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Momentum-resolved electronic excitations in the Mott insulator Sr_2IrO_4 studied by resonant inelastic x-ray scattering

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We report an Ir L_3 edge resonant inelastic x-ray scattering measurement of the low-lying electronic excitations in Sr_2IrO_4 over the complete Brillouin zone of the IrO_2 plane. A remarkably strong inelastic signal which exceeds the elastic scattering in intensity is observed. Peaks observed at 0.5, 3.2, and 6.0 eV are respectively ascribed to an interband transition across the Mott gap and charge-transfer excitations from the O $2p$ band to the Ir $5d$ bands. The dispersion of the Mott gap excitation is found to be particularly weak. This is interpreted as a signature of the narrow $5d$ bands of the novel Mott insulating state of Sr_2IrO_4 induced by the strong spin-orbit interaction.

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I. INTRODUCTION

Strongly correlated electron systems, in particular transition metal oxides, are a constant source of fascination in modern condensed matter physics. They exhibit a rich panoply of exotic physical properties such as high temperature superconductivity for cuprates and colossal magnetoresistance for manganites. Among the transition metal oxides, $3d$ electron systems are the most extensively studied, due to the drastic correlation effects brought about by the narrow $3d$ bandwidth (W) and the strong on-site Coulomb repulsion (U). In contrast, the $5d$ orbitals have a larger spatial extent, and the correlation effects are rather moderate due to the wider bandwidth and the weaker Coulomb repulsion ($U \approx W$).

Recently, a novel type of Mott insulating state has been brought to light in the $5d$ transition-metal oxide Sr_2IrO_4. The spin-orbit interaction, whereas usually neglected in $3d$ transition metal oxides, is believed to be crucial for this unusual Mott insulator.[1,2] Sr_2IrO_4 has a K_2NiF_4-type layered perovskite structure and its five $5d$ electrons occupy the t_{2g} orbitals which are split into lower fourfold $J_{\rm eff} = \frac{3}{2}$ states and higher twofold $J_{\rm eff} = \frac{1}{2}$ states by the strong spin-orbit interaction. The $J_{\rm eff} = \frac{1}{2}$ states are represented as $|J_{\rm eff} = \frac{1}{2}, m_{J_{\rm eff}} = \pm\frac{1}{2}\rangle = \frac{1}{\sqrt{3}}(|yz, \pm\sigma\rangle \mp i|zx, \pm\sigma\rangle \mp |xy, \mp\sigma\rangle)$, where σ is the spin state.[1] Within the half-filled $J_{\rm eff} = \frac{1}{2}$ states, the Coulomb repulsion opens a band gap, thereby inducing the Mott insulating state. Clarifying the effects of the electron correlation and the spin-orbit interaction on the electronic structure is of fundamental importance for understanding this novel Mott insulating state. While the interband excitation across the Mott gap at zero momentum transfer has been identified at 0.5 eV by an optical conductivity measurement,[3] its dispersion relation still remains to be explored.

In this article, we report a study of the momentum dependence of the electronic excitations in Sr_2IrO_4 utilizing resonant inelastic x-ray scattering (RIXS) at the Ir L_3 edge. RIXS has previously been applied at the K edge of $3d$ transition metals to measure electronic excitations in oxides, including the excitation across the Mott gap, in a momentum-resolved fashion.[4–10] In the case of K edge RIXS, the excited core electron stays in a p state in the intermediate state and can be regarded as a spectator in most cases, as the valence electron is scattered predominantly through the Coulomb interaction with the core hole. It is referred to as *indirect* RIXS.[11] A consequence of the indirect excitation process is the much weaker intensity of the inelastic signal than the elastic scattering, which often hampers the observation of low-lying excitations. On the other hand, in L-edge RIXS, or *direct* RIXS, the excited core electron occupies a d state and participates in the scattering process directly. A strong inelastic signal ensues, which sometimes exceeds the elastic scattering. However, accessible momentum space in L-edge RIXS of $3d$ transition metals is limited due to the long wavelength of the x rays, e.g., one cannot span the whole Brillouin zone of the CuO_2 plane in the high-T_c superconducting cuprates at the Cu L edge. RIXS at the L edge of $5d$ transition metals benefits from both advantages, i.e. strong intensity and a wide accessible momentum space. To our best knowledge, there is only one published RIXS study at the L edge of a $5d$ transition metal, where the measurement of the RIXS signal from Pt nanoparticles was reported but its momentum dependence was not.[12] We here take a step forward and measure the momentum dependence of the Ir-L_3 RIXS spectrum of Sr_2IrO_4. Three excitations are identified as being the excitation across the Mott gap at 0.5 eV and charge-transfer excitations from the O $2p$ band to the upper Hubbard band at 3.2 eV and to the $3z^2$-r^2 band at 6.0 eV. All three excitations are found to be weakly dispersive.

II. EXPERIMENTAL DETAILS

The experiments were carried out at BL11XU at SPring-8. Incident x rays were monochromatized by a double-crystal Si(111) monochromator and a secondary Si(400) channel-cut monochromator. Horizontally scattered x rays were analyzed in energy by a spherically bent Si(844) crystal. The total energy resolution estimated from the full width at half maximum of the elastic peak was about 1 eV.

Single crystals of Sr_2IrO_4 were grown by the flux method using $SrCl_2$ flux. A mixture of $SrCO_3$, IrO_2, and $SrCl_2$ with a

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molar ratio of 2.0 : 1.0 : 15 was melted at 1300° C. The melt was slowly cooled down to 900° C at a rate of 8° C/h and then furnace cooled to room temperature. The surface normal to the stacking direction (c axis) was irradiated by x rays. The crystal was so mounted that the [110] and [001] directions span the scattering plane. We use the Miller indices of the tetragonal unit cell of $a = 5.4979$ Å and $c = 25.798$ Å with the space group $D_{4h}^{20} - I4_1/acd$[13] to represent the momentum transfer ($\boldsymbol{Q}$). The points of interest in the momentum space were selected so the scattering angle (2θ) is close to 90° in order to reduce the elastic scattering. The incident and scattered polarizations are therefore orthogonal to each other, which is the so-called depolarized configuration. All spectra were measured at room temperature.

III. RESULTS AND DISCUSSION

A. Incident energy dependence and assignment of the excitations

The incident photon energy dependence of the RIXS spectra across the Ir L_3 edge is shown in Fig. 1(a) as a two-dimensional mapping, where the intensity is plotted as a function of both the incident energy and the energy loss of the photon. The momentum transfer is fixed at $\boldsymbol{Q} = (0,0,33)$. Polarization($\epsilon$)-dependent x-ray absorption spectra (XAS) measured by the total fluorescence yield method are presented in Fig. 1(b). The peak of the while line shows nearly no shift between the in-plane ($\epsilon \parallel [110]$) and out-of-plane ($\epsilon \parallel [001]$) polarizations. It is in marked contrast to the Cu K-edge spectrum of La_2CuO_4 which is strongly polarization dependent,[14] even though both Sr_2IrO_4 and La_2CuO_4 have a similar K_2NiF_4-type structure. Because the final state of XAS corresponds to the intermediate state of the RIXS process, the RIXS intensity is usually enhanced near the peak of the while line. This is the case for Sr_2IrO_4, where the RIXS intensity is at its strongest when the incident energy is tuned near 11217 eV. We can discern three inelastic features, located at 0.5-, 3.2-, and 6.0-eV energy loss.

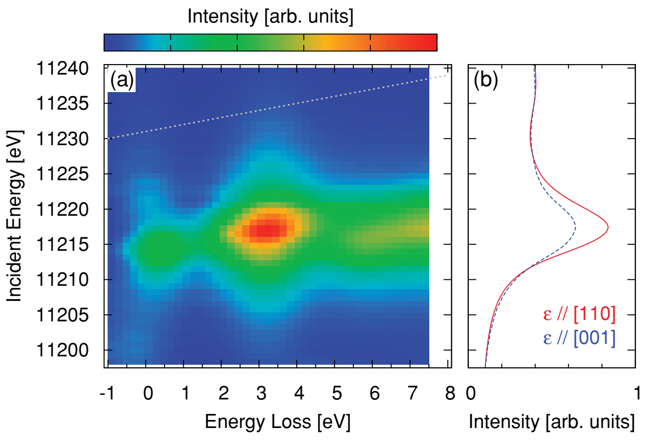

FIG. 1. (Color online) (a) Incident energy dependence of the RIXS spectrum. Absolute momentum transfer is $\boldsymbol{Q} = (0,0,33)$. The white dotted line with a slope of 1 is shown to represent at typical fluorescence behavior. Its position on the intensity map is arbitrary. (b) Polarization (ϵ) dependent x-ray absorption spectra at the Ir L_3 edge.

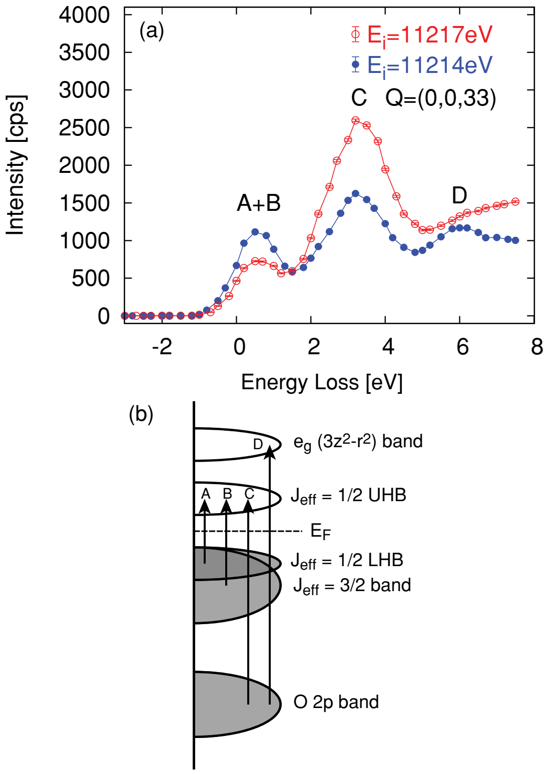

FIG. 2. (Color online) (a) RIXS spectra of Sr_2IrO_4 measured at two representative incident photon energies. (b) Schematic band diagram based on Refs. 18 and 1.

The RIXS spectra measured for two representative values of the incident energy (E_i), respectively, corresponding to the resonance of the 0.5- and 3.2-eV features ($E_i = 11214$, 11217 eV), are presented in Fig. 2(a). Strikingly enough, the intensity of the inelastic signal on resonance reaches almost 3000 counts per second (cps), which is two to three orders of magnitude higher than at the K edge of $3d$ materials using the same spectrometer. The count rate at the elastic position (0-eV energy loss) is comparable to that of K-edge RIXS, therefore the elastic peak is hidden by the tail of the strong 0.5-eV feature.

We now take a moment to examine the incident energy dependence in Fig. 1(a). The excitations, especially those at 0.5 and 3.2 eV, form elongated streaks parallel to the E_i axis. The fact that the energy loss is independent of the incident energy is a direct signature of the Raman character of the excitations, that is, the absorption and emission processes of the photon occur coherently. On the other hand, the excitations in Pt metal were found to mostly form a diagonal pattern,[12] which corresponds to constant final photon energy and reflects fluorescence-like character. The difference of character in the incident energy dependence of RIXS can be attributed to the rate of incoherent scattering of the $5d$ electrons. If the scattering rate is high, the photoelectron in the $5d$ band quickly loses the information of the phase in the intermediate state of the RIXS process. In the metallic systems, such incoherent scattering is likely to be enhanced through the coupling with various low-energy modes and hence the fluorescence character is dominant in Pt. On the contrary, such coupling is small for the localized $5d$ electrons in the insulating Sr_2IrO_4.

We turn to the the assignment of the observed excitations. Two peaks have been recently observed by optical conductivity at 0.5 and 1.0 eV[1,3] and were ascribed to the Mott gap excitation between the $J_{\text{eff}} = \frac{1}{2}$ bands and an interband transition from the $J_{\text{eff}} = \frac{3}{2}$ band to the upper Hubbard band (quartet-doublet transition), respectively. Theoretical calculations of by the same group confirmed that the upper Hubbard band consists of the $J_{\text{eff}} = \frac{1}{2}$ state.[15] In contrast, another theoretical group proposed in their calculation of RIXS spectra that the local quartet-doublet transition should appear at $\frac{3}{2}\lambda = 0.5$-0.6 eV,[16] where λ is the spin-orbit coupling constant. In this work, it is also mentioned that the quartet-doublet transition will strongly mix with the charge excitation across the Mott gap by the intersite hopping. Very recently, Watanabe *et al.* clearly demonstrated in their theoretical work that the highest occupied band along the M-X line is composed of the $J_{\text{eff}} = \frac{1}{2}$ state while the bands with $J_{\text{eff}} = \frac{3}{2}$ character are located closer to the Fermi energy along the Γ-X and Γ-M lines, namely the highest occupied band is a mixture of the $J_{\text{eff}} = \frac{1}{2}$ and $J_{\text{eff}} = \frac{3}{2}$ states.[17] It is noted that the calculated band structure in the two theoretical works[15,17] are similar to each other. Focusing on the band gap in the theories, it approximates to 0.5 eV which coincides with the energy of the low-lying peak in our RIXS spectra. Therefore we ascribe the peak at 0.5 eV to the excitation across the band gap. Because the electron correlation is one of the important factors leading to the insulating state of Sr_2IrO_4, we refer the band gap to the Mott gap though the lower Hubbard band is a mixture of the $J_{\text{eff}} = \frac{1}{2}$ and $J_{\text{eff}} = \frac{3}{2}$ states. As for the peaks at 3.2 and 6.0 eV in the RIXS spectra, their excitation energies concur with the optical conductivity results[18] and they can therefore be ascribed to the charge-transfer excitations from the O $2p$ band to the upper Hubbard band and the $3z^2$-r^2 band, respectively. These excitations are respectively labeled A–D in Fig. 2(a) and a schematized band diagram is shown in Fig. 2(b) with the appropriate labels.

B. Momentum dependence

In order to discuss the momentum dependence, we describe the Brillouin zone and reduced momentum transfer ($\boldsymbol{q}$) used below. Because Sr_2IrO_4 has a layered-perovskite structure, momentum dependence along the c^* axis is expected to be small, as in high-T_c cuprates.[19] We therefore focus on the momentum dependence in the IrO_2 plane. In the chemical unit cell based on the space group $D_{4h}^{20} - I4_1/acd$, there are two crystallographically equivalent Ir atoms. Since the spectra at $\boldsymbol{Q} = (0,0,33)$ and $\boldsymbol{Q} = (0,1,33)$ are different albeit corresponding to equivalent momenta in the chemical unit cell, we take the unfolded Brillouin zone with one Ir atom in the unit cell to denote the reduced momentum transfer. In this notation, $\boldsymbol{Q} = (0,0,33)$ and $\boldsymbol{Q} = (0,1,33)$ correspond to $\boldsymbol{q} = (0,0)$ and $\boldsymbol{q} = (\pi,\pi)$, respectively.

The momentum dependence of the RIXS spectrum measured at $E_i = 11214$ eV is shown as a two-dimensional map in Fig. 3(a). The raw RIXS spectra of the high-symmetry points in the Brillouin zone are also shown in Fig. 3(b). We first discuss the excitation across the Mott gap which is closely related to the atypical Mott insulating state of Sr_2IrO_4. Along the $(0,0)$-$(\pi,0)$ direction, the peak position of the excitation is unchanged though the intensity varies slightly. Except for the substantial intensity enhancement at $\boldsymbol{q} = (\pi,\pi)$ which is discussed later, the momentum dependence is also weak along the $(0,0)$-(π,π) direction. Therefore we conclude that the dispersion relation of the Mott gap excitation in Sr_2IrO_4 is peculiarly weak.

In general, the RIXS spectra can be approximated as a convolution of the occupied and unoccupied bands. According to band calculation in the local density approximation including both the spin-orbit and the on-site Coulomb interactions (LDA + SO + U),[1] the dispersion of both occupied lower Hubbard band and unoccupied upper Hubbard band is much smaller than the present energy resolution. This is in concordance with the small momentum dependence observed for the 0.5-eV excitation in the RIXS spectrum. Interestingly, in the calculations without the spin-orbit interaction, the bands near the Fermi energy have larger dispersion irrespective of the inclusion of the Coulomb repulsion, as usually expected in $5d$ electrons, and this should correspond to a large momentum dependence of the Mott gap excitation in RIXS. In other words, the small momentum dependence of the Mott gap excitation is an intrinsic characteristic of the novel, spin-orbit-driven Mott insulating state of Sr_2IrO_4.

To extract the spectral weight corresponding to the enhancement at $\boldsymbol{q} = (\pi,\pi)$, we subtract the spectrum of $\boldsymbol{q} = (\frac{\pi}{2},\frac{\pi}{2})$, which can be regarded as a representative spectrum of the momentum-independent Mott gap excitation, from that of $\boldsymbol{q} = (\pi,\pi)$. The remainder, plotted in Fig. 3(c) as closed circles, is located near zero energy loss. The momentum point $\boldsymbol{q} = (\pi,\pi)$, which corresponds to $\boldsymbol{Q} = (1,0,33)$ in the absolute momentum transfer scale, is structurally forbidden in the space group $I4_1/acd$.[13] It is not a magnetic Bragg point either.[2] One possible origin of the spectral weight increase is the occurrence of a magnetic excitation. It was very recently demonstrated that magnetic excitations including the single magnon excitation can be observed by RIXS at the L edge.[20,21] If the IrO_2 plane is regarded as a two-dimensional antiferromagnet, $\boldsymbol{q} = (\pi,\pi)$ is the antiferromagnetic Bragg point. By analogy with La_2CuO_4,[22] antiferromagnetic correlations in the IrO_2 plane can be thought of as surviving even above T_N. We note that a canted antiferromagnetic transition occurs in Sr_2IrO_4 at $T_N = 250$ K[2,13] while the spectra were measured at room temperature. Thus, diffuse scattering from slow magnetic fluctuations or antiferromagnetic spin waves are likely to appear near zero energy loss at $\boldsymbol{q} = (\pi,\pi)$. Theoretically, the possibility to observe magnetic excitations with RIXS at the Ir L_3 edge has been suggested and the intensity of the magnetic excitation diverges at $\boldsymbol{q} = (\pi,\pi)$ in the theory,[16] which is consistent with our observation. The two-dimensional character of the excitation is supported by the experimental fact that the spectrum at $\boldsymbol{q} = (\pi,\pi)$ is independent of the momentum transfer along the c^* axis; in Fig. 3(b), the spectrum at $\boldsymbol{Q} = (0,1,30)$ (crosses) is completely identical to the one at $\boldsymbol{Q} = (0,1,33)$ (open circles). Because magnetic excitations are expected to appear around 100 meV,[16] improvement of the energy resolution is indispensable to identify the excitations as separated features from the elastic peak. In addition, polarization analysis may be able to ascertain the magnetic origin since magnetic excitations have a characteristic polarization dependence.[20,23]

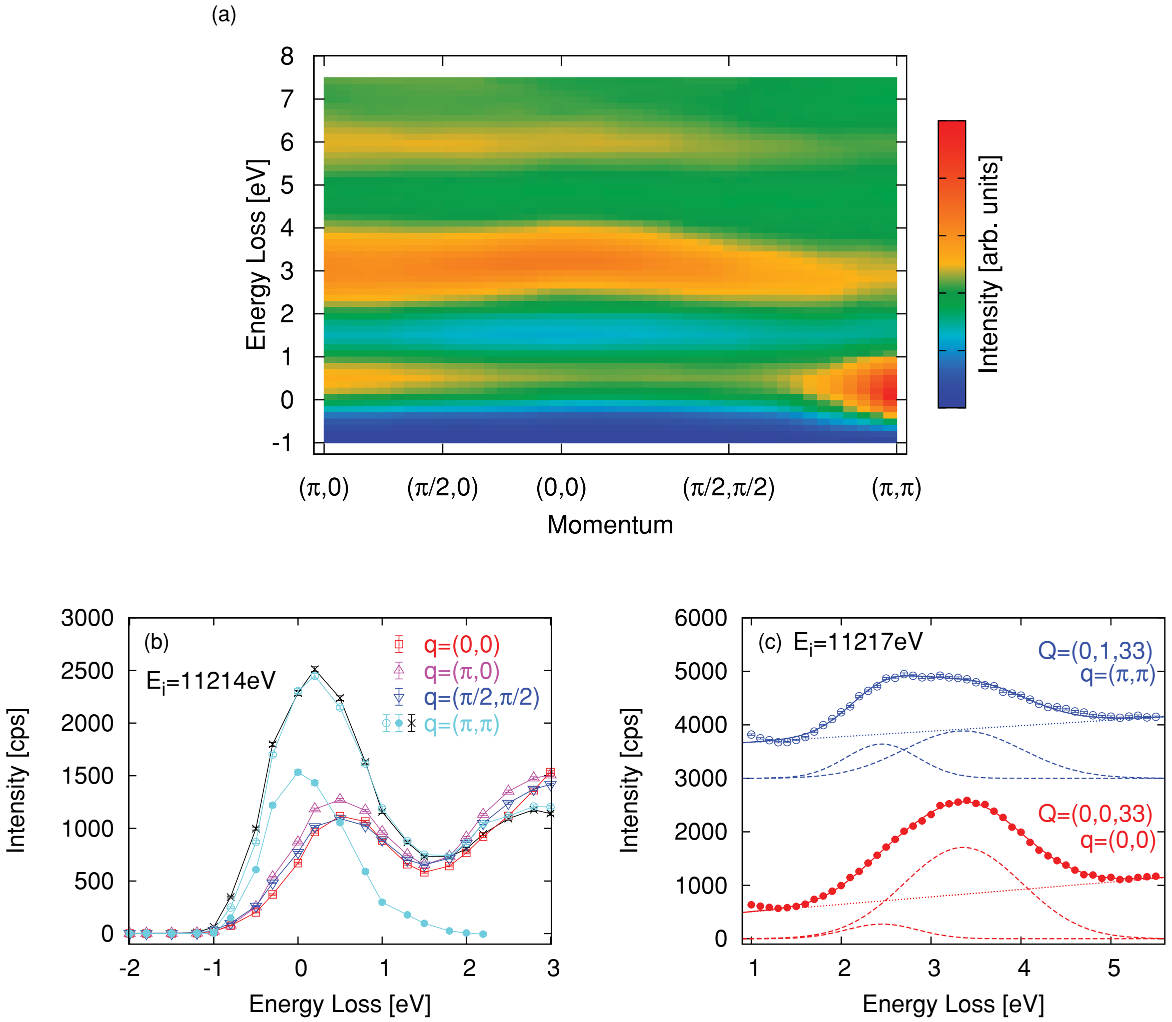

FIG. 3. (Color online) (a) Momentum dependence of RIXS spectrum plotted as a function of the reduced momentum transfer. The incident photon energy is 11214 eV and the absolute momentum transfer is $\boldsymbol{Q} = (h,k,33)$. (b) Enlarged view of the excitation across the Mott gap at the high-symmetry points of the Brillouin zone. Circles, triangles, and squares are the data taken at $\boldsymbol{Q} = (h,k,33)$ and the crosses are the spectra at $\boldsymbol{Q} = (0,1,30)$. Open symbols and crosses are the raw data. Filled circles are the difference spectrum obtained by subtracting the spectrum at $\boldsymbol{q} = (\frac{\pi}{2},\frac{\pi}{2})$ from the spectrum at $\boldsymbol{q} = (\pi,\pi)$. (c) Comparison of the charge-transfer excitation between $\boldsymbol{q} = (0,0)$ and (π,π). The latter spectrum is vertically shifted by 3000 cps. Circles are the experimental data. Dashed lines are the spectral weight of the charge-transfer excitation decomposed into two Gaussians. Solid lines are the sum of the two Gaussians and the background (dotted lines).

The charge-transfer excitation at 3.2 eV also depends on the momentum along the (0,0)-(π,π) direction while it does not along the (0,0)-$(\pi,0)$ direction. The momentum dependence along the (0,0)-(π,π) direction can be explained by the folding of the Brillouin zone. We compare the two spectra at $\boldsymbol{q} = (0,0)$ and (π,π) in Fig. 3(c). After subtracting a linear background from the experimental data, the spectral shape is well reproduced by two Gaussian components whose peak position and width are common for the two momenta. It means that the two momenta (0,0) and (π,π) are equivalent in terms of excitation energy and therefore the dispersion relation is folded at $(\frac{\pi}{2},\frac{\pi}{2})$ which is on the Brillouin zone boundary of the chemical unit cell. Only the intensity is different between the two momenta. On the other hand, such folding of the momentum dependence of RIXS has not been discussed so far in La_2CuO_4[6,24,25] though the unfolded Brillouin zone was taken to represent the reduced momentum transfer in the CuO_2 plane. In both Sr_2IrO_4 and La_2CuO_4, the Brillouin zone is folded in half by the $\sqrt{2}a \times \sqrt{2}a$ superstructure in the MO_2 ($M =$ Cu, Ir) plane, where a (∼3.8 Å) is the lattice parameter of the primitive unit cell. The superstructure is caused by the displacement of the oxygen atoms; the displacement is parallel to the plane in the iridate while it is perpendicular to the plane in the cuprate. As a result, the M–O–M bond angle departs from 180°. Because the bond angle of Sr_2IrO_4 (∠Ir–O–Ir ∼ 158°)[13] is smaller than that of La_2CuO_4 (∠Cu–O–Cu ∼ 174°),[26] the folding in the momentum dependence becomes apparent in the iridate. Similarly, the momentum dependence of the excitation at 6.0 eV may be explained by the folding of the Brillouin zone though decomposition of the peak into two components is impossible in the present spectra.

IV. SUMMARY

We have performed a resonant inelastic x-ray scattering study of Sr_2IrO_4. A particularly strong resonance effect is observed for the low-lying charge excitations near the Ir L_3 edge. The Mott gap excitation is observed at 0.5 eV and the momentum dependence of the excitation is found to be small. This is ascribed to the narrow $5d$ band governed by the spin-orbit interaction, which results in the novel Mott insulating state of Sr_2IrO_4. In addition, the data are suggestive of the occurrence of a magnetic excitation near $\boldsymbol{q} = (\pi,\pi)$. We also observed charge-transfer excitations at 3.2 and 6.0 eV, which show weak dispersion as well.

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