Observation of Nanometric Square Skyrmion Lattice in Centrosymmetric Tetragonal Magnet GdRu$_2$Si$_2$

Topological objects in condensed matter have attracted much attention as a source of rich emergent phenomena and functions. One typical example is the magnetic skyrmion, a vortex-like swirling spin texture with stable particle nature in magnetic materials. In this work, we report the discovery of a square skyrmion lattice state in the centrosymmetric tetragonal magnet GdRu$_2$Si$_2$ via resonant X-ray scattering and Lorentz transmission electron microscopy experiments. The results establish a new route to stabilize skyrmions even without geometrical frustration or inversion symmetry-breaking, and suggest a new platform for designing topological spin textures in single-component systems.

Topological objects in condensed matter have attracted much attention as a source of rich emergent phenomena and functions. A typical example is the magnetic skyrmion, a vortex-like swirling spin texture with stable particle nature in magnetic materials [Fig. 1(A)]. Because of its small size and unique electromagnetic response, the skyrmion is now being intensively studied as a novel information carrier for high-density data storage devices [1].

Previously, skyrmions have been mostly found in noncentrosymmetric systems with Dzyaloshinskii–Moriya (DM) interaction. However, the latest theories suggest that skyrmions can also be stabilized in centrosymmetric systems by different microscopic mechanisms, although their experimental verification in real materials has rarely been achieved. Very recently, the emergence of a hexagonal skyrmion lattice was discovered in the centrosymmetric frustrated magnets Gd$_2$PdSi$_3$ and Gd$_3$Ru$_4$Al$_{12}$, where geometrical frustration was considered to be the key for skyrmion formation [2, 3]. Nevertheless, it remains an open question whether such geometrical frustration or inversion symmetry-breaking is indispensable for stabilizing skyrmions or not.

Here, we report the experimental discovery of a square skyrmion lattice in the centrosymmetric tetragonal magnet GdRu$_2$Si$_2$ without the presence of geometrical frustration. By performing detailed magnetic structure analysis based on polarized resonant soft X-ray scattering and Lorentz transmission electron microscopy techniques, the emergence of the square skyrmion lattice state has been clearly established. Notably, the observed skyrmion size is as small as 1.9 nm, which is the smallest value ever reported for single-component materials. The origin of the observed skyrmion formation is discussed in terms of the recently proposed four-spin interaction mechanism mediated by itinerant electrons [4].

The target material GdRu$_2$Si$_2$ crystallized in a typical ThCr$_2$Si$_2$-type structure with centrosymmetric tetragonal space group I4/mmm. The crystal structure consists of alternate stacking of square Gd layers and Ru$_2$Si$_2$ layers as shown in Fig. 1(B). The magnetism is governed by Gd^{3+} ($S = 7/2$, $L = 0$) ions with Heisenberg magnetic moment. Previous works revealed that GdRu$_2$Si$_2$ exhibits incommensurate magnetic order below $T_N \sim 46$ K, with magnetic modulation vector $\boldsymbol{Q} = (0.22, 0, 0)$ confined within the tetragonal basal plane. By applying an external magnetic field $\boldsymbol{B}$ along the tetragonal axis [001], various magnetic phase transitions can be observed [Fig. 1(C)], although the detailed spin texture of each phase has not been elucidated. Interestingly, in the intermediate range of magnetic field (phase II) characterized by multiple steps in magnetization as well as anomalies in magneto-transport behavior, the enhancement of Hall resistivity may indicate the formation of the topological spin texture.

To investigate the detailed magnetic structure, we carried out magnetic X-ray scattering experiments in resonance with the Gd L_2 edge at 5 K. By exploring the magnetic satellite peaks around the nuclear Bragg reflection, the magnetic modulation vector $\boldsymbol{Q}$ in each phase can be directly investigated. Due to the tetragonal nature of the crystal structure, the appearance of equivalent magnetic modulation vectors $\boldsymbol{Q}_1 = (q, 0, 0)$ and $\boldsymbol{Q}_2 = (0, q, 0)$ was observed. Interestingly, only in phase II, we successfully identified superlattice reflection $\boldsymbol{Q}_1 + \boldsymbol{Q}_2$ modulations, which are allowed to appear only when the double-$\boldsymbol{Q}$ magnetic state is formed [Fig. 2(B)]. Therefore, we conclude that the magnetic structure of phase II is a double-$\boldsymbol{Q}$ state [Fig. 2(A)]. Further investigation of the polarization of scattered X-rays suggests that the spin texture in phase II can be described by the superposition of two screw spin textures with orthogonal magnetic modulation vectors, or a Bloch-type square skyrmion lattice. The spin texture can be more directly confirmed by real-space imaging with Lorentz transmission electron microscopy (LTEM), which clearly visualizes the square-lattice-like magnetic superstructure with a period of 1.9 nm formed on the atomic lattice [Fig. 2(C, D)].

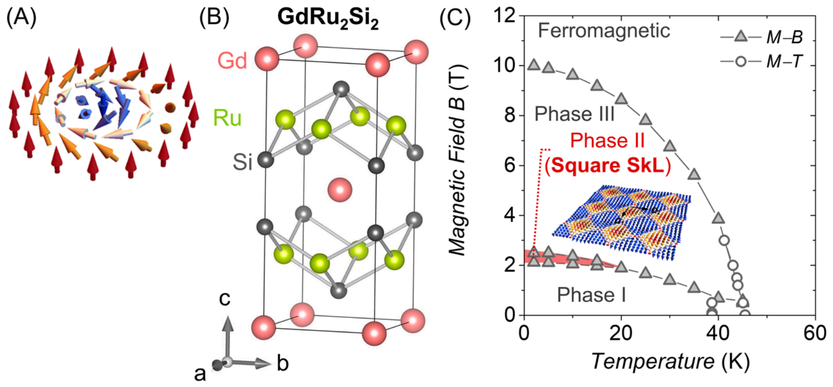

Figure 1: (A) Schematic illustration of magnetic skyrmions. (B) Crystal structure of GdRu$_2$Si$_2$. (C) Magnetic field – Temperature phase diagram for B // [001]. The inset shows the square lattice of skyrmions.

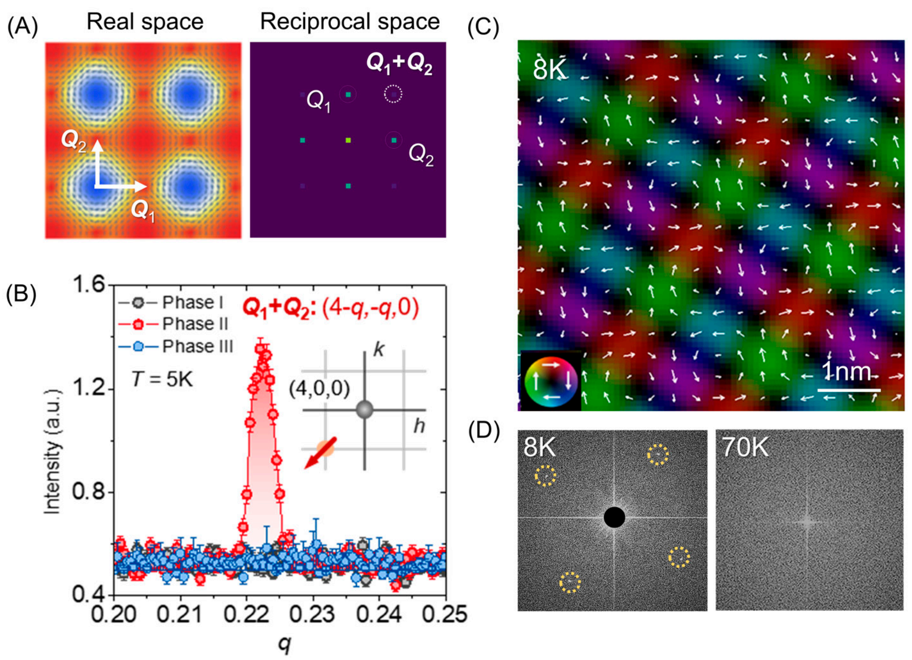

Figure 2: (A) Illustration of square lattice of skyrmions obtained by super-positioning two screw modulations in real space and corresponding calculated Fourier transform pattern. (B) The line scan profile along the (4-q,-q, 0) scan corresponds to the $Q_1 + Q_2$ magnetic satellite peaks around the fundamental Bragg spot (4, 0, 0) in B = 0T (phase I), B = 2.1T (phase II) and B = 3T (phase III). (C) Lateral magnetization distribution obtained from analysis of the L-TEM images. White arrows show the direction of in-plane magnetization and the background color stands for the direction (hue) and relative magnitude (brightness) of the lateral magnetization. (D) Fast Fourier Transform pattern of spin texture in (C) at low temperature T = 8K and high temperature T = 70K.

Since the crystal structure of GdRu$_2$Si$_2$ is centrosymmetric, the contribution from the DM interaction is not relevant in this case. Moreover, GdRu$_2$Si$_2$ is characterized by a tetragonal crystal lattice and its Curie–Weiss temperature almost coincides with the magnetic ordering temperature, which excludes geometrical frustration as the driving force behind skyrmion formation. Therefore, the present case of a square skyrmion lattice can potentially be ascribed to the four-spin interaction mediated by itinerant electrons [5, 6]. The present results establish that skyrmion formation is possible even without geometrical frustration or inversion symmetry-breaking. This highlights four-spin interaction as a new route to realize skyrmions with extremely small size, and will dramatically expand the candidate materials to host skyrmions. Our finding will promote further investigations of similar highly-symmetric rare-earth intermetallics as a unique material platform to realize nanometric skyrmions.

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