Nano-to-micro spatiotemporal imaging of magnetic skyrmion’s life cycle

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Magnetic skyrmions are self-organized topological spin textures that behave like particles. Because of their fast creation and typically long lifetime, experimental verification of skyrmion's creation/annihilation processes has been challenging. Here, we successfully track skyrmion dynamics in defect-introduced Co9Zn9Mn2 by using pump-probe Lorentz transmission electron microscopy. Following the nanosecond photothermal excitation, we resolve 160-nm skyrmion's proliferation at <1 ns, contraction at 5 ns, drift from 10 ns to 4 µs, and coalescence at ~5 µs. These motions relay the multiscale arrangement and relaxation of skyrmion clusters in a repeatable cycle of 20 kHz. Such repeatable dynamics of skyrmions, arising from the weakened but still persistent topological protection around defects, enables us to visualize the whole life of the skyrmions and demonstrates the possible high-frequency manipulations of topological charges brought by skyrmions.

INTRODUCTION

Skyrmion is a topologically stable particle first proposed in nuclear physics (1). It is characterized by a topological charge representing an integer how many times the field configuration wraps a sphere. Recently, it has been proven that this concept can be applied to condensed matter physics (2–5). Previous Lorentz transmission electron microscopy (LTEM) (6) studies revealed winding spin textures in non-centrosymmetric chiral magnets, referred to as magnetic skyrmions. In these materials, the competition between the relativistic Dzyaloshinskii-Moriya (DM) (7, 8) and ferromagnetic exchange interactions stabilizes the triangular skyrmion lattice (SkL) under the external magnetic field. Here, the energy scale of atomic exchange energy J is required for creation (annihilation) of a skyrmion. When J is much larger than the thermal fluctuation of kB T, where kB is Boltzmann constant, it generally ensures the skyrmion’s long lifetime at a certain temperature T, which is associated with the topological protection (9).

The diameter of skyrmions in chiral magnets typically ranges from ~5 to ~100 nm depending on the competition of the magnetic interactions. Since the size is much larger than the lattice constant, the skyrmions are free from the commensurate pinning and thus obtain high mobility (9). When there are any imperfections in the system, which inevitably exist as defects, disorders, edges, interface roughness, etc. in real materials, skyrmions tend to deform (10, 11) and become rather flexible due to the weakened topological protections (12). Understanding the effect of such imperfections on skyrmion dynamics has been an important issue in skyrmion-based applications (13, 14). The combination of the flexible particle character with high internal degrees of freedom is expected to bring about the multiscale dynamics analogous to the soft matters (15, 16). Although skyrmion dynamics such as creation/annihilation (17, 18), deformation and self-vibrations (19, 20), drift motion (21, 22), and many-body interactions (23, 24) have been independently investigated by specific experimental probes, how these dynamics sequentially evolve in the multiple spatiotemporal scales is not yet clarified. This issue should be highly relevant in operating the skyrmion-based spintronic devices (9, 13, 14), where the kinetics of skyrmions are playing the pivotal roles for the performance. Nevertheless, its comprehensive investigation has been a challenging issue, particularly because of difficulties in achieving a wide spatiotemporal range down to nanoscale for the magnetic imaging techniques.

Here, we report nano-to-micro spatiotemporal imaging of the skyrmions in a chiral magnet Co9Zn9Mn2 thin film with defects introduced. We use pump-probe LTEM (Fig. 1A) with high spatiotemporal resolutions of 10 nm and 10 ns. By irradiating the nanosecond-photothermal pulses, we continuously resolve proliferation at <1 ns, contraction at 5 ns, drift from 10 ns to 4 µs, and coalescence at ~5 µs, which relay the multiscale ordering and relaxation of skyrmions cluster in a repeatable cycle of 20 kHz. The flexibility and short lifetime of the skyrmions are attributed to the weakened but still persistent topological protection around the local defects. Our observations visualize the unique life cycle of the photothermally induced metastable skyrmions around the defects. While majority of the past research on magnetic alloys had been rather focusing on clean skyrmion crystals, our present result offers important aspects in skyrmion physics, which are associated with the major processes of manipulating skyrmions.

RESULTS

Pump-probe LTEM on metastable skyrmions

For the pump-probe LTEM measurements, we prepared the partially disordered skyrmions condensed state by introducing crystal defects. First, by using the focused ion beam (FIB) method, we shaped a 100-nm thin plate from a single-crystalline Co9Zn9Mn2 (25, 26). The thin-plate Co9Zn9Mn2 exhibits a helical spiral magnetic structure and triangular SkL in the equilibrium phase diagram (Fig. 1B), as represented by the magnetization textures obtained by analyzing LTEM images with a transport-of-intensity equation at 370 K under 0- and 35-mT magnetic fields, respectively (Fig. 1C and D). The magnetic modulation period (~160 nm) is determined by 2πJ/D, where D is the DM interaction. With field cooling, the equilibrium...
Overviewing one-cycle dynamics of metastable skyrmions

In the pump-probe measurements, we homogeneously irradiated the thin plate with nanosecond-pulsed laser with the fluence of $F = 5.1$ mJ cm$^{-2}$ to induce the skyrmion dynamics. First, we estimate the change in the sample (lattice) temperature (32) caused by the laser irradiation from the transient intensity of the Bragg spots (Fig. 2A) obtained by the time-resolved electron diffraction measurements (Fig. 2B). The rapid decrease at time zero and the exponential recovery with a time constant of 1.4 $\mu$s represent the temperature jump up to 380 K in 1 ns (pulse duration of pump laser) and the subsequent relaxation back to RT in 1.4 $\mu$s, respectively (thick black curve in Fig. 2B).

The LTEM images before and after the temperature jump show that the disordered (locally seven bonded) skyrmion state ($\sim$430 ns; Fig. 2C) changes into the quasi-hexagonal skyrmion cluster (270 ns; Fig. 2D), suggesting the photothermal-induced disorder-order transformation. A series of the LTEM images in Fig. 2 (E1 to E8; taken from the region depicted by broken-line squares in Fig. 2, C and D) indicates that the magnetic contrast gradually transforms as a function of delay time $t$, showing the one-cycle dynamics of the skyrmions. Transient sample temperatures are indicated at the bottom of each panel. Here, we focus on adjacent two elliptical skyrmions ($\sim$350 nm $\times$ 160 nm) presented in Fig. 2E1 ($t = -15$ ns), colored by blue (skA) and red (skB) in Fig. 2F1, at RT under 17-mT magnetic field. As shown in the series of LTEM images (Fig. 2E) and the schematics (Fig. 2F), skA and skB exhibit different but correlated dynamics. Within the time resolution, skB splits into two smaller and more isotropic skyrmions (skB1 and skB2 with diameters of $\sim$160 nm), i.e., skyrmion proliferation. On the other hand, skA rapidly shrinks in 10 ns, accompanying the shift of the center of mass, and then realizes the nearly circular shape of $\sim$160 nm. After that, skB1 moves upward for $\sim$100 ns to where the distances among the skyrmions become uniform, thereby forming a rather isotropic skyrmion cluster. These subsequent dynamics in Fig. 2 (E1 to E5) can be interpreted as the self-organization process associated with the disorder-order transformation. After $\sim$300 ns, on the other hand, the relaxation process sets in, and the skB1 starts to move back to its initial position with slight deformation. Last, skB1 and skB2 recombine to form the elliptical skyrmion around $\sim$5000 ns (skyrmion coalescence) and skA simultaneously elongates to recover its initial shape. Almost identical magnetic patterns at $\sim$15 (Fig. 2E1) and 7820 ns (Fig. 2E8) prove that the observed skyrmion dynamics are repeatable in this time scale. Here, we closely look at individual skA and skB with quantitative analysis of the transient LTEM images.
Skyrmion proliferation, reshaping, and self-organization toward ordered cluster (0 to 300 ns)

First, we show the initial change of the skyrmion caused by the thermal jump from RT to 380 K. Figure 3 (A1 and A2) exhibits the LTEM images obtained at −10 and 10 ns, respectively, which indicate a suppression of the magnetic contrast after photoexcitation. Detailed t dependence of its spatial integration as shown in Fig. 3D suggests that the suppression reflects the ultrafast demagnetization process. It has been known that the demagnetization caused by the jump in the electron temperature occurs in a short time [e.g., ~100 fs for ferromagnetic alloy (33)]. Although the present observation is naturally limited by the time resolution (gray area in Fig. 3D), we can define the time zero as the reference to the subsequent slower skyrmion dynamics as follows.

Next, we discuss the skyrmion proliferation and reshaping processes occurring in 20 ns from time zero. To investigate the proliferation of skB, we show the intensity profiles of the LTEM image along line 1 (Fig. 2E1). As shown in Fig. 3B, an additional signal shows up in the middle of skB around time zero (double-headed arrows B → B1 and B2). It corresponds to the emergence of the magnetic wall in skB indicated by the black arrows in Fig. 2 (E3 and F3), suggesting the proliferation process of skB (~330 nm) to skB1 (~160 nm) and skB2 (~160 nm). Detailed t dependence of the magnetic wall formation, estimated by the intensity in the white dashed rectangle in Fig. 3B, suggests that the proliferation occurs concurrently with the demagnetization around time zero (see gray area in Fig. 3E). On the other hand, skA contracts to form a circular shape. In the line profile of the LTEM image along line 2 (Fig. 2E1) in Fig. 3C, the contraction seems to be completed in ~10 ns (see double-headed arrow A). We quantitatively analyze the width of skA (W_A) from the contour plots in Fig. 2G obtained from the LTEM images in Fig. 2E (section S3). Detailed t dependence of W_A indicates that the shrinkage from ~310 to ~160 nm occurs ~5 ns behind the demagnetization and skyrmion proliferation (gray area in Fig. 3F). These results reveal the proliferation (time resolution limited) and contraction (5-ns delayed) processes of the flexible skyrmions, which realize the circular skyrmions with a diameter comparable to that at equilibrium ~380 K (160 nm in Fig. 1D).

Difference in the response time of these two processes can be understood by considering the underlying spin dynamics. The skyrmion proliferation can be regarded as the topological transformation accompanied by the skyrmion nucleation. In general, once the local magnetic moment is reversed by external stimuli with an energy scale of J, the DM interaction sequentially aligns the neighboring spins and lastly forms the skyrmion as a winding spin texture (9, 34). Numerical simulation suggested that such skyrmion...
nucleation typically completes in subnanosecond by a pulse laser heating (35), which is consistent with the resolution-limited proliferation around time zero (Fig. 3E). The retardation behavior in the contraction might be understood by the theoretical studies focusing on the skyrmion motion associated with the internal spin degrees of freedom (36, 37). Here, when a skyrmion is driven by a time-dependent external force, the internal spins start precessions giving rise to a large effective mass and delayed drift motion (37). Numerical simulation (35) demonstrated that the spin orientations in the skyrmion are highly disturbed just after the pulse laser heating. The disorder in the spin orientation is then reduced, showing a damped oscillation to form stable and smooth winding texture. The damping time (retardation time) can be several orders of magnitude larger than the time required for skyrmion nucleation (subnanosecond), depending on the Gilbert damping constant of the material (35). We can thus expect that the delayed response in the skyrmion contraction (shift of the center of mass) is caused by the peculiar response in the internal degrees of freedom, which is detectable in the present time resolution.

After acquiring the isotropic circular shapes, the skyrmions rather seem to move as particles. Analysis on the contour plots for skB/skB1 indicates the shift along $S_B$ as functions of time. We found that skB1 starts to move behind the contraction of skA (gray areas in Fig. 3, F and G), which could be naturally understood as the skB1 moving toward a space produced around skA. The increase in $S_B$ continues until ~100 ns. We found that at 100 to 300 ns where $S_B$ takes maximum (orange areas in Fig. 3, H and I), the quasi-hexagonal skyrmion cluster is realized around skA (Fig. 2D), in contrast to the initial state (~430 ns) where skA is surrounded by seven distorted skyrmions (Fig. 2C). Observed skyrmion cluster rearrangement is consistent with an attractive force between skyrmions, which can drive skyrmion cluster formation in a helical background (24). These results indicate that the proliferation, contraction, and drift motion of respective skyrmions (in ~10-ns, ~100-nm scales) are well correlated and develop into the better-ordered skyrmion cluster of larger spatiotemporal scales (~300 ns, submicrometer).

**Relaxation process and skyrmion coalescence (300 ns to 10 μs)**

After ~300 ns, the quasi-hexagonal skyrmion cluster starts to relax back to the initial disordered state. As shown in Fig. 4 (A and B), the relaxations of $W_A$ and $S_A$ in the $t < 4 \mu s$ region are reproduced by assuming the exponential decay with a time constant obtained from the transient temperature in Fig. 4C (1.4 μs) (section S4). Around 4 to 6 μs, on the other hand, the system shows a complicated behavior and sudden recovery to the initial state. To investigate the “mean” dynamics of the skyrmions around 5 μs, we analyze the intensity profile along line 3 (Fig. 2E3). Figure 4D exhibits the splitting of the intensity across the time zero, indicated by the double-headed arrow B1 representing the skB1 part, suggesting the proliferation process of skB to skB1 and skB2 as abovementioned. The size of proliferated skB1 remains constant until ~4 μs (Fig. 4D) and collapses at around 5 μs (skyrmion coalescence) as shown in Fig. 4E. Such discontinuity should be reflecting the glimpse of the “topological protection” by which the skyrmion annihilation cannot be achieved by the adiabatic deformation. Considering that the sample temperature is settled back close to ~300 K already at around >3 μs (Fig. 4C), the lifetime of the proliferated skyrmion ~5 μs may not be simply following right after the cooling of transient temperature.

By more closely looking at the magnetic images, we can discuss the coalescence process occurring stochastically around the time

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**Fig. 3. Skyrmion proliferation, contraction, and self-organization.** (A1) and (A2) LTEM images at ~10 and 10 ns, respectively. Decrease in the magnetic contrast indicates a demagnetization due to the thermal excitation from 300 to 380 K. (B) Time dependence of the intensity profile along line 1 in Fig. 2E1. (C) The same as (B) but along line 2 in Fig. 2E1. (D) Time dependence of the integrated intensity of the LTEM images in (A), a.u., arbitrary units. (E) Time dependence of the LTEM intensity in (B) (white dotted rectangle) showing the emergence of the magnetic wall between skB1 and skB2. (F and G) Time dependences of the width along $a$ of skA ($W_A$) and the shift along $b$ for skB (filled circles) and skB1 (open circles) ($S_B$), respectively, estimated from the contour plots in Fig. 2G. (H) The same as (F) but on a longer time scale. Blue and light blue markers are obtained from the datasets 1 (from ~20 to 24 ns) and 2 (from ~430 to 3970 ns), respectively. (I) The same as (G) but on a longer time scale. Red and pink markers are obtained from the datasets 1 and 2, respectively. The orange areas in (H) and (I) represent the time scale to form the quasi-hexagonal skyrmion cluster.
Fig. 4. Relaxation process and skyrmion coalescence. (A and B) Time evolutions of $W_b$ and $S_b$ on a linear time scale. Open and filled markers are obtained from the datasets 2 (from −430 to 3970 ns) and 3 (from −1280 to 15870 ns), respectively. The black curves in (A) and (B) represent the fitting functions assuming an exponential decay with a time constant of 1.4 μs and a constant background (section S5). (C) Transient temperature deduced from the electron diffraction data in Fig. 2B. (D) Time dependence of the intensity profile along line 3 in Fig. 2E obtained from the dataset 2. (E) The same as (D) but on a longer time scale obtained from the dataset 3. (F to H) Typical contour plots for the LTEM images in regions I, II, and III, respectively. Color bars in (A) to (C) represent the three characteristic states shown in (F) to (H) obtained from the dataset 3. (I) Schematics for the probability of the presence of proliferated skB1 and skB2 as a function of $t$.

DISCUSSION

We summarize our observations in Fig. 5. By irradiating the nanosecond optical pulse, the flexible skyrmions found in the present setup behave as the following. (i) The ultrafast demagnetization occurs following the rapid heating (<1 ps). (ii) Skyrmion proliferates in <1 ns. (iii) Skyrmion contracts by shifting its position with a

range of $5 \pm 1$ μs. As indicated in Fig. 4 (A to C), we can define the time regions I, II, and III according to the contour plot analysis (section S5). In region I, three skyrmions (skA, skB1, and skB2) are well separated (Fig. 4F). In region III, two large elliptical skyrmions (skA and skB) exist similarly as in the initial state (Fig. 4H). On the other hand, the contour plots in region II show that these relevant skyrmions do exist but are connected or overlapped to each other (Fig. 4G). These observations can be explained by considering the event probability of respective skyrmion dynamics. As stated, in the present pump–probe measurements, 36 million events (20 kHz for a 30-min acquisition time) are integrated into a single data at $t$. Therefore, the images presented here should represent the average of these 36 million events. Taking this into account, the plots in region II should be interpreted as the weighted mean of those in regions I and III just on the verge of region II. On the basis of this consideration, we depict a schematic viewgraph showing the probability of the presence of proliferated skB1 and skB2 as a function of $t$ (Fig. 4I). We note that this $t$ dependence is also similar to the image in Fig. 4E.

From these results, we can understand the relaxation process as follows. Starting from the quasi-hexagonal skyrmions cluster at ~300 ns, skB1 moves back toward skB2 with slight elongation following the thermal relaxation down to RT (Fig. 4B). When the lower edges of skB1 and skB2 are aligned (~4 μs), the coalescence starts to occur stochastically in region II. Rapid decrease in the mean size of skB1 around $5 \pm 1$ μs (Fig. 4E) can be interpreted as an exponential decay of the probability of the presence of proliferated skB1 and skB2 with microsecond time constant (Fig. 4I), which is attributed to weakened but still persistent topological protection around the local defects (I2). Probability distribution averaged over 36 million events in Fig. 4I represents the repeatable nature of the skyrmion’s whole life.

Fig. 5. Repeatable life cycle of the magnetic skyrmions in the present setup. Blue, red, and gray wheels represent the skA, skB/skB1/skB2, and skyrmions surrounding skA and skB, respectively. Orange circle represents the time scale required to form the quasi-hexagonal skyrmion cluster.
time delay of ~5 ns (damping), thus forming a circular shape. (iv) Quasi-hexagonal skyrmion cluster is formed in ~100 ns, i.e., self-organization, driven by the drifts of the reshaped skyrmions (orange circle in Fig. 5). (v) Hexagonal skyrmion cluster maintains to ~300 ns, and the relaxation process toward the disordered state follows. (vi) Last, the skyrmion coalescence occurs as a stochastic topological transition at 5 ± 1 μs. We note that, although these events are well resolved both in time and space, they should not be extrapolated to the general picture. Details of the dynamics, especially the lifetime of skyrmion, should strongly depend on materials, boundary conditions, temperature, etc.

In the present Co₉Zn₉Mn₂ thin plate, we thus established the skyrmions’ life cycle showing the successive disorder-order transformations. It is notable that the self-organization to the higher-symmetric skyrmion cluster is not instantaneously realized by the thermal excitation. Instead, the skyrmion-reshaping processes occur and then they start moving as particles, leading to the skyrmion cluster formation in a longer time scale. Such multiscale spatiotemporal structure akin to soft matters (15, 16) is reasonably understood by the retarded responses related to the damping of internal spins and directly reflects the hierarchy of internal microscopic spin magnetic moments and mesoscopic particle nature. The weakened but still persistent topological protection further adds the microsecond lifetime of the skyrmions. These results provide good initial insights into the kinetics of the flexible skyrmions around the imperfections, which inevitably exist in real materials and may limit the performance of the skyrmion-based spintronic devices (13, 14). Further pump–probe LTEM experiments at lower fluences for investigating elastic dynamics of magnetic textures as well as the possible repeatable transformation between helical stripes and skyrmions will be important future works. The present results visualize the unique life cycle of the photothermally induced metastable skyrmions and also propose previously unidentified concept for skyrmion-based spintronic applications based on the repeatable creation, annihilation, and transportation of topological charge, such as high-frequency undulation of the emergent magnetic field in nanoscale spintronic circuit.

MATERIALS AND METHODS

Sample preparation

Bulk Co₉Zn₉Mn₂ single crystals were synthesized using procedures described elsewhere (25, 26). The thin plates for the LTEM observations were prepared from the bulk sample by using dual-beam FIB instrument (Hitachi, FB-5000). After the thinning process, we further irradiated the thin plate with Ga ion (acceleration voltage of 40 kV) at a grazing angle about 2° to randomly introduce local crystal defects.

Pump-probe LTEM

Pump-probe LTEM measurements were performed at RIKEN Center for Emergent Matter Science (Thermo Fisher Scientific, Tecnai Femto). For the excitation of the sample, we used the laser pulse (1064 nm and 1-ns duration) delivered from a Q-switched laser operating at 20-kHz repetition (Bright Solutions, Wedge-HF). Its diameter at the sample position was set to 100 μm. For generating the electron packets, we used the laser pulse up-converted to 266 nm (10-ns duration) delivered from a Q-switched laser (Advanced Optowave, AWave-532). In this work, we obtained three datasets by pump-probe LTEM for the same sample. We used the dataset 1 (from ~20 to 24 ns) for Figs. 2 (E1 to E3) and 3 (A to I), dataset 2 (from ~430 to 3970 ns) for Figs. 2 (C, D, and E to E7), 3 (H and I), and 4 (A, B, and D) and figs. S2 and S3, and the dataset 3 (from ~1280 to 15870 ns) for Figs. 2E8 and 4 (A, B, and E) to hand the fig. S4. We integrated the pulsed electrons for 30 min to obtain an LTEM image at a certain delay t.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/7/25/eabg1322/DC1

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