Magnetic Skyrmion Lattice in Gd-Based Centrosymmetric Magnets

Magnetic skyrmions, nanometric spin defects carrying a topological winding number, have mostly been explored in noncentrosymmetric magnets. Two papers published in Science and Nature Communications last year have expanded the research fields for skyrmion hunts to centrosymmetric magnets supported by magnetic frustration. The compounds featured in the studies are Gd-based Gd_2PdSi_3 and $Gd_3Ru_4Al_{12}$, which are characterized by two archetypical geometrically frustrated lattices, triangular and (distorted) kagome lattice, respectively. Polarization analysis of resonant X-ray scattering has revealed the triangular lattice of skyrmions in both centrosymmetric magnets. These results afford proof-of-principle examples of the application of resonant X-ray diffraction to identifying spin structures in Gd-based compounds, and show promise for facilitating the future discovery of exotic spin textures in broader classes of magnets.

Since the discovery of the magnetic skyrmion lattice state in chiral magnets, increasing numbers of noncentrosymmetric magnets have been investigated as platforms to explore magnetic skyrmions. This is because in noncentrosymmetric systems relativistic effects enable the antisymmetric exchange interactions, known as Dzyaloshinskii-Moriya interactions, between spins, which provide the magnets with instability towards a modulated spin structure such as the helimagnetic state. The size of magnetic skyrmions stabilized by this scheme, however, is limited to typically larger than 10 nm due to the weak strength of the relativistic effect. As a result, the emergent magnetic field originating from the topological nature of skyrmions inversely proportional to the square of the size of skyrmions is small in noncentrosymmetric skyrmions. This prompted us to look for different mechanisms for the stabilization of more spatially confined skyrmions to harness the emergent responses such as topological Hall effect for potential spintronics applications.

We focused instead on geometrical frustration as a source of modulated spin structure. Such frustration is present even in centrosymmetric magnets and magnetic modulations induced as such are in principle not limited in the length scale other than the atomic distance of the underlying lattices. It is well-known that the competing magnetic interactions in, for example, triangular and kagome lattices, lead to various exotic spin states such as spin liquids, helimagnetic states, and noncoplanar spin states. Recent theoretical studies suggested the potential of stabilizing the skyrmion lattice state in frustrated systems under certain circumstances.

The first experimental discovery of the emergence of the skyrmion-lattice state driven by frustration was reported in Ref. [1], where we investigated the target compound Gd_2PdSi_3. Figure 1 shows the basic crystal structure of Gd_2PdSi_3, where triangular lattices of Gd atoms stack together with honeycomb lattices of Pd/Si atoms. We performed extensive measurements of magnetization and magnetotransport properties to identify the topological Hall effect in the intermediate magnetic phase under an out-of-plane magnetic field. The origin of the topological Hall effect is ascribed to the emergence of the skyrmion lattice state on the order of 3 nm (Fig. 1). This conclusion was corroborated by magnetic structure analysis using the resonant X-ray diffraction technique. Geometrical frustration in the Gd-triangular-lattice combined with higher-order spin exchange interactions is considered to be the driving force to stabilize the skyrmion state in this compound.

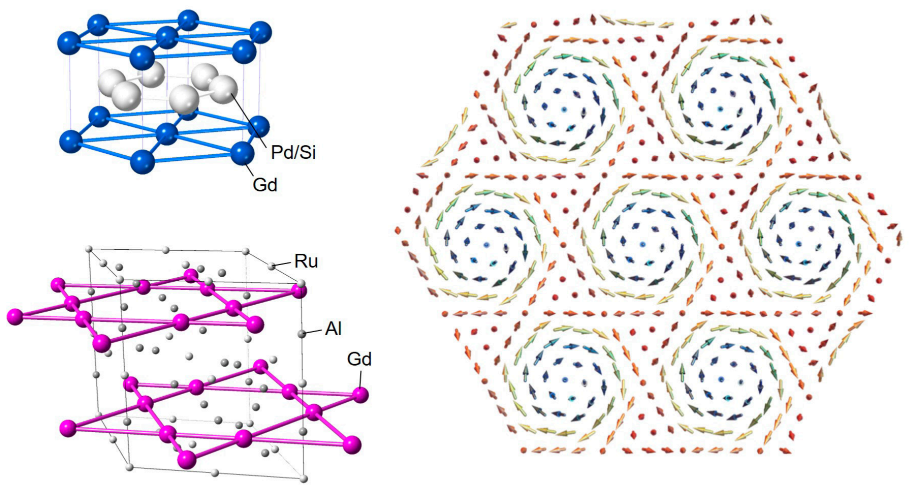

Figure 1: (Left panel) Crystal structures of Gd-based magnets. (Right panel) magnetic moment configuration for a triangular lattice of magnetic skyrmions.

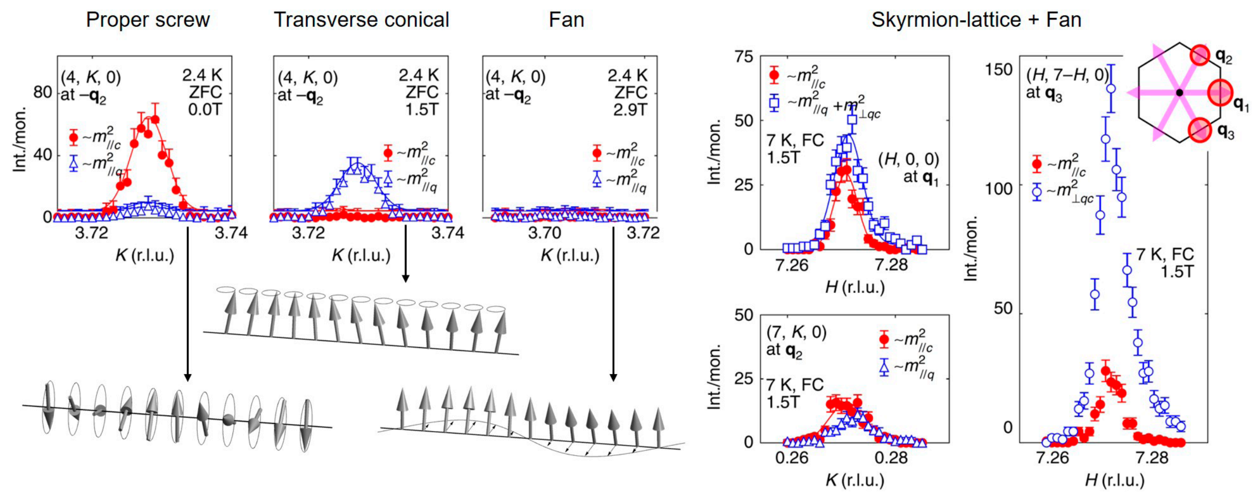

Figure 2: (Left panel) Polarization-resolved intensity profiles obtained in each magnetic state as depicted in the lower part. (Right panel) Intensity profile for skyrmion lattice phase with fan domain contamination. Excerpt from Ref. [3].

In order to clarify the universality of our design principle for centrosymmetric skyrmions, we investigated another frustrated magnet, $Gd_3Ru_4Al_{12}$. Figure 1 shows the crystal structure of $Gd_3Ru_4Al_{12}$. Gd atoms are aligned in the ab-plane to form a so-called breathing kagome lattice, composed of two types of inequivalent triangles. Magnetic frustration is expected to play a key role in the magnetic structure in this compound as a preceding X-ray resonant diffraction study [2] reported helical ordering of spin trimers of Gd atoms. We synthesized single crystals of $Gd_3Ru_4Al_{12}$ and uncovered a complicated magnetic phase diagram under a magnetic field perpendicular to the kagome lattice plane via magnetization, electrical transport, neutron diffraction, Lorentz transmission electron microscopy as well as resonant X-ray diffraction. We identified the skyrmion lattice state with topological Hall effect demonstrating the prevailing topological spin textures in centrosymmetric magnets with magnetic frustration. The wavelength here is comparable with that of Gd_2PdSi_3.

Figure 2 summarizes a key measurement of resonant X-ray diffraction to reveal various spin structures including the skyrmion state in $Gd_3Ru_4Al_{12}$. We performed polarization analysis to clarify the spin orientation in each magnetic phase. Due to the geometrical configuration of the diffractometer and polarization of incident X-rays, we can resolve the in-plane and out-of-plane components of modulated magnetic moments. For the proper screw state, both components were observed while the out-of-plane component was absent for transverse conical and fan structures. We observed scattering intensity for both components in three different satellite peaks under a magnetic field, which is consistent with the skyrmion-lattice state.

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