Elliptical Bloch skyrmion chiral twins in an antiskyrmion system

Jagannath Jena1,4, Börge Göbel1,2,4, Tianping Ma1, Vivek Kumar3, Rana Saha1, Ingrid Mertig2,1, Claudia Felser3 & Stuart S.P. Parkin1

Skyrmions and antiskyrmions are distinct topological chiral spin textures that have been observed in various material systems depending on the symmetry of the crystal structure. Here we show, using Lorentz transmission electron microscopy, that arrays of skyrmions can be stabilized in a tetragonal inverse Heusler with $D_{2d}$ symmetry whose Dzyaloshinskii-Moriya interaction (DMI) otherwise supports antiskyrmions. These skyrmions can be distinguished from those previously found in several B20 systems which have only one chirality and are circular in shape. We find Bloch-type elliptical skyrmions with opposite chiralities whose major axis is oriented along two specific crystal directions: [010] and [100]. These structures are metastable over a wide temperature range and we show that they are stabilized by long-range dipole-dipole interactions. The possibility of forming two distinct chiral spin textures with opposite topological charges of ±1 in one material makes the family of $D_{2d}$ materials exceptional.
Non-collinear spin textures range in size from atomic scale to microscopic and have attracted much interest because of their unique physical properties and potential applications in spintronic devices such as the racetrack memory. Of these spin textures, skyrmions and antiskyrmions are particularly fascinating because they are magnetic objects with distinct chiral magnetic boundaries. Both are characterized by opposite topological charges of ±1 and have only been predicted to coexist in non-chiral systems like in frustrated magnets. However, experimentally, this coexistence has not been observed, yet. Which object is actually found in a given magnetic material depends on the underlying symmetries of its crystal structure.
These objects are surrounded by a black line with a white contrast in the center at higher fields. Upon decreasing the field, these objects become larger and a gray contrast appears in their center. The objects exist at zero field (Fig. 3e) and are partially metastable even for fields as large as $-128$ mT, where a transition to a helical phase sets in (Fig. 3f). These objects would appear to look like Bloch-like skyrmions, as previously observed, but these
conventional skyrmions have been found to be circular and not elliptical in shape as we observe here. Note that a similar behavior is also found at 250 and 150 K (cf. Supplementary Fig. 3). At 100 K, we only observe a few isolated elliptical skyrmions that directly transition to the helical phase (see Fig. 4a–c). However, in a thicker sample, a periodic lattice of elliptical skyrmions has been stabilized even at such low temperatures (Fig. 4d–k) pointing towards the relevance of dipole–dipole interactions in this material.

The $D_{2d}$ symmetry of the tetragonal Heusler system gives rise to an anisotropic DMI, which strictly favors the stabilization of antiskyrmions over that of conventional skyrmions. In order to account for the unusual elliptically shaped nano-objects that we see at low temperatures in Mn$_{1.4}$Pt$_{0.9}$Pd$_{0.1}$Sn, we have

**Fig. 3 Formation of elliptical skyrmions.** The figure shows the evolution of the LTEM contrast with decreasing field applying the modified experimental protocol (applying reversible tilting along [110] at high fields until a lattice of objects has formed) at the temperature of 200 K. Elliptic objects are observed that are identified as elongated Bloch skyrmions. One observes a few skyrmions at high fields (a) and then a transition to a densely packed array of skyrmions (b–e), which transforms into the helical phase only at negative fields (f). The black horizontal line in each LTEM image corresponds to bending contours. The scale bar corresponds to 500 nm.

**Fig. 4 Elliptical skyrmions at 100 K for two different sample thicknesses.** In a–c LTEM measurements in the previously investigated sample (170 nm thick) are shown. The field is reduced coming from the ferromagnetic phase, with only a few skyrmions shown in a. In b helices have formed that constitute the ground state at zero field in c. In d–k the sample has a thickness of 280 nm. Starting again at high fields (d), a dense array of elliptical skyrmions forms (e), which persists upon decreasing the field (f–g), even down to zero field (h) and below (i). Finally, at $-104$ mT (j), the helical phase starts to dominate. At $-144$ mT (k), the helical ground state is restored. The scale bars correspond to 500 nm.
carried out micromagnetic simulations (see Methods) for this material that include a DMI interaction consistent with the $D_{2d}$ symmetry, and take into account dipole–dipole interactions.

**Explanation and micromagnetic simulations.** To explore possible metastable magnetic configurations, we carry out micromagnetic simulations that are initialized with circularly shaped, topologically different magnetic states. The process of relaxation is shown in Supplementary Fig. 4; in the following, we discuss the relaxed, metastable configurations (labeled as “simulation” in Fig. 5a–d).

When the initial state is set to be an antiskyrmion, we find, after relaxation, that this object is indeed a metastable configuration for the magnetic parameters chosen (Fig. 5c). When the initial state is set to be a particular Bloch skyrmion with a given chirality (here clockwise), we observe that the object converges to an object with an elliptical shape, as we find experimentally (Fig. 5a). Note that, similar to the protocol dependence of the experiments, the metastabilized states in the simulations depend on the initial magnetic texture. This means, for example, that not every initial Bloch skyrmion configuration leads to the stabilization of a metastable object. However, if a nano-object is successfully stabilized, it is an elliptically elongated Bloch skyrmion. A very interesting point that we find from these simulations is that the chirality of the initial object determines along which direction the major axis of the ellipse is oriented. The ellipses are always oriented with their major axis along one of two perpendicular in-plane directions ($[100]$ or $[010]$).

The elliptical objects are Bloch-like because of dipole–dipole interactions, since costly volume charges $\text{div}(\mathbf{m})$ are absent in Bloch walls. The dipole–dipole interactions are of relevance in this system since the sample thickness and the saturation magnetization are considerable. As mentioned above, one of the relaxed Bloch skyrmions is elongated along $[100]$ (Fig. 5a, b) as in our experiments, which we attribute to the anisotropic DMI. This type of DMI favors Néel configurations along the in-plane $\{11\}$ crystallographic directions and Bloch configurations along $\{01\}$.
the in-plane \{1 0\} directions. Importantly, the favored chirality of the Bloch parts differs locally: a clockwise state is favored along [0 1 0] and [0 1 0] (green and pink arrows), while counterclockwise parts (blue and yellow) are favored along [1 0 0] and [1 0 0]. Consequently, the favorable parts of the Bloch skyrmion are enlarged, effectively elongating the skyrmion along the [1 0 0] direction here. The simulated LTEM images for such a configuration (Fig. 5a, b) well describe the experimental observations even at different magnetic fields strengths (Fig. 5e, f). Simulated LTEM images for different defocus values are shown in Supplementary Fig. 5. When the initial Bloch skyrmion is chosen to have a counter-clockwise chirality (cf. Supplementary Fig. 4c), the relaxed state is again an elliptical object, but which is now elongated along the perpendicular [0 1 0] direction (see Fig. 5i). This time the blue and yellow highlighted Bloch parts are favored by the anisotropic DMI and are consequently enlarged.

We have indeed observed experimentally that the elliptical skyrmions are oriented along just two crystal directions, and indeed those that our micromagnetic simulations showed were energetically favored (Fig. 5i–k). Moreover, the LTEM images show that the elliptical skyrmions in \( \mathrm{Mn}_{1.4} \mathrm{Pt}_{0.9} \mathrm{Pd}_{0.1} \mathrm{Sn} \) have specific and opposite chiralities for these two crystal directions. Thus, these elliptical skyrmions come as “chiral twins.”

This makes elliptical skyrmions fundamentally different from DMI-stabilized skyrmions in B20 materials, where only one chirality is allowed, determined by the chirality of the material’s cubic B20 symmetry is used, show circularly shaped Bloch skyrmions, even after taking into account dipole–dipole interactions. Note that there exist reports of deformed or distorted skyrmions in several B20 materials. However, the deformations in these cases are irregular, and are either a result of anisotropic -vectors or confinement in nano-structured materials, from nano-sized grains or laterally patterned nano-structures. These deformations are not tied to the underlying crystal structure and, thus, are not an intrinsic property as we find here.

The influence of dipole–dipole interactions also becomes apparent in the micromagnetic simulations of an initial antiskyrmion after relaxation to a metastable configuration (Fig. 5c, d). Interestingly, here we find two distinct shapes after relaxation. In larger magnetic fields, where the object is smaller, the original circular shape is maintained but, in lower magnetic fields, a square deformation takes place: the Bloch parts of the antiskyrmion along the family of in-plane [1 0] directions (pink, blue, green, and yellow arrows) are favored by the dipole–dipole interactions, leading to an enlargement of these parts. On the contrary, the Néel parts are unfavorable, leading to a spatial shrinking. The result is a square-shaped antiskyrmion with a calculated LTEM signal (see Methods) similar to the experimental observations (cf. Fig. 5d, h). In higher magnetic fields, the antiskyrmion decreases in size (Fig. 5c) so that dipole–dipole interactions become less important due to the shortened wall lengths. Since the domain wall width is now comparable to the size of the magnetic object, Bloch and Néel parts become almost energetically equivalent and the square deformation is not noticeable anymore.

In this regard, we note that there is a clearly noticeable transformation process between them as observed in Fig. 1h–j. In Supplementary Fig. 7, we show that round antiskyrmions can transform to square-shaped antiskyrmions when the magnetic field is simply decreased. However, when the field is increased again, the state remains square shaped. The transition to round antiskyrmions is only possible when an in-plane field is temporarily provided. There appears to be a small energy barrier between the round and square antiskyrmion lattices in our experiments. In the case of isolated objects in an ideal sample, as presented in the micromagnetic simulations, this barrier is absent. For this reason, the interaction between neighboring objects also seems to have an influence on the metastability of magnetic objects in \( D_{2d} \) materials. The possibility for controlled switching between both states is promising for future applications.

Since we observed antiskyrmions near room temperature and elliptical skyrmions at 250 K and below, we also investigated the critical behavior at intermediate temperatures. Near 268 K we find coexisting skyrmions and antiskyrmions (round at high fields in Fig. 6a and square shaped at low fields in Fig. 6b), indicating that both nano-objects have a comparable energy at this temperature with a presumably decreased energy barrier.

**Discussion**

The reported metastability of round- and square-shaped anti-skyrmions, as well as elliptically deformed Bloch skyrmions with two different chiralities arises from the anisotropic DMI in Heusler materials compared to traditional skyrmion hosts. The anisotropic DMI favors the stabilization of antiskyrmions, while the dipole–dipole interactions favor Bloch-type skyrmionic objects. In Fig. 7 we show the phase diagram, which summarizes our results. Comparing this phase diagram with our initial publication shows how important the experimental protocol is for the stabilized magnetic phase. In the present protocol, we find elliptical skyrmions at low temperatures (250 K and below) and round- and square-shaped antiskyrmions near and above room temperature (see the phase diagram in Fig. 7). At a critical temperature near 268 K, we observe the coexistence of antiskyrmions and elliptical skyrmions (Fig. 6).

We relate the temperature dependence to the decreased net magnetization at higher temperatures, which reduces the importance of dipole–dipole interactions. To further investigate the role of the magnetization on the stability of elliptical skyrmions, we have measured a thicker sample (Fig. 4). While in the previously investigated sample (thickness 170 nm), only a very few individual skyrmions could be stabilized at 100 K (Fig. 4a–c), in the thicker sample (280 nm thickness), we observe a well-ordered skyrmion lattice (Fig. 4d–k).

Besides the coexistence of skyrmions and antiskyrmions in a single material that we observe for the first time, we also observe two different elliptically deformed Bloch skyrmions with opposite
chiralities. While our theoretical results predict equal probabilities for the formation of the two distinct elliptical skyrmions, in our experiments we mostly find Bloch skyrmions that are elongated along [1 0 0] (see Fig. 3) at temperatures between 150 and 250 K. Even when repeating the experiment with tilted magnetic fields along a different in-plane direction and tilting back, the result is the same (see Supplementary Fig. 8). This implies that the elongation direction is not much influenced by the direction of the in-plane field or the temperature in our case. However, in a different part of the sample (shown in Fig. 5i), we observe a mixed state of both skyrmion types. Therefore, we conclude that in the first case, the chirality of the skyrmions is fixed by the lattice itself. This could, for example, be caused by strain; even a small lattice deformation would lead to an inequivalence of the two elliptical-skyrmion chiralities. In the second part of the sample, the deformation could be negligible, which is why both chiralities would be allowed. Furthermore, it is worth mentioning that in this part of the sample, we even observe what appears to be a topologically trivial object; a bubble that is the combination of a skyrmion and an antiskyrmion (highlighted in Fig. 5i).

In summary, we have observed two topologically distinct phases in a material with $D_{2d}$ symmetry: elliptical skyrmions and antiskyrmions. The observed antiskyrmions come in two varieties, square and round shaped, which can be switched reversibly. We observed individual objects of all three types and observed lattices of elliptical skyrmions and square-shaped antiskyrmions over a wide range of temperature even in zero magnetic field. The elliptical skyrmions are stabilized by dipole–dipole interactions, which leads, via an interplay with the anisotropic DMI in this $D_{2d}$ system, to their elongation along the [1 0 0] or [0 1 0] directions, namely the propagation direction of the ground state helical phase. Moreover, the elliptical skyrmions possess either a left- or right-handed chirality, depending on their elongation direction. This makes elliptical skyrmions distinct from the rotational-symmetric DMI-stabilized Bloch skyrmions in B20 materials. In the latter case, dipole–dipole interactions reinforce the DMI to favor circularly shaped skyrmions, without the possibility of creating spin textures with distinct topologies, as we find here in a $D_{2d}$ material. Our observation of magnetic non-collinear chiral spin textures with opposite topological charges in Mn$_{1.4}$Pt$_{0.9}$Pd$_{0.1}$Sn show that $D_{2d}$ materials have greater functionalities as compared to, for example, B20 materials. One can anticipate that their magnetization dynamics may also show greater complexity with the eventual possibility of switching between different topological textures.

Note added: During the review process we became aware of ref. 33, in which the existence of skyrmions, antiskyrmions, and topologically trivial bubbles in an unspecified non-centrosymmetric Heusler magnet is shown. The antiskyrmion shows a square deformation and the skyrmion is elliptically deformed along a single direction.

**Methods**

**Experiment.** The Heusler compound Mn$_{1.4}$Pt$_{0.9}$Pd$_{0.1}$Sn is a bulk polycrystalline sample, which was prepared by the arc-melting method as described in an earlier report. The single crystalline lamella of [0 0 1] orientation were prepared in a Ga$^+$ ion contained dual beam system of FIB (FEI Nova Nanolab 600 SEM/FIB) operating at an accelerated voltage of 30 keV. To find [0 0 1]-oriented grains, an electron backscattering diffraction (Quantax, Bruker) experiment is performed in a TESCAN GAIA 3 system. Various magnetic textures are investigated in an aberration-corrected high resolution transmission electron microscope [FEI TITAN 80-300] operated at an acceleration voltage of 300 keV in LTEM mode. The magnetic field was applied along the electron beam direction of the TEM by partially exciting the objective lens. An in-plane component of the magnetic field was applied by tilting the lamella a few degrees away from the zone axis [0 0 1] using a double tilt liquid nitrogen holder that allows a maximum tilting angle of up to ±40°. The under-focus value for the LTEM images is 1 mm. Periodic vertical lines in the LTEM images are non-magnetic contrasts caused by the FIB specimen preparation.

**Micromagnetic simulations.** For the micromagnetic simulations the Landau–Lifshitz–Gilbert equation was propagated for every discretized magnetization cell (normalized moment $m$) according to:

$$m = -\gamma_e m \times B_{eff} + \alpha m \times m.$$

Here, $\gamma_e$ is the gyromagnetic ratio of an electron, and $\alpha = 0.3$ is the Gilbert damping parameter. $B_{eff}$ is the effective magnetic field derived from the free energy density $F$, taking into account Heisenberg exchange, dipole–dipole interactions, magnetic anisotropy, external magnetic field, and the DMI. Tetragonal Heusler compounds with a $D_{2d}$ symmetry exhibit an “anisotropic DMI” as follows:

$$H_{DMI} = D \left( m_x \frac{\partial m_z}{\partial x} - m_z \frac{\partial m_x}{\partial z} - m_y \frac{\partial m_z}{\partial y} + m_z \frac{\partial m_y}{\partial y} \right).$$

In Mn$_{1.4}$Pt$_{0.9}$Pd$_{0.1}$Sn this form of the DMI emerges due to the layered nature of the crystal structure where the Pt atoms break inversion symmetry of a magnetic Mn layer differently along the [1 0 0] and [0 1 0] directions.
We take the following parameters from ref. 14, where antiskyrmions were initially reported: saturation magnetization $M_s = 445 \text{ kA/m}$ and exchange interaction $A_{ex} = 1.2 \times 10^{-10} \text{ J/m}$. The anisotropic DMI is characterized by the constant $D$. In ref. 14 $D$ was assumed to be 0.006 J/m², which was an overestimate, since the dipole–dipole interaction was neglected. Thus, here we use $D = 0.003 \text{ J/m}^2$. A magnetic field $B_z$ was applied along the predominant magnetization orientation (+z) and an easy-axis anisotropy along $z$ was considered with $K_a = 0.2 \text{ Ml/m}^3$. A sample size of 1200 nm × 1200 nm × 175 nm was simulated at a cell size of 5 nm × 5 nm × 5 nm. The simulations were carried out using the GPU (graphics processing unit)-accelerated program Mumax36,37, which was modified to include an anisotropic DMI.

**Simulation of LTEM images.** For the calculation of the LTEM images, we use a first-order approximation considering only a single layer (average magnetization of all layers) and a deflection of the incoming electron beams by the Lorentz force. The incoming electrons at the position $(x',y')$ are modeled by a Gaussian smearing function with a smearing factor $a = 60 \text{ nm}$. The transmitted electron flux at $(x,y)$ is proportional to:

$$\exp\left\{\frac{-[(x - d \cdot m_x(x',y') - x)^2 + (y + d \cdot m_z(x',y') - y)^2]}{a^2}\right\} \text{d}x\text{d}y.'$$

Here, $d = 2 \text{ nm}$ characterizes the maximum deflection of electrons by the Lorentz force.

**Data availability**

The data that support the findings of this study are available from the corresponding author upon request. The micromagnetic simulation code is available at https://github.com/mumax/3/releases/tag/v3.9.3. Anisotropic DMI has been included in this code. The modified version is available from B.G. upon reasonable request.

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**Author contributions**

J.J. and S.S.P.P. conceived the project. J.J. prepared the lamella, performed the LTEM measurement and skyrmion by Lorentz TEM. Microsc. Microanal. 25, 1840–1841 (2019).

**Competing interests**

The authors declare no competing interests.

**Additional information**

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**Correspondence and requests for materials should be addressed to S.S.P.P.**

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