# Anomalous magnetic correlation in mixed valence state of $EuRuSn_3$

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Various magnetic textures and correlations have been investigated in, for example, skyrmion formations and spin liquid states, which are caused by a combination of magnetic interactions on characteristic lattice geometry [1, 2]. It is known that magnetic order in $NdRuSn_3$ is suppressed down to low temperatures due to geometrical magnetic frustration originating from the characteristic crystal lattice formed by magnetic Nd ions [3, 4]. On the other hand, its analogous compound, $EuRuSn_3$, is attracting attention because of magnetic ordering below approximately 11 K [5]. Previous studies, based on magnetic susceptibility measurements and Mössbauer spectroscopy, pointed out the existence of a complex magnetic structure and a static mixed-valence state comprising magnetic $Eu^{2+}$ and nonmagnetic $Eu^{3+}$ ions. Furthermore, magnetization measurement results suggested non-collinear ferromagnetism or ferrimagnetism. Therefore, $EuRuSn_3$ is a promising candidate for a unique magnetic ground state affected by mixed valence and geometrical frustration, and there is a strong demand for elucidating its physical properties.

In this study, we succeeded in growing high-quality single crystals of $EuRuSn_3$ using the same synthesis method in previous study [5] and attempted to elucidate its magnetic ground state through bulk property measurements and quantum beam scattering experiments. Magnetic property measurements between 2 and 300 K were performed using a SQUID magnetometer (MPMS of Quantum Design, Inc.) installed at the Neutron Science and Technology Center, Comprehensive Research Organization for Science and Society (CROSS Lab). Neutron diffraction (ND) measurements were performed using a powder diffractometer HERMES installed at the T1-3 beam hole and a triple-axis thermal neutron spectrometer HQR installed at the T1-1 beam hole of the Japan Research Reactor 3 (JRR-3) owned by the Japan Atomic Energy Agency (JAEA). We performed x-ray absorption measurements at BL-3A and 4C of Photon Factory, KEK.

In the magnetic susceptibility measurements, we observed successive anomalous phase transitions exhibiting a ferromagnetic correlation below approximately 11 K. Additionally, magnetizations under zero-field and field cooling processes below 7.5 K deviated from each other, indicating a random magnetic behavior. The XAS measurement near the Eu $L_2$ and $L_3$ edge revealed that approximately 4% of Eu ions are nonmagnetic trivalent, which is expected to act as impurities breaking the local symmetry of the Eu sublattice. Using a ND technique, we succeeded in observing an increase in magnetic scattering intensity. Figure 1 shows temperature dependence of integrated intensity obtained by longitudinal (black circles) and rocking-curve (red squares) scans through $\boldsymbol{Q} = (3, 3, 1)$. We also detected magnetic peaks at several reciprocal lattice points. These results indicate a long-range antiferromagnetic correlation superimposes on the random magnetic state. The critical temperature is close to the random magnetic transition temperature. The above results indicate that $EuRuSn_3$ is a material exhibiting anomalous magnetic correlation emergence associated with randomness induced by the mixed-valence state.

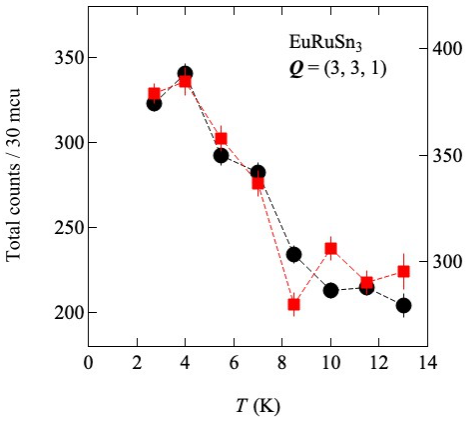

Fig. 1. Emergence of magnetic Bragg reflection at $\boldsymbol{Q}$ = (3, 3, 1) of $EuRuSn_3$.

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