3A/2009S2-008

All-in-all-out Magnetic Order in the Insulating Phase of a Pyrochlore Iridate $Eu_2Ir_2O_7$

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1 Introduction

Ir oxide compounds can exhibit a novel J_{eff}=1/2 state with unquenched orbital momentum due to strong spin-orbit coupling.[1-3] For example, there is an intense debate about the role of the J_{eff}=1/2 state in a metal-insulator transition, which several Ir oxide compounds undergo. Rare-earth (R) iridium oxide compounds $R_2Ir_2O_7$ with the geometrically frustrated pyrochlore lattice exhibit a metal-insulator transition accompanying an anomaly in magnetic susceptibility except R=Pr.[4-6] The origin of the transition is yet unclear partly because of a lack of information on the magnetic structure in the insulating phase.

Here we study magnetic order in the low-temperature insulating phase of $Eu_2Ir_2O_7$, which undergoes this metal-insulator transition at T_{MI}~120 K.[5,6] Large neutron absorption coefficients of Eu and Ir make a neutron study on the compound difficult. On the other hand, resonant x-ray diffraction is a useful tool for studying magnetic order in iridium compounds, because the $L_{2,3}$ absorption includes the excitation of an electron from 2p core states to partially filled 5d valence states, which are directly associated to the magnetism.[2] The wavelength of $L_{2,3}$ absorption edge is near 0.1 nm, which is suitable for diffraction.

2 Experimental

A single crystal was grown from a flux and characterized by J. J. Ishikawa and S. Nakatsuji, ISSP, Univ. Tokyo.[6] The crystal has shiny triangle surfaces of a dimension of about 0.5 mm. The resonant x-ray diffraction measurement was carried out by using a multi-circle diffractometer at BL-3A. The sample was mounted in a ^{4}He closed-cycle refrigerator. The incident beam was polarized perpendicular to the scattering plane (σ). Mo (200) reflection was used to analyze the polarization of scattered beam.

3 Results and Discussions

A previous muon spin rotation study suggests that Ir moments should be arranged with some commensurate propagation vector.[7] Several line scans along (*qkl*), (*qql*), and (*qqq*), where *k* and *l* are integers, at 10 K indicate the absence of extra reflection at incommensurate positions with relatively high symmetries. Hereafter, we discuss resonant x-ray reflections at (4*n*+2, 0, 0), which are ordinarily forbidden in pyrochlore systems. Figure 1 shows temperature dependence of the integrated intensity of (10 0 0) reflection at 11.23 keV (Ir L_3 edge) at 10 K. Here the scattering plane is set perpendicular to the [0 1 -1] axis, and σ' (π') denotes the scattered beam polarized perpendicular (parallel) to the scattering plane. In the σ-π' channel, the reflection emerges only below T_{MI}, while the reflection in the σ-σ' channel remains above T_{MI}. It is well known that the reflection condition originating from a screw axis or glide plane can be relaxed for anomalous scattering terms in general. In the pyrochlore iridate, each IrO_6 octahedron has trigonal distortion of compressed type. The distortion induces an anisotropy in Ir atomic anomalous scattering factor, and makes the (4*n*+2 0 0) reflections allowed. In the present configuration, the anomalous-tensor-susceptibility (ATS) scattering of (4*n*+2 0 0) may appear only with the σ' polarization. The π' component is hence assigned to the magnetic scattering. Spectra of the intensity for (10 0 0) and (6 0 0) reflections shown in Fig. 2 also confirm the magnetic origin of the π' component. The scattering in the σ-π' channel shows a resonant peak at 11.230 keV. On the other hand, the spectrum in the σ-σ' channel is characterized by a resonant peak at 11.238 keV as well as a shoulder at around 11.23 keV. The difference in spectrum is well explained in terms of ligand-field splitting of 5d state. The octahedral ligand field splits the Ir 5d orbitals into e_g and t_{2g} states. The L_3-edge $2p_{3/2}$-5d absorption is hence composed of two bands. Five 5d electrons in the lower-lying t_{2g} state are the source of a magnetic moment of each Ir^{4+} ion, while the empty e_g band is irrelevant to the magnetism. As a consequence, it is predicted that the magnetic scattering should be resonant only with the lower-lying $2p_{3/2}$-t_{2g} excitation, whilst the anomalous tensor susceptibility of Ir atom should be enhanced at both the absorption energies.

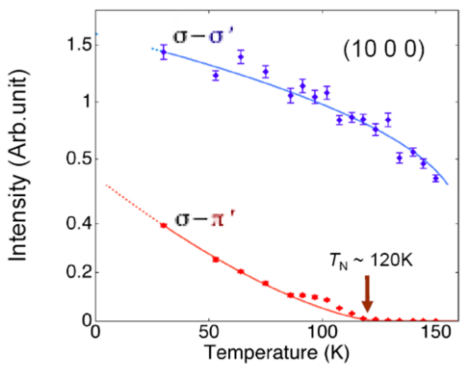

Fig. 1: Integrated intensity of (10 0 0) reflection as a function of temperature. The x-ray photon energy is 11.230 keV. Blue and red dots show intensities in the s-s' and s-p' channels, respectively.

The emergence of (4*n*+2 0 0) magnetic reflections clearly indicates that a magnetic unit cell should contain four Ir^{4+} moments. Here we propose the most probable arrangement of the magnetic moments among several candidates of four-sublattice magnetic order. The moments on four vertices of each Ir tetrahedron point to the center of the tetrahedron (inwards) or in the opposite direction (outwards), which can be termed 'all-in-all-out' structure. Only this magnetic structure can retain the cubic symmetry[8], which is in accord with a detail x-ray analysis of the crystal structure at low temperatures.

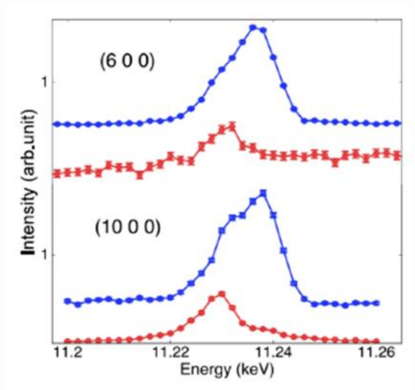

Fig. 2: Spectra of the peak intensity for (6 0 0) and (10 0 0) reflections near Ir L_3 absorption edge. Blue and red dots show intensities in the σ-σ' and σ-π' channels, respectively.

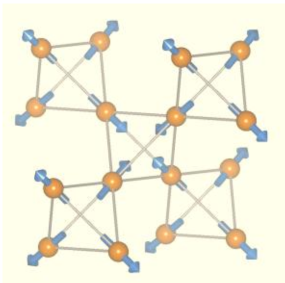

Fig. 3: All-in-all-out magnetic structure in the pyrochlore lattice.

Acknowledgment

The sample crystal was provided by J. J. Ishikawa and S. Nakatsuji.

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