Electronic and Magnetic Structures of Spin-Orbit Coupling Induced Mott Insulator Sr_2IrO_4

Perovskite-type transition metal oxides (TMOs) show many fascinating phenomena, such as high-T_c superconductivity in cuprates and colossal magnetoresistance in manganites. 3*d* TMOs have been extensively studied since their rather narrow *d* band can induce a Mott instability. Valence *d* orbitals of a transition metal ion in an octahedral crystal field are split into the t_{2g} and e_g levels, where the d_{xy}, d_{yz} and d_{zx} orbitals are lower in energy than the $d_{3z^2-r^2}$ and $d_{x^2-y^2}$. When the *d* orbitals are partially and selectively occupied (e.g., by Jahn-Teller effect), the orbital angular momentum of the transition metal ion is totally quenched because the orbitals that are stabilized by the Jahn-Teller distortion are expressed using real wave functions. With increasing period number of an element, the spatial extent of *d* orbitals tends to increase, and therefore, their bandwidth is expected to be broad. Indeed, Sr_2RhO_4 ($4d^5$) is a fairly good metal compared with Sr_2CoO_4 ($3d^5$). Sr_2IrO_4 ($5d^5$) is a magnetic insulator [1], whereas it would have a t_{2g}^5 low-spin state and become a metal with a partially filled wide t_{2g} band. Recently, it has been predicted that the strong spin-orbit coupling recovers the orbital motion of 5*d* electrons; the resultant spin-orbit integrated states form two narrow bands, so that even a small on-site Coulomb repulsion opens a Mott gap [2].

To confirm the realization of a spin-orbit coupling induced Mott insulator, we have conducted resonant X-ray magnetic scattering (RXMS) experiments at beamlines **BL19LXU** and **BL29XU**. We used highly brilliant SR with a photon energy corresponding to Ir *L*-edge (2*p*-5*d*) to explore unconventional electronic states. A selection rule of RXMS identifies a 5*d* state as a complex spin-orbit integrated state and not as a crystal field state [3]. Figure 1 shows RXMS spectra of the magnetic reflection (1 0 22) at the Ir *L* edge. The reflection intensity at the L_2 edge is less than 1% of that at the L_3 edge, which is a direct consequence of a selection rule for RXMS. Figure 2 illustrates 5*d* level splitting by a crystal field and a spin-orbit coupling. RXMS at the L_2 edge becomes forbidden for the spin-orbit integrated state, whereas equal resonant intensities are expected at the L_2 and L_3 edges for the crystal field state. The spin-orbit integrated state well explains the experimentally obtained RXMS spectra.

The magnetic structure of Sr_2IrO_4 remained undetermined, because Ir is a strong neutron absorber. Using the enhancement at the L_3 edge, we have tried to determine the magnetic structure of Sr_2IrO_4. The crystal structure is of the K_2NiF_4 type and shown in Fig. 3(a). Our RXMS results revealed that the magnetic structure is canted antiferromagnetic (see Fig. 3(b)). In zero field, magnetic reflections are observed at (1 0 4*n*+2), (0 1 4*n*) and (0 0 *odd*) as shown in Figs. 3(c)-3(e). By representational analysis with the extinction rule for magnetic reflections, a symmetrically allowed possible magnetic structure is considerably restricted to two candidates. One is compatible with metamagnetism of Sr_2IrO_4 and the other is not, so that the magnetic structure is uniquely specified. The application of a magnetic field induces metamagnetic transition and rearrangement of magnetic moments. The determined magnetic structure also well explains the appearance of a net magnetic moment in the metamagnetic state and an accompanying change in the extinction rule for magnetic reflections [3].

The experimental establishment of the spin-orbit integrated states of valence electrons is a first step for developing a new research field of relativistic materials. For materials characterized by a spin-orbit coupling, RXMS is an ideal method to probe the phase of the outer electron wave function. Our study opens up a new exclusive feature of X-ray magnetic scattering.

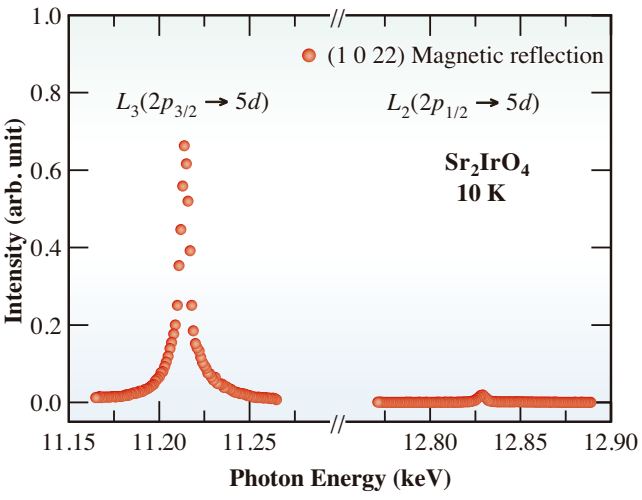

Fig. 1. RXMS spectra of the magnetic reflection (1 0 22) at the *L* edge.

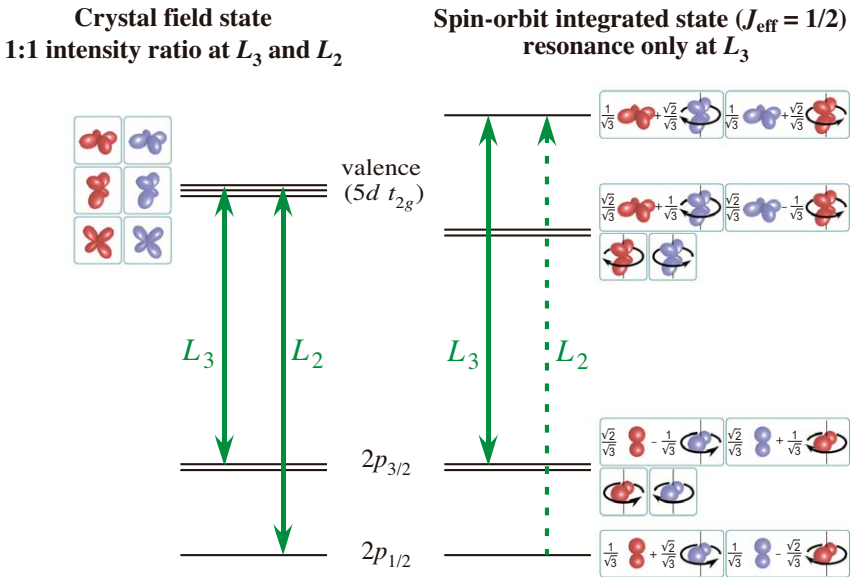

Fig. 2. 5*d* level splitting diagram of (left) a tetragonal crystal field and (right) the spin-orbit coupling. Equal resonant intensities are expected at the L_2 and L_3 edges for the crystal field state. For the spin-orbit integrated state (J_{eff}=1/2), RXMS at the L_2 edge becomes forbidden. Wave functions of 5*d* electrons are depicted with their orbital form and spin. Red and blue correspond to up and down spins, respectively.

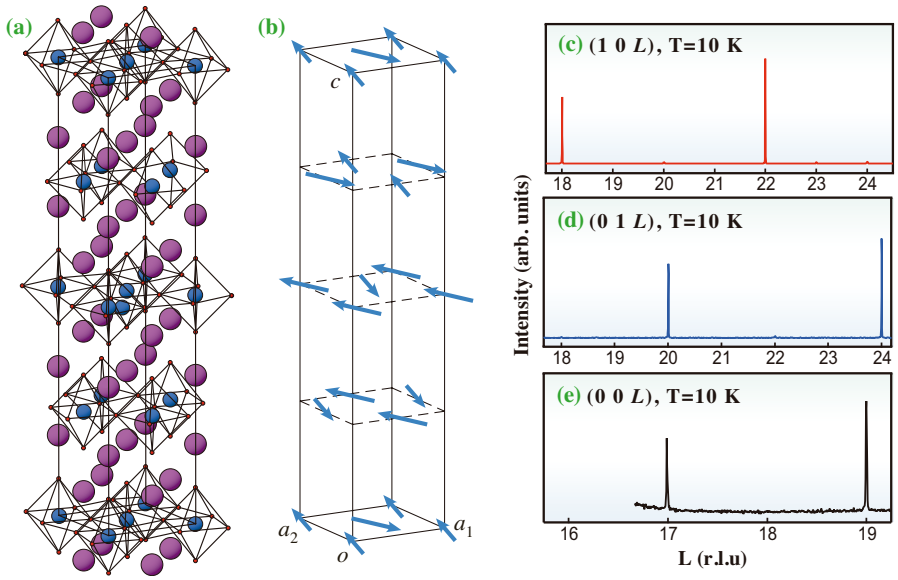

Fig. 3. Crystal and magnetic structures of Sr_2IrO_4. **(a)** Crystal structure of Sr_2IrO_4 (space group $I4_1/acd$). The blue, red and purple spheres represent Ir, O and Sr, respectively. **(b)** Magnetic structure of Sr_2IrO_4 in zero magnetic moment. Arrows represent J_{eff}=1/2 moments. **(c)** Scan profile along the (1 0 *L*) direction at 10 K in zero magnetic field. **(d)** Scan profile along the (0 1 *L*) direction at 10 K in zero magnetic field. **(e)** Scan profile along the (0 0 *L*) direction at 10 K in zero magnetic field.

Hiroyuki Ohsumi[a,*], Taka-hisa Arima[a,b] and Hidenori Takagi[c,d]

[a] SPring-8 / RIKEN
[b] Institute of Multidisciplinary Research for Advanced Materials, Tohoku University
[c] Department of Advanced Materials Science, The University of Tokyo
[d] RIKEN Advanced Science Institute (Wako)

*E-mail: ohsumi@spring8.or.jp

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