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

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

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13 Nanometric topological spin textures, such as magnetic skyrmions (Sks) and antiskyrmions
14 (antiSks), have attracted much attention in condensed matter physics and spintronics. To date,
15 most studies have focused on two-dimensional Sks and antiSks in thin films with inherent or
16 synthetic antisymmetric spin exchange interaction, termed Dzyaloshinskii–Moriya interaction,
17 for their topological features and information-carrier functions. Although three-dimensional
18 (3D) spin textures are ubiquitous, previous studies have focused mostly on cylindrical Sks or
19 other non-topological magnetic vortices. Nontrivial 3D spin textures, such as antiSks composed
20 of alternating Bloch- and Néel-type spin spirals, chiral bobbars carrying emergent magnetic
21 monopoles, and deformed Sk strings, have rarely been identified, particularly on their
22 topological nature. To elucidate these textures, we have developed a 3D nanometric magnetic
23 imaging technique - tomographic Lorentz transmission electron microscopy. This approach
24 enables not only the visualisation of 3D shape of magnetic objects, but also their 3D vector field
25 mapping. The present research will lead to discoveries and advanced understanding of fertile
26 3D magnetic structures in a broad class of magnets, providing insight into 3D topological
27 magnetism.

28

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 functionalities¹⁻⁵. The topological charge N of spin textures with magnetic moment distribution $\mathbf{m}(\mathbf{r})$ is defined by¹

$$N = \int \frac{d^2r}{4\pi} \mathbf{n}_r \cdot \left(\frac{\partial \mathbf{n}_r}{\partial x} \times \frac{\partial \mathbf{n}_r}{\partial y} \right), \quad (1)$$

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

$$\varphi(\mathbf{r}) = m\phi + \gamma. \quad (2)$$

Here, ϕ is the azimuthal angle of the point $\mathbf{r}$, 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 ($r \rightarrow \infty$). The γ 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)¹, 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 π) 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 modeling¹⁻² as well as real-space imaging in several magnets³⁻⁷ with inherent broken inversion symmetry and in heterostructure films⁸. 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 materials⁹⁻¹¹. 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 bobbles¹² and spin-hedgehogs¹³ carrying emergent magnetic monopoles, and 3D chiral solitons termed hopfions¹⁴, for the interpretation of 2D micrography observations^{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^{17, 18} and Nernst¹⁹ 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 system⁸. Similar hybridised Bloch- and Néel-type spirals are also proposed in multilayered films with interfacial DMI²⁰. 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 magnet²¹ and Heusler alloy with a non-centrosymmetric lattice structure²². 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 heterostructure²³, a cobalt

77 nanowire²⁴, and a chiral-lattice magnet FeGe hosting Sk arrays below room temperature (RT)²⁵. The
78 3D vector field (**B**; hereafter, the field **B** indicates magnetic induction) mapping directly revealed
79 vortices in formers, whereas possible Néel-type spin spirals were indicated near the sample surface
80 in the latter. A few studies have also reported 3D reconstruction algorithms and their application to
81 micrometre-scale artificial vortices^{26–27}.

82 Despite the exploitation of 3D imaging for magnetic twisted structures by nano-tomography, the
83 difficulty in the 3D vector field mapping and insufficient spatial resolution of X-ray tomography, as
84 well as the complicated hardware and reconstruction process required for electron holography,
85 hinders the topologically-resolved mapping of 3D spin textures in magnets. The purpose of this study
86 is to develop an *in-situ* tomographic imaging technique and an algorithm for 3D vector field
87 reconstruction to facilitate the real-space observations of 3D topological spin textures, such as antiSk,
88 Sk strings, and the chiral bobber carrying an emergent magnetic monopole, at zero magnetic field and
89 RT. The realizations of atomic-scale spin spirals, such as magnetic monopoles and Bloch lines,
90 require the 3D imaging technique with the high spatial resolution, which is challenging for the
91 established X-ray tomographic technique. To solve the spatial resolution, we first developed an
92 automatically reconstructed 3D scalar field imaging based on Lorentz transmission electron
93 microscopy (TEM) with spatial resolution below 2 nm. In contrast with the conventional tomographic
94 TEM technique for visualizing 3D crystal structures at in-focus mode, such atomic-scale tomographic
95 Lorentz TEM for magnetic imaging should be performed at defocused mode (the magnetic induction
96 in principle is zero at the in-focus mode). However, the blurred images obtained by systematically
97 varying the sample orientation to build the 3D scalar field map are challenging for the 3D
98 reconstruction. To overcome critical challenges, such as stabilizing antiskyrmions and skyrmion
99 strings at zero field in TEM samples, image blurring, and overlapping the neighborhood spin textures
100 at the relatively high tilt angle, we optimized the proper sample geometry and the defocused
101 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 $\text{Fe}_{1.9}\text{Ni}_{0.9}\text{Pd}_{0.2}\text{P}$ (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 $[1\bar{1}0]$ -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 $[1\bar{1}0]$ -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 spirals²⁹.

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° to $+50^\circ$ at 2° 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

127 TEM images and zero-tilt induction field map for a single antiSk. The changes of projected Lorentz
 128 TEM images of the antiSk with varying the tilt angle qualitatively agree with the simulations
 129 (Extended Data Fig. 2). Then, we construct the 3D magnetic configurations from the tilt series of the
 130 2D Lorentz TEM images by using the TEM tomography package “Composer” and followed by a 3D
 131 viewer “Visualizer” (see details in Methods section). Figures 1e and 1f show top and oblique views
 132 extracted from the 3D shape of magnetic textures (see Movie S1 in the supplementary information
 133 (SI), displaying 3D images from arbitrary angles), respectively, which are represented by the
 134 difference of intensities $|I(xyz + \Delta f) - I(xyz - \Delta f)|$ ($\Delta f = 100 \text{ } \mu\text{m}$) between overfocused and
 135 underfocused Lorentz TEM corresponding to the phase shift $\varphi(xyz)$ of electron wave. The magnetic
 136 twists composed of magnetic moments to appear in green, whereas the linear spin textures (magnetic
 137 domains) show dark colour. Notably, the tomographic Lorentz TEM results (Fig. 1f) clearly reveal
 138 that the magnetic twists align almost uniformly through the entire thickness of the sample. Although
 139 the shape of spin texture does not change along the z-axis, intensity difference appears between the
 140 surfaces and centre, as shown in Figs 1j–l: the intensity is weaker around the top/bottom surface (Fig.
 141 1j, 1l), while becomes stronger at the deep in the bulk (Fig. 1k). The variation in the intensity between
 142 the surface and the bulk inside suggests a hybridised magnetic structure of Bloch-type twists in the
 143 bulk and Néel-type twists at the surfaces, which should decrease the intensity of magnetic signals due
 144 to the cancellation of the in-plane magnetic moments projected in Lorentz TEM images. Another
 145 possible reason for the reduction of magnetic signal at the sample surfaces would be the damaged
 146 layers with several-nanometres thickness lying close to each surface, which is caused by the focused
 147 ion beam during the thinning process. Such crystalline imperfections on surfaces should reduce more
 148 or less the local ordered moments and weaken related intensities.

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

150 To reconstruct 3D vector field maps of the antiSk from series of 2D field maps obtained by
 151 sequentially tilting the sample around the x- and y-axes, we prepared a needle-like sample, as shown

152 in Fig. 2a. The 2D Lorentz TEM image (**a**, underfocused) and corresponding magnetic induction field
153 map (**b**) show a mixture of antiSkSs and in-plane helices at zero field and RT. We recorded tilt series
154 of underfocused and overfocused Lorentz TEM images, with tilt angles α and β around the y- and x-
155 axes (see details in Extended Data Fig. 1), respectively, ranging from -50° to $+50^\circ$ with a 5° increment.
156 We then extracted the corresponding phase images (Extended Data Fig. 4) and magnetic induction
157 field at every tilt angle from -35° to $+35^\circ$ by performing TIE analyses; note here that the magnetic
158 contrast became extremely weak due to the loss of electron wave amplitude with increasing effective
159 sample thickness when the tilt angle was greater than 35° . The 3D phase and field maps were obtained
160 using the software “TomoPy” (see details in Extended Data Fig. 3 and the related description of the
161 3D reconstruction procedures in the Methods). Movie S2 shows the 3D phase configurations from
162 arbitrary view angles, and Extended Data Fig. 5 shows the 3D vector-field maps (**a**) and
163 corresponding (**b**) xy map at $z = 100$ nm of the surrounding area in Fig. 2a composed of two antiSkSs
164 and two elongated spin textures, respectively. The 3D phase images demonstrate that 1) the Bloch-
165 type spirals (black colours in the phase image of Movie 2) continuously extend from the top surface
166 to the bottom; 2) four Bloch lines (light grey in the phase images) between Bloch-type spirals
167 constitute antiSkSs. The 3D vector field maps reveal complicated spin textures (Extended Data Fig.
168 5a) while twisted textures in the antiSk (the surrounded area with a dashed rectangle in Extended
169 Data Fig. 5b) can be discerned. To show the vector field map of the antiSk clearer, we plot the 3D
170 field map for one antiSk (surrounded by white dashed lines in Fig. 2b) in Figs. 2c-d, representing the
171 downward moments in the core as indicated by dark arrows and the upward moments at the periphery
172 as indicated by bright arrows. The twisted moments (gradational purple and green arrows in Figs. 2
173 c-d) between the downward (the $-z$ direction) and upward (the $+z$ direction) moments, and four Bloch
174 lines (Néel-type spirals) at the four corners (Figs. 2c, e-g), are discerned. Between the neighbouring
175 Bloch lines, the gradational red arrows display the magnetic moments pointing to the right, the blue
176 arrows show the magnetic moments pointing to the left, and the green and purple arrows indicate the

177 magnetic moments pointing to the +y and the -y directions, respectively; see also the xy-plane images
 178 in Figs. 2e–g. The field distributions at the top (e, $z = 200$ nm) and bottom (g, $z = 0$ nm) surfaces are
 179 almost the same as those at the centre (g) ($z = 100$ nm), except for that at edge corners of field maps
 180 (possibly induced by the neighbouring spin textures in the sample), indicating a simple and ideal
 181 antiSk configuration, in good agreement with the theoretical predictions. Note that the difference
 182 between the vector field map and the magnetisation map of an ideal antiSk is observable by
 183 micromagnetic simulations (Extended Data Figs. 6a–b) and confirmed by Lorentz TEM observations
 184 (Extended Fig. 6c) due to the existence of stray fields around Bloch lines, where the divergence of
 185 magnetisation, and hence magnetic charge should not be zero. However, such stray fields hardly
 186 affect the fundamental 3D topological features of the antiSk identified here (Fig. 2).

187 **Identification of Sk strings and chiral bobber carrying the magnetic monopole via 3D magnetic** 188 **imaging**

189 We apply the aforementioned tomographic Lorentz TEM to examine topological defects, such as
 190 emergent magnetic monopoles involved in broken Sk strings^{9, 10, 13, 16}. One such example, the chiral
 191 bobber¹² (schematically shown in Fig.3h) carrying an emergent magnetic monopole at its end, attracts
 192 much attention as the most typical 3D topological defect, and yet has not been confirmed by 3D real-
 193 space imaging. Here, we used the aforementioned FC process to form metastable zero-field Sks in a
 194 chiral lattice ($P4_132$ -space group) magnet $\text{Co}_9\text{Zn}_9\text{Mn}_2$ ^{30–31} with a dimension of $1 \times 0.2 \times 0.25$ (μm)³. We
 195 confirmed the isolated 3D string-like structures by tomographic Lorentz TEM, as shown by an xy top
 196 view (Fig. 3a) and an xz cross-section view at $y = 100$ nm (Fig. 3 b). Skyrmionic circular domains
 197 with an average diameter of about 80 nm are initially observed in the xy-plane. The Sks numbered
 198 #1–4 are discerned as curved strings (see a schematic of the deformed Sk string in Fig. 3k) in the xz
 199 cross-section image. In particular, a short one (#2) ends near the centre of the magnet ($z = -150$ nm),
 200 exhibiting the chiral bobber-like feature (Fig. 3h). Such chiral bobber-like structures and deformed
 201 strings are also confirmed by several xy-slices at different z-values, as shown in Figs. 3c–e. The slice

202 at $z = -50$ nm close to the top surface exhibits only three circular domains corresponding to strings
203 #1, #3, and #4, whereas chiral bobber #2 is absent. Meanwhile, the slice at $z = -200$ nm close to the
204 bottom surface displays all circular domains with comparable intensities, and the slice at $z = -250$
205 nm at the bottom surface reveals the strongest intensity for chiral bobber #2 but extremely weak
206 intensities for Sk strings #1, #3, and #4. These observations directly demonstrate the 3D aspects of
207 deformed Sk strings #1, #3, and #4 and chiral bobber #2 close to the bottom surface. To confirm the
208 topological aspects of the deformed Sk string and chiral bobber, we analysed Lorentz TEM images
209 of top and oblique views of the string and bobber by the TIE. The induction field maps at zero tilt
210 (Fig. 3f, 3i) and the projected field maps at 50° tilt for the bobber (Fig. 3g) and deformed string #1
211 (Fig. 3j) show that there is no significant difference for these spin textures as viewed from the
212 perpendicular direction to the sample plate (zero tilt). However, the oblique field maps discern
213 topological distinctions between the string and the bobber (Fig. 3g, 3j): the string extends to the top
214 surface, whereas the bobber ends deep within the bulk, revealing monopole textures at the bobber's
215 end.

216 To fully identify the spin textures of the deformed string in the bulk, we reconstructed the 3D
217 vector field map of Sk string #4, which was isolated in the ferromagnetic background and did not
218 overlap with other Sk strings even at tilt angles greater than 45° . Extended Data Fig. 7 shows a series
219 of phase images of the Sk string obtained by sequentially tilting the sample around both the x- and y-
220 axes in a 5° -step increment. Note here again that a tilting angle above 50° increases the effective
221 thickness of the sample to about 300 nm, resulting in the loss of the electron wave amplitude. We
222 used this series of phase images to reconstruct the 3D vector field map for the Sk string. The field
223 maps through the entire thickness of the Sk string (from the top to bottom), as shown in Fig. 3l,
224 demonstrate that the spin swirls keep at depth in the bulk while collapsing at both the top and bottom
225 surfaces (see changes of coloured arrows through the thickness). Figure 3l also reveals that the string
226 is highly deformed deep within the bulk, while maintaining its vorticity and helicity throughout the

227 string. Our tomographic Lorentz TEM results provide the first experimental evidence of the 3D
228 topological spin textures associated with highly-deformed metastable Sk strings in a chiral-lattice
229 magnet at zero field and RT.

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

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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.

309 developed the algorithm for the 3D vector field map reconstruction and wrote the related description
310 in the Methods section. F. S. Y. contributed to the antiskyrmion simulations and wrote the
311 corresponding description in the Methods section. L. C. P. and K. N. prepared the TEM sample. K.
312 K. and Y. Taguchi synthesised the bulky single-crystal samples. All authors discussed the data and
313 commented on the manuscript.

314 **Competing interests**

315 The authors declare no competing interests.

316 **Author Information**

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318 **Data availability**

319 The data that support the findings of this study are available from the corresponding author upon
320 reasonable request.

Figure 1: Tomographic Lorentz transmission electron microscopy (TEM) images of antiskyrmions accompanied by orthogonal helices in a non-centrosymmetric magnet $\text{Fe}_{1.9}\text{Ni}_{0.9}\text{Pd}_{0.2}\text{P}$ (FNPP) with the M_3P -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 $|I(\text{xyz} + \Delta f) - I(\text{xyz} - \Delta f)|$.**

330 $|\Delta f|$ ($\Delta f = 100 \text{ } \mu\text{m}$) between overfocused and underfocused Lorentz TEM of 3D magnetic
 331 configurations composed of antiskyrmions (square-shape domains) and helices (stripes) in a (001)
 332 thin plate of FNPP. The colour bar shows the arbitrary intensity. **g**, The 2D field map of the area
 333 inside the dashed rectangle in **e**. **h-i**, Enlarged views of the field map in **g** for (**h**) orthogonal helices
 334 (corresponding to the region surrounded by dashed lines in **g**) and (**i**) antiskyrmion #2. The colour
 335 wheel and white arrows indicate the magnitude and direction of the in-plane fields. Two yellow and
 336 two black arrows in **i** indicate Bloch lines in the antiskyrmion. **j-l**, Tomographic images of (**j**) a slice
 337 around the top surface, (**k**) a middle, and (**l**) around the bottom surface of the magnified 3D magnetic
 338 configurations for the surrounding area in **e**. The sample orientations are indicated by the red
 339 ($x//[110]$ -axis)/green ($y//[1\bar{1}0]$ -axis)/blue ($z//[001]$ -axis) arrows.

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342 **Figure 2: Three-dimensional (3D) field maps of an antiskyrmion observed in a needle-like**
 343 **FNPP. a**, Under-focus Lorentz TEM image and **b**, its magnetic induction field map. The colour wheel
 344 shows the in-plane-field direction at every point. **c**, 3D vector field map of an antiskyrmion,
 345 corresponding to the area surrounded by white dashed lines in **b**. The direction of the coloured arrows
 346 is indicated by a colour ball (the right-upper corner of **c**). **d**, Field maps of the antiskyrmion in the xz
 347 plane at $y = 100$ nm. Dark arrows show magnetic fields pointing down ($-z$ direction) at the
 348 antiskyrmion core, while bright arrows indicate the fields pointing up at the peripheral of the
 349 antiskyrmion, respectively. **e–g**, Field maps of the antiskyrmion in the xy plane at (**e**) $z = 200$ nm
 350 (top-surface slice), (**f**) $z = 100$ nm (central slice), and (**g**) $z = 0$ nm (bottom-surface slice).

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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

360 dashed lines are eye guides for strings. The coloured arrows in **a-e** indicate the x (red), y (green) and
361 z (blue) axes, respectively. **f-g, i-j**, Two-dimensional projections of field maps at tilt angles of (**f, i**)
362 0° and (**g, j**) 50° for (**f-g**) the bobber #2, and (**i-j**) a skyrmion string #1. **h** and **k**, Schematics of (**h**)
363 the chiral bobber carrying a magnetic monopole (a blue dot at the end), (**k**) a deformed skyrmion
364 string, respectively. **l**, Several xy-slices of a 3D vector field map of deformed skyrmion string #4 from
365 top to bottom. The coloured arrows encoded by the colour ball show the magnetic field swirling while
366 dark and white arrows indicate the down and up field directions, respectively.

367 **Methods**

368 **1. Sample preparation**

369 Single crystals of $\text{Fe}_{1.9}\text{Ni}_{0.9}\text{Pd}_{0.2}\text{P}$ (FNPP) and $\text{Co}_9\text{Zn}_9\text{Mn}_2$ were grown by a self-flux method⁷
370 and the Bridgman method followed by water quenching^{30, 31}, respectively. The crystal orientations
371 were checked by X-ray Laue diffraction as well as electron diffraction. To perform tomographic
372 Lorentz TEM, the micron-sized samples were sliced along the (001) plane from bulk crystals and
373 shaped using a focused ion beam system (Hitachi NB5000).

374 **2. Generation of zero-field antiskyrmions and skyrmions**

375 To excite zero-field antiskyrmions in FNPP micron-size magnets, as shown in Figs. 1 and 2, we
376 performed *in-situ* field cooling (FC) the sample in a transmission electron microscope (JEM2800,
377 JEOL). Firstly, we attached a micron-size sample to a heating holder (HC3500, Gatan), which was
378 placed on the sample stage above an objective lens coil. Next, a current of approximately 1 A was
379 passed through the objective lens coil to generate a field of 50 mT antiparallel to the incident electron
380 beam and normal to the (001) plane of the FNPP sample. Then, we cooled the FNPP sample from
381 400 K to a room temperature in the 50 mT field and then turned off the objective lens current to obtain
382 the zero-field magnetic configuration. Finally, we moved the FNPP sample to a high-tilt sample
383 holder (JEOL), allowing the TEM sample to be tilted by $\pm 80^\circ$.

384 Metastable skyrmion strings involving a chiral bobber were generated in a micron-size magnet of
385 $\text{Co}_9\text{Zn}_9\text{Mn}_2$ using the aforementioned FC procedure.

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

387 Extended Data Figs. 1a, c–j display a series of projected 2D Lorentz TEM images in the under-
388 focused mode by sequentially varying the tilt angles β around the x-axis (c–f) and α around the y-axis
389 (g–j), while Extended Data Fig. 1b presents the 2D magnetic induction field map at zero tilt angle
390 obtained by TIE analyses²⁸. Extended Data Figs. 2a, c–j present the simulated Lorentz TEM images

391 at the same tilt angles as those in Extended Data Fig. 1a, c–j, while Extended Data Fig. 2b shows the
 392 simulated magnetic field map at zero tilt angle. The colour wheel with white arrows encodes the
 393 direction of the in-plane field at every point in the antiskyrmion. Red, blue-green, yellow-green, and
 394 blue-magenta represent magnetic fields pointing to the right, to the left, up, and down, respectively;
 395 the relatively dark colour indicates the dominance of out-of-plane fields, which are prominent on the
 396 core and periphery. The simulation results qualitatively agree well with the experimental observations.

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 398 **Extended Data Figure 1: Series of 2D projected Lorentz TEM images at tilt angles of (α, β) and**
 399 **an in-plane magnetic induction field (B) map (colour image) at zero tilt of an antiskyrmion in**
 400 **the $\text{Fe}_{1.9}\text{Ni}_{0.9}\text{Pd}_{0.2}\text{P}$ (FNPP) thin plate (thickness $t \approx 200$ nm).** The tilt angles (α, β) and a colour
 401 wheel for encoding the in-plane field direction are indicated in insets of the figure, respectively. The
 402 yellow arrow shows the incident electron beam antiparallel to the z-axis. The coordinate systems (x,
 403 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^{32, 33} with added Poisson noise. The images in Extended Data Figs. 2 **a**, **c-j** were simulated at a defocus distance Δf of $-50 \mu\text{m}$. 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 $128 \times 128 \times 40 \text{ pixels}^3$ corresponding to a sample size of $640 \times 640 \times 200 \text{ nm}^3$.

Extended Data Figure 2: Series of simulated Lorentz TEM images at tilt angles of (α, β) and an in-plane magnetic induction field (B) map (colour image) at zero tilt for an antiskyrmion. The tilt angles (α, β) and a colour wheel for encoding the in-plane field direction are indicated in insets of the figure, respectively.

416 **5. Tomographic Lorentz TEM imaging and reconstruction of 3D vector field maps for** 417 **antiskyrmions and skyrmion strings**

418 **5-1 Tomographic Lorentz TEM**

419 Tomographic Lorentz TEM was performed in a low magnification mode of a commercial
420 transmission electron microscope (JEM2800, JEOL), where the objective lens current was turned off
421 to realize the zero external magnetic field state in the micron-size sample.

422 Firstly, we used software “Recorder” (tomography.com) to acquire a series of projected 2D Lorentz
423 TEM images automatically by varying the tilt angle α around the y-axis from -50° to $+50^\circ$ in a 2°
424 increment, as illustrated in Extended Data Fig. 3a. A fast-speed camera (Oneview, Gatan) with an
425 exposure time of 20 ms was used to collect data. Drift of images with varying tilt angles was corrected
426 by referring to markers in the sample, and the focus correction was checked out during sequential
427 Lorentz TEM observation. The aforementioned procedure was carried out by varying the tilt angle β
428 around the x-axis. Then, the obtained images were stacked by a software package “Composer,” which
429 enables the reconstruction of 3D Lorentz TEM images from previous 2D Lorentz TEM images
430 obtained. The procedure for 3D reconstruction is as follows:

- 431 1) Align the images for the dual-axis tilt series by taking the correct correlation using the markers,
432 which enables the reduction of artifacts, such as diffraction contrasts, during tilt processes;
- 433 2) Reconstruct the 3D image by using the simultaneous iterative reconstruction technique;
- 434 3) Check the accuracy of the cross-correlations of the stacked images by using the line alignment
435 cross-correlation.

436 Finally, the 3D viewer “Visualizer” (tomography.com) was employed to determine the volume
437 function of the 3D magnetic configuration according to the real sample geometry. The colour and
438 transparency maps as well as the multislice and movie of the 3D object were obtained, as shown in
439 the main text and Supplementary movies at arbitrary angles and positions.

440 **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)^{28, 34-35} described as

$$\frac{2\pi}{\lambda} \frac{\partial I(xyz)}{\partial z} = -\nabla_{xy} [I(xyz) \nabla_{xy} \varphi(xyz)] . \quad (3)$$

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

$$\nabla_{xy} \varphi(xyz) = -\frac{e}{\hbar} (\vec{B} \times \vec{n}) t , \quad (4)$$

we can obtain the magnetic inductions B_x and B_y accumulated through the sample thickness from the phase φ . 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 technique³⁶⁻³⁸. The third magnetic field component, B_z , was calculated from the zero-divergence condition $\partial B_x / \partial x + \partial B_y / \partial y + \partial B_z / \partial z = 0$. 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° . 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 axes³⁶. 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}.

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475 configurations of a single skyrmion string (a) and the flowchart of an algorithm for 3D vector
476 field reconstruction (b).

477 5-3 Acquisition of phase image series by sequentially tilting the needle-like FNPP sample

478 Extended Data Fig. 4 shows a tilt series of 2D phase images of needle-like FNPP that were used to
 479 reconstruct the 3D vector field map. At zero tilt, a mixture of antiskyrmions and elongated spin
 480 textures parallel to the sample edges in the FNPP can be discerned. When the sample is tilted around
 481 the x-axis, both the projected 2D antiskyrmions and elongated spin textures extend or shorten along
 482 the y-axis, whereas they expand or shorten along the x-axis as the sample is tilted around the y-axis.
 483 Some projected phase images (see the images at tilt angles of -14° and -18° around the x-axis and
 484 of 28° and -22° around the y-axis) show extremely weak signals due to the local strain, diffraction
 485 effects or interference between the incident electrons and crystal lattice during sample holder tilting.
 486 We reconstructed the 3D phases using the data presented in Extended Data Fig. 4 for two
 487 antiskyrmions and two partial extended spin textures (enclosed by a yellow rectangular in the phase
 488 image at zero tilt), as shown in Supplementary Movie 2 for the view from arbitrary angles.

489
 490
 491 spin textures obtained at various tilt angles about the x- and y-axes. The boxed area in the left

492 image was selected to reconstruct 3D vector field maps of the spin textures, including two
 493 numbered antiskyrmions.

494 5-4 The 3D vector field maps for antiskyrmions in the FNPP

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496 **Extended Data Fig. 5: 3D vector field maps of magnetic configurations squared in the Extended**

497 **Data Fig. 3 observed in the FNPP. a, 3D field map. b Field map within the xy-plane at $z = 100$ nm.**

498 Dashed lines in **a** and **b** indicate the antiskyrmion shown in Fig. 2.

499 5-5 The spatial distribution of the magnetisation and field maps for the antiskyrmion at zero 500 tilt

501 Extended Data Figs. 6**a-b** show the simulated magnetisation (**a**) and field (**b**) distributions in an
 502 antiskyrmion at zero tilt, revealing stray fields around Bloch lines. Such stray field arising from the
 503 nonzero divergence of magnetisation has been discerned in the present experimental observation of
 504 the magnetic induction field map (**c**) for the antiskyrmion at $z = 0$.

505

506 **Extended Data Fig. 6: Simulated magnetisation (a) and field (b) maps, and experimentally**
 507 **obtained field (c) maps of the antiskyrmion at zero tilt.** The white arrows and colours in **a** indicate
 508 the magnetisation directions, while those in **b-c** indicate the induction-field directions. Dark/bright
 509 colour in **a** shows up/down magnetisation, while dark colour in **b-c** indicates out-of-plane magnetic
 510 induction fields.

511 **5-6 Acquisition of phase image series by sequentially tilting a deformed skyrmion string**

512 The same 3D reconstruction procedure as described in 5.3 section was employed for a single
 513 skyrmion string by using a series of 2D phase images acquired by sequentially tilting the micron-size
 514 magnet $\text{Co}_9\text{Zn}_9\text{Mn}_2$, as shown in Extended Data Fig. 7. The series of phase images show the extension
 515 of the skyrmion string in the x- and y-directions for both the tilt axes due to the deformation of the
 516 skyrmion string (see the tomographic Lorentz TEM image in Fig. 3b). The reconstructed 3D vector
 517 field maps (Fig. 3I) confirm the deformation of the skyrmion string, showing good agreement with
 518 our tomographic Lorentz TEM results (Figs. 3a–e and Supplementary Movie 3).

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

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