Magnetic excitations of Sr$_3$Ir$_2$O$_7$ observed by inelastic neutron scattering measurement

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In this short note, we report on the first inelastic neutron scattering (INS) study on high-energy magnetic excitations in Sr$_3$Ir$_2$O$_7$. We observed excitations between ~80 meV and ~180 meV. The peak position of the excitations is consistent with the dispersion of a single magnon determined by resonant inelastic X-ray scattering (RIXS) measurement. Thus, our results demonstrate that INS and RIXS observe identical excitations in iridate oxides, as in La$_2$CuO$_4$.

Iridate oxides, Sr$_2$IrO$_3$ and Sr$_3$Ir$_2$O$_7$, are Mott insulators with spin-orbit entangled magnetic state bearing the effective total angular moment, $J_{\text{eff}} = 1/2$. A new root for high-transition-temperature (high-$T_c$) superconductivity is expected for iridate oxides. Accordingly, their magnetic excitations have received significant attention analogous to the spin dynamics of high-$T_c$ cuprate oxides. Resonant inelastic X-ray scattering (RIXS) is a powerful tool to investigate magnetic excitations owing to its accessibility to high-energy regions even for small samples. The excitations of Sr$_2$IrO$_3$ and Sr$_3$Ir$_2$O$_7$ comprise a single magnon branch below 300 meV and a novel high magnetic mode (spin-orbit exciton with $J_{\text{eff}} = 3/2$) approximately 600 meV.

Inelastic neutron scattering (INS) that directly measures the dynamical structure factor, $S(Q, \omega)$, in the momentum ($Q$) and energy ($\omega$) spaces is also an indispensable method for investigating spin dynamics. For cuprate oxides, INS provides a considerable amount of valuable information on magnetic excitations such as hourglass-shaped excitations and resonance excitations in the superconducting phase. In addition, the determination of $f(Q)$, which is proportional to the neutron scattering intensity, can present information on orbital character and spin-orbit entangled excitations in iridate oxides. Herein, $f(Q)$ is the magnetic form factor that is a Fourier transform of spin density distribution in real space. However, INS measurement of iridium oxides is challenging owing to the significant neutron absorption cross-section of the iridium nuclei. Report of excitations exceeding 100 meV in iridate oxides by INS is scarce following our review of the literature, whereas observation below ~40 meV was recently achieved for monolayer Sr$_2$IrO$_4$. In this short note, we present an INS investigation of bilayer Sr$_3$Ir$_2$O$_7$. By using high-energy neutrons with a reduced absorption effect, we observed a magnon branch existing between ~80 meV and ~180 meV, and verified the consistency in dispersion with that obtained by RIXS.

For the experiment, we prepared single crystals by the flux-method. The maximum size of the crystals was 2 mm × 2 mm × 1.5 mm. Approximately 500 crystals with a total mass of 2.1 grams were assembled on aluminum plates (with 0.1 mm thickness) such that the crystallographic c-axis is perpendicular to each plate. We performed INS measurements on 4 SEASONS at the Materials and Life Science Experimental Facility (MLF) at J-PARC, Japan. A 250 meV incident neutron energy was selected to determine the magnon excitations below 200 meV. The signals were collected for two days under 300 kW beam power operation at 6 K, that is significantly below the magnetic ordering temperature (~280 K). To minimize the absorption effect, the crystals were placed with the c-axis parallel to the incident neutron beam. In this study, we use the tetragonal notation to describe the Miller index.

Figure 1 depicts the INS spectra along the energy direction at $(h, k)$ equal to (0.25, 0.25) and (0.90, 0.10). The former position corresponds to the antiferromagnetic (AF) zone boundary (ZB). The spectra at the latter position wherein the magnetic structure factor is weak, is shown as the background. The intensity around 100 meV is higher at (0.25, 0.25) than that at (0.90, 0.10). From the difference in the spectral intensities at the two positions, the peak-energy in the spectra at (0.25, 0.25) was evaluated to be 107(8) meV. This energy is consistent with the results reported in RIXS. Note that the peaks at ~170 meV are independent of $Q$ and were observed even in the measurement after removing the crystals. Thus, this signal is background from outside the sample.

To further examine the excitation spectra, we sliced the spectra at several energies. Figure 2 shows the spectra along the $k$-direction through (0.5, ±0.5) corresponding to the AF zone center (ZC). An arch-shaped signal centered at $k = 0$ is observed in Fig. 2 (a). The spectra sliced at $h\omega = 180$ meV with an energy band of ± 10 meV (Fig. 2 (b)) shows a broad peak at $k = 0$. In decreasing $h\omega$, the signal splits into two peaks that are symmetric around $k = 0$, and the distance between the peaks increases. The peak positions at $h\omega = 80$ meV are close to ZC (Fig. 2 (e)), and no visible magnetic signal below 70 meV was detected, indicating the existence of a large energy gap in the spin excitations. The excitations in energies from ~80 meV to ~180 meV is consistent with the single

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Fig. 1. (Color online) INS spectra slices at (0.25, 0.25) and (0.9, 0.1) with the $h$- and $k$-width of 0.1. The inset is a photo of the assembled samples.
single magnon branch, which is indicated using RIXS.\(^3\) Therefore, we detected the magnetic excitations above \(\sim 80\) meV in iridates using high-energy neutrons with sufficient beam flux. From the figure, the scattering intensity for \(|k| > 0.5\) is weak or absent, leading to the asymmetric spectra around ZC (\(k = \pm 0.5\)). Although the cause of the intensity unbalance is unknown, \(f(Q)\) can be damped rapidly against \(Q\) compared to that for cuprate oxides, owing to the extended nature of the 5d orbitals. In fact, in the neighborhood of (1,5,1,5), which is ZC with a large \(|Q|\) value, the magnon branch was not observed. Thus, the intensity unbalance could be due to a particular \(f(Q)\), and determining the intensity distribution via INS can yield information on the orbital character.

In Fig. 3, the momentum dependence of the peak-position and intensity are presented. The open (closed) circles represent the results obtained from the constant-momentum (constant-energy) spectra. The horizontal (vertical) bars for the open (closed) circles are the sliced momentum (energy) width of the spectra, and the vertical (horizontal) bars represent the evaluated peak-width in energy (momentum) in full-width at half-maximum. The peak positions follow a gray line representing the dispersion relation determined by RIXS. This indicates that our experiment is foundational for future complementary studies. Figure 3 (b) shows the bare INS intensity that is not corrected with \(|f(Q)|^2\) and the absorption coefficient depending on \(\hbar \omega\). Accordingly, the values have complementary depending on \(\hbar \omega\). The peak positions agree well with the dispersion of a single magnon observed using RIXS. Therefore, the high energy neutrons aids in the investigation of the magnetic excitations of iridate oxides in the region of a hundred meV.

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