongated spin textures obtained at various tilt angles about the x and y axes. The boxed area in the left image was selected to reconstruct 3D vector field maps of the spin textures, including two numbered antiskyrmions.

7) 3D vector field maps for antiskyrmions in the FNPP

Figure S7. 3D vector field maps of magnetic configurations observed in the FNPP (a-b), and the simulated 3D vector fields for an ideal antiskyrmion (c). a, 3D field map. b, Field map within
the xy-plane at z = 100 nm. Dashed lines in a and b indicate the antiskyrmion shown in Fig. 2c. c, 3D vector field map for an ideal antiskyrmion reconstructed from a tilt series of Lorentz TEM images simulated in the tilt range ±35°.

8) Spatial distribution of the magnetization and field maps for the antiskyrmion at zero tilt

Figures. S8a-b show the magnetization (a) and field (b) distributions in an antiskyrmion at zero tilt, simulated with Mumax3 and summed along the sample thickness. The maps reveal stray fields around Bloch lines. Such stray field arising from the nonzero divergence of magnetization has been discerned in the present experimental observation of the magnetic induction field map (c) for the antiskyrmion at z = 0.

![Figure S8. Simulated magnetization (a) and field (b) maps, and experimentally obtained field (c) maps of the antiskyrmion at zero tilt.](image)
The white arrows and colors in a indicate the magnetization directions, while those in b-c indicate the induction-field directions. Dark/bright color in a shows up/down magnetization, while dark colour in b-c indicates out-of-plane magnetic induction fields.

9) Acquisition of phase image series by sequentially tilting a deformed skyrmion string

The same 3D reconstruction procedure as described above was employed for a single skyrmion string by using a series of 2D phase images acquired by sequentially tilting the micron-size magnet
Co$_9$Zn$_9$Mn$_2$, as shown in Fig. S9. The series of phase images show the extension of the skyrmion string in the x- and y-directions for both the tilt axes due to the deformation of the skyrmion string (see the tomographic Lorentz TEM image in Fig. 3b). The reconstructed 3D vector field maps (Fig. S10) confirm the deformation of the skyrmion string, showing good agreement with our tomographic Lorentz TEM results (Figs. 3a–e and supporting Movie S2).

Figure S9. Series of projected phase images of a single skyrmion obtained at various tilt angles around the x and y axes.

Figure S10. Slices through the thickness of 3D field map for a deformed Sk string. The left-right image order corresponds to the top-bottom order in Fig. 3l.

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