Direct imaging of magnetic field-driven transitions of skyrmion cluster states in FeGe nanodisks

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Magnetic skyrmion is a nanosized magnetic whirl with nontrivial topology, which is highly relevant for applications on future memory devices. To enable the applications, theoretical efforts have been made to understand the dynamics of individual skyrmions in magnetic nanostructures. However, directly imaging the evolution of highly geometrically confined individual skyrmions is challenging. Here, we report the magnetic field-driven dynamics of individual skyrmions in FeGe nanodisks with diameters on the order of several skyrmion sizes by using Lorentz transmission electron microscopy. In contrast to the conventional skyrmion lattice in bulk, a series of skyrmion cluster states with different geometrical configurations and the field-driven cascading phase transitions are identified at temperatures far below the magnetic transition temperature. Furthermore, a dynamics, namely the intermittent jumps between the neighboring skyrmion cluster states, is found at elevated temperatures, at which the thermal energy competes with the energy barrier between the skyrmion cluster states.

Significance

The rapid growth of data volume demands faster and denser storage devices. The noncoplanar swirling spin texture, known as magnetic skyrmion, has potential application in future memory devices. To realize such applications, it is essential to understand the properties of individual skyrmion in patterned nanoelements. While quite a number of theoretical efforts have been made in this field, direct experimental demonstration in such a real modeling system is a challenge. Here, we report the direct visualization of skyrmion cluster states in FeGe nanodisks. We determine the common relationship among the temperature, magnetic field, and disk size. These results have an immediate implication for designing future skyrmion-based devices.

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transitions between different skyrmion cluster states. A representative TEM image of the nanodisk is shown in Fig. 1A. An external magnetic field, \( \mathbf{H} \), is applied perpendicular to the disk plane with its direction pointing upward (marked as a red circled dot). Fig. 1B shows the intact magnetic contrast in ground state under zero magnetic field cooling conditions at the temperature \( T = 100 \text{ K} \), where the dark and light stripes represent the different planar magnetic moment distributions. By using the magnetic transport-intensity equation (TIE) analysis of the Lorentz TEM data (19), the in-plane components of the spin textures in the disk are constructed, as shown in Fig. 1C. Obviously, the Lorentz TEM images reveal a distorted helical ground state with four turns (Fig. 1C and E). The period of the helix is about 68 nm, similar to the value in bulk \( \lambda \approx 70 \text{ nm} \) (18). However, the spatial confinement effect in the lateral direction yields an incomplete helix around the disk edge due to the specific form of the boundary conditions (20, 21).

When a magnetic field is applied, the distorted helical state rapidly adjusts its spin orientation. At \( H \approx 1.42 \text{ kOe} \), it transfers into a confined helix with three turns, with period remaining the featured value \( \lambda \approx 70 \text{ nm} \) (Fig. 1D) (18). With further increase of the magnetic field, the helix shrinks and eventually changes into three elongated skyrmions, i.e., bimerons (22), positioned along the edge (Fig. 1E and N). Meanwhile, the remaining spins around the edge form a big vortex according to theoretical prediction and simple geometry analysis, although the Fresnel fringe partially smears out the real magnetic contrasts of edge vortex (8, 13, 20, 21). At \( H \approx 1.96 \text{ kOe} \), the elongated skyrmions shrink into circles and assemble along the edge (Fig. 1F), indicating the attractive interaction among them (8, 17).

Once the skyrmions are formed around the edge, the elevated magnetic fields shrink these individual skyrmions and drive them gradually toward the interior of the nanodisk, due to repulsions between edge spins and skyrmions (Fig. 1G–J) (4, 17). Finally, this triskyrmion cluster with triangular arrangement is formed in a wide interval of magnetic fields (\( 1.96 \text{ kOe} < H < 3.70 \text{ kOe} \)), although the position of each individual skyrmion in the cluster can adjust correspondingly with the varied \( H \) (23). At \( H \approx 3.91 \text{ kOe} \), two skyrmions annihilate instantaneously almost at the same time (Fig. 1K), leaving one skyrmion sitting nearly at the center of the disk to minimize the skyrmion–edge interaction (4). The single-skyrmion state is eventually terminated at \( H \approx 3.98 \text{ kOe} \) (Fig. 1L). The magnetic field intervals for hosting cluster states with skyrmion numbers \( N_s = 1 \) are much smaller than that of \( N_s = 3 \) (Fig. 1Q). These results are consistent with recent experimental observations on thin MnSi nanowires by MR measurements (15), in which the magnetic field intervals of hosting maximum number of skyrmions \( N_{s_{\text{max}}} \) is always wider than those with \( N_s < N_{s_{\text{max}}} \) if the same dynamical procedure is followed. By contrast, this observation is quite different from the theoretical calculation that the magnetic intervals of hosting different skyrmion clusters are comparable (8, 13). These differences probably come from the fact that the theoretical model only considers the equilibrium states (13), and the magnetization dynamics of skyrmions were not included.

**Fig. 1.** Variations of spin texture with magnetic field in a 270-nm FeGe nanodisk at \( T = 100 \text{ K} \). (A) TEM image of the FeGe nanostripe surrounded by an amorphous PtC layer. (B) The intact magnetic contrast in ground state under underfocused conditions of Lorentz TEM with a defocus value of –192 \( \mu \text{m} \). The magnetic field is applied perpendicular to the stripe plane. (C–L) Magnetic-field dependence of the spin texture at (C) 0 kOe, (D) 1.42 kOe, (E) 1.62 kOe, (F) 1.96 kOe, (G) 2.76 kOe, (H) 3.04 kOe, (I) 3.40 kOe, (J) 3.70 kOe, (K) 3.91 kOe, and (L) 3.98 kOe. The color wheel in L indicates the direction and strength of in-plane magnetization at each point. For clarity, some typical spin textures in C, D, and I are zoomed in M, N, and O, respectively. (M) A single skyrmion. The white arrows represent the in-plane magnetization at each point. (Q) The phase diagram in \( H \) space. \( N_s \), skyrmion numbers.
Magnetization Dynamics of Skyrmion Cluster States at High Temperature. The field-driven discrete transition of skyrmion cluster states persists at elevated temperatures for \( T < T_c \). Because the thermal fluctuation promotes the formation of skyrmions (14, 24), \( N_m \) for the 270-nm disk increases from 3 at \( T = 100 \) K to 6 at \( T = 220 \) K, as shown in Fig. 2D. The corresponding magnetic field-driven evolution of the spin textures is displayed in Fig. 2A–L. At \( H \approx 0 \) Oe, the distorted helical ground state shows a similar spin configuration to that at \( T = 100 \) K. At \( H \approx 0.47 \) kOe, a skyrmion is isolated from the spin helices, bringing out a mixed state composed of bimerons, skyrmions, and spin helices (Fig. 2C). With further increase of the magnetic field, the mixed state transits to six skyrmions (Fig. 2D). This magnetization process follows previously reported results in 2D films, including FeGe (18), Fe\(_{0.5}\)Co\(_{0.5}\)Si (19), and numerical calculation (13). In this case, one skyrmion lies in the center of the disk, while five other skyrmions circle around, forming a pentagon cluster state. This \( N_s = 6 \) cluster state transits to an \( N_s = 5 \) state at \( H \approx 1.01 \) kOe, where the central skyrmion vanishes, leaving an \( N_s = 5 \) cluster with the regular pentagon arrangements.

Further increasing magnetic fields, the \( N_s = 5 \) cluster state, in turn, transfers into \( N_s = 4, 3, 2, \) and 1 cluster states with a square (Fig. 2F), triangle (Fig. 2I), double bell (Fig. 2J), and target (Fig. 2K) geometry, respectively, via a step-by-step annihilation of an individual skyrmion and the final field-polarized ferromagnetic state (Fig. 2L). Each transition is accompanied by symmetry changes of the cluster states. The position of the skyrmion in the one-skyrmion state is not fixed at the center of the disk, which is in contrast to the expectation according to the theory, suggesting the pinning effects, probably originating from the defects, overwhelm the skyrmion−edge repulsion interaction (Fig. 2K).

Besides the aforementioned properties of skyrmion cluster states, a more striking feature is that we observed a series of quantized jumps during the transition between the two cluster states at a defined magnetic field \( H \). In other words, there exists a quantization intermittent fluctuation region for the transition from one cluster state to another. For example, Fig. 2M shows a
typical \( N_s(t) \) versus time at a fixed \( H \approx 0.98 \) kOe (see Movie S1), where the two cluster states with \( N_s = 6 \) and 5 change by the annihilation or creation, alternatively, of the skyrmion at the center of the disk, and such two-level jumps can survive in the wide range from 0.56 kOe to 1.01 kOe (Fig. 2 E–G). This phenomenon is widely observed during the field-driven transition of the cluster states (see Fig. S2 and Movies S1–S5) at temperatures close to \( T_{C0} \), as shown in Fig. 2N, where the intermittent jumps of cluster states with \( N_s = 2 \) and 1 were clearly seen at \( H \approx 1.41 \) kOe. A frequency spectrum of the jumped behavior by Fourier analysis is shown in Fig. S3, exhibiting no notable regularities within the time resolution limit of the Lorentz TEM. The observations and analysis indicate that the system exhibits the telegraph noise, which occurs due to the thermal fluctuation at the first-order transition between cluster states with different skyrmion numbers.

Aside from the telegraph noise, for an \( N_s = 4 \) cluster with a square symmetry, four skyrmions appear to change their positions with a clockwise or anticlockwise rotation so that the Lorentz TEM images become blurred, with less visible contrast. Such rotational motion is more apparent in the 330-nm disk (see Movie S3 and Fig. S2), where the eight skyrmions circling the disk rotate around the central one or two skyrmions. Unfortunately, the time resolution of Lorentz TEM cannot grasp the detail of skyrmion dynamics in the cluster states, which is highly reminiscent of the thermally driven ratchet motion, as observed in other B20 compounds, including MnSi and Cu2OSeO3 (24). The phase diagram in the \( H \) space is illustrated in Fig. 2O. Unlike the tiny magnetic field intervals for \( N_s < N_s^{m} \) states at \( T = 100 \) K, elevated temperature brings dramatic thermal fluctuations, overwhelming the magnetization dynamics. Thus, the magnetic field intervals for all of the cluster states are comparable.

**Temperature–Field Phase Diagram for the Nanodisk.** We have also investigated the \( H \)-driven spin texture in the whole temperature region from 100 K to 260 K; a typical temperature–field (\( T-H \)) phase diagram for a 310-nm nanodisk is shown in the Fig. 3. At the temperature below 190 K, where skyrmions are in sparse distribution forms of cluster state, the maximum number of skyrmions, \( N_s^{m} \), is almost unchanged (see Fig. S4). Conversely, closely packed skyrmions appear at \( T > 190 \) K. With the current experimental accuracy of temperature control, we don’t catch a crossover region in which the skyrmion number \( N_s^{m} \) varies from sparse cluster state at low temperatures to closed packed cluster state at high temperatures. This observation possibly indicates that the crossover temperature region might be very narrow or that the transition happens instantly, which needs to be clarified in future work.

**Relation Between the Maximal Skyrmion Number and the Disk Diameter.** To thoroughly understand the magnetic properties in the confined geometry, we systematically investigated the dependence of skyrmion arrangements on the disk size at two typical temperatures, 100 K and 220 K (the sample parameters are shown in Table S1).

At high temperature, 220 K, a close packing of skyrmions in the \( N_s^{m} \) skyrmion state is expected. Following the same magnetization dynamics for a 270-nm disk (Fig. S2), we clearly observe the prediction that the maximal number of skyrmions, \( N_s^{m} \), increases with the increase of the disk diameter, \( D \) (Fig. 4, filled and open blue circles, \( T = 220 \) K). However, the variation of \( N_s^{m} \) with \( D \) is discrete due to the particle property of skyrmions. In this case, the arrangement of skyrmions in the disk resembles the packing of congruent circles in a circular container; that is, with the raising of container size, the arrangement of circles varies in the pattern sequence of triangular, square, regular pentagon, five around one, six around one, etc. (SI Maximal Number of Skyrmions and Closest Packing in a Circle and Fig. S5). Considering the fact that the maximum circle number is generally proportional to the area of the container, the variation of the circle number \( N \) with container diameter \( D \) can be approximately fitted by a phenomenological curve in the form of \( n = k(D/d - q)^2 \), where \( k \) is the packing density constant, \( D \) and \( d \) are the diameter of the container and small circles, respectively, and constant \( q \) comes from the drop of packing density near the circumference of the container. For dense packing, it provides \( k = 0.861 \) and \( q = 0.32 \) (SI Maximal Number of Skyrmions and Closest Packing in a Circle). Because a

**Fig. 3.** \( H-T \) phase diagram of 310-nm FeGe nanodisk. At temperatures lower than 190 K, skyrmion numbers reached a maximum of 4 in the form of sparse skyrmion. At \( T > 200 \) K, skyrmion numbers reached a maximum of 7 or 8 in the form of packed skyrmions.

**Fig. 4.** The maximal number of skyrmions as a function of disk size at two representative temperatures, 100 K and 220 K. The bright or dark spots represent skyrmions obtained by direct Lorentz TEM imaging. The difference of bright–dark contrast is due to the magnetic chirality inversion of the skyrmions in different crystal chirality. The open blue circles represents the data that are not be used in the fitting parameters at \( T = 100 \) K. The \( N_s^{m}(D) \) curves show square and linear relationships at high temperature and low temperature, respectively, reflecting two different mechanisms to create skyrmions in confined geometry.
small disk may deviate significantly from this rule, the experimental data at larger size \( D > 240 \text{ nm} \) are fitted. We obtained \( k = 0.86 \pm 0.06, q = 0.7 \pm 0.1 \), assuming that \( d \) is equal to the featured skyrmion lattice constant \( a_2 = 2/\sqrt{3} \approx 11 \text{ nm} \); \( q \approx 0.7 \) is larger than that of dense packing situation, which can be interpreted as the presence of edge state, and the deviation of 0.38 implies the size of the edge vortex is on the order of \( 0.38 a_2/2 \approx 15 \text{ nm} \) (8, 13), which is also a reasonable estimation.

At low temperatures below 190 K, the evolution of the distorted helical state followed the process for a 270-nm disk at \( T = 100 \text{ K} \), where one complete distorted spin helix changes into one skyrmion (Fig. S6). Because the helical state possesses a relatively fixed period \( A \), the maximal skyrmion number, \( N^m \), is directly proportional to the reduced size \( D/A \) (Fig. 4, black dots, \( T = 100 \text{ K} \)), i.e., linearly dependent on \( D \). The yielded compensated constant 0.6 indicates the size of the edge vortex is on the order of \( 0.6/2A = 21 \text{ nm} \), which implies that the region of the edge vortex at low temperatures is larger than that at high temperature.

**Discussion**

Since the discovery of magnetic skyrmion in 2009, quite a few theoretical and experimental works have been performed on the spin textures and magnetization process in confined helimagnets including nanodisks, nanowires with circular or parallelogram cross-sections (8, 13, 15); the elemental results lie in the emergent skyrmion cluster states and discrete transitions between them due to the confined geometry. These predictions were directly confirmed by our Lorentz TEM study. Meanwhile, several previously unidentified properties were found beyond the conventional predictions. For example, the magnetic field intervals for these different cluster states depend on the temperature, and show remarkable deviation from the theoretical results (8, 13). In addition, the temperature effect is not fully considered theoretically, so the telegraph noise is not predicted. Moreover, recent works have successively discovered several skyrmion materials at or above room temperature, including beta-Mn-type CoZnMn alloys (25), CoFeB/Ta bilayers (26), ultrathin Pt/Co/MgO nanostructures (10), and Ir/Co/Pt multilayers (27). These advances unambiguously promote real application. However, the thermal telegraph noise observed in the present work would provide another perspective on these efforts, because thermal fluctuations may impose limits on the development of skyrmion-based devices.

Skyrmions have many features in common with Abrikosov vortices in superconductors (28, 29). They are both condensed into a triangular lattice in bulk or 2D films due to the minimization of their repulsive interactions. It is well-established that size confinement would lead to certain novel quantum objects, such as the giant vortex in a superconducting nanodisk, which cannot survive in a macroscopic sample. Similar to the giant vortex, a new topological state, named target skyrmion, characterized by a skyrmionic core with a series of circular spin stripes, has been suggested in nanodisks or nanowires under certain conditions (8, 30). Especially, the target skyrmion may be the spontaneous ground state, as the disk size is close to the featured helical period (31). Similar target domain structure has been observed in Ni and FePt nanodots (32, 33). However, the theory-predicted target skyrmion in the chiral magnets is not observed in the present experiment. We have tried to image the magnetic structure on the disk with a diameter around 100 nm, but the weak magnetic contrast and Fresnel fringe make it impossible to obtain valuable information so far. The search for the target skyrmion is still an interesting issue.

The skyrmion is a particle-like spin configuration with an integer topological charge. Its particle behavior makes it instructive to compare the ordering and dynamics of skyrmion cluster states in confined geometries (34–38) with the broader class of systems exhibiting mesoscopic ordering in similar geometries. The observed behavior of magnetic skyrmion clusters in a nanodisk shows a similarity to various other phenomena such as the charge ordering in quantum dots, ion crystals, colloidal ordering in traps (37), and dusty plasmas (35). However, different from these orderings, the skyrmion number is not conserved, and thus the dynamical transitions between the cluster states with different skyrmion numbers is unique.

In conclusion, we have addressed the problem of the quantum skyrmion phase transitions in strongly confined helimagnets, i.e., nanodisks with comparable size to the helical period. All of the disks show a distorted helical ground state. On an external magnetic field aligned perpendicular to the disk plane, the helical ground state split into individual skyrmions, with the maximum number determined by the disk size and temperature. The phase transitions between these cluster states are abrupt, with the appearance of telegraph noise at high temperature. These results reveal unique physics of the skyrmion cluster states in confined geometries, and can guide the development of skyrmion-based memory devices in which the individual skyrmions could be used for multibit memory cells.

**Materials and Methods**

**Sample Fabrication.** The B20-type FeGe samples were synthesized with a cubic anvil-type high-pressure apparatus with the detailed description in ref. 15. The nanodisks for TEM observation were prepared by using an FIB and scanning electron microscope (SEM) dual beam system (Helios Nanolab 600i; FEI) combined with a gas injection system (GIS), and a micromanipulator (Omniprobe 200+; Oxford Instruments). The details of the sample fabrication processes are shown in Fig. S1. The parameters of fabricated samples with varied diameters are summarized in Table S1.

**Lorentz TEM Measurements.** The high-resolution lateral magnetization distribution map was obtained by TIE analyses of the Lorentz TEM images. Thin-plate thickness was measured using electron energy loss spectroscopy (EELS). A double-tilt liquid-nitrogen cooling holder (Cryo-Transfer Holder; Gatan) was used to detect the phase transition below the Curie transition temperature \( T_c \). This enables the specimen temperature to be reduced to 100 K, with the measured temperature displayed on the cooling holder controller. The magnetic field applied normal to the thin plate was induced by the magnetic objective lens of the TEM (JEM2100F, JEOL).

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