Triangular-to-Square Lattice Transformation of Metastable Skyrmions

A magnetic skyrmion is a topologically protected particle-like object and usually crystallizes into a close-packed triangular lattice. In contrast with the case of conventional atoms or molecules, the size and shape of skyrmions can be flexibly tuned by an external magnetic field. In this study, we have discovered that such a magnetic-field-induced size/shape change of a skyrmion particle induces the triangular-to-square structural transition of the skyrmion lattice in a chiral cubic magnet, Cu_2OSeO_3, by means of small-angle resonant soft X-ray scattering. Our results demonstrate that the size/shape degree of freedom of skyrmion particles is an important factor in determining their stable lattice form.

Topological solitons, i.e., stable defect structures that cannot be erased by continuous transformation, have attracted much attention as novel building blocks of exotic ordered phases. One typical example is a magnetic skyrmion with nanometric swirling spin texture, which behaves as a topologically protected particle. Such particle-like objects are expected to form a periodic lattice in a similar manner to atomic or molecular crystals; a better understanding of their crystallization and structural transition process is greatly needed.

Magnetic skyrmions usually crystallize into a close-packed triangular-lattice form [Fig. 1(a)] in the equilibrium condition. Recent experiments have shown that the triangular-to-square transformation of the skyrmion lattice (SkL) can be induced by a magnetic field in the non-equilibrium condition [1, 2], although its detailed mechanism remains an open question. In this study, we investigated the microscopic origin of such a symmetry change of SkL in a chiral-lattice insulator, Cu_2OSeO_3 [3], by means of a small-angle resonant soft X-ray scattering (RSXS) experiment [4].

A bulk single crystal of Cu_2OSeO_3 was grown by the chemical vapor transport method. A thin plate of Cu_2OSeO_3 with a thickness of about 800 nm was fabricated by using the focused ion beam (FIB) technique. To block the transmission beam, the backside of the Si_3N_4 membrane window was covered with gold film, and a pinhole of ~6 μm in diameter was drilled. The sample was mounted to cover the pinhole and fixed to the membrane with single tungsten contact to avoid tensile strain. The RSXS measurement was performed on BL-16A using circularly polarized X-ray with the resonance energy of 931 eV at the Cu L_3 edge. A magnetic field was applied parallel to the incident X-ray beam (|| [001]) by a Helmholtz coil.

Figure 1 summarizes the magnetic-field (B) variation of the quenched metastable SkL state, which is stabilized by field cooling (FC) passing through the equilibrium SkL phase. Just after the FC process, a six-spot diffraction pattern was observed [Fig. 1(e)], which indicates the realization of the metastable triangular SkL characterized by the three Q-vectors within a plane perpendicular to the B direction. In the B-increasing process, the metastable triangular SkL state with the six-spot pattern survives [Fig. 1(d)] just before entering the conical phase, where the Q-vector is aligned parallel to the magnetic field and diffraction spots disappear. In the B-decreasing process, the six-spot pattern remains even down to negative fields and suddenly changes into a four-spot pattern at –40 mT [Fig. 1(f)]. In another field-sweeping process, we also observed a reversible change between the six-spot and four-spot patterns, and thus the observed four-spot pattern is assigned to a square SkL state [Fig. 1(b)], endowed with the topological charge.

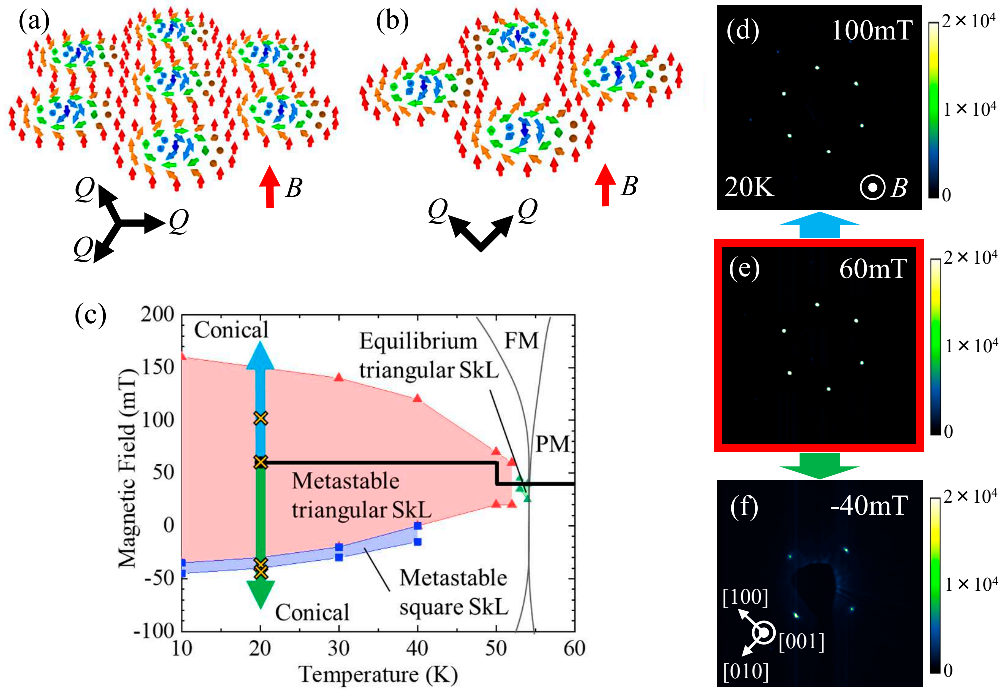

Figure 1: (a), (b) Schematics of triangular (a) and square (b) SkL. (c) Magnetic field-temperature phase diagram determined from the field-sweeping scans after the FC process shown by the arrows. (d)–(f) RSXS diffraction patterns in the field-sweeping process after FC.

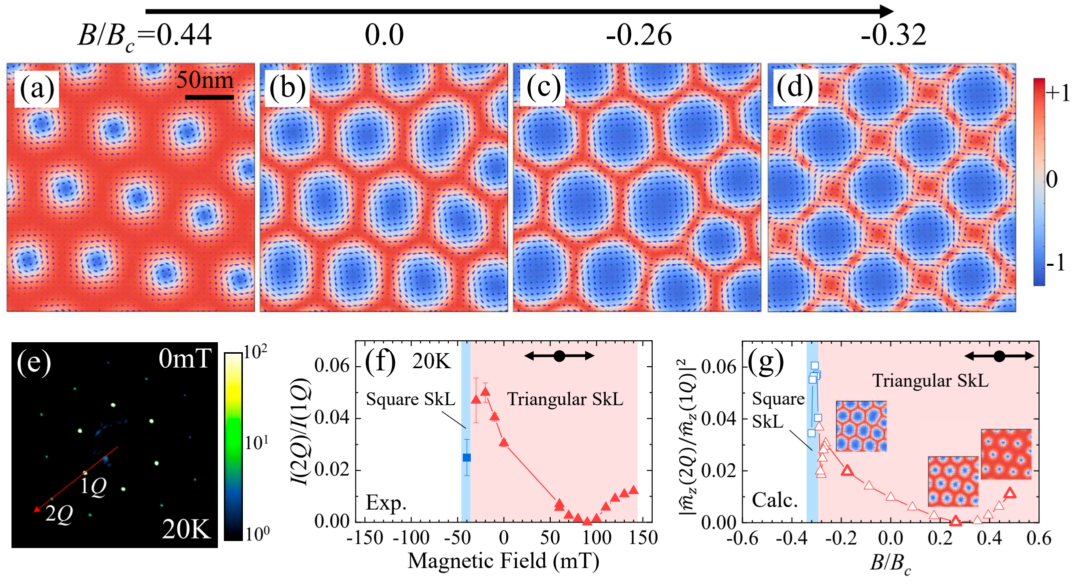

Figure 2: (a)–(d) Real-space distribution of local magnetization in the field-decreasing process. The arrows correspond to the in-plane component of local magnetization and the background color represents the out-of-plane component of local magnetization. (e) RSXS diffraction pattern for the metastable triangular SkL after FC (20 K, 0 mT). (f), (g) Magnetic field dependences of $I(2Q)/I(1Q)$ evaluated from the RSXS result (f) and calculated from the results of micromagnetic simulations (g). The spin textures for the selected B-value are shown in the inset.

To investigate the microscopic origin of lattice transformation of SkL, we performed micromagnetic simulations based on the Landau–Lifshitz–Gilbert (LLG) equation by using MuMax3 software. By decreasing the B value, the diameter of the skyrmion particle core gradually expands and the intervening region between skyrmions squeezes, which finally triggers the triangular-to-square lattice transformation of metastable skyrmions [Fig. 2(a)–(d)] to minimize the energy cost at the skyrmion–skyrmion interface region. To confirm such B-induced change of skyrmion core size, we calculated the relative amplitude of the higher harmonics in the magnetic modulation. As shown in Fig. 2(e), the second harmonic magnetic reflections (2Q) are clearly observed in the metastable triangular SkL state; we evaluated $I(2Q)/I(1Q)$, i.e., the integrated intensity of 2Q reflection normalized by the 1Q one. As seen in Fig. 2(f), $I(2Q)/I(1Q)$ reaches the minimum at around +90 mT, where the skyrmion core diameter (d) is half of the core-to-core distance (a) and the magnetic modulation is almost sinusoidal. By changing the B-value from the minimum, d/a deviates from 1/2 and the 2Q intensity is increased. Such B-dependence is well reproduced by the micromagnetic simulations [Fig. 2(g)], which suggests that the observed transformation of SkL is triggered by the B-induced change of skyrmion core diameter.

The above results demonstrate that the flexible size/shape of skyrmion particles and its sensitivity to a magnetic field plays a crucial role in the transition of lattice structure. The observed phase transition process is rather distinct from the conventional atomic crystal cases, suggesting that the topological solitons with flexible size/shape degree of freedom may ubiquitously host nontrivial manner of crystallization and field-induced structural transition. Our present finding will contribute to a better understanding of the ordering process of such topological soliton ensembles, and may trigger further searches for unique hierarchical organizations in various physical contexts.

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