Mechanisms of Skyrmion and Skyrmion Crystal Formation from the Conical Phase

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Abstract
Real-space topological magnetic structures such as skyrmions and merons are promising candidates for information storage and transport. However, the microscopic mechanisms that control their formation and evolution are still unclear. Here, using in situ Lorentz transmission electron microscopy, we demonstrate that skyrmion crystals (SkXs) can nucleate, grow, and evolve from the conical phase in the same ways that real nanocrystals form from vapors or solutions. More intriguingly, individual skyrmions can also “reproduce” by division in a mitosis-like process that allows them to annihilate SkX lattice imperfections, which is not available to crystals made of mass-conserving particles. Combined string method and micromagnetic calculations show that competition between repulsive and attractive interactions between skyrmions governs particle-like SkX growth, but nonconservative SkX growth appears to be defect mediated. Our results provide insights toward manipulating magnetic topological states by applying established crystal growth theory, adapted to account for the new process of skyrmion mitosis.

Keywords
Magnetic skyrmion, skyrmion crystal, nucleation, growth mechanism, Lorentz-TEM

Disciplines
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Mechanisms of Skyrmion and Skyrmion Crystal Formation from the Conical Phase

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Abstract

Real-space topological magnetic structures such as skyrmions and merons are promising candidates for information storage and transport. However, the microscopic mechanisms that control their formation and evolution are still unclear. Here, using in-situ Lorentz transmission electron microscopy, we demonstrate that skyrmion crystals (SkXs) can nucleate, grow, and evolve from the conical phase in the same ways that real nanocrystals form from vapors or solutions. More intriguingly, individual skyrmions can also “reproduce” by division in a mitosis-like process that allows them to annihilate SkX lattice imperfections, which is not available to crystals made of mass-conserving particles. Combined string method and micromagnetic calculations show that competition between repulsive and attractive interactions between skyrmions governs particle-like SkX growth, but non-conservative SkX growth appears to be defect-mediated. Our results provide insights towards manipulating magnetic topological states by applying established crystal growth theory, adapted to account for the new process of skyrmion mitosis.

Keywords: Magnetic skyrmion, Skyrmion crystal, Nucleation, Growth mechanism, Lorentz-TEM,
Crystal formation mechanisms play critical roles in controlling materials microstructures. Advances in in situ microscopy techniques have contributed significantly to our understanding of the interplay between the thermodynamics and kinetics of the processes of crystallization and crystal growth\textsuperscript{1–5}. It is now widely accepted that the growth of a crystal proceeds either through monomer-by-monomer addition (MA) or by particle attachment (PA), or both mechanisms concurrently\textsuperscript{2}. The formation of quasiparticle assemblies, such as skyrmion crystals (SkXs), presents new opportunities to explore phenomena that are not reflected in the growth of crystals made of real particles such as atoms or molecules.

A skyrmion is a nanoscale vortex-like spin structure with topological charge\textsuperscript{6–10}, and magnetic skyrmions are promising information carriers for spintronic applications\textsuperscript{11}. Different mechanisms, such as: magnetic dipolar interaction, Dzialoshinskii-Moriya interaction (DMI), frustrated exchange interaction, and four-spin exchange interactions, have been proposed for their formation\textsuperscript{12–14}. In particular, the skyrmions that generated by DMI has been widely studied, including Néel-type skyrmion, Bloch-type skyrmion et. al\textsuperscript{15,16}. The discovery of room temperature skyrmion in Co\textsubscript{x}Zn\textsubscript{y}Mn\textsubscript{z} (x + y + z = 20) alloys (space group $P4_132$ or $P4_332$) provided an important step towards applications\textsuperscript{17}. Recent studies in this system have revealed zero-field metastable skyrmion\textsuperscript{15,18}, magnetic modulation period and a critical temperature change with different Mn concentration\textsuperscript{17}, a spin-liquid state\textsuperscript{19}, and a spin-glass state in $3 \leq z \leq 7$ region\textsuperscript{20,21}.

In general, an external magnetic field perpendicular to the wavevector of the helical structure may induce a conical state, followed by formation of SkXs\textsuperscript{16,22}. Skyrmions and SkXs form at thermodynamic equilibrium, depending on external magnetic fields, electric currents and temperature\textsuperscript{12,15,16,23–27}. The equivalents of crystal lattice defects such as grain boundaries and dislocations have been observed in SkXs\textsuperscript{28–30}. 

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Despite extensive imaging of SkXs \cite{10,27,31} and a variety of proposed mechanisms for individual skyrmion formation \cite{7,14,32}, real-space observation of skyrmion winding from matrix phases remains challenging, due to the narrow range of temperature and external magnetic field over which the transition occurs. In this study, we have achieved real-time monitoring of SkX nucleation in $\beta$-Mn-type Co$_8$Zn$_{10}$Mn$_2$ thin films using Lorentz transmission electron microscopy (LTEM). We discover that the formation of SkX includes two processes that overlap in time. The first is nucleation of skyrmions by coordination of spin, which proceeds through an intermediate state. The second is growth of SkXs, which follows a nucleation and growth mechanism of typical first-order phase transformation, by the addition of subsequent skyrmions. SkXs grow through the addition of individual skyrmions on their peripheries, the addition of skyrmion clusters, and by a new non-conservative growth mechanism of internal skyrmion splitting. Micromagnetic simulation together with string method reveals that the inter-skyrmion attractive forces promote the formation of skyrmion clusters, while the skyrmion self-splitting (SS) process is due to anisotropic stretching forces from its miscoordinated neighbors.

**Results and Discussion**

**Nucleation of skyrmion and SkX**

Nucleation of the first skyrmion is a crucial step toward SkX formation and represents transformation from the conical phase with a corresponding change of topological number from zero to one. It is also the most challenging process to monitor. We have overcome the experimental difficulty by slowing down the kinetics using precisely controlled temperature, magnetic field, and a rapid camera speed of 50 ms per frame. First, we systematically examined the temperature and external magnetic field dependence of different thermal-equilibrium states. A phase diagram with a contour plot of skyrmion phase density based on LTEM observation (fig. S1) shows that
skyrmions grow from the conical phase above 80°C at an external field higher than 100 mT in a ~200 nm thick (110) plate (fig. S2). Accordingly, the sample was heated up quickly to 84°C under 135 mT so that the conical-to-skyrmion transition proceeded through a first-order process. The helical phase observed at room temperature under zero field (Fig. 1A) transformed into a single conical phase upon reaching the targeted experimental condition (Fig. 1B). A skyrmion embryo, with about 60% of the contrast of a fully-formed skyrmion, appeared after 1.85 s of incubation time (Fig. 1C), developing into a full-contrast skyrmion with a slight shift from the initial position at 2.6 s (Fig. 1D). Both the skyrmion embryo and the skyrmion were mobile, undergoing random movements akin to Brownian motion.

The region around the first skyrmion then acted as a preferred nucleation site for a second skyrmion, and a two-skyrmion complex was formed with a specific separation after 5.75 s (Fig. 1E). Subsequent skyrmions grew at a faster rate (Figs. 1F-H), possibly due to formation of more skyrmion-skyrmion bonds. Skyrmion trimer forms with a triangular structure and skyrmion tetramer takes the shape of a rhombus. The oligomers grow to clusters with further addition of skyrmions by MA mechanism. As with the first skyrmion, contrast evolution was observed for the third and fourth ones, as shown in intensity profile comparison (Fig. 1O, see fig. S3 for time-resolved evolution). Because all Lorentz images were taken under the same defocus condition, the contrast transformation of the same “particle” is approximately proportional to the projected magnetization change along the electron beam direction, suggesting the formation of intermediate states, during nucleation of skyrmion, similar to skyrmion lattice decay.

**Propagation of SkX through MA and PA mechanisms**

When there were more than two skyrmions in the cluster, SkX lattice is observed to rotate and translate, along with the formation and addition of more skyrmions (Figs. 1H-K). The growth
of the SkX cluster results in pronounced facet development (Fig. 1M-N), corresponding to \{100\} of the hexagonal system. The initial skyrmion cluster develops by the formation of single skyrmions at or very close to their coordination sites, in what is essentially a MA mechanism (Fig. 1D-N), as described in classical crystal growth theory. The lattice parameter, i.e., the core to core distance between adjacent skyrmions, was almost constant (138 ± 5 nm) after adding additional skyrmions. Movie S1 shows the nucleation and growth of this cluster.

“Particle” attachment events were observed when two growing skyrmion clusters were close to each other. The merging of two clusters typically includes two processes: connection and lattice reorientation. Depending on the cluster size, they occur either concurrently or sequentially (movie S2). When both clusters are small, the skyrmion lattices rotate rapidly, and they rotate to a small misorientation during attachment (Figs. 2A-C). The misorientation angle between two clusters reduced from 26° to 3.5° within 50 ms when the slightly smaller cluster jumped into the larger one which was initially ~400 nm away (Figs. 2B-C). If a small cluster merges into a much larger one, attachment occurs first, forming a grain boundary in the SkX, followed by reorientation of the smaller grain. In the case shown in fig. S4, the initial misorientation angle of 21° was reduced to 6°, 2.1 s after the attachment. These types of cluster attachment are very similar to nanoparticle agglomeration, via the so-called oriented attachment mechanism, which includes three-dimensional rotation to reduce interparticle misalignment, and atom-by-atom reorientation after attachment through dislocation or grain boundary migration. When both skyrmion grains are large and immobile, merging typically starts with the formation of a bridge between them, followed by skyrmion growth and lattice rearrangement (movie S3). As shown in Figs. 2D-E, a bridge with two skyrmion lattice planes was first created when the grains were ~300 nm apart. Subsequently, skyrmions grew around the high-curvature regions of the bridge than growing near
the flat facet (Fig. 2F). At the same time, SkX rearrangement was accompanied, and slight misorientation reduction (from 7.8° to 5.4°) was observed. SkX thus also grows through the PA pathway.

Dislocation annihilation through SS mechanism

Lattice defects can be formed either at the surface or inside the SkX. Surface kinks with fivefold coordination provide preferred sites for MA skyrmion growth (fig. S5, movie S4), which is the major pathway for SkX propagation. The defects inside the SkX such as edge dislocations, formed either at grain boundary by the merger of SkX particles or by structural relaxation, are geometrically observed as adjacent fivefold and sevenfold-coordinated skyrmions (5-7 defect, Fig. 3A), which can be described equivalently as wedge disclination dipoles. This type of imperfect skyrmion coordination is able to evolve by SS mechanism that is not possible in a crystal made of real molecules (movie S5): a skyrmion with seven neighbors (Fig. 3A) rapidly divides into two skyrmions (Fig. 3B). The intensity evolution of the central skyrmion clearly demonstrates SS process (fig. S6). As a result, the position of the disclination dipole or edge dislocation shifts closer to the cluster edge, like a dislocation climbing by the attachment of an interstitial atom. The newly created skyrmion triggered local SkX lattice rearrangement (Fig. 3C), just as an edge dislocation climbing in a grain boundary toward the surface will reduce the misorientation of two crystals. In this case, however, the climb is facilitated by the spontaneous creation of a new crystal lattice quasi-particle rather than the addition or subtraction of an atom by diffusion from or to the surface.

Structural relaxation

SkX lattice rearrangement continues after skyrmions fully replace the conical phase. Splitting of the FFT spots was clearly visible right after the phase transition completed, implying the existence of differently oriented domains (fig. S7). Single domain SkX was formed 30s later,
as confirmed by the disappearance of the satellite FFT spots (movie S6). This process proceeds through grain boundary defect reduction by internal skyrmion growth or rearrangement\textsuperscript{28,30}.

**Interaction force between asymmetric skyrmions**

The skyrmion-skyrmion interaction is dependent on the surrounding parental state\textsuperscript{32,38–40}. The skyrmion in the conical phase exhibits asymmetric screw-like modulation along the skyrmion core\textsuperscript{41}. The interaction force that controls crystallization of SkX from the conical phase is directly calculated by gradually bringing two well-separated skyrmions together using a combined micromagnetic and string method (supplementary 2.1-3), which is capable of searching the lowest transition path between two states. Fig. 4A shows the calculated energy profiles as a function of distance between two skyrmions separated in $x$ or $y$ direction with different sample thicknesses. All interaction profiles exhibit minima similar to the repulsive core and attractive tail Lennard-Jones functions. The attractive part is driven by reducing of cone-skyrmion interface energy. As shown in Fig. 4B, isolated skyrmions are twisted by the conical phase along $z$ direction, which results in high cone-skyrmion interfacial energy\textsuperscript{33}. When two skyrmions touch and form a skyrmion-skyrmion interface, both of them are less twisted. At the same time, the cone-skyrmion interface energy is minimized (Figs. 4C-E). Note that, due to the spiral nature of the conical phase, the interaction is orientation dependent as well. Thus, a slight difference between the energy profiles along the $x$ and $y$ direction is observed when the film thickness decreases from $3.8L_D$ to $1.9L_D$, where $L_D$ is the periodicity of the helical phase. Particles or quasi-particles in 2D systems with this kind of interaction favor clusters with hexagonal coordination\textsuperscript{42}, similar to ferrofluid, colloidal systems, and vortices in MgB$_2$ or low-$\kappa$ superconductor\textsuperscript{43}, except that the skyrmion system belongs to the hardcore interaction. Similarly, merging of separated clusters reduced cone/skyrmion interfacial energy and promotes cluster attachment, as observed in Fig. 2.
Micromagnetic simulation of SS process

To study SS process similar to our experiment, we use a combined micromagnetic and string method to search for the lowest energy transition path for forming a new skyrmion at 5-7 defect. Our calculation shows that SS is the energetically favored path. We create an initial state with a cluster containing 5-7 defect (Fig. 5B), mimic our experiment. Note that, the sevenfold-coordinated skyrmions is elongated due to anisotropic stretching forces from its miscoordinated neighbors. We then replace this skyrmion with two skyrmions and relax the system to get another cluster, which is the target state after a new skyrmion is added. After that, we use our string method to search for the lowest energy transition pathway between these two clusters.

Fig. 5A shows the evolution of energy and skyrmion number $N_s = \frac{1}{4\pi} \int \int m \cdot (\frac{\partial m}{\partial x} \times \frac{\partial m}{\partial y}) dxdy$, where $m$ is a unit vector parallel to magnetization direction during SS. Representative spin configurations during the splitting process are shown in Fig. 5B-D. The SS starts at the surface of the sample by creating a magnetic monopole (Fig. 5D). The monopole then moves toward the opposite surface, and unzip the skyrmion into two (Figs. 5E-F). This process has some similarity to the skyrmion unwinding procedure described in Ref. 44. However, the SS is triggered by imbalanced force from defected skyrmion lattice, while skyrmion merging is due to decreasing of magnetic field (details in supplementary 2.4).

Conclusions

In summary, the nucleation and growth of skyrmion and SkX from the conical phase in β-Mn-type Co$_8$Zn$_{10}$Mn$_2$ thin films are directly observed by precise field and temperature control. Skyrmions grow through intermediate states before penetrating through the whole film. Analogous to crystallization in molecular systems, both MA and PA mechanisms are clearly demonstrated.
Moreover, skyrmion addition can occur inside a growing SkX by a SS mechanism that has no analog in molecular crystals. Micromagnetic simulation combined with string method demonstrates that the crystal-like SkX formation is driven by a Lennard-Jones like skyrmion-skyrmion interaction forces. The SS of a skyrmion at 5-7 defect is energetically favorable and proceed by creating a magnetic monopole near the surface, and then unzips the skyrmion into two. Our discovery provides an essential step toward understanding and manipulating the evolution of magnetic topological states for applications in quantum information technologies.

**Materials and Methods**

The bulk Co\textsubscript{8}Zn\textsubscript{10}Mn\textsubscript{2} sample with critical temperature T\textsubscript{c} \sim 370 K was prepared by first sealing individual metals (all > 99.9% metals basis) in a quartz ampoule backfilled with ultra-high purity argon. The ampoule was placed into a furnace and heated to 1000 °C for 12 hours, then cooled at 1 °C/hr to 925 °C and held for 96 hours before quenching into water. Magnetic measurements were performed using a Quantum Design VersaLab\textsuperscript{TM} vibrating sample magnetometer. A thin, polycrystalline piece was polished so the sample dimensions were greater than 5:1 aspect ratio. The sample was field cooled at a rate of 2 K/min under an applied field of H = 20 Oe. The polycrystalline sample shows a Curie temperature of 362 K and magnetization of 0.17 µB/f.u. under an applied field of H = 20 Oe.

A Co\textsubscript{8}Zn\textsubscript{10}Mn\textsubscript{2} (110) thin plate was fabricated using focused ion beam system (FIB, FEI Helios NanoLab G3). The crystal orientation was verified by electron backscattered diffraction analysis before lift-out. The plate was approximately 200 nm thick. The thickness map of the sample (fig. S2) was obtained by electron energy loss spectroscopy with Gatan Quantum ER 965.
In-situ Lorentz transmission electron microscopy (LTEM) observation was conducted on an FEI Titan Themis by using FEI NanoEx-Tm-i/v in-situ TEM holder which enabled rapid heating and precise temperature control under isothermal conditions. The external magnetic field was applied along the electron beam direction by partially exciting the objective lens. LTEM videos were taken at 20 fps by FEI Ceta camera to record skyrmion dynamics. The number of skyrmions was counted using Image J. The in-plane magnetization maps of the magnetic structure were obtained by the LTEM Fresnel images with a phase-retrieval QPt software on the basis of the transport of intensity equation\textsuperscript{45}.

Supplementary Materials:

Section 1. Supplementary figures and videos
Figures S1-S7, Movies S1-S6

Section 2. Simulation details
Figure S8-S9, Movies S7-S9
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Figures

Fig. 1 Skyrmion and SkX nucleation. (A) Helical phase with periodicity of ~115 nm at room temperature under 0 mT. (B-N) Sequence of LTEM images from movie S1. Time indicates time progress after sample reached 84 °C under 135 mT. (B) Conical phase. (C) Skyrmion embryo with weak contrast. (D) Formation of an isolated skyrmion. Note the skyrmion position change from C to D. (E-H) SkX growth through MA with obvious intensity evolution of each skyrmion. (I-L) SkX growth accompanied with rearrangement. (M, N) Cluster of 9 and 23 skyrmions with facet. (O) Comparison of intensity profiles obtained from blue line in (G) and red line in (H).
**Fig. 2 Coalescence by oriented attachment and relaxation of SkX.** A-C and D-F are sequences of LTEM images from movie S2 and S3, taken at 88°C under 100 mT, respectively. (A) Two small SkX clusters. Insets show FFT obtained from each cluster. White and red arrows in FFT correspond to planes indicated by white and red lines in LTEM images. (B) Two clusters approached close to 400 nm and the misorientation angle changed to 26° by oscillation. (C) A small cluster jumped and merged into a larger cluster within 0.05 s and misorientation angle reduced to 3.5° simultaneously. The misorientation angle was measured using the rotation angle between the corresponding FFT patterns. (D) Two large clusters are ~300 nm apart. (E) Two clusters were attached by a bridge formation. (F) Skyrmions grow around the bridge. Misorientation was slightly reduced.
Fig. 3 Self-splitting (SS) mechanisms of SkX growth. Sequences of LTEM images from movie S5, taken at 88°C under 100 mT, with schematic illustrations. Black circles indicate skyrmion positions in corresponding LTEM images. (A) Fivefold (pentagon) and sevenfold (heptagon) disclination inside the cluster. The green circle shows the center skyrmion of sevenfold disclination. (B) Splitting of the center skyrmion into two skyrmions (red). Hexagonal lattice (hexagon) was formed by splitting. The initial disclinations climbed closer to the surface of the cluster (the right side). (C) SkX lattice rearrangement to hexagonal lattice. Unfilled green circles are SkX lattice position of (B) before rearrangement and filled black circles with green outlines are after rearrangement.
Fig. 4. Interaction force and magnetic structure of two skyrmions. (A) The energy density $E$ as a function of inter-skyrmion distance $r$ for separation in $x$ ($e_x$) or $y$ ($e_y$) direction. $E$ is calculated with respect to the conical phase ($E_{\text{cone}}$) and single skyrmion ($E_{\text{IS}}$) energy under an external field $H$ of $H/H_D = 0.5$, where $H_D$ is the saturation field. The $E$ exhibits a decrease-increase Lennard-Jones like behavior with a minimal value at a distance slightly larger than the conical phase periodicity ($L_D$). (B-E) The magnetic structure of two interacting skyrmions marked by $b$-$e$ in (A). The white and color streamlines in (B-E) are surfaces with $z$ component of the magnetization $m_z = -0.4M_s$ and $m_z = 0.9M_s$, respectively, where $M_s$ is the saturation magnetization. The color indicates $x$ component of the magnetization $m_x$. 
Fig. 5. Energy profile and magnetic configuration during SS of a skyrmion at 5-7 defect. (A)
The energy density $E$ (left coordinate) and skyrmion number ($N_{sk}$) as a function of image number.
$E$ is calculated with respect to the conical phase ($E_{cone}$) and initial state ($E_0$) energy under an
external field ($H$) of $H/H_D = 0.5$, where $H_D$ is the saturation field. $N_{sk}$ is averaged along z direction.
(B) Top view of the SkX with a 5-7 defect used in calculation. The SS skyrmion is marked by
green arrow. Fivefold (white line) and sevenfold (black line) disclination is formed similar to
experiment. (C) 2D view of magnetic structure of the splitting skyrmion (marked by white circle)
with a magnetic monopole in the middle. (D-F) 3D view of magnetic structure of a SS skyrmion
conforming to images marked by d-f in (A), respectively. The color plots in (B-F) indicate the
$z$ component of the magnetization $m_z$; arrows in (C-F) are direction of spins; Streamline in (D-F)
is a surface with $m_z = 0.2$. 