Significance of the inter-site Coulomb interaction between the O 2p and Cu 3d holes revealed by resonant inelastic x-ray scattering of Sr$_{14}$Cu$_{24}$O$_{41}$

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Abstract. We have carried out resonant inelastic x-ray scattering (RIXS) measurements at the Cu K-edge on Sr$_{14}$Cu$_{24}$O$_{41}$, which has two kinds of layers, namely, the ladder layer and the chain layer. The excitation from the Zhang–Rice singlet band to the upper Hubbard band (UHB) shows a large dispersion with a periodicity of the ladder layer. The excitation corresponding to the ligand-to-metal charge transfer is nearly dispersionless. Comparison between the RIXS data and theoretical calculations shows that the inter-site Coulomb interaction between the Cu 3d and O 2p holes plays a prominent role in the RIXS process. This interaction is expected to be important in the hole state in the ladder layer of this compound.

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1. Introduction

A variety of interesting phenomena such as superconductivity and sliding density waves which are related to spin and charge degrees of freedom of quasi-one-dimensional ladders and chains have been found in Sr$_{14-x}$Ca$_x$Cu$_{24}$O$_{41}$. In this material, two kinds of layers composed of copper and oxygen are alternately stacked along the $b$-axis direction [1]–[3]. One contains two-legged Cu$_2$O$_3$ ladders composed of corner-sharing CuO$_4$ plaquettes, and the other contains CuO$_2$ chains composed of edge-sharing CuO$_4$ plaquettes. Both layers are quasi-one-dimensional along the $c$-axis with different lattice periodicity, $c_{\text{ladder}}$ and $c_{\text{chain}}$, where $7c_{\text{ladder}} \simeq 10c_{\text{chain}}$.

Superconductivity has been found under pressure for $x \geq 11.5$ in Sr$_{14-x}$Ca$_x$Cu$_{24}$O$_{41}$, whereas smaller $x$ tends to make the system insulating [4]–[6]. Properties of the insulating systems in the lower $x$ range have been studied experimentally by such methods as nuclear magnetic resonance (NMR) and neutron diffraction [7]–[23]. In the ground state of the insulating systems, it is considered that some kind of superstructure (dimerization, charge density wave (CDW), or hole ordering) is formed and motion of this superstructure is expected at higher temperatures (sliding density wave). Although the nominal valence (+2.25) of Cu is independent of $x$, the holes are transferred to the ladder layer from the chain layer with increasing $x$ [9]. The increase of the hole concentration in the ladder layer suppresses the insulating CDW phase and induces superconductivity.

Resonant inelastic x-ray scattering (RIXS) is a powerful tool for studying strongly correlated electron systems because it can clarify bulk electronic states and probe the momentum dependence of low-energy excitations across the Fermi level ($E_F$) in solids [24]–[32]. Since RIXS provides combined information about occupied and unoccupied states, it is complementary to angle-resolved photoemission spectroscopy (ARPES), which detects the occupied states. RIXS can be applied not only to metals but also to insulators, which cannot be simply studied by photoemission spectroscopy. Another merit of RIXS is its bulk sensitivity since its probing depth is more than several micrometres. RIXS is hence more bulk sensitive than the bulk sensitive soft x-ray ARPES, whose probing depth can reach 15 Å.

In this paper, we report on the results of RIXS measurements for a single crystal of the parent material Sr$_{14}$Cu$_{24}$O$_{41}$ in order to elucidate the electronic states by keeping in mind that contributions of both ladder and chain layers coexist in the spectrum. We first present the excitation energy ($h\nu$) dependence of the RIXS spectrum in a wider energy-loss region than the previous report [32]. By doing so, we can show the presence of a strong $h\nu$ dependence in the spectral lineshape and discuss the contributions from the two layers. We next present the momentum dispersion of the RIXS spectrum and compare the experimental results with a theoretical calculation based on the d–p model. It is found that Coulomb interaction between...
the Cu 3d hole and the O 2p hole \((U_{pd})\) is playing an essential role in the RIXS process in this material in contrast to the previous report [32]. This is in accordance with the discussions taking \(U_{pd}\) into account based mainly on optical studies of cuprates [33]–[38].

2. Experimental

We carried out the RIXS measurements at BL19LXU of SPring-8 with a 27 m long x-ray undulator [39]. The undulator radiation tuned to a proper energy range was monochromatized by two Si (111) crystals. We have used two channel-cut Si (220) crystals to further monochromatize the x-ray. This light was incident onto the polished (001) surface of a \(\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}\) crystal kept at room temperature in a vacuum chamber. The scattering plane was set to the (010) plane of the sample and the polarization vector of the incident light was in this plane. The scattered radiation with the wavenumber transfer \((\Delta k)\) parallel to the CuO chain and the ladder’s leg direction, i.e. the [001] direction, was energy analyzed by a spherically bent Si (553) crystal which is mounted on a goniometer. The focused and energy-analyzed scattered light was detected by a NaI scintillator and counted by single-channel analysis. The total energy resolution, judged from the full width at half maximum of the elastic peak, was about 0.4 eV.

3. Results and discussion

Figure 1(a) shows the Cu 1s x-ray absorption spectrum (XAS) measured by fluorescence yield. Labels (A)–(E) show the photon energies at which RIXS measurements were carried out. The tiny peak located at the photon energy \((h\nu)\) of 8.981 keV (A) is attributed to the so-called ‘Cu 1s–3d quadrupole’ transition, to which contribute not only the genuine Cu 1s–3d quadrupole transition but also the transition from the Cu 1s to the Cu 3d and 4p mixed state through the Cu 1s–4p dipole allowed transition [40]. On-site Cu 3d–4p mixing can take place if the Cu site is not at the center of the inversion symmetry. When the Cu site is at the center of the inversion symmetry, on the other hand, the Cu 4p state can be mixed with the 3d state on the neighboring Cu site. In the present case, both channels of dipole allowed transition may be contributing since violation of inversion symmetry is seen on the ladder layer.

The shoulder feature located at around \(h\nu = 8.990\) keV (B) mainly reflects the Cu 1s–4p\(\pi\) transition and the main peak centered around \(h\nu = 9.000\) keV (D) mainly reflects the 1s–4p\(\sigma\) absorption [41]. Shoulder features are also observed at \(h\nu = 8.996\) (C) and 9.004 (E) keV. Calculation of XAS by the d–p model [42] indicates that, for each of the 1s–4p\(\pi\) and 1s–4p\(\sigma\) excitations, there are two types of XAS final states (intermediate states of the RIXS process) characterized by different screening processes of the 1s core hole [31]. One is the poorly screened state, in which the excited Cu site is in the \(cd^9\) state, where \(c\) represents the 1s hole. The other is the well-screened state, in which the d hole is filled by an O 2p electron and the excited Cu site is in the \(cd^{10}\) state. The poorly screened state is roughly 7 eV higher in energy than the well-screened state. Therefore, for the 1s–4p\(\pi\) excitation, the well-screened intermediate state corresponds to the structure (B) and the poorly screened state contributes to the region around (C) and (D). For the 1s–4p\(\sigma\) excitation, the well-screened state makes the main contribution to the main peak around (C) and (D) and the poorly screened state contributes to the higher \(h\nu\) region around (E).

Figures 1(b) and (c) show the \(h\nu\) dependence of the RIXS spectra at \(\Delta k = 3.0\pi/c_{\text{ladder}}\) and \(\Delta k = 3.0\pi/c_{\text{chain}},\) where \(c_{\text{ladder}}\) and \(c_{\text{chain}}\) are 3.923 and 2.726 Å, respectively [21]. In these

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Figure 1. (a) XAS near the Cu K-edge of Sr$_{14}$Cu$_{24}$O$_{41}$. Labels (A)–(E) indicate the excitation photon energies $h\nu$ at which RIXS spectra in figures 1(b) and 1(c) were measured, where (A) $h\nu = 8.981$ keV, (B) 8.990 keV, (C) 8.996 keV, (D) 8.999 keV and (E) 9.006 keV. (b) and (c) $h\nu$ dependence of the RIXS spectra of Sr$_{14}$Cu$_{24}$O$_{41}$ at room temperature measured at $\Delta k = 3.0\pi/c_{\text{ladder}}$ and $\Delta k = 3.0\pi/c_{\text{chain}}$, respectively. Labels (A)–(E) correspond to the $h\nu$ shown in figure 1(a). The vertical lines indicate the structures in the spectra.

In order to understand the origin of the different intensity ratio, we compare our results with the $h\nu$-dependent RIXS spectra of Sr$_2$CuO$_3$ and CuGeO$_3$ [31], which have
Figure 2. (a) Δk dependence of the RIXS spectra at room temperature measured at $h\nu = 8.996$ keV. (b) The second derivative plot based on 2(a). The two black dotted lines are guides to the eye.

corner-sharing CuO$_3$ chains and edge-sharing CuO$_2$ chains, respectively, corresponding to the ladder and chain layers of the present system. In the corner-sharing Sr$_2$CuO$_3$, the RIXS spectra are composed mainly of two structures at around 3 and 5 eV. The 3 eV structure is much stronger than the 5 eV structure at $h\nu = 8.997$ keV. With increasing $h\nu$ in this region, the 3 eV structure becomes weaker and the 5 eV structure becomes stronger. At $h\nu = 9.005$ keV, the 3 eV structure disappears. In the edge-sharing CuGeO$_3$, the spectra show three structures around 1.6, 3.7 and 6.3 eV. The structure at around 6.3 eV is much stronger than the other structures at all $h\nu$. Both 3.7 and 6.3 eV structures increase with increasing $h\nu$. In the present system, the structure at around 5.5 eV in both of figures 1(b) and (c) is strong at $h\nu = 8.999$ keV (D) and its intensity decreases with decreasing $h\nu$. This tendency is in agreement with the behavior of the high energy-loss feature of both the corner-sharing Sr$_2$CuO$_3$ (∼5 eV) and the edge-sharing CuGeO$_3$ (∼6.5 eV). The structure near 4 eV is stronger than the structure near 5.5 eV at $h\nu = 8.996$ keV (C), whereas it decreases with increasing $h\nu$. This tendency rather resembles that of the 3 eV structure of the corner-sharing Sr$_2$CuO$_3$. Therefore, the 4 eV structure is mainly attributed to the ladder layer and the 5.5 eV structure is attributed to both ladder and chain layers. The low energy-loss structure around 2 eV at 8.981 keV (A) in figures 1(b) and (c) can be interpreted to correspond to the 1.6 eV structure in CuGeO$_3$.

Next, we measured the momentum dependence of the RIXS spectra at $h\nu = 8.996$ keV (C) as shown in figure 2. Figure 2(a) is the raw data of the momentum dependence. The normalization is performed by the intensity of the elastic peak at 0 eV. In this figure, at least two branches are recognized. One is located between 5.5 and 6 eV and is nearly dispersionless.
The other is located in the region between 4.5 and 3 eV. It is difficult to directly determine the dispersions from figure 2(a) alone. Therefore, the second derivative of figure 2(a) is plotted in figure 2(b) as a function of $\Delta k$, where interpolation in terms of $\Delta k$ and a smoothing procedure are applied. The white zone corresponds to the $\Delta k$ region not investigated because of the strong Bragg reflection. Broken black lines are guides to the eye, indicating dispersions.

In figure 2(b), the branch between 3 and 4 eV shows a relatively large dispersion with energy loss maxima at around $\Delta k = 3.0\pi/c_{\text{ladder}}$ and $5.0\pi/c_{\text{ladder}}$ (corresponding to $\Delta k = 2.1\pi/c_{\text{chain}}$ and $3.5\pi/c_{\text{chain}}$). Namely, the periodicity of the dispersion indicates that this branch comes from the ladders. As discussed above, both the ladders and the chains are expected to contribute to the dispersionless 5.5 eV branch. Since only one branch is seen even in the second-derivative plot (figure 2(b)), we interpret that the RIXS peaks of the ladders and the chains coexist near 5.5 eV.

In order to understand the dispersions in these RIXS spectra, we compare our results with the d–p model theoretical calculations. We have calculated the RIXS process of the corner-sharing structure by using the finite-size cluster d–p model [37, 42], which exactly takes into account the electron correlation. The electron correlation is important in the intermediate state as well as in the ground and final states. In this calculation, the Cu$_6$O$_{18}$ cluster, namely the corner-sharing one-dimensional array of six CuO$_4$ plaquettes as shown in figure 3(a), is used as a model for the ladder layer of Sr$_{14}$Cu$_{24}$O$_{41}$. We consider the Cu 3d$_{x^2−y^2}$ and O 2p orbitals in the d–p model Hamiltonian. The parameters used were the charge-transfer (CT) energy $\Delta (\equiv \varepsilon_d − \varepsilon_p)$, d–p hybridization ($dp\sigma$), p–p hybridization ($pp\sigma$ and $pp\pi$, where $pp\pi = −0.3pp\sigma$), d–d on-site Coulomb interaction $U_{dd}$ and Coulomb repulsion between the Cu 3d and Cu 1s holes ($Q$). We cannot reproduce the RIXS spectra by using only these parameters and thus take the d–p inter-site Coulomb interaction ($U_{dp}$) into account as a new parameter. For consistency, we also take into account the O 2p on-site Coulomb interaction ($U_{pp}$). The parameter values used in the calculation are as follows: $\Delta = 3.5$ eV, ($dp\sigma) = −1.5$ eV, ($pp\sigma) − (pp\pi) = 0.65$ eV, $U_{dd} = 9.0$ eV, $U_{pp} = 4.0$ eV, $U_{pd} = 1.5$ eV and $Q = 7.0$ eV.

It should be noted that these parameters include renormalization due to the screening by electrons that are not considered in the present model although the screening is less efficient than that in metallic systems. Reductions of $U_{dd}$ and $Q$ at the Mn site in CdTe, and the difference in the screening mechanisms between $U_{dd}$ and $Q$, are discussed by Gunnarsson et al [43]. It should also be noted that the present $U_{dd}$, which is the repulsive interaction between two holes in one 3d$_{x^2−y^2}$ orbital, is larger by roughly 2 eV than the mean value of the repulsive interaction between two holes in any of the five 3d orbitals on one site [44].

Cu 1s XAS calculated with the cluster of figure 3(a) is shown in figure 3(b). The origin of the relative photon energy ($\omega$) is arbitrary in the figure. The figure shows a two-peak structure separated by about 7 eV. The low-(high-)energy peak located at around $\omega \sim 3(10)$ eV is the well-(poorly-)screened peak. In the present model, the 4p band is considered as a single level. In reality, there are two 4p bands, 4$\sigma$ and 4$\pi$ whose energy difference is 6–7 eV, as mentioned above. Therefore, in order to compare with the experimental XAS result shown in figure 1(a), we should consider another spectrum, of the same shape but different overall intensity, shifted by 6–7 eV and add it to the original spectrum. Since both the 4$\sigma$ and 4$\pi$ bands have certain bandwidths, these two XAS spectra have to be broadened before being added.

Figure 3(c) shows the RIXS spectra calculated by using the above mentioned parameters and the incident photon energy of the arrow in figure 3(b). The obtained spectra are broadened by a Lorentz function with a full width at half maximum of 1.0 eV by taking into account

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the lifetime of the excited final states. In the figure, the spectra for four $\Delta k$-points $(0, 1/3\pi, 2/3\pi, \pi)$ are shown, where $\Delta \omega$ corresponds to the energy loss in the experimental spectra. Two main features are found as indicated by the arrows. The low energy-loss feature has a large dispersion with the lowest energy loss of $\Delta \omega \sim 2.8$ eV at $\Delta k = 0(2n\pi)$ and the highest energy loss of $\Delta \omega \sim 3.8$ eV at $\Delta k = \pi((2n + 1)\pi)$. The higher energy-loss structure is nearly dispersionless and is located at about 5.6 eV. The experimentally observed low energy-loss dispersion between $\sim 3$ and $\sim 4$ eV with the periodicity of $2\pi/c_{\text{ladder}}$ (see figure 2(b)) corresponds closely to the dispersion feature between 2.8 and 3.8 eV in figure 3(c). On the other hand, the 5.5 eV feature in figure 2(b) corresponds to the 5.6 eV structure in figure 3(c), which is nearly dispersionless. From the calculation, the low-energy excitation with $\sim 1$ eV dispersion is due to the excitation from the Zhang–Rice singlet band (ZRSB) [45] to the upper Hubbard band.
(UHB). The dispersionless structure around 5.5 eV corresponds to the excitation described as ‘ligand-to-metal charge transfer’ excitation [42].

We could successfully reproduce the experimental RIXS spectra by taking into account $U_{pd}$ in the calculation. As in the optical excitation [33]–[38], the low energy edge of the ZRSB–UHB excitation in RIXS is given by $\Delta + U_{pd}$ in the limit of vanishing d–p hybridization. Accordingly, if we set $U_{pd}$ to zero in the present calculation, the RIXS spectra as a whole are shifted to the elastic line by about 1.5 eV, which cannot reproduce the experimental results at all. When we increase $\Delta$ in order to reproduce the energy position of the experimental result without $U_{pd}$, the dispersion of the calculated result becomes too small compared with the experimental results. By introducing $U_{pd}$, the energy position shifts to higher energy loss without much reducing the dispersion. Therefore, we can conclude that the observed size of the dispersion of the ZRSB→UHB excitation of about 1 eV can be explained only by considering $U_{pd}$ of the order of 1.5 eV. This value of $U_{pd}$ is in semiquantitative agreement with $U_{pd}$ inferred from the excitons observed by Raman experiments in insulating layered cuprates [34]–[36].

As mentioned above, the importance of $U_{pd}$ has already been suggested in the optical study, where its effect is to enlarge the interband excitation energy. Another effect of $U_{pd}$ is to bind excitonically the hole in the ZRSB and the electron in the UHB, which exist in the RIXS final state belonging to the ZRSB→UHB branch. The hole in the ZRSB is mainly composed of the $d^9 L$ state, where $L$ represents a hole at the oxygen site, and the electron in the UHB is mainly represented by the $d^{10}$ state. If we consider a cluster of Cu–O–Cu–O–Cu–O–Cu, the ground state is approximately $|d^9 0 d^9 0 d^9 0 d^9\rangle$, where ‘0’ represents a p$^6$ state on the O site. Just after the ZRSB→UHB excitation, the state $|d^9 0 d^{10} L d^9 0 d^9\rangle$ is realized, increasing the system energy by $\Delta + U_{pd}$ relative to the ground state because there is a pair of neighboring p and d holes ($L$ and $d^9$). If the hole in the ZRSB and the electron in the UHB propagate independently, a state $|d^9 0 d^{10} 0 d^9 L d^9\rangle$ with an energy of $\Delta + 2U_{pd}$ is realized because there are two pairs of $L$ and $d^9$. Therefore, $U_{pd}$ tends to bind together the hole in the ZRSB and the electron in the UHB.

4. Conclusions

In conclusion, we carried out RIXS measurements to probe the electronic excitations in Sr$_{14}$Cu$_{24}$O$_{41}$. By a comparison with the finite-size large cluster calculation based on the d–p model, the observed features are well interpreted, strongly suggesting that the O 2p–Cu 3d inter-site Coulomb interaction $U_{pd}$ plays an essential role. $U_{pd}$ is expected to play an important role in the behavior of the holes introduced in the ladder layer since the hole exists mainly on the oxygen site ($L$ state). Because the hole state controls the various interesting properties of Sr$_{14-3x}$Ca$_x$Cu$_{24}$O$_{41}$ as mentioned above, the present result confirming that $U_{pd}$ is of the order of 1.5 eV is expected to have a significant meaning in the physics of this material.

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