Room-temperature antiskyrmions and sawtooth surface textures in a non-centrosymmetric magnet with $S_4$ symmetry

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Topological spin textures have attracted much attention both for fundamental physics and spintronics applications. Among them, antiskyrmions possess a unique spin configuration with Bloch-type and Néel-type domain walls owing to anisotropic Dzyaloshinskii–Moriya interaction in the non-centrosymmetric crystal structure. However, antiskyrmions have thus far only been observed in a few Heusler compounds with $D_{5d}$ symmetry. Here we report a new material, Fe$_{1.9}$Ni$_{0.9}$Pd$_{0.2}$P, in a different symmetry class ($S_4$), in which antiskyrmions exist over a wide temperature range that includes room temperature, and transform into skyrmions on changing magnetic field and lamella thickness. The periodicity of magnetic textures greatly depends on the crystal thickness, and domains with anisotropic sawtooth fractals were observed at the surface of thick crystals and attributed to the interplay between the dipolar interaction and the Dzyaloshinskii–Moriya interaction as governed by crystal symmetry. Our findings provide an arena in which to study antiskyrmions, and should stimulate further research on topological spin textures and their applications.

Magnetic skyrmions and antiskyrmions with vortex-like topological spin textures have recently attracted increasing attention as a source of various emergent phenomena and their potential applications to spintronics devices, such as racetrack memory. One of the microscopic origins for these topological textures is the competition between the ferromagnetic exchange interaction and the Dzyaloshinskii–Moriya interaction (DMI), the latter of which is derived from relativistic spin–orbit interaction in the absence of spatial inversion symmetry. Theoretical proposals have been made for several types of DMI-based topological spin textures. Skyrmions of both Bloch type (Fig. 1a,b) and Néel type (Fig. 1c,d) are characterized by an integer topological number. A Bloch-type skyrmion is constructed by helical spin propagations (Bloch walls) with either of clockwise (CW) (Fig. 1a) or anticlockwise (ACW) (Fig. 1b) rotation, and is observed in cubic chiral magnets that belong to the $T$ and $O$ symmetry classes. A Néel-type skyrmion is produced by cycloidal spin propagations (Néel walls), also with either a CW (Fig. 1c) or ACW (Fig. 1d) orientation, being observed in heterostructures with interfacial DMI and bulk polar magnets with $C_n$ symmetry.

An antiskyrmion, however, is characterized by a topological number with the opposite sign and composed of spin spirals, which cover all helicities (Fig. 1e,f). This unique spin configuration is possible only when the DMI along two orthogonal axes have opposite signs, and thus antiskyrmions are theoretically predicted to form in $D_{5h}$ and $S_4$ symmetry classes with a fourfold rotoinversion axis. Experimentally, antiskyrmions have been observed in inverse Heusler compounds with $D_{5h}$ symmetry (space group $I4_22m$), such as Mn-deficient Mn$_2$P$_{1-x}$Pd$_x$Sn (and Mn$_2$PtSn) and stoichiometric Mn$_2$Rh$_{0.98}$Ir$_{0.02}$Sn. In contrast to extensive studies of skyrmions in various magnets, previous experimental studies for antiskyrmions were confined to these $D_{5h}$ Heusler alloys, and no other materials have been found. Here we report our discovery of a new material, Fe$_{1.9}$Ni$_{0.9}$Pd$_{0.2}$P, with $S_4$ symmetry, in which antiskyrmions and skyrmions are observed above room temperature by Lorentz transmission electron microscopy (LETEM). In conjunction with magnetic force microscopy (MFM), we also describe the evolution of bulk and surface magnetic textures, which reflect the anisotropic DMI and underlying $S_4$-type lattice symmetry as the crystal thickness is increased.

Basic magnetic properties of Fe$_{1.9}$Ni$_{0.9}$Pd$_{0.2}$P

Our target compound M,P (M, transition metal) crystallizes in a non-centrosymmetric tetragonal structure with the space group $I4$ in the $S_4$ symmetry class (Fig. 1g). The structural symmetry is characterized by only 4 around the [001] axis, and thus lower than $D_{5h}$. Among M,P compounds, Fe,P is a ferromagnet with a high Curie temperature, $T_C \approx 700$ K and saturation moment $M_s \approx 1.84 \mu_B$ / Fe, whereas Ni,P is a Pauli paramagnet without showing magnetic orders. In their solid solutions (Fe,Ni), P (schreibersite), $T_C$ and $M_s$ linearly decrease as the Ni concentration is increased.

We obtained high-quality bulk single crystals of Fe$_{1.9}$Ni$_{0.9}$Pd$_{0.2}$P (see Supplementary Note 1, Supplementary Tables 1 and 2, and Supplementary Fig. 1 for sample characterization). The Pd doping to the schreibersite was anticipated to enhance the spin–orbit coupling and hence DMI, and it also turned out to change the magnetic anisotropy from an in-plane to a uniaxial one (see Supplementary Note 2 and Supplementary Fig. 2 for magnetization data in Fe$_{1.9}$Ni$_{1.4}$P). The temperature dependence of the magnetization at 0.01 T (Fig. 1h) showed a ferromagnetic transition at $T_C \approx 400$ K. As presented in Fig. 1i, the magnetization at 2 K reached a saturation value of $M_s \approx 3.5 \mu_B$ per formula unit (f.u.) at 0.35 T for [001] but at 0.64 T...
for [110], which clearly indicates easy-axis anisotropy. There was no hysteresis for both directions. We derived the uniaxial anisotropy constant \( K_u \approx 106 \text{kJ m}^{-3} \) from the area enclosed between the [001] and [110] magnetization curves for \( H > 0 \). The quality factor \( Q \), defined as the ratio between uniaxial anisotropy and magnetostatic energies \( K_u/\mu_0 M_s^2/2 \) (where \( \mu_0 \) is the vacuum permeability), was found to be \( Q \approx 0.4 \) at 2 K (0.28 at 300 K). Therefore, Fe\(_{1.9}\)Ni\(_{0.9}\)Pd\(_{0.2}\)P is a soft magnet with a relatively strong dipolar interaction and weak uniaxial anisotropy, both of which are important for the formation of antiskyrmions\(^{16,20}\), similarly to the Heusler alloys\(^{5,21}\).

**LTEM observation of antiskyrmions and skyrmions**

For the real-space observation of magnetic structures, we performed LTEM measurements on (001) thin plates. Figure 2 summarizes the LTEM results at 295 K for a fixed sample thickness \( t \approx 130 \text{nm} \). The magnetic structures strongly depend on measurement protocols, especially the tilting of the sample plate under magnetic fields (see details in Supplementary Note 3 and Supplementary Fig. 3). At zero field, a helical stripe pattern with a periodicity of \( \lambda \approx 280 \text{nm} \) was observed (Fig. 2b). Here helical propagation vectors (\( q \)) are pinned almost along the [110] and [110] axes, and the helicity of the Bloch-type domain wall is reversed between them. This result evidences the presence of anisotropic DMI, as expected for \( S_z \) crystal symmetry. As the [110] and [110] axes were not experimentally distinguished, in this article we used [110] for the CW rotation axis and [110] for the ACW one.

Under magnetic fields perpendicular to the plate, antiskyrmions sparsely formed (Supplementary Fig. 3a). When the sample plate was tilted by 12° and then a magnetic field applied, a large number of magnetic bubbles with zero topological number, here termed non-topological (NT) bubbles as in Peng et al.\(^{22}\), were created above 350 mT (Supplementary Fig. 3b). Next, as the plate was tilted back towards the initial orientation at 350 mT, the NT bubbles deformed into bullet-like shapes (Fig. 2c) and eventually converted to antiskyrmions. As shown in Fig. 2d, the antiskyrmions were square-shaped and the longer Bloch-wall parts along the [110] and [100] directions, respectively, whereas there is random site disorder among the three transition metals at the M sites. However, on average, the overall crystal symmetry in this mixed structure is preserved and the magnetic properties of this itinerant material is minimal. h. Temperature (\( T \)) dependence of magnetization (\( M \)) under the magnetic field \( H=0.01 \text{T} \) in a bulk single crystal of Fe\(_{1.9}\)Ni\(_{0.9}\)Pd\(_{0.2}\)P. Magnetizations for \( M \parallel [001] \) and \( M \parallel [110] \) are presented with blue and red lines, respectively. The data taken during field cooling (FC) and field warming after a zero-field cooling (ZFC) are denoted with solid and dotted lines, respectively. i. Magnetic field dependence of magnetization in a bulk single crystal of Fe\(_{1.9}\)Ni\(_{0.9}\)Pd\(_{0.2}\)P at 2 K for \( H \parallel [001] \) (blue line) and \( H \parallel [110] \) (red line).

**Fig. 1** | Antiskyrmions and basic properties of Fe\(_{1.9}\)Ni\(_{0.9}\)Pd\(_{0.2}\)P. a–f. Schematics of Bloch-type skyrmions (a,b), Néel-type skyrmions (c,d) and antiskyrmions (e,f). Black arrows denote the directions of the in-plane magnetic moments. Along the red and blue lines, the magnetic structures are CW and ACW Bloch walls, respectively, whereas along the green and yellow lines, they are CW and ACW Néel walls, respectively. For antiskyrmions, the four different walls alternately appear every 45° along the azimuthal angle direction in the order CW Bloch (red) → ACW Bloch (blue) → CW Néel (yellow), ACW Néel (green). Schematic of non-centrosymmetric tetragonal crystal structure of M\(_3\)P with the space group of I4 (\( S_z \) symmetry) as viewed along the [100], [010] and [001] axes from the top to the bottom, respectively. There are three inequivalent crystallographic M sites denoted as M1 (red), M2 (green) and M3 (blue). As discerned in these schematics, a twofold rotation symmetry around the [100] and [010] axes and a mirror symmetry with respect to [001] and [110] planes are absent in the \( S_z \) symmetry, which results in a symmetry lower than \( T_d \). According to the sample characterization, as detailed in Supplementary Note 1, Supplementary Tables 1 and 2, and Supplementary Fig. 1, our target compound Fe\(_{1.9}\)Ni\(_{0.9}\)Pd\(_{0.2}\)P crystallizes in the M\(_3\)P structure, whereas there is random site disorder among the three transition metals at the M sites. However, on average, the overall crystal symmetry in this mixed structure is preserved and the magnetic properties of this itinerant material is minimal. h. Temperature (\( T \)) dependence of magnetization (\( M \)) under the magnetic field \( H=0.01 \text{T} \) in a bulk single crystal of Fe\(_{1.9}\)Ni\(_{0.9}\)Pd\(_{0.2}\)P. Magnetizations for \( M \parallel [001] \) and \( M \parallel [110] \) are presented with blue and red lines, respectively. The data taken during field cooling (FC) and field warming after a zero-field cooling (ZFC) are denoted with solid and dotted lines, respectively. i. Magnetic field dependence of magnetization in a bulk single crystal of Fe\(_{1.9}\)Ni\(_{0.9}\)Pd\(_{0.2}\)P at 2 K for \( H \parallel [001] \) (blue line) and \( H \parallel [110] \) (red line).
the creation and annihilation of a Bloch line pair. These LTEM measurements were performed at different temperatures, and similar field-induced transformations from antiskyrmions to skyrmions were observed at almost all the measured temperatures below \( T \) down to 100 K, as summarized in Fig. 2g.

Similar square antiskyrmions and elliptical Bloch-type skyrmions with different helicities have been reported previously in Mn\(_{1.4}\)Pt\(_{0.9}\)Pd\(_{0.1}\)Sn (refs. 22–23) and explained in terms of the competition between anisotropic DMI and dipolar interaction. In general, to minimize the magnetic volume charges, dipolar interactions prefer...
Although the shape of (anti)skyrmions is similar to those in the case of Mn$_{1.4}$Pt$_{0.9}$Pd$_{0.1}$Sn, in which the two phases are separated by the magnetic field over the whole temperature region below $T_c$, and the skyrmion phases in the present compound is controllable at various thicknesses, which ranged from 50 to 210 nm. The LTEM images at 295 K and 400 mT after a similar tilting procedure are shown in Fig. 3a–c for lamella thicknesses of 50 nm (a), 100 nm (b) and 210 nm (c). The scale in (c) also applies to the other panels.

As the magnetic textures are strongly influenced by dipolar interactions, we carried out LTEM measurements for a lamella with various thicknesses, which ranged from 50 to 210 nm. The LTEM images at 295 K and 400 mT after a similar tilting procedure are presented in Fig. 3a–c for $t \approx 50$, 100 and 210 nm, respectively. At the thinnest area with $t \approx 50$ nm, elliptical skyrmions were observed (Fig. 3a). However, square antiskyrmions were discerned at thick areas above $t \approx 130$ nm up to 210 nm (Fig. 3c). At an intermediate thickness of $t \approx 100$ nm, elliptical skyrmions and square antiskyrmions coexisted (Fig. 3b). Clearly, the size of the (anti)skyrmions became larger as the lamella thickness increased, similar to the increase in helical periodicity at zero field (Supplementary Fig. 5). The thickness-dependent transformation between the two topological textures is also understood in terms of the competition between dipolar interaction and DMI. In a sufficiently thin plate with a large surface-to-volume ratio, to reduce the magnetostatic energy, Bloch-type skyrmions without a Néel-like configuration are favourised. However, in a thick sample with a large volume, dipolar interactions are less important than DMI, and thus antiskyrmions are preferred.

**MFM observation of sawtooth domain patterns**

Next, we performed MFM measurements to observe the surface magnetic textures, such as helical stripes and antiskyrmions, for thicker samples that ranged from $t \approx 0.52$ up to 240 μm. Figure 4b–f shows MFM images for different thicknesses, taken at 295 K and 0 mT after the application of magnetic fields with tilting of the samples, as illustrated in Fig. 4a. For $t \approx 0.52$ μm (Fig. 4b), square objects were clearly observed in addition to stripe domains. Although MFM detects the out-of-plane component of the stray magnetic field from the domains on the surface, the MFM images bear a strong resemblance to those of the LTEM images in zero field (Supplementary Fig. 3d), and hence we can safely conclude that the square objects correspond to antiskyrmions that persist at zero field. As the thickness was increased, the stripe periodicity and the antiskyrmion size further increased, and their domain structures became more complex. The domain boundaries exhibited clear undulations for $t \approx 2.2$ μm (Fig. 4c), which turned into prominent sawtooth waves for $t \approx 5.3$ μm (Fig. 4d), at which core structures with reversed magnetization were also observed, as indicated with a dotted circle. These complex domain textures are further branched and characterized as fractals, that is, self-similar objects in multiple length scales, for $t \approx 50$ μm (Fig. 4e,f). Note that the square objects enclosed by the vertical and horizontal sawtooth stripes with $q \| [110]$ and $q \| [110]$ are characterized by 4 symmetry, as schematically illustrated in Fig. 4g, which is identical to the symmetry of the underlying crystal lattice.

Here we discuss the observed complex domain pattern. In general, magnetic domains are formed so that the magnetostatic energy due to magnetic surface charges is reduced at the cost of increased domain wall energy. It has been known since the 1960s that, as the crystal thickness increases, magnetic domains at the surface become more complex, and show corrugations and even fractalizations to further screen the surface magnetic charges on larger scales. Such wavy or branched flower-like domain textures have been observed in a broad range of uniaxial ferromagnets, such as Co (refs. 26–28), Nd–Fe–B alloys (refs. 26,30,31), hexagonal ferrites (24,32) and garnet ferrites33. In spite of such a long history of domain wall physics over 60 years, there are no reports of the anisotropic sawtooth pattern governed by 4 symmetry observed in this antiskyrmion material, which is thus attributed to the anisotropic DMI in the $S_4$ crystal lattice.

To understand the role of the DMI in the formation of the sawtooth pattern, we performed micromagnetic simulations for a three-dimensional system using the program MuMax	extsuperscript{33} (see Methods and Supplementary Note 9 for the details). In the simulation, we incorporated exchange stiffness, $K_e$, and demagnetization energy using the experimentally evaluated values at 300 K, and introduced the anisotropic DMI as a small perturbation. We found that a slightly asymmetric wave appears near the surface without DMI, but the DMI clearly enhances the asymmetric feature of the wave and leads to the sawtooth pattern, as presented in Extended Data Fig. 1, which is similar to the MFM result for the crystal with $t \approx 5.3$ μm. The sawtooth pattern can be well understood in terms of the helicity of the domain walls uniquely determined by the anisotropic DMI in the bulk, and the additional modification caused by the competition between the domain wall energy and demagnetization energy.
Fig. 4 | Thickness evolution of topological spin textures. 

A schematic of the sample-tilting protocol under magnetic fields prior to the MFM measurements. A magnetic field of 400 mT was applied parallel to the [001] direction, followed by tilting the sample plate to α = 30° and then back to 0°. This procedure was repeated at different fields down to 200 mT (by 10 or 20 mT steps) and then the magnetic field was removed. MFM images (b–f) were taken at 295 K (∼3 K) and 0 mT after this protocol. b–f, MFM images for thicknesses of 0.52 μm (b), 2.2 μm (c), 5.3 μm (d) and 50 μm (e and f). Bright yellow and dark purple colours in the MFM images correspond to the magnetizations that point out of the plane (parallel to [001]) and into the plane (antiparallel to [001]), respectively. a.u., arbitrary units. The dotted circle in d indicates a domain with an inner core structure of reversed magnetization. f is an enlarged view for the area indicated by a dotted square in e. g, Schematic of the sawtooth waves with 4 symmetry. The dotted arrows indicate the direction of each sawtooth wave. All the MFM images at the initial states, which include those for other thickness samples, are presented in Supplementary Fig. 6. h, The t dependence of the magnetic stripe periodicity (λ) of Fe1.9Ni0.9Pd0.2P at 295 K and 0 mT. Data points determined by LTEM and MFM measurements are denoted with orange circles and green squares, respectively. Error bars correspond to the difference between the maximum and minimum values observed within the same thickness region. The data points are fitted to a power law, λ ∝ t^{0.65} for t ≤ 10 μm (purple line) and λ ∝ t^{2.3} for t ≥ 10 μm (pink line). The blue and red shades in the background correspond to the thickness regions in which Bloch-type skyrmions and antiskyrmions, respectively, were observed (Fig. 3). The yellow shade represents the thickness region at which sawtooth patterns were found at the surface.

Thick near the surface (see Supplementary Note 10 and Supplementary Fig. 7 for a detailed discussion). The micromagnetic simulation indicates that the sawtooth pattern reverses for the top surface and the bottom surface when viewed from the same direction, which is attributed to the opposite direction of the stray fields for the two surfaces. This was confirmed by additional MFM measurements on the opposite side of the crystal (Supplementary Fig. 7).

Thickness dependence of magnetic periodicity

In Fig. 4h, the magnetic periodicity λ determined from the LTEM and MFM measurements is plotted against the thickness t in a wide range from 50 nm to 240 μm. λ shows a dramatic increase from the order of 100 nm up to the order of 10 μm as t is increased. Such substantial dependence of λ on t up to the micrometre-scale is a typical consequence of dipolar interaction, as commonly observed in the conventional uniaxial ferromagnets(26–30) or in the antiskyrmion–host Heusler compound, as recently reported(31). The observed λ(t) can be fitted to a power law as λ ∝ t^{0.66} below t = 10 μm, and λ ∝ t^{2.3} above t = 10 μm. The obtained power-law exponents are in accordance with those in theoretical models for uniaxial ferromagnets. As described in Supplementary Note 8 in more detail, the well-known Kittel’s model for sufficiently thin films gives λ ∝ t^{1/2} (ref. 26), and other theories applicable to thick crystals with wavy or spiky domain structures near the surface predict λ ∝ t^{3/2} (ref. 27) or λ ∝ t^{2/3} (refs. 26,28). Although the DMI is not considered in these theories, the overall feature of the thickness evolution as governed by the dipolar interaction can be captured. On the basis of these theoretical models, we calculated the critical thickness t∗ = 0.4 μm at 300 K, above which undulations of the surface domain walls take place, in good agreement with the MFM data. This value is very small as compared with those previously reported for uniaxial ferromagnets (typically t∗ = 10 μm (ref. 30)) due to the relatively weak uniaxial anisotropy (Q ∼ 1) in the present material. In a recent MFM study for Mn1.4PtSn (ref. 20), surface patterns of magnetic stripes and antiskyrmions remained simple and clear undulations of domain walls were absent up to t = 4 μm, probably because the Q value is relatively large in the Heusler compounds. Associated with the weak magnetic anisotropy, another vortex-like structure within (anti)skyrmions was observed by LTEM (Figs. 2 and 3), which is also absent in the Heusler alloys. The magnetic softness of the present material as compared with that of the Heusler compounds is another interesting point in these two different antiskyrmion systems.

Summary and outlook

The present work demonstrates that the Pd-doped schreibersite, Fe1.9Ni0.9Pd0.2P, is a new class of room-temperature antiskyrmion material with S_{1} symmetry, and that antiskyrmions and skyrmions can be easily interconverted by changing magnetic field and crystal
thickness over a wide temperature region. The interplay between anisotropic DMI and long-range dipolar interactions provides a rich variety of magnetic textures with variable size, shape and topology, including surface sawtooth fractals as dominated by the crystal symmetry. These new findings accelerate the studies of antiskyrmions and related topological spin textures, and open an entirely new chapter for domain wall physics, where the DMI-induced asymmetric features have been overlooked for over 60 years. Furthermore, these topological and anisotropic spin textures with versatile tunability at room temperature are promising for spintronics applications.

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**Methods**

**Sample preparation.** Single-crystalline bulk samples of Fe$_{1.9}$Ni$_{0.9}$Pd$_{0.2}$P and Fe$_{1.6}$Ni$_{1.4}$P were synthesized by a self-flux method from pure Fe, Ni and Pd metals and red phosphorous sealed in an evacuated quartz tube. The target phase of tetragonal M$_2$P was isolated from the ingot. Phase purity of the M$_2$P structure was confirmed by powder X-ray diffraction with Cu Kα radiation, as detailed in Supplementary Fig. 1. Crystal orientations were checked by an X-ray Laue diffraction method. Chemical compositions were examined by a scanning electron microscope equipped with an energy dispersive X-ray analyser (Supplementary Tables 1 and 2).

**Magnetization measurement.** For the magnetization measurement, a single-crystalline bulk piece was cut to a rectangular shape (approximately 0.3 mm x 0.5 mm x 1.0 mm) with almost the same surface areas of the (001) and (110) planes. Magnetization measurements were performed by a superconducting quantum interference device magnetometer (MPMS3, Quantum Design) equipped with an oven option.

**LTEM measurement.** For the LTEM measurements, (001) thin plates of various thickness ($t$=30, 70, 100, 130, 160 and 210 nm) were thinned from the bulk crystals by a focused ion beam (FIB) system (NBS5000, Hitachi). LTEM measurements were performed with a transmission electron microscope (JEM-2100F, JEOL) equipped with a double-tilt liquid-nitrogen holder. An in-plane component of the field was applied by tilting the plate, as illustrated in Fig. 2a. LTEM defocused images with bright and dark contrasts reflected the in-plane component of the magnetic induction fields averaged over the plate thickness. The lateral magnetic induction field distribution was obtained by a TIE analysis of the over-focus and under-focus LTEM images and displayed by colour coding.

**DPC-STEM measurement.** For additional information to support the TIE data, we prepared the DPC imaging in STEM (JEM-2100F, JEOL) equipped with a double-tilt liquid-nitrogen holder (Gatan 636) and a double-tilt heating holder (Fusion select, Protochips). External magnetic fields were applied perpendicular to the plates and parallel to the incident electron beam by tuning the objective lens current. An in-plane component of the field was applied by tilting the plate, as illustrated in Fig. 2a. LTEM defocused images with bright and dark contrasts reflected the in-plane component of the magnetic induction fields averaged over the plate thickness. The lateral magnetic induction field distribution was obtained by a TIE analysis of the over-focus and under-focus LTEM images and displayed by colour coding.

**MFM measurement.** For the MFM measurements, a staircase-shaped plate with (001) surfaces and various thickness ($t$=520 nm, 2.2 µm, 5.3 µm and 10.3 µm) was prepared from a bulk piece by FIB. In addition, bulk crystals with a thickness of $t$=50, 140 and 240 µm were prepared, and their (001) surfaces were treated by chemical mechanical polishing with colloidal silica. For the purpose of the MFM observations of metastable antiskyrmions at zero magnetic field, a magnetic field was applied perpendicular to the (001) plane in the Physical Properties Measurement System (Quantum Design), and an in-plane component of the field was applied by tilting the plate as depicted in Fig. 4a. MFM measurements were performed at room temperature (295 ± 3 K) and zero field with a commercial scanning probe microscope (MFP-3D, Asylum Research). We used a MFM cantilever (MFM-300, cantilever) with a resonance frequency of ~72 kHz and Co coating on the whole tip to keep the magnetization of the tip along the direction perpendicular to the sample surface. We used the two-pass technique, in which we mapped a surface topography by tapping the sample in the first pass, and then mapped an MFM image while lifting the cantilever from the sample (distance 30–50 nm) in the second pass. For the analysis, the flattening procedure was used. The colour of the MFM images shows the phase shift of the oscillating cantilever, which typically ranges from +2° (bright yellow) to −2° (dark purple) in this work, whose sign and amplitude correspond to those of the second derivative of the stray magnetic field mainly produced by the magnetization perpendicular to the sample plane.

**Micromagnetic simulation.** For the micromagnetic simulation of the sawtooth domain pattern, we used the well-established GPU-accelerated program MuMax3 and personalized the code to incorporate the asymmetric DMI and an improved numerical accuracy. The energy functional is given by:

$$E [m] = \int_V \left( A (\nabla m)^2 + D (m \cdot (x \times \partial_x m) - m \cdot (y \times \partial_y m)) \right) - K_u (m \cdot \nabla m)^2 - \frac{1}{2} m \cdot B_s dV$$

where $m$ is the normalized magnetization and $V$ is the volume of the system. $A$ and $D$ correspond to the exchange stiffness and the DMI constant, respectively. $B_s$ represents the demagnetizing field for a large-scale structure.

For the simulation, we used the experimentally evaluated parameters at 300 K as $A = 8.1 \text{ pJ m}^{-1}$, $K_u = 31 \text{ kJ m}^{-2}$ and $M_s = 417 \text{kA m}^{-1}$, and treat the DMI as a small perturbation with various values of $D$ (0, 0.05, 0.1 and 0.2 mJ m$^{-2}$) (see Supplementary Note 9 for more details). The simulated sample size of 1.6 µm x 0.8 µm x 5.3 µm with periodic boundary conditions in the x−y plane was chosen according to the experimental observations and was discretized to 512 x 256 x 256 cuboids. The simulated sawtooth pattern for $D = 0.2 \text{ mJ m}^{-2}$ is presented in Extended Data Fig. 1.

**Data availability**

All the data presented in the article and Supplementary Information are available from the corresponding authors upon reasonable request.

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**Author contributions**

K.K., X.T., Y. Tokura and Y. Taguchi jointly conceived the project. K.K. synthesized the bulk crystals and performed the magnetization measurements. L.P. fabricated the FIB samples and performed the LTEM and DPC-STEM measurements. MFM measurements were performed by K.K. with the support of F.K. J.M. theoretically considered the experimental results and performed micromagnetic simulations. The results were discussed and interpreted by all the authors.

**Competing interests**

The authors declare no competing interests.

**Additional information**

Extended data is available for this paper at https://doi.org/10.1038/s41563-020-00898-w. Supplementary information is available for this paper at https://doi.org/10.1038/s41563-020-00898-w.

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**Extended Data Fig. 1 | Micromagnetic simulations of the sawtooth magnetic texture.** The panels show the magnetization in various layers at different depth of a film as obtained from a three-dimensional micromagnetic simulation. The simulated sample measures 1.6 µm × 0.8 µm × 5.3 µm where periodic boundary conditions are applied in the x-y-plane to mimic an extended plate. The colour encodes the direction of the magnetization in the plane and black/white encodes the out-of-plane component, as indicated by the square-shaped antiskyrmion on the bottom right panel, which also sketches the DMI-preferred helicities. In addition, small arrows also show the direction of the in-plane components of the magnetization.