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Real-space observations of three-dimensional antiskyrmions and skyrmion strings

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Nanometric topological spin textures, such as magnetic skyrmions (Sks) and antiskyrmions (antiSks), have attracted much attention in condensed matter physics and spintronics. To date, most studies have focused on two-dimensional Sks and antiSks in thin films with inherent or synthetic antisymmetric spin exchange interaction, termed Dzyaloshinskii–Moriya interaction, for their topological features and information-carrier functions. Although three-dimensional (3D) spin textures are ubiquitous, previous studies have focused mostly on cylindrical Sks or other non-topological magnetic vortices. Nontrivial 3D spin textures, such as antiSks composed of alternating Bloch- and Néel-type spin spirals, chiral bobbers carrying emergent magnetic monopoles, and deformed Sk strings, have rarely been identified, particularly on their topological nature. To elucidate these textures, we have developed a 3D nanometric magnetic imaging technique - tomographic Lorentz transmission electron microscopy. This approach enables not only the visualisation of 3D shape of magnetic objects, but also their 3D vector field mapping. The present research will lead to discoveries and advanced understanding of fertile 3D magnetic structures in a broad class of magnets, providing insight into 3D topological magnetism.
Electron spins and their manipulation have been extensively investigated in a broad area of science. In particular, vortex-like spin textures carrying an integer topological charge, such as the magnetic skyrmion (Sk) (Fig. 1a), have seen intensive research interest because of their emergent electromagnetic properties and information-carrier functionalities\textsuperscript{1–5}. The topological charge $N$ of spin textures with magnetic moment distribution $m(r)$ is defined by\textsuperscript{1}

$$N = \int \frac{d^2 r}{4\pi} \mathbf{n}_r \cdot \left( \frac{\partial n_x}{\partial x} \times \frac{\partial n_y}{\partial y} \right),$$

where $\mathbf{n}_r = m(r)/|m(r)| = (\cos \varphi(r) \sin \Theta(r), \sin \varphi(r) \cos \Theta(r), \cos \Theta(r))$ is a unit polarization vector, and $\varphi(r)$ is described by

$$\varphi(r) = m\phi + \gamma.$$  

Here, $\phi$ is the azimuthal angle of the point $r$, and $\Theta(r)$ is $0/\pi$ at $r = 0/\infty$, assuming the downward spin at the spin texture’s core ($r = 0$) and the upward spin at its periphery ($r \to \infty$). The $\gamma$ determines the in-plane spin rotation direction (helicity) related to the sign of the Dzyaloshinskii–Moriya interaction (DMI), and $m$ is the spin texture’s vorticity, $+1$ and $-1$ for Sks and antiskyrmions (antiSks), respectively, resulting in opposite-sign topological charges for Sk ($N = -1$) and antiSk ($N = +1$).

The ideal two-dimensional (2D) topological features of Sk and antiSk, namely the spin-swirling of Bloch type spiral ($\gamma = \pm \pi/2$) or Néel type spiral ($\gamma = 0$ or $\pi$) for a Sk with $m = +1$ and the alternating Bloch- and Néel-type spirals constituting a circular or square-shaped magnetic domain for antiSks with $m = -1$, have been confirmed by theoretical modeling\textsuperscript{1–2} as well as real-space imaging in several magnets\textsuperscript{3–7} with inherent broken inversion symmetry and in heterostructure films\textsuperscript{8}. However, the features of ideal Sks and antiSks often collapse or deform due to the effects of crystalline imperfections, local intrinsic magnetic anisotropy, and demagnetisation on the sample surface in actual materials\textsuperscript{9–11}. Such irregular topological spin textures cannot be well understood through the 2D magnetic imaging. Theoretical studies have predicted many intriguing 3D spin
textures, such as chiral bobbers\textsuperscript{12} and spin-hedgehogs\textsuperscript{13} carrying emergent magnetic monopoles, and 3D chiral solitons termed hopfions\textsuperscript{14}, for the interpretation of 2D micrography observations\textsuperscript{14–16}. However, these 3D topological models have not been directly visualized so far. Recent discoveries of novel transport properties such as giant topological Hall\textsuperscript{17, 18} and Nernst\textsuperscript{19} effects in chiral-lattice or centrosymmetric magnets hosting nanometric Sks also urge the detailed imaging of 3D magnetic configurations. From the perspective of applications, on the other hand, the genuinely-longitudinal Sk motion without Hall motion along the electric current flow direction is favourable for Sk-based devices. To realise the longitudinal motion of Sks with electric current flow, 3D hybrid topological spin textures with $N_{\text{total}} = 0$ ($N = +1$ and $-1$ for top- and bottom-layer Sks) are proposed in an antiferromagnetically exchange-coupled bilayer system\textsuperscript{8}. Similar hybridised Bloch- and Néel-type spirals are also proposed in multilayered films with interfacial DMI\textsuperscript{20}. In contrast to a simple Bloch-type or Néel-type spin whirl of skyrmions, antiskyrmion’s texture composed of alternating Bloch- and Néel-type spin spirals is theoretically predicted as a more detailed 3D spin texture. Bloch lines with Néel-type spin spirals at four corners of the squared antiskyrmion are crucial to determine its topology. However, these nontrivial 3D spin textures have not yet been confirmed due to the lack of 3D magnetic imaging technique for nanometric spin textures. Accordingly, the current trend aims to establish a 3D magnetic imaging technique to identify novel topological objects, such as 3D antiskyrmions and skyrmion strings.

Thus far, the state-of-the-art 3D magnetic imaging has been performed using X-ray magnetic circular dichroism, which is sensitive to magnetic components parallel to the X-ray. Two pioneering studies mapped scalar fields in 3D magnetic structures in a centrosymmetric magnet\textsuperscript{21} and Heusler alloy with a non-centrosymmetric lattice structure\textsuperscript{22}. The former established 3D magnetization maps for the conventional vortex and antivortex, and the latter revealed the 3D shape of Sk strings carrying topological defects in related micron-size magnets. As opposed to X-ray nano-tomography, electron holographic 3D imaging has also been performed for a magnetic thin film heterostructure\textsuperscript{23}, a cobalt
nanowire$^{24}$, and a chiral-lattice magnet FeGe hosting Sk arrays below room temperature (RT)$^{25}$. The 3D vector field ($\mathbf{B}$; hereafter, the field $\mathbf{B}$ indicates magnetic induction) mapping directly revealed vortices in formers, whereas possible Néel-type spin spirals were indicated near the sample surface in the latter. A few studies have also reported 3D reconstruction algorithms and their application to micrometre-scale artificial vortices$^{26-27}$.

Despite the exploitation of 3D imaging for magnetic twisted structures by nano-tomography, the difficulty in the 3D vector field mapping and insufficient spatial resolution of X-ray tomography, as well as the complicated hardware and reconstruction process required for electron holography, hinders the topologically-resolved mapping of 3D spin textures in magnets. The purpose of this study is to develop an *in-situ* tomographic imaging technique and an algorithm for 3D vector field reconstruction to facilitate the real-space observations of 3D topological spin textures, such as anti-SkS, Sk strings, and the chiral bobber carrying an emergent magnetic monopole, at zero magnetic field and RT. The realizations of atomic-scale spin spirals, such as magnetic monopoles and Bloch lines, require the 3D imaging technique with the high spatial resolution, which is challenging for the established X-ray tomographic technique. To solve the spatial resolution, we first developed an automatically reconstructed 3D scalar field imaging based on Lorentz transmission electron microscopy (TEM) with spatial resolution below 2 nm. In contrast with the conventional tomographic TEM technique for visualizing 3D crystal structures at in-focus mode, such atomic-scale tomographic Lorentz TEM for magnetic imaging should be performed at defocused mode (the magnetic induction in principle is zero at the in-focus mode). However, the blurred images obtained by systematically varying the sample orientation to build the 3D scalar field map are challenging for the 3D reconstruction. To overcome critical challenges, such as stabilizing antiskyrmions and skyrmion strings at zero field in TEM samples, image blurring, and overlapping the neighborhood spin textures at the relatively high tilt angle, we optimized the proper sample geometry and the defocused conditions to suppress the image blurring. In addition, we installed the scripts in the TEM control
center to automatically build the 3D scalar field map in skyrmion strings carrying the magnetic monopole and the Bloch lines constituting antiskyrmions.

**Identification of topological features of antiSks via tomographic Lorentz TEM**

First, to reveal the 3D spin textures of antiSks, we have examined the magnetic configurations of single-crystal Fe$_{1.9}$Ni$_{0.9}$Pd$_{0.2}$P (FNPP) at zero field and RT by the tomographic Lorentz TEM. FNPP has a tetragonal structure with $S_4$ symmetry (Fig. 1b)$^7$. The broken inversion symmetry of the crystal structure gives rise to DMI. The $I\bar{4}$ space group ($S_4$ symmetry) provides two orthogonal spin-spiral wavevectors with opposite spin helicities, right-handed (CCW) and left-handed (CW) screws, along the [110] and $[\bar{1}10]$-axes (Fig. 1c), respectively. The competition among the ferromagnetic exchange interaction, dipolar interaction and DMI enables the formation of antiSks (Fig. 1d) above RT when an external magnetic field is applied along the [001]-axis. We adopted field cooling (FC, see details in Methods) to stabilise a mixture of in-plane helices (stripes in Figs. 1e-1f) parallel to the [110]- and $[\bar{1}10]$-axes and antiSks (square-shape textures in Fig. 1e) at zero field in a (001) FNPP thin plate with a dimension of $1.53 \times 3.36 \times 0.2$ (μm)$^3$. Prior to the investigation of the 3D magnetic configurations in FNPP, we identified antiSks and orthogonal helices. We analysed a series of defocused Lorentz TEM images for the surrounding region in Fig. 1e to obtain their magnetic induction maps (Fig. 1g) by performing transport-of-intensity (TIE)$^{28}$ analyses (see details in Methods). The magnified field maps for the helices (Fig. 1h) and antiSk#2 (Fig. 1i) clearly demonstrate the perpendicular nature of helices with CW (blue and yellow stripes) and CCW (red and green stripes) helicities and an antiSk composed of four alternately-arranged Bloch lines with Néel-type spirals and Bloch-type spirals$^{29}$.

To reveal the 3D vector field distribution in the antiSk and hence to identify its topological nature, we developed an *in-situ* tomographic Lorentz TEM technique. Firstly, tilt series of 2D projected Lorentz TEM images around the x-/y-axis were recorded for tilt angles ranging from $-50^\circ$ to $+50^\circ$ at $2^\circ$ intervals by using the script package “Recorder” (see details in Methods) installed in the TEM control unit. Extended Data Fig. 1 presents the dual-axis (x and y) tilt series of projected 2D Lorentz...
TEM images and zero-tilt induction field map for a single antiSk. The changes of projected Lorentz TEM images of the antiSk with varying the tilt angle qualitatively agree with the simulations (Extended Data Fig. 2). Then, we construct the 3D magnetic configurations from the tilt series of the 2D Lorentz TEM images by using the TEM tomography package “Composer” and followed by a 3D viewer “Visualizer” (see details in Methods section). Figures 1e and 1f show top and oblique views extracted from the 3D shape of magnetic textures (see Movie S1 in the supplementary information (SI), displaying 3D images from arbitrary angles), respectively, which are represented by the difference of intensities $|I(xyz + \Delta f) - I(xyz - \Delta f)|$ ($\Delta f = 100 \mu \text{m}$) between overfocused and underfocused Lorentz TEM corresponding to the phase shift $\varphi(xyz)$ of electron wave. The magnetic twists composed of magnetic moments to appear in green, whereas the linear spin textures (magnetic domains) show dark colour. Notably, the tomographic Lorentz TEM results (Fig. 1f) clearly reveal that the magnetic twists align almost uniformly through the entire thickness of the sample. Although the shape of spin texture does not change along the z-axis, intensity difference appears between the surfaces and centre, as shown in Figs 1j–1l: the intensity is weaker around the top/bottom surface (Fig. 1j, 1l), while becomes stronger at the deep in the bulk (Fig. 1k). The variation in the intensity between the surface and the bulk inside suggests a hybridised magnetic structure of Bloch-type twists in the bulk and Néel-type twists at the surfaces, which should decrease the intensity of magnetic signals due to the cancellation of the in-plane magnetic moments projected in Lorentz TEM images. Another possible reason for the reduction of magnetic signal at the sample surfaces would be the damaged layers with several-nanometres thickness lying close to each surface, which is caused by the focused ion beam during the thinning process. Such crystalline imperfections on surfaces should reduce more or less the local ordered moments and weaken related intensities.

**Visualization of the 3D vector field map in the antiSk**

To reconstruct 3D vector field maps of the antiSk from series of 2D field maps obtained by sequentially tilting the sample around the x- and y-axes, we prepared a needle-like sample, as shown
in Fig. 2a. The 2D Lorentz TEM image (a, underfocused) and corresponding magnetic induction field map (b) show a mixture of antiSks and in-plane helices at zero field and RT. We recorded tilt series of underfocused and overfocused Lorentz TEM images, with tilt angles α and β around the y- and x-axes (see details in Extended Data Fig. 1), respectively, ranging from −50° to +50° with a 5° increment. We then extracted the corresponding phase images (Extended Data Fig. 4) and magnetic induction field at every tilt angle from −35° to +35° by performing TIE analyses; note here that the magnetic contrast became extremely weak due to the loss of electron wave amplitude with increasing effective sample thickness when the tilt angle was greater than 35°. The 3D phase and field maps were obtained using the software “TomoPy” (see details in Extended Data Fig. 3 and the related description of the 3D reconstruction procedures in the Methods). Movie S2 shows the 3D phase configurations from arbitrary view angles, and Extended Data Fig. 5 shows the 3D vector-field maps (a) and corresponding (b) xy map at z = 100 nm of the surrounding area in Fig. 2a composed of two antiSks and two elongated spin textures, respectively. The 3D phase images demonstrate that 1) the Bloch-type spirals (black colours in the phase image of Movie 2) continuously extend from the top surface to the bottom; 2) four Bloch lines (light grey in the phase images) between Bloch-type spirals constitute antiSks. The 3D vector field maps reveal complicated spin textures (Extended Data Fig. 5a) while twisted textures in the antiSk (the surrounded area with a dashed rectangle in Extended Data Fig. 5b) can be discerned. To show the vector field map of the antiSk clearer, we plot the 3D field map for one antiSk (surrounded by white dashed lines in Fig. 2b) in Figs. 2c–d, representing the downward moments in the core as indicated by dark arrows and the upward moments at the periphery as indicated by bright arrows. The twisted moments (gradational purple and green arrows in Figs. 2c–d) between the downward (the −z direction) and upward (the +z direction) moments, and four Bloch lines (Néel-type spirals) at the four corners (Figs. 2c, e–g), are discerned. Between the neighbouring Bloch lines, the gradational red arrows display the magnetic moments pointing to the right, the blue arrows show the magnetic moments pointing to the left, and the green and purple arrows indicate the
magnetic moments pointing to the +y and the −y directions, respectively; see also the xy-plane images in Figs. 2e–g. The field distributions at the top (e, z = 200 nm) and bottom (g, z = 0 nm) surfaces are almost the same as those at the centre (g) (z = 100 nm), except for that at edge corners of field maps (possibly induced by the neighbouring spin textures in the sample), indicating a simple and ideal antiSk configuration, in good agreement with the theoretical predictions. Note that the difference between the vector field map and the magnetisation map of an ideal antiSk is observable by micromagnetic simulations (Extended Data Figs. 6a-b) and confirmed by Lorentz TEM observations (Extended Fig. 6c) due to the existence of stray fields around Bloch lines, where the divergence of magnetisation, and hence magnetic charge should not be zero. However, such stray fields hardly affect the fundamental 3D topological features of the antiSk identified here (Fig. 2).

**Identification of Sk strings and chiral bobber carrying the magnetic monopole via 3D magnetic imaging**

We apply the aforementioned tomographic Lorentz TEM to examine topological defects, such as emergent magnetic monopoles involved in broken Sk strings\(^9,10,13,16\). One such example, the chiral bobber\(^12\) (schematically shown in Fig. 3h) carrying an emergent magnetic monopole at its end, attracts much attention as the most typical 3D topological defect, and yet has not been confirmed by 3D real-space imaging. Here, we used the aforementioned FC process to form metastable zero-field Sks in a chiral lattice (\(P4_{1}32\)-space group) magnet \(\text{Co}_9\text{Zn}_9\text{Mn}_{230-31}\) with a dimension of \(1\times0.2\times0.25\ (\mu\text{m})^3\). We confirmed the isolated 3D string-like structures by tomographic Lorentz TEM, as shown by an xy top view (Fig. 3a) and an xz cross-section view at \(y = 100\ \text{nm}\) (Fig. 3b). Skyrmionic circular domains with an average diameter of about 80 nm are initially observed in the xy-plane. The Sks numbered #1–4 are discerned as curved strings (see a schematic of the deformed Sk string in Fig. 3k) in the xz cross-section image. In particular, a short one (#2) ends near the centre of the magnet \((z = -150\ \text{nm})\), exhibiting the chiral bobber-like feature (Fig. 3h). Such chiral bobber-like structures and deformed strings are also confirmed by several xy-slices at different z-values, as shown in Figs. 3e–e. The slice
at $z = -50$ nm close to the top surface exhibits only three circular domains corresponding to strings #1, #3, and #4, whereas chiral bobber #2 is absent. Meanwhile, the slice at $z = -200$ nm close to the bottom surface displays all circular domains with comparable intensities, and the slice at $z = -250$ nm at the bottom surface reveals the strongest intensity for chiral bobber #2 but extremely weak intensities for Sk strings #1, #3, and #4. These observations directly demonstrate the 3D aspects of deformed Sk strings #1, #3, and #4 and chiral bobber #2 close to the bottom surface. To confirm the topological aspects of the deformed Sk string and chiral bobber, we analysed Lorentz TEM images of top and oblique views of the string and bobber by the TIE. The induction field maps at zero tilt (Fig. 3f, 3i) and the projected field maps at $50^\circ$ tilt for the bobber (Fig. 3g) and deformed string #1 (Fig. 3j) show that there is no significant difference for these spin textures as viewed from the perpendicular direction to the sample plate (zero tilt). However, the oblique field maps discern topological distinctions between the string and the bobber (Fig. 3g, 3j): the string extends to the top surface, whereas the bobber ends deep within the bulk, revealing monopole textures at the bobber’s end.

To fully identify the spin textures of the deformed string in the bulk, we reconstructed the 3D vector field map of Sk string #4, which was isolated in the ferromagnetic background and did not overlap with other Sk strings even at tilt angles greater than $45^\circ$. Extended Data Fig. 7 shows a series of phase images of the Sk string obtained by sequentially tilting the sample around both the x- and y-axes in a $5^\circ$-step increment. Note here again that a tilting angle above $50^\circ$ increases the effective thickness of the sample to about 300 nm, resulting in the loss of the electron wave amplitude. We used this series of phase images to reconstruct the 3D vector field map for the Sk string. The field maps through the entire thickness of the Sk string (from the top to bottom), as shown in Fig. 3l, demonstrate that the spin swirls keep at depth in the bulk while collapsing at both the top and bottom surfaces (see changes of coloured arrows through the thickness). Figure 3l also reveals that the string is highly deformed deep within the bulk, while maintaining its vorticity and helicity throughout the
string. Our tomographic Lorentz TEM results provide the first experimental evidence of the 3D topological spin textures associated with highly-deformed metastable Sk strings in a chiral-lattice magnet at zero field and RT.

The 3D tomographic Lorentz TEM imaging technique developed here will enable the visualization and magnetic vector mapping of unknown nanometric 3D magnetic configurations with non-trivial topology in magnetic materials, such as hopfions, and provide profound insight into 3D topological magnetism.
References


**Supplementary Information** is available with the online version of the paper.

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**Author Contributions**

X. Y., Y. Taguchi, T. Arima and Y. Tokura conceived the project. X. Y. performed tomographic Lorentz TEM, analysed the experimental data, and wrote the manuscript with Y. Tokura. K.V. I.
developed the algorithm for the 3D vector field map reconstruction and wrote the related description in the Methods section. F. S. Y. contributed to the antiskyrmion simulations and wrote the corresponding description in the Methods section. L. C. P. and K. N. prepared the TEM sample. K. K. and Y. Taguchi synthesised the bulky single-crystal samples. All authors discussed the data and commented on the manuscript.

Competing interests

The authors declare no competing interests.

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Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.
Figure 1: Tomographic Lorentz transmission electron microscopy (TEM) images of antiskyrmions accompanied by orthogonal helices in a non-centrosymmetric magnet 

Fe_{1.9}Ni_{0.9}Pd_{0.2}P (FNPP) with the M_{3}P-structure (M: transition metal). a–d, Schematics of (a) Bloch-type skyrmion, (b) the crystal structure of FNPP viewed from the [001] direction, where the sites $M_1$, $M_2$ and $M_3$ are occupied by Fe, Ni and Pd. (c) orthogonal helices along the [110] and [1\bar{1}0] axes, and (d) an antiskyrmion. Colour arrows in a-d indicate the direction of electron spins, and CW and CCW stand for clockwise (left-handed) and counter-clockwise (right-handed) helicities, respectively. e-f, Top (e) and oblique views (f) of the difference of intensities $|I(xyz + \Delta f) - I(xyz - \Delta f)|$. 

!![](image)
$\Delta f$ (\(\Delta f = 100 \, \mu m\)) between overfocused and underfocused Lorentz TEM of 3D magnetic configurations composed of antiskyrmions (square-shape domains) and helices (stripes) in a (001) thin plate of FNPP. The colour bar shows the arbitrary intensity. \(g\), The 2D field map of the area inside the dashed rectangle in \(e\). \(h-i\), Enlarged views of the field map in \(g\) for (\(h\)) orthogonal helices (corresponding to the region surrounded by dashed lines in \(g\)) and (\(i\)) antiskyrmion #2. The colour wheel and white arrows indicate the magnitude and direction of the in-plane fields. Two yellow and two black arrows in \(i\) indicate Bloch lines in the antiskyrmion. \(j-l\), Tomographic images of (\(j\)) a slice around the top surface, (\(k\)) a middle, and (\(l\)) around the bottom surface of the magnified 3D magnetic configurations for the surrounding area in \(e\). The sample orientations are indicated by the red (\(x\)/[110]-axis)/green (\(y\)/[1\(\bar{1}\]0]-axis)/blue (\(z\)/[001]-axis) arrows.
Figure 2: Three-dimensional (3D) field maps of an antiskyrmion observed in a needle-like FNPP. a, Under-focus Lorentz TEM image and b, its magnetic induction field map. The colour wheel shows the in-plane-field direction at every point. c, 3D vector field map of an antiskyrmion, corresponding to the area surrounded by white dashed lines in b. The direction of the coloured arrows is indicated by a colour ball (the right-upper corner of c). d, Field maps of the antiskyrmion in the xz plane at y = 100 nm. Dark arrows show magnetic fields pointing down (-z direction) at the antiskyrmion core, while bright arrows indicate the fields pointing up at the peripheral of the antiskyrmion, respectively. e–g, Field maps of the antiskyrmion in the xy plane at (e) z = 200 nm (top-surface slice), (f) z = 100 nm (central slice), and (g) z = 0 nm (bottom-surface slice).
Figure 3: Tomographic Lorentz TEM images and corresponding vector field maps of a chiral bobber and deformed skyrmion strings in a chiral-lattice micromagnet $\text{Co}_9\text{Zn}_9\text{Mn}_2$ with dimensions of $1 \times 0.2 \times 0.25 \, (\mu\text{m})^3$. a, Top and b, cross-section views of three isolated skyrmion strings (numbered #1, #3 and #4 in Fig. 3a) and one chiral bobber (#2 in Fig. 3a) observed by tomographic Lorentz TEM. c–e, Slices of the 3D magnetic configurations of skyrmion strings and the bobber at different thicknesses marked by white dashed lines in b: (c) near the top-surface ($z = -50 \, \text{nm}$), (d) close to the sample centre ($z = -200 \, \text{nm}$), and (e) near the bottom-surface ($z = -250 \, \text{nm}$). Orange
dashed lines are eye guides for strings. The coloured arrows in **a-e** indicate the x (red), y (green) and z (blue) axes, respectively. **f-g, i-j**, Two-dimensional projections of field maps at tilt angles of (f, i) $0^\circ$ and (g, j) $50^\circ$ for (f-g) the bobber #2, and (i-j) a skyrmion string #1. **h and k**, Schematics of (h) the chiral bobber carrying a magnetic monopole (a blue dot at the end), (k) a deformed skyrmion string, respectively. **l**, Several xy-slices of a 3D vector field map of deformed skyrmion string #4 from top to bottom. The coloured arrows encoded by the colour ball show the magnetic field swirling while dark and white arrows indicate the down and up field directions, respectively.
Methods

1. Sample preparation

Single crystals of Fe$_{1.9}$Ni$_{0.9}$Pd$_{0.2}$P (FNPP) and Co$_9$Zn$_9$Mn$_2$ were grown by a self-flux method$^7$ and the Bridgman method followed by water quenching$^{30,31}$, 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. Generation of zero-field antiskyrmions and skyrmions

To excite zero-field antiskyrmions in FNPP micron-size magnets, as shown in Figs. 1 and 2, we performed in-situ 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. 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.

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

Extended Data Figs. 1a, 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 Extended Data Fig. 1b presents the 2D magnetic induction field map at zero tilt angle obtained by TIE analyses$^{28}$. Extended Data Figs. 2a, c–j present the simulated Lorentz TEM images
at the same tilt angles as those in Extended Data Fig. 1a, c–j, while Extended Data Fig. 2b shows the simulated magnetic field map at zero tilt angle. The colour 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 colour 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.

Extended Data Figure 1: Series of 2D projected Lorentz TEM images at tilt angles of (α, β) and an in-plane magnetic induction field (B) map (colour image) at zero tilt of an antiskyrmion in the Fe_{1.9}Ni_{0.9}Pd_{0.2}P (FNPP) thin plate (thickness t ≈ 200 nm). The tilt angles (α, β) and a colour 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\textsuperscript{32,33} with added Poisson noise. The images in Extended Data Figs. 2 a, c-j were simulated at a defocus distance $\Delta f$ of $-50$ µm. The simulated field map is represented in Extended Data Figs. 1 b. All images were simulated via micromagnetic simulations using material parameters from Ref. [7] with a mesh size of $128 \times 128 \times 40$ pixels\textsuperscript{3} corresponding to a sample size of $640 \times 640 \times 200$ nm\textsuperscript{3}.

Extended Data Figure 2: Series of simulated Lorentz TEM images at tilt angles of $(\alpha, \beta)$ and an in-plane magnetic induction field (B) map (colour image) at zero tilt for an antiskyrmion. The tilt angles $(\alpha, \beta)$ and a colour wheel for encoding the in-plane field direction are indicated in insets of the figure, respectively.
5. Tomographic Lorentz TEM imaging and reconstruction of 3D vector field maps for antiskyrmions and skyrmion strings

5-1 Tomographic Lorentz TEM

Tomographic Lorentz TEM was performed in a low magnification mode of a commercial transmission electron microscope (JEM2800, JEOL), where the objective lens current was turned off to realize the zero external magnetic field state in the 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 Extended Data Fig. 3a. A fast-speed camera (Oneview, Gatan) with an exposure time of 20 ms was used to collect data. Drift of images with varying tilt angles was corrected by referring to markers in the sample, and the focus correction was checked out during sequential Lorentz TEM observation. The aforementioned procedure was carried out by varying the tilt angle $\beta$ around the x-axis. Then, the obtained images were stacked by a software package “Composer,” which enables the reconstruction of 3D Lorentz TEM images from previous 2D Lorentz TEM images obtained. The procedure for 3D reconstruction is as follows:

1) Align the images for the dual-axis tilt series by taking the correct correlation using the markers, which enables the reduction of artifacts, such as diffraction contrasts, during tilt processes;

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 colour and transparency maps as well as the multislice and movie of the 3D object were obtained, as shown in the main text and Supplementary movies at arbitrary angles and positions.

5-2 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 utilises two open-source Python packages, PyLorentz and TomoPy (see Extended Data Fig. 3b 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.

Firstly, we aligned, cropped, and filtered tilt series of 2D projections of Lorentz TEM images (along the x-/y-axis) accumulated intensities of electron beam through the sample. Then, we proceeded in analyses of images with PyLorentz, yielding 2D field maps of lateral magnetic field components $B_x$ and $B_y$ via the transport of intensity equation (TIE)\textsuperscript{28, 34-35} described as

$$\frac{2\pi}{\lambda} \frac{\partial I(\text{xyz})}{\partial z} = -\nabla_y [I(\text{xyz})] \nabla_y \varphi(\text{xyz}).$$

\hspace{1cm} (3)

TIE gives a relationship of the quantitative phase $\varphi$ and the difference of intensities $\frac{\partial I(\text{xyz})}{\partial z}$ between overfocused and underfocused Lorentz TEM images. Because of the Maxwell-Ampére equation

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

\hspace{1cm} (4)

we can obtain the magnetic inductions $B_x$ and $B_y$ accumulated through the sample thickness from the phase $\varphi$. In equation (4), $t$ is the plate thickness and $\vec{n}$ is the unit vector parallel to the incident electron beam. These 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{36-38}. The third magnetic field component, $B_z$, was calculated from the zero-divergence condition $\frac{\partial B_x}{\partial x} + \frac{\partial B_y}{\partial y} + \frac{\partial B_z}{\partial z} = 0$. All $B$ values are presented in arbitrary units, as it is not possible to extract the absolute $B$ 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 this limitation were the interaction and consequent overlap among neighbourhood spin textures, which occurred at relatively large angles. The range of experimentally accessible tilt angles (known as the "missing-wedge" problem $^{36-38}$) hinders 3D reconstruction via common techniques, wherein iterations are performed sequentially for each tilt axis. In contrast, in our TomoPy-based reconstruction algorithm, iteration was performed simultaneously over two tilt axes$^{36}$. Thus, the requirement for the maximum tilt angle was significantly reduced. Note that the use of two (rather than three) tilt axes is sufficient for self-consistent 3D vector reconstruction$^{36-37}$.

configurations of a single skyrmion string (a) and the flowchart of an algorithm for 3D vector field reconstruction (b).
**5-3 Acquisition of phase image series by sequentially tilting the needle-like FNPP sample**

Extended Data Fig. 4 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.

We reconstructed the 3D phases using the data presented in Extended Data Fig. 4 for two antiskyrmions and two partial extended spin textures (enclosed by a yellow rectangular in the phase image at zero tilt), as shown in Supplementary Movie 2 for the view from arbitrary angles.

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.

5-4 The 3D vector field maps for antiskyrmions in the FNPP

Extended Data Fig. 5: 3D vector field maps of magnetic configurations squared in the Extended
Data Fig. 3 observed in the FNPP. 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. 2.

5-5 The spatial distribution of the magnetisation and field maps for the antiskyrmion at zero
tilt

Extended Data Figs. 6a-b show the simulated magnetisation (a) and field (b) distributions in an
antiskyrmion at zero tilt, revealing stray fields around Bloch lines. Such stray field arising from the
nonzero divergence of magnetisation has been discerned in the present experimental observation of
the magnetic induction field map (c) for the antiskyrmion at z = 0.
Extended Data Fig. 6: Simulated magnetisation (a) and field (b) maps, and experimentally obtained field (c) maps of the antiskyrmion at zero tilt. The white arrows and colours in a indicate the magnetisation directions, while those in b-c indicate the induction-field directions. Dark/bright colour in a shows up/down magnetisation, while dark colour in b-c indicates out-of-plane magnetic induction fields.

5-6 Acquisition of phase image series by sequentially tilting a deformed skyrmion string

The same 3D reconstruction procedure as described in 5.3 section 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 Extended Data Fig. 7. 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. 3l) confirm the deformation of the skyrmion string, showing good agreement with our tomographic Lorentz TEM results (Figs. 3a–e and Supplementary Movie 3).

Extended Data Figure 7: Series of projected phase images of a single skyrmion obtained at various tilt angles around the x- and y-axes, respectively.

References


Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- movieS1.mp4
- movieS2.mp4
- movieS3.mp4