Resonant Inelastic X-ray Scattering of High-T_c Superconducting Cuprates

Recently, resonant inelastic X-ray scattering (RIXS) has been developed as a new spectroscopic technique to measure the electronic structure of materials utilizing intense synchrotron radiation X-rays [1]. Especially, RIXS in the hard X-ray regime has the great advantage that the momentum dependence of the excitations can be measured unlike conventional optical methods. While angle-resolved photoemission spectroscopy, which also gives momentum-resolved spectra, essentially yields the one-particle spectra for the occupied states below the Fermi energy, RIXS yields two-particle (electron-hole pair) excitation from which both occupied and unoccupied states are elucidated. We have applied the RIXS technique to strongly correlated electron systems, such as manganites and cuprates, in order to understand the electronic structure and underlying correlation effects. Here, we show two RIXS studies at the Cu *K*-edge on high-T_c superconducting cuprates, $YBa_2Cu_3O_{6.93}$ (YBCO, T_c = 93 K) and $Nd_{1.85}Ce_{0.15}CuO_4$ (NCCO, T_c = 25 K). The carrier concentration of both compounds is at the optimal condition for the superconducting transition temperature.

The RIXS experiments were carried out at beamline **BL11XU**. A Si(400) channel-cut monochromator and a Ge (733) analyzer were used. The total energy resolution estimated from the FWHM of the elastic scattering is about 400 meV. The energy of the incident X-ray was tuned near the Cu *K*-edge (~ 8990 eV) where the excitation across the Mott gap is resonantly enhanced. More precisely, the Mott gap in the cuprates is the charge-transfer gap between the Zhang-Rice singlet band and the upper Hubbard band.

YBCO has a unique crystal structure and two crystallographically inequivalent Cu sites exist in the unit cell as shown in Fig. 1. One (dark blue) is in the CuO_2 plane, which is a common unit in the most superconducting cuprates. The other (light blue) forms a CuO chain running along the ***b***-axis. Substantial anisotropic character in both normal and superconducting states indicates that the CuO chain also contributes to the bulk electronic properties. A separate measurement of the electronic structure between the CuO chain and the CuO_2 plane is necessary to understand the role of the chain and we could succeed in determining it from the momentum dependence of a twin-free crystal. Figure 2 shows the RIXS spectra of YBCO in the *ab* plane and two characteristic excitations can be seen. One is an excitation at 2 eV, which is prominent at the zone boundary of the ***b****-direction; that is, the intensity is enhanced near (0,π) and (π,π). The other is a broad excitation at 1.5-3.5 eV, which is observed at all the momenta. Clear momentum dependence along the ***b****-direction is direct evidence that the peak at 2 eV is the excitation across the Mott gap of the chain because the excitation along the ***b****-axis should be equivalent to that along the ***a****-axis in the plane. On the other hand, the momentum-independent broad feature corresponds to the excitation in the CuO_2 plane. These characters are well reproduced by a theoretical calculation based on the one-dimensional Hubbard model for the chain and the two-dimensional one for the plane when different values of the on-site Coulomb energy are assumed. This means that the Mott gap of the chain is found to be smaller than that of the plane [2].

While the carrier in the most superconducting cuprates is a hole, superconductivity in $Nd_{2-x}Ce_xCuO_4$ occurs by electron-doping. Therefore $Nd_{2-x}Ce_xCuO_4$ is an important material to study the asymmetry

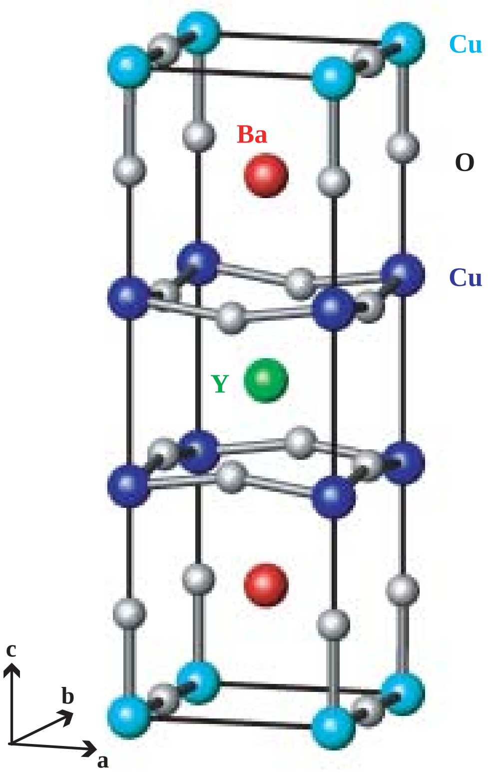

Fig. 1. Crystal structure of $YBa_2Cu_3O_7$. Two crystallographically inequivalent Cu sites exist in the unit cell. One (dark blue) is in the CuO_2 plane and the other (light blue) forms a CuO chain running along the -axis.

of hole- and electron-doping. In general, an interband excitation across the Mott gap and an intraband excitation by doped carriers are observed when carriers are introduced in the Mott insulators. The intraband excitation in hole-doped $La_{2-x}Sr_xCuO_4$ appears as a continuous spectral weight below the Mott gap [3]. In contrast, intraband excitation in electron-doped $Nd_{2-x}Ce_xCuO_4$ is more prominent and shows a clear dispersion relation. Figure 3 shows the RIXS spectra of NCCO. The spectral shape near the Brillouin zone center is insensitive to the carrier concentration (x), and the 2 eV peak at (0,0) is the interband excitation across the Mott gap. Another excitation in the spectra is broad but dispersive. As a function of the momentum transfer, the excitation shifts to higher energy up to 2-2.5 eV at the Brillouin zone boundary accompanied by an increase in spectral width. Because its intensity is roughly proportional to x, it can be identified as the intraband excitation by the doped electrons. It is noted that the larger resonance of the intraband excitation in NCCO comes from the difference in resonance process; namely, the intraband excitation in the electron-doped system is fairly enhanced when a core hole is screened by a doped electron in the intermediate state of the resonant scattering, and this process is absent in the hole-doped systems. These experimental results are consistent with a theoretical calculation. Furthermore, we found that the intraband excitation in the RIXS spectra is quantitatively similar to the dynamical charge correlation function of the doped carrier in the CuO_2 plane [4].

In summary, our results on YBCO and NCCO demonstrate that the excitation across the Mott gap persists even in the carrier-doped metallic state. This is an important character of strongly correlated metals. We also observed the dispersion relation of the intraband excitations in NCCO by taking advantage of the resonant process of the electron-doped system.

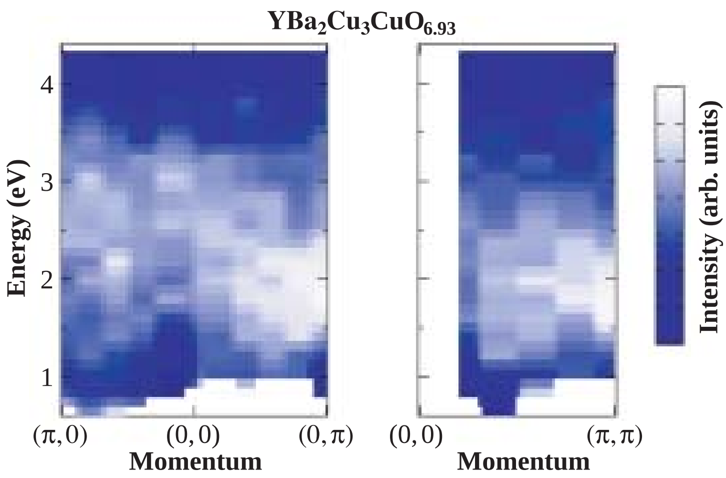

Fig. 2. RIXS spectra of $YBa_2Cu_3O_{6.93}$. The tail of the quasielastic scattering and the excitations at higher energy are subtracted. The excitation across the Mott gap of the CuO chain is enhanced at 2 eV at (0,π) and (π,π), while that of the CuO_2 plane is broad at 1.5-3.5 eV and almost independent of the momentum transfer.

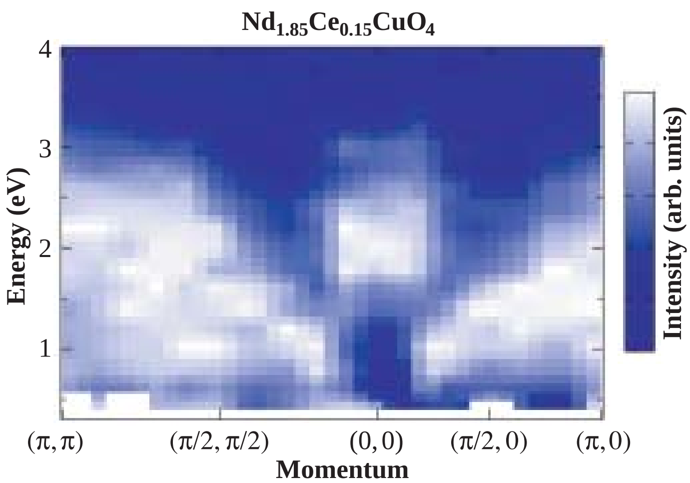

Fig. 3. RIXS spectra of $Nd_{1.85}Ce_{0.15}CuO_4$. The tail of the quasielastic scattering and the excitations at higher energy are subtracted. The peak at 2 eV at the Brillouin zone center is the interband excitation across the Mott gap, and the dispersive excitation with momentum-dependent width is the intraband excitation in the upper Hubbard band.

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