Momentum Dependence of Charge Excitations in the Electron-Doped Superconductor Nd$_{1.85}$Ce$_{0.15}$CuO$_4$: a RIXS Study

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We report a resonant inelastic x-ray scattering (RIXS) study of charge excitations in the electron-doped high-$T_c$ superconductor Nd$_{1.85}$Ce$_{0.15}$CuO$_4$. The intraband and interband excitations across the Fermi energy are separated for the first time by tuning the experimental conditions properly to measure charge excitations at low energy. A dispersion relation with $q$-dependent width emerges clearly in the intraband excitation, while the intensity of the interband excitation is concentrated around 2 eV near the zone center. The experimental results are consistent with theoretical calculation of the RIXS spectra based on the Hubbard model.

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The asymmetric features of the electronic phase diagram of the doping dependence of the Mott insulating Cu oxides between hole- and electron-doping have been an issue of debate for a long time. Their exploration is very important for the understanding not only of the mechanism of high-$T_c$ superconductivity but also of the effects of doping on a Mott insulator. Experimental studies investigating the reconstruction of the electronic bands by the carrier doping have extensively been pursued [1, 2], and now the comprehension was reached that the Mott gap feature remains up to considerable doping levels. Combined with an extensive theoretical analysis, we can observe not only the interband excitation across the Mott gap but also the intraband excitation within the UHB throughout the whole Brillouin zone.

The RIXS experiments were carried out on the IXS spectrometer installed at the beam line 11XU of SPring-8. A Si (111) double-crystal monochromator and a Si (400) channel-cut secondary monochromator were utilized. Horizontally scattered x-rays were analyzed in energy by a bent Ge (733) crystal. The overall energy resolution is about 400 meV estimated from the full width at half maximum (FWHM) of the quasielastic scattering. Single crystals of Nd$_{2-x}$Ce$_x$CuO$_4$ of $x = 0.15$ and 0.075 were prepared, which show superconductivity below $T_c = 25$ K, and antiferromagnetic order below $T_N \sim 120$ K, respectively. The surface of the crystal is normal to the $c$-axis, which was kept in the scattering plane so as to be scanned in the reciprocal lattice space spanned by either the [100]-[001] or the [110]-[001] axes. All spectra were collected at room temperature.

Figures 1 show the incident energy ($E_i$) dependence of RIXS, together with the fluorescence spectra. The absolute momentum transfer is fixed at $Q = (0.5, 0.12)$. Resonantly enhanced peaks at around 2 eV and 6 eV can be seen. The latter peak was also observed in the undoped Nd$_2$CuO$_4$, and it was identified as a charge transfer excitation to the antibonding state [3]. We find two resonances at around 8990 eV and 9000 eV for the 6 eV peak. These energies correspond to the resonance energies for $\epsilon_i \parallel c$ and $\epsilon_i \parallel ab$, $\epsilon_i$ being the polarization of
As shown in Fig. 2(a), the momentum dependence of RIXS along the c-axis is weak, as expected from the strong two-dimensionality of the CuO$_2$ plane. However it should be noted that the quasielastic tail is appreciably suppressed for the scan at $l = 12.5$. In our experimental condition (π-polarization of the incident x-ray), the intensity of the elastic scattering, whose major component is Thomson scattering, is mostly proportional to $\cos^2 \theta$, where $\theta$ is the scattering angle and thus decreases when $\theta$ is close to 90 degree. It is crucially important to reduce the elastic scattering to measure the low energy excitations in RIXS. For this reason, we selected $l = 12.5$ to measure the momentum dependence in the CuO$_2$ plane, though it is not a high symmetry plane.

Figures 2(b) and (c) show the momentum dependence of the RIXS spectra along a line in the $q = (\pi,0)$ and $(\pi,\pi)$ directions, respectively, where $q$ represents the reduced momentum transfer in the $ab$-plane. Except for the spectrum at $Q = (0.75,0,12.5)$, all the spectra were measured at $h < 0.5$ of $Q = (h,0,12.5)$ or $Q = (h,h,12.5)$. The spectrum at $Q = (0.75,0,12.5)$ lies between those of $Q = (0.2,0,12.5)$ and $Q = (0.3,0,12.5)$, which indicates that the electronic structure is symmetric with respect to $h = 0.5$. In order to examine the momentum dependence more clearly, we subtract the elastic contribution near 0 eV and the high-energy contribution above 4 eV from the raw data (dashed lines in Figs. 2(b) and (c)). The former is estimated from the anti-Stokes region, and the latter is treated as a tail of the excitation at 6 eV by extrapolating smoothly to the lower-energy region. The open symbols in Figs. 2(b) and (c) are the resulting spectra, where the data below 0.6 eV are not shown due to the uncertainty in the assignment of the quasielastic contributions. The spectra are replotted in Fig. 2(d) as a contour map, where the maximum intensity at each momentum point is normalized to unity and a smoothing procedure is applied. We can clearly see two characteristic excitations. One is the excitation at 2 eV observed at the zone center. Its intensity rapidly decreases with increasing $q$. The other one is a broad but dispersive excitation along the $(\pi,0)$ and $(\pi,\pi)$ directions. As a function of $q$, the latter excitation shifts to higher energy up to 2-2.5 eV at the zone boundary, accompanied by an increase of the spectral width. The upper edges of the excitations are dispersive with a width of more than 2 eV from the zone center to the zone boundary.

Two characteristic excitations just described above have been elucidated further by duplicating the similar scans for $x = 0.075$ crystal. Typical data of scans are shown in Fig. 2(c). The excitation spectra at the zone center superpose each other and it is essentially independent of $x$. On the other hand, the spectra at finite $q$s show the weaker intensities for $x = 0.075$ in lower-energy region and the intensity seems to be proportional to $x$. Such dependence on doping indicates different nature between two excitations which is identified by the follow-

FIG. 1: (a) Resonant inelastic x-ray scattering spectra of Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ as a function of energy loss at some representative incident x-ray energies $E_i$. The scattering vector is fixed at $Q = (0.5,0,12)$. The strong enhancement at around 8991 eV which corresponds to the 1$s$ transition, while the next two are the 1$s$ - 4$p_x$ transition. The intensity of the 2 eV peak shows an enhancement at around 8991 eV which corresponds to the absorption edge for 4$p_x$. Hereafter $E_i$ is fixed at 8991 eV to focus on the low energy excitations.
FIG. 2: Momentum dependence in Nd$_{1.85}$Ce$_{0.15}$CuO$_4$. (a) Along the $c$-axis, (b) $(\pi, 0)$, and (c) $(\pi, \pi)$ directions. $E_i = 8991$ eV. The filled symbols are raw data, and the open ones in (b) and (c) are data from which the elastic scattering and the scattering at higher energy (dotted lines) are subtracted. (d) Contour plot of the RIXS intensity. After subtraction of the elastic and high-energy contributions (open symbols in (b) and (c)) the data are normalized for the maximum intensity in each momentum and interpolated smoothly. (e) Comparison of the RIXS spectra to Nd$_{1.925}$Ce$_{0.075}$CuO$_4$.

...ing theoretical analysis of RIXS from the electron-doped CuO$_2$ plane.

Keeping in mind the nearly monotonic dispersion like excitation mode, we performed calculations of the RIXS spectrum using the numerically exact diagonalization technique on a $4 \times 4$ cluster of a Hubbard model with the electron density $18/16 = 1.125$. The model includes the hopping of the electrons between first, second, and third nearest neighbor sites ($t$, $t'$, and $t''$, respectively) and the on-site Coulomb interaction $U$. The RIXS spectrum is expressed as a second-order process of the dipole-transition between Cu 1$s$ and 4$p$ orbitals, where a Coulomb interaction between a 1$s$ core-hole and a 3$d$ electron, $U_{c1}$, is explicitly included. We use $t'/t = -0.25$ and $t''/t = 0.12$, which are obtained from shape of the Fermi surface. For other parameters, we take $U/t = 8$, $U_{c1}/t = 10$, and $t = 0.3$ eV. The inverse of the life time of the intermediate state is assumed to be $\Gamma = 3t$.

Figure 4 shows the calculated RIXS spectrum, where $E_i$ is set to a value denoted by the arrow in the absorption spectrum shown in the inset. The RIXS spectrum shows two characteristic excitations similar to the observed ones: One is a 2 eV excitation at $q = (0, 0)$, and the other is a broad band of excitations up to $\sim 3$ eV for all momenta except (0, 0). The 2eV excitation is the Mott gap excitation from LHB to UHB, as discussed in Ref. 12. The broad excitations can be assigned to the intraband excitation. To confirm this, we calculated the dynamical density response function $N(q, \omega)$, which can describe the momentum-dependent intraband and interband density fluctuations separately when $U$ is large. Comparing RIXS and $N(q, \omega)$ (broken lines), we find qualitatively similar behavior for all momentum and energy regions except for the 2 eV excitation at $q = (0, 0)$. This means that the broad and dispersive excitations observed in Fig. 2 come from the charge fluctuations in the metallic phase.

The energy position of the highest peak in RIXS and $N(q, \omega)$ is plotted in the inset of the right panel of Fig. 4. In addition to the positions taken from the main panels, we plot peak positions obtained from a $\sqrt{18} \times \sqrt{18}$ cluster with an electron density 20/18. The resulting momentum dependence is found to trace out the center of both spectra in the contour plot of Fig. 2(d). This agreement justifies the assignment of the observed two structures to the Mott-gap and intraband excitations.

As emphasized above, the intraband excitations show broad features. Such a broadness comes from strong correlation common to Hubbard-type models, and thus it is independent of the presence of the long-range hoppings. Instead, the effect of $t'$ and $t''$ may appear in the very low-energy region, where excitations are predominately controlled by the Fermi surface topology inducing 2$k_F$ and 4$k_F$ branches, $k_F$ being the Fermi momentum. Unfortunately, at present the energy resolution of RIXS is limited so that it is difficult to resolve the branches. We continue our efforts to improve the energy resolution and this will open a new view of the intraband charge excitations in the high-$T_c$ cuprates.
in the future.

Although we made use of electron-doped Nd_{1.85}Ce_{0.15}CuO_{4}, it may also be possible to use hole-doped materials such as La_{2−x}Sr_{x}CuO_{4} to detect the intraband excitation. However, it is important to notice that there is an advantage of electron doping over hole doping. As seen in the inset of the left panel of Fig. 3, the absorption spectrum shows three components: ω − ε_{1s−4p} = −17t, −12t, and −8t. The latter two components are also seen in the undoped system. On the other hand, the former appears upon electron doping only, corresponding to a final state where the core hole attracts a doped electron on the same site. Therefore, the final state hardly contains any pair of empty and doubly occupied sites, i.e., no excitations across the Mott gap. This means that the intraband charge excitations dominate the RIXS spectrum if E_{i} is tuned to the lowest-energy peak. As long as we fix E_{i} to the absorption-edge region, the contribution from the lowest-energy peak induces large intraband charge excitations. In our case we select the incident energy to the absorption edge for 4p_{x} as shown in Fig. 3(b), and this condition is satisfied. In contrast, a corresponding final state in the hole-doped system, which emerges after hole doping, exists in a much higher-energy region at around ω − ε_{1s−4p} = 0. Experimentally tuning the incident photon energy to this region seems to be difficult because of the overlap of other absorption processes.

Finally, we compare the interband excitations across the Mott gap between hole- and electron-doping. Recently RIXS experiments for La_{2−x}Sr_{x}CuO_{4} have been reported [1, 2]. Their results showed that the spectral shape is almost independent of the momentum transfer except for small shifts in energy. On the other hand, the interband excitation of Nd_{1.85}Ce_{0.15}CuO_{4} concentrates on an energy (∼2 eV) at the zone center and becomes broad in energy with increasing momentum transfer. Such a difference in momentum dependence is consistent with a previous theoretical result [3], where the difference in the strength of antiferromagnetic correlation plays a crucial role.

In summary, we have performed a RIXS study for the electron-doped superconductor Nd_{1.85}Ce_{0.15}CuO_{4}, and found characteristics of the intraband and interband excitations. The interband excitation shifts to higher energy with the increase of the peak width as a function of momentum transfer. The spectral shape of the intraband excitation has a similarity to N(q, ω) of the two-dimensional Hubbard model. This demonstrates that RIXS is a good tool to measure momentum-dependent density fluctuations in strongly correlated metallic systems. On the other hand, the interband excitation across the Mott gap is enhanced in intensity at the zone center, which is in contrast to the momentum-independent spectral shape in hole-doped La_{2−x}Sr_{x}CuO_{4}.

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