Magnetic structure analysis of $EuAl_4$ by polarized neutron scattering

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Magnetic skyrmions are noncoplanar spin textures with topologically stable particle-like nature and is attracting attention as a candidate of high-density information carriers. While magnetic skyrmions were originally discovered in noncentrosymmetric systems, in which Dzyaloshinskii-Moriya interaction is active, nanometric skyrmion lattices have recently been found in centrosymmetric rare-earth alloys and a novel formation mechanism mediated by itinerant electrons has been newly proposed.

The title compound $EuAl_4$ is characterized by centrosymmetric tetragonal crystal structure ($I4/mmm$), and the magnetism is governed by the Eu^{2+} ions [1]. This compound shows multiple-step magnetic transitions as sweeping the magnetic field $H \parallel [001]$ (Fig. 1). Preceding neutron scattering experiment at zero field revealed incommensurate magnetic orders below $T_N = 15.4$ K [2]. To elucidate the magnetic structure for each phase in magnetic fields, we have performed small-angle neutron scattering experiments under $H \parallel [001]$ on a single-crystal sample and found the rhombic and square lattices of nanometric skyrmions in the intermediate field region (phases II and III) [3].

To further investigate the magnetic structures in the course of phase transitions at zero field (I → V → VI → IV), we performed neutron diffraction measurements with longitudinal polarization analysis at various temperatures with a triple-axis spectrometer PONTA. The experiments were carried out in $(h,0,l)$ and (h,h,l) scattering planes, and we measured line scan profiles of the magnetic Bragg reflections at $(q,0,0)$ and $(q,q,0)$ along the $(h,0,0)$ and $(h,h,0)$ directions, respectively (Fig. 2). In both setups, neutron spin polarization vector was set to be perpendicular to the scattering plane, and spin-flip (SF) and non-spin-flip (NSF) scattering signals detect the out-of-plane and in-plane components of spin modulation, respectively.

It is clearly confirmed that phases I and V are single-Q states hosting screw spin textures with different amplitudes of Q-vector. In phase VI, we have identified not only fundamental magnetic reflection $\boldsymbol{Q}_1$ but also $\boldsymbol{Q}_1+\boldsymbol{Q}_2$ reflection, proving its double-Q character. From the polarization analysis, $\boldsymbol{Q}_1$ and $\boldsymbol{Q}_1+\boldsymbol{Q}_2$ reflections are characterized by in-plane and out-of-plane sinusoidal spin modulations, respectively. Thus, phase VI is regarded as a double-Q state described by the superposition of four sinusoidal modulations. For phase IV, we have identified very weak but discernible $\boldsymbol{Q}_1+\boldsymbol{Q}_2$ reflection in the preceding SANS data, suggesting that phase IV is also a double-Q state. Combined with the results of the polarization analysis that the $\boldsymbol{Q}_1$ reflection corresponds to the in-plane sinusoidal spin modulation, phase IV is assigned as a vortex lattice state. These results suggest a new route to realize various double-Q spin textures in a simple crystal lattice with itinerant electrons.

[1] A. Nakamura *et al.*, JPSJ **84**, 124711 (2015).
[2] K. Kaneko *et al.*, JPSJ **90**, 064704 (2021).
[3] R. Takagi *et al.*, Nat. Commun. **13**, 1472 (2022).

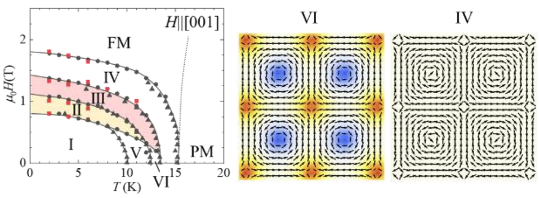

Fig. 1. Magnetic phase diagram of $EuAl_4$ and schematic spin textures for phases VI and IV.

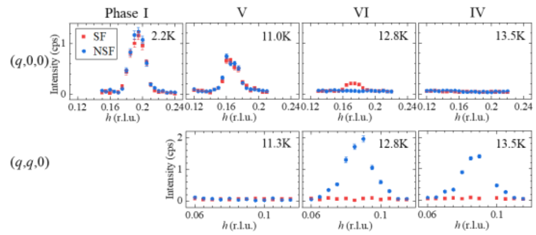

Fig. 2. Line scan profiles of SF and NSF scatterings for each phase at zero field.