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Canted magnetic structure in a dipole-quadrupole coupled magnet DyAuGe

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1 Introduction

The search for new quantum materials with emergent electrodynamic responses is essential for future applications in spintronic technologies. Rare-earth-based frustrated magnets are a group of materials attracting growing interest due to the coupled magnetic dipole and electric quadrupole degrees of freedom enabled by the strong spin-orbit interaction in 4f electrons. Recent material searches along these lines have led to a rich series of discoveries highlighted by the topological spin texture called skyrmions in Gd-based magnets [1] and unconventional spiral spin states in Eu-based magnets [2-3]. More exotic magnetic states beyond classical Heisenberg spins are expected to show up in the system characterized by multipolar degrees of freedom.

In this study, we performed the x-ray diffraction of Dy-based triangular-lattice magnets DyAuGe, and clarify a unconventional canted spin configuration induced by antiferroquadrupole ordering [4].

2 Experiment

We performed the resonant x-ray scattering experiment of single crystals of DyAuGe grown by Au-Ge flux. The measurement was carried out at BL-3A by using the horizontally polarized x-ray in resonance with Dy L_3 absorption edge (7.794 keV). The scattering plane was set to be (H, 0, L). The sample was loaded into a vertical-field superconducting magnet with the b axis parallel to the field direction.

3 Results and Discussion

We observe Bragg scattering at (1/2, 0, L) for L = 5 and 6 as shown in Fig. 1(a). The same L = 6 peak is observed in isostructural HoAuGe, where collinear two-sublattice spin configuration was observed [5]. The observation of L = 5 in DyAuGe suggests the magnetic structure is more complicated due to canting of Dy magnetic moments. Such noncollinear nature has also been observed in many Dy-based compounds, where quadrupole degrees of freedom play an important role.

We performed the energy scan of these Bragg peaks (Fig. 1(b)) and found that the scattering is dominated by lattice. This is in contrast to the resonant magnetic scattering of (1/2, 0, 6) peak in HoAuGe. These contrasting features are also evidence of the antiferroquadrupole ordering coupled with dipole ordering in DyAuGe.

We also found (0, 0, 7) peak (Fig. 1(c)), which is forbidden by the lattice scattering, below the transition temperature. The energy scan (Fig. 1(d)) indicates that the scattering resonates at Dy L_3 edge, suggestive of the antiferromagnetic dipole ordering in the out-of-plane.

We performed the measurement of magnetic susceptibilities and specific heat of DyAuGe, and analyzed the crystal electric field (CEF). We have found that the low-energy CEF levels are composed of quasi-quartet with energy gap comparable to the exchange interaction between Dy ions. Due to the quartet nature, the tilt of the magnetic moment towards the c axis induces quadrupole moment in the ab-plane. Alternative canting of each moment on each layer explains the antiferroquadrupole ordering, as shown in Fig. 1(e). The atomic shift of Au and Ge is expected by electrostatic potential from the quadrupole moment, which is consistent with the lattice dominant Bragg scattering at (1/2, 0, L) for L = 5, 6.

In summary, the unconventional canted magnetic structure in DyAuGe has been revealed by the resonant x-ray scattering. Such a spin configuration is induced

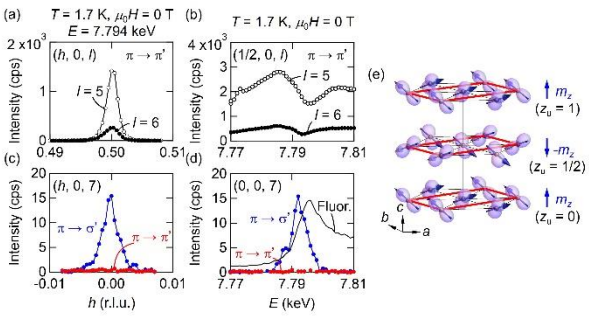

Fig. 1: (a) h scan around (h, 0, L) for L = 5 and 6 at 1.7 K and zero field. The measurement was performed with a single crystal of DyAuGe. (b) Incident-photon energy scan for each Bragg peak near Dy L_3 edge. (c)-(d) Corresponding scans for (0,0,7) Bragg peak. (e) Schematic magnetic structure of the ground state in DyAuGe. Blue arrows are magnetic dipole moments and purple dumbbells are quadrupole moments induced by a tilt of the dipole moment.

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