Electronic and Magnetic Structures of
Spin-Orbit Coupling Induced Mott Insulator Sr$_2$IrO$_4$

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 $d_{xy}$, $d_{xz}$, and $d_{yz}$ 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$_2$RhO$_4$ (4$d^5$) is a fairly good metal compared with Sr$_2$CoO$_4$ (3$d^5$). Sr$_2$IrO$_4$ (5$d^5$) 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$_2$IrO$_4$ 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$_2$IrO$_4$. The crystal structure is of the K$_2$NiF$_4$ type and shown in Fig. 3(a). Our RXMS results revealed that the magnetic structure is canting 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$_2$IrO$_4$ 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. 5d level splitting diagram of (left) a tetragonal crystal field and (right) the spin-orbit coupling. Equal resonant intensities are expected at the L₂ and L₃ edges for the crystal field state. For the spin-orbit integrated state \( J_{\text{eff}} = 1/2 \), RXMS at the L₂ edge becomes forbidden. Wave functions of 5d 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 Sr₂IrO₄. (a) Crystal structure of Sr₂IrO₄ (space group I₄₁/acd). The blue, red and purple spheres represent Ir, O and Sr, respectively. (b) Magnetic structure of Sr₂IrO₄ in zero magnetic moment. Arrows represent \( J_{\text{eff}} = 1/2 \) 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.

References

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