Skyrmion phase on a frustrated breathing kagomé lattice

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1 Abstract

Recently, our group has experimentally realize [1-3] theoretical predictions [4,5] of a new class of skyrmion host compounds with centrosymmetric space groups. In contrast to skyrmions stabilized by Dzyaloshinskii-Moriya interactions [6], skyrmions here emerge due to frustrated interactions [4] or due to higher order terms included in the Kondo Hamiltonian describing local moment magnets coupled to a Fermi sea [5]. $Gd_3Ru_4Al_{12}$ is a good metal with Heisenberg-type Gd^{3+} moments, where competing interactions are realized due to the symmetry of the lattice. We have experimentally observed the skyrmion phase and competing magnetic orders using resonant elastic x-ray scattering (REXS) at the Gd-L₂ edge using beamline BL-3A [2].

2 <u>Main</u>

Centrosymmetric materials with relatively high (hexagonal or tetragonal) symmetry are an emerging focus in the hunt for topological spin textures [1-3]. Here, competing Heisenberg-like interactions or four-spin terms (e.g. in the RKKY-expansion of the Kondo Hamiltonian) can realize complex magnetic orders such as spirals and skyrmion vortices. As compared to skyrmions in centrosymmetric materials (e.g. MnSi [6]) or at interfaces [7], Dzyaloshinskii-Moriya interactions (DMI) are absent or cancel out globally. Characteristically, spin textures in centrosymmetric materials condense at much shorter length scales ($\sim 2-3$ nanometers) as compared to the typical DMI case ($\sim 10 - 300$ nm), and generate a very large emergent magnetic field B_{em} . The latter is thought to drive large transport and magneto-optical responses [8], as well as new functionality [9].

Gd₃Ru₄Al₁₂ crystallizes in a quasi-layered structure of P6₃/mmc space group, with perfectly planar sheets of Gd³⁺ magnetic ions in a distorted ('breathing') Kagome lattice (Fig. 1). The Ru sublattice, forming a triangular net, is largely responsible for electronic conduction. As compared to another compound, Gd₂PdSi₃, the present material is more metallic and boasts a qualitatively different phase diagram, where a transverse conical state competes with the skyrmion lattice at low temperature *T* (Fig. 2). This indicates that manipulation of magnetic anisotropy is important in the control of topological spin textures in centrosymmetric magnets.



 PG_3 /mmc space group. (b) Illustration of the 'breathing Kagome' rare earth sublattice dominating the magnetic properties. Excerpt from Ref. [2].

We studied REXS at BL-3A of Photon Factory using large, polished single crystals in reflection geometry inside the 8 Tesla superconducting magnet [2]. With the aid of polarization analysis, scattering from the q = (0.23, 0, 0)reflection was observed in several phases. At the lowest temperature (T = 2.4 K), we identified helical order in zero magnetic field (H = 0), followed by a transverse conical and a fan-like state as H is increased. At higher temperature $(T \sim 8 \text{ K})$, within the boundaries of the SkL phase as determined from bulk characterization, transport, and Lorentz-transmission electron microscopy [2], we observed REXS intensity consistent with two volume fractions: a helical-type state (the skyrmion lattice) coexists with fan-like order. As the helical nature of the skyrmion state cannot be tested in LTEM, REXS provided very important input for the interpretation of our combined experimental results.



Figure 2: (a-c,j) Illustration of magnetic orders in various phases.(d-i) REXS with polarization analysis, HK0 scattering plane, Gd-L₂ edge for phases in (a-c). $\pi - \pi'$ (red) and $\pi - \sigma'$ (blue) components of the magnetic satellite at q = (0.23, 0, 0) were extracted and associated with in-plane and out-of-plane components of the ordered moment. (k-m) same for the skyrmion lattice. The $\pi - \pi'$ intensity was found to be comparable for three reflections, consistent with multi-q magnetic order [inset of (m)]. The magnetic field was applied parallel to the *c*-axis

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