Charge Excitations in Nd$_{2-x}$Ce$_x$CuO$_4$ Observed with Resonant Inelastic X-ray Scattering: Comparison of Cu K-edge with Cu L$_3$-edge

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We report a Cu K-edge resonant inelastic x-ray scattering (RIXS) study of momentum-dependent charge excitations in Nd$_{2-x}$Ce$_x$CuO$_4$ ($x = 0.075$ and $0.18$). The peak position and width of the excitations coincide excellently with those observed in Cu L$_3$-edge RIXS. It demonstrates that the same charge excitations are observed at the two edges.

Low-energy electron dynamics in high-$T_c$ cuprates is characterized by the motion of charge and spin. Among the various experimental technique for studying the electron dynamics, resonant inelastic x-ray scattering (RIXS) has gained a great deal of attention because one can measure electronic excitation spectra with momentum resolution and element selectivity.$^{1,2}$ Cu K- and L$_3$-edges are mostly used for the RIXS study of the cuprates. Energy resolution has been improved significantly in the last two decades and it reaches a few tens of meV at best at the edges.$^{3,4}$ While spin excitation (single spin-flip process) is allowed only at the $L_3$-edge, charge excitation can be observed at both edges.

In the electron-doped cuprate Nd$_{2-x}$Ce$_x$CuO$_4$ (NCCO), momentum-dependent charge excitations, which are located at higher energy than the spin excitations, are indeed observed in the K-$^5$-$^7$ and $L_3$-edge RIXS spectra,$^7$-$^9$ even though different interpretations, intraband particle-hole excitations$^5$-$^7$ and a certain mode associated with a symmetry-breaking state,$^8$ were proposed. Recently, the charge excitations in the $L_3$-edge RIXS were found to depend not only on the the in-plane momentum but also on the out-of-plane one and ascribed to a plasmon mode$^9$ which was proposed theoretically.$^{10,11}$ In order to further investigate the character of the charge excitations, for example, contrasting behavior at high temperature

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within a slight difference of carrier concentration (≈0.02 electron per Cu atom),\(^8\) it is important to verify whether the same excitation is observed at the two edges because each edge has a suited energy-momentum range for observing the excitations. In Ref. 7, the charge excitations at the \(K\)-edge are compared with the one at the \(L_3\)-edge. However, the comparison is made only at a few momentum points and the out-of-plane momentum is not considered. In this short note, we report a Cu \(K\)-edge RIXS study of NCCO in comparison with the \(L_3\)-edge and conclude that the momentum-dependent charge excitations in the Cu \(K\)-edge RIXS spectra are the same as in the \(L_3\)-edge.

RIXS experiments were performed at BL11XU of SPring-8. Incident x rays were monochromatized by a Si(111) double-crystal monochromator and a Si(444) channel-cut monochromator, and horizontally scattered x rays were analyzed in energy by a Ge(733) analyzer. Experimental geometry was the same as the previous works;\(^5,7\) \(\pi\)-polarized incident photons with 8991 eV were irradiated on the \(ac\)-plane of NCCO, but the energy resolution of 100 meV was improved better than the works.\(^5–7\) All the spectra were taken at 10 K. We use Miller index \((H, K, L)\) of the body-centered-tetragonal crystallographic unit cell for momentum transfer \((Q)\).

Figure 1 shows the Cu \(K\)-edge RIXS spectra of NCCO. A peak at 2 eV and a momentum-dependent feature below the peak are consistent with the previous work,\(^5\) but the improved energy resolution enables us to observe the latter more clearly, especially at low in-plane momenta \((H, K)\). The 2-eV peak is an interband excitation across the charge-transfer gap and it is also observed in parent Nd\(_2\)CuO\(_4\).\(^6\) On the other hand, the momentum-dependent feature appears when electrons are doped. We fit the spectra by the sum of elastic scattering, the momentum-dependent charge excitation, the interband excitation, and a tail of high energy charge excitation peaked around 6 eV.\(^12\) The experimental resolution is considered for the momentum-dependent charge excitation and the interband excitation. Filled circles in Fig. 2(a) and (b) are the peak position and width (full-width at half maximum) of the momentum-dependent feature obtained from the fitting analysis, respectively. Because intensity of the feature is roughly proportional to the carrier density, the feature of \(x = 0.075\) forms continuum-like spectral shape in contrast to the salient peak in \(x = 0.18\). It makes the fitting analysis of \(x = 0.075\) difficult and this is the reason why the error is larger than \(x = 0.18\).

Open circles in Fig. 2(a) are the peak positions in our Cu \(L_3\)-edge study,\(^7\) where the crystals from the same batch as this work were used. Momentum transfer along the \(c^*\)-direction \((L)\) varies between 1.55 and 1.65 and it is almost equivalent to that in the present data in Fig. 1, considering that the dispersion is folded at even numbers of \(L\).\(^9\) The peak positions
Fig. 1. (Color Online) RIXS spectra of NCCO for (a) $x = 0.075$ and (b) $x = 0.18$. Filled circles are the experimental data and solid lines are the fitting results of elastic (light gray), momentum-dependent charge excitation (red), interband excitation across the charge-transfer gap (blue), tail of high-energy excitation (dark gray), and sum of the all components (black). Peak position of the momentum-dependent charge excitation is indicated by vertical bars in each spectrum.
Fig. 2. (Color Online) Peak position and width of the momentum-dependent charge excitations in the electron-doped cuprates. Filled circles are the fitting results of the Cu K-edge RIXS in Fig. 1. Open circles and squares are those of the Cu L$_3$-edge taken from Refs. 7 and 9, respectively. The width cannot be evaluated reliably in the spectra of Ref. 7 because the tail of the excitation overlap considerably with the spin and dd excitations due to the low energy resolution and the experimental condition which is suitable for observing the spin excitations.

are consistent between the Cu K- and L$_3$-edges. In addition, we plot the peak positions of La$_{2-x}$Ce$_x$CuO$_4$ (LCCO) of $x = 0.175$ and $L = 1.65$ (open squares) taken from a recent high-energy-resolution work at the Cu L$_3$-edge. The positions of LCCO $x = 0.175$ agree very well with those of NCCO $x = 0.18$.

In Fig. 2(b), the width of the peak is compared between the K-edge and the L$_3$-edge. The momentum dependence of the width of LCCO $x = 0.175$ at the L$_3$-edge is connected smoothly to that of NCCO $x = 0.18$ at the K-edge. It means that the broadening of the peak comes from intrinsic electronic properties of the cuprates rather than some effects in the RIXS process. While momentum-independent lifetime of electron is phenomenologically taken into account in a theoretical work, RIXS can provide experimental data to discuss microscopic origin of the broadening quantitatively.

The excellent agreement of the peak position and width between NCCO $x = 0.18$ and LCCO $x = 0.175$ proves that the momentum-dependent charge excitation in the Cu K-edge RIXS spectra has the same origin as the dispersive mode observed at the Cu L$_3$-edge. Our result approves the complementary use of the two edges for exploring the charge excitations throughout the energy-momentum space. In general, the L$_3$-edge is advantageous for the ex-
citations at low energy due to weak elastic scattering while it has limitation of the accessible Brillouin zone. In the case of cuprates, huge $dd$ excitations above 1.5 eV hampers the observation of the charge excitations at the energy. On the other hand, the $K$-edge does not have the shortcomings of the $L_3$-edge, but it is difficult to measure the inelastic signal at very low energy. Even though the lower limit in the energy resolution of this study is 0.4-0.5 eV, we will have change to access lower energy if the best resolution (25 meV) is achieved.$^3$

Another finding is that the peak position of the charge excitations shifts to higher energy with increasing electron doping. Although the data of $x = 0.075$ scatter, Fig. 2(a) shows that the peak position of $x = 0.18$ is higher in energy than that of $x = 0.075$. Such doping dependence has been reported in the $L_3$-edge RIXS work$^9$ as a character of plasmon excitation and we confirm it here at the $K$-edge. It is noted that doping dependence was measured at higher in-plane momentum in the previous $K$-edge work$^5, 6$ and it just shows increase of intensity with increasing doping. It may indicate that the high-energy shift is limited at low in-plane momentum and doping effect of the charge excitations changes at a certain momentum.

In summary, we performed a Cu $K$-edge RIXS study of NCCO and analyzed the momentum-dependent charge excitations below the charge transfer gap. The peak position and width agree very well with those at the Cu $L_3$-edge and we conclude that the same charge excitations are observed at the two edges.

Acknowledgment

The authors would like to thank T. Tohyama and H. Yamase for fruitful discussion. The synchrotron radiation experiments were performed at the BL11XU of SPring-8 with the approval of the Japan Synchrotron Radiation Research Institute (JASRI) (Proposals No. 2013B3502 and 2014A3502). This work was financially supported by JSPS KAKENHI Grants No. 25400333 and No. 16H04004.
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