Electronic Excitations in $La_{1-x}Sr_xMnO_3$ Studied by Resonant Inelastic X-ray Scattering

Strongly correlated electron systems, such as transition metal oxides, have been an attractive issue in recent condensed matter physics [1]. Strong on-site Coulomb repulsion separates a partially filled band into the occupied lower Hubbard band (LHB) and the unoccupied upper Hubbard band (UHB), and the system becomes an insulator with a charge gap, known as the Mott gap. In the vicinity of the metal-insulator transition by carrier doping, these systems show not only novel physical properties such as high-temperature superconductivity in cuprates and colossal magnetoresistance (CMR) in manganites, but also a variety of electronic phases from the interplay of different degrees of freedom of charge, spin, orbital and lattice. The carrier doping in the strongly correlated electron systems affects the electronic structure in a wide energy range, and its reconstructions occur up to a few eV across the metal-insulator transition as well as at the magnetic or orbital ordering. Therefore, it is quite important to examine the electronic excitations in a wide energy-momentum space to understand the electronic properties of these compounds. In this respect, the resonant inelastic X-ray scattering (RIXS) in the hard X-ray regime is an ideal tool for the elucidation of the electron excitations around a few eV since it gives momentum-dependent spectra, unlike the conventional optical experiments. Here we report on a RIXS study of CMR materials, $La_{1-x}Sr_xMnO_3$.

The excitation across the Mott gap in the RIXS process at the Mn *K*-edge is schematically shown in Fig. 1. The initial state is (a). When the incident X-ray is absorbed, a core hole is created in the 1*s* orbital of an Mn site (b). Then, an electron transfers from surrounding atoms to an unoccupied Mn orbital to screen the core hole potential (c). After emitting the X-ray, the transferred electron remains in the Mn orbital as a final state (d).

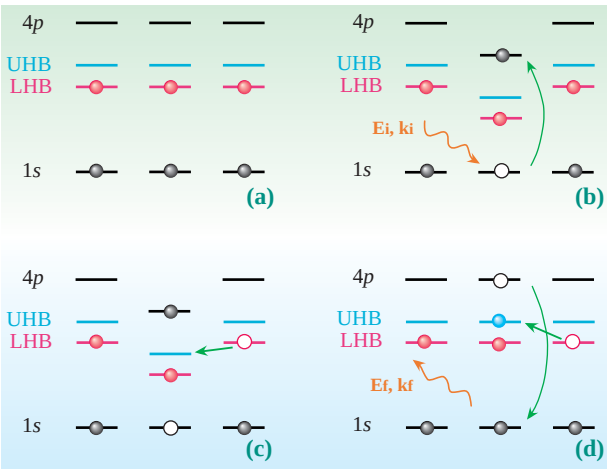

Fig. 1. Schematic picture of the Mn *K*-edge RIXS process.

The RIXS experiments were carried out at beamline **BL11XU**. A spectrometer for inelastic X-ray scattering is installed in this beamline [2]. The total energy resolution was 230-500 meV, depending on the selection of monochromators. The energy of the incident X-ray was tuned at 6556 eV, where the excitation across the Mott gap is most enhanced.

RIXS spectra of $LaMnO_3$ and $La_{0.6}Sr_{0.4}MnO_3$ are shown in Fig. 2. $LaMnO_3$ is a Mott insulator, while $La_{0.6}Sr_{0.4}MnO_3$ has enough holes to be metallic. In $LaMnO_3$, the peak at 2.5 eV corresponds to the excitation from the effective LHB hybridized with the O 2*p* orbital to UHB across the Mott gap, which shows a weak dispersion in momentum dependence and an apparent polarization dependence of scattering intensity. These experimental facts are related to the orbital order in $LaMnO_3$ and are consistent with theoretical calculation. The peaks at higher energies of 8 and 11 eV are the charge transfer excitations from O 2*p* to Mn 3*d* and from O 2*p* to Mn 4*s*/4*p*, respectively [3]. It should be emphasized that a salient peak remains even in metallic $La_{0.6}Sr_{0.4}MnO_3$, as seen in Fig. 2(a). We show the spectra at a higher scattering angle in Fig. 2(b), where the elastic scattering decreases and the spectral shape at low energy can be seen more clearly. Since we confirmed that the momentum dependence of energy dispersion for the Mott gap excitation is small in both compounds, the difference of momentum between two spectra is not important. A gap feature was observed in insulating $LaMnO_3$, while the gap is partially filled in metallic $La_{0.6}Sr_{0.4}MnO_3$. Even though the Mott gap is filled in $La_{0.6}Sr_{0.4}MnO_3$, a fairly large spectral weight remains at the excitation from the LHB to the UHB even in the metallic state. The momentum and polarization dependences of the Mott gap excitation are qualitatively similar to those of $LaMnO_3$, even though a static orbital order disappears in $La_{0.6}Sr_{0.4}MnO_3$. These results indicate that the strong Coulomb repulsion and the orbital correlation robustly persist

even in the metallic state of $La_{0.6}Sr_{0.4}MnO_3$.

Figure 3 shows the temperature dependence of the Mott gap excitation in $La_{0.8}Sr_{0.2}MnO_3$, which shows a metal-insulator transition accompanied by a ferromagnetic transition at Tc = 309 K [4]. The RIXS intensity of Q = (2.7, 0, 0) increases with decreasing temperature, whereas that of Q = (2.2, 2.2, 0) is independent of temperature. The temperature dependence of intensity depends on the direction of the scattering vector, and the intensity along the <h00> direction roughly accords with the bulk magnetization. We consider that the difference in temperature dependence between the <h00> and <hh0> directions may be attributed to the anisotropy of the magnetic interaction related to the orbital correlation, as is often discussed. The present result indicates again that the orbital degree of freedom is important for electronic properties in $La_{1-x}Sr_xMnO_3$.

In summary, our results demonstrate that the momentum-dependent spectra of RIXS are important in studying electronic structure and in elucidating the underlying interactions of electronic degrees of freedom in strongly correlated electron systems.

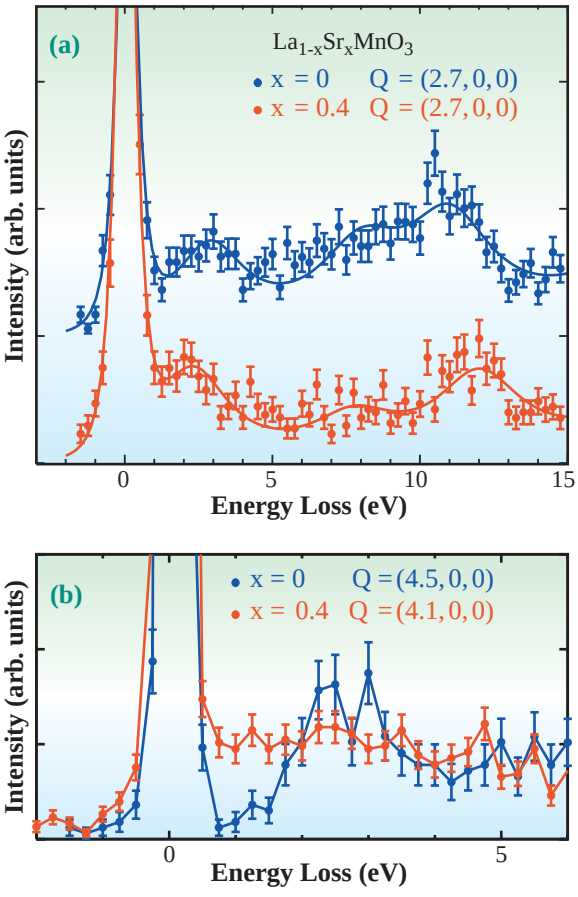

Fig. 2. RIXS spectra of $LaMnO_3$ and $La_{0.6}Sr_{0.4}MnO_3$ at Q = (2.7, 0, 0) **(a)** and at higher scattering angle **(b)**. The energy resolution is 230 meV.

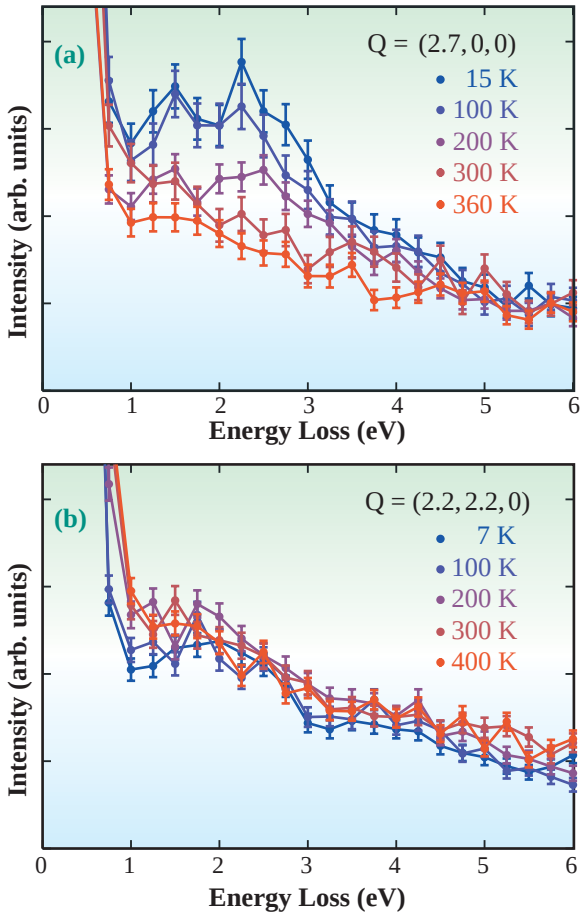

Fig. 3. RIXS spectra of $La_{0.6}Sr_{0.4}MnO_3$ at some temperatures. The scattering vectors are Q = (2.7, 0, 0) for **(a)** and Q = (2.2, 2.2, 0) for **(b)**, respectively. The energy resolution is about 500 meV.

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