Electron-doping evolution of spin and charge excitations in cuprate superconductors

Superconductivity, which is a state of matter where the electrical resistance disappears completely, is a fascinating phenomenon for many researchers in condensed matter physics and materials science. In particular, high-T_c superconductivity of cuprates has attracted great interest since its initial discovery more than a quarter century ago.

The parent compound of cuprate superconductors is an antiferromagnetic Mott insulator, where electrons cannot move in the crystal due to their strong Coulomb repulsion. Upon doping either the electrons or holes as mobile charge carriers, the compound becomes metallic and the antiferromagnetic spin order fluctuates. Superconductivity occurs in the doped metallic state. Additionally, the antiferromagnetic interaction, which causes the spin order in the parent compound, is intimately related to the superconductivity. Such a complicated spin and charge behavior is a characteristic of strongly correlated electrons. Both spin and charge excitations should be clarified for a definitive understanding of the electron dynamics in cuprates, and the differences or similarities between electron- and hole-doped compounds have been a central issue in the study of cuprate superconductors.

In this study [1], we used resonant inelastic X-ray scattering (RIXS) to measure the spin and charge excitations in the electron-doped superconductor, $Nd_{2-x}Ce_xCuO_4$. Very recently, RIXS at the transition-metal *L*-edge became a viable alternative to inelastic neutron scattering (INS) to measure momentum-resolved spin excitations. On the other hand, charge excitations can be observed at both the *L*- and *K*-edges. Our RIXS experiments at the Cu L_3-edge and *K*-edge were performed at ID08 of the ESRF and **BL11XU** of SPring-8, respectively. In the principal publication, we also measured the INS spectra using the chopper spectrometer, 4SEASONS at J-PARC.

Figure 1(a) shows the Cu L_3-edge RIXS spectra of the parent Nd_2CuO_4 (x = 0) at three momenta in the CuO_2 plane ($\mathbf{q}_{||}$). The spectra can be univocally decomposed into the elastic (green), single spin-flip (single-magnon, blue), multiple spin-flip (multi-magnon, yellow), and *dd* (local electron transition between the Cu 3*d* orbitals, gray) components. The single-magnon is the dominant component. In the electron-doped superconductor, $Nd_{1.82}Ce_{0.18}CuO_4$ (x = 0.18) shown in Fig. 1(b), the single-magnon component is still dominant, but additional spectral weight (red) appears at $\mathbf{q}_{||}$ = (0.09,0) and (0.18,0). Unlike the almost dispersionless multi-magnon in the parent compound, the additional weight has a steep dispersion. Figures 2(a) and 2(b) show the Cu L_3-edge RIXS intensity maps of x = 0 and 0.18, respectively, where the blue squares are the superimposed peak positions of the spin excitations (single-magnon). The spin excitations of x = 0 have sinusoidal dispersion, as expected for antiferromagnetic spin-wave excitations. Upon electron doping (x = 0.18), the spin excitation moves to a higher energy and is accompanied by a broadening of the width. We also confirmed the high-energy shift of the spin excitations near the magnetic zone center by INS. This doping evolution of the spin excitations is in distinct contrast of the hole-doped case, where the spectral distribution of the spin excitations broadens but its energy position is almost unchanged [2].

Figure 1(c) shows the Cu *K*-edge RIXS spectra of x = 0.18. We observed two types of charge excitations: interband excitations across the charge-transfer gap

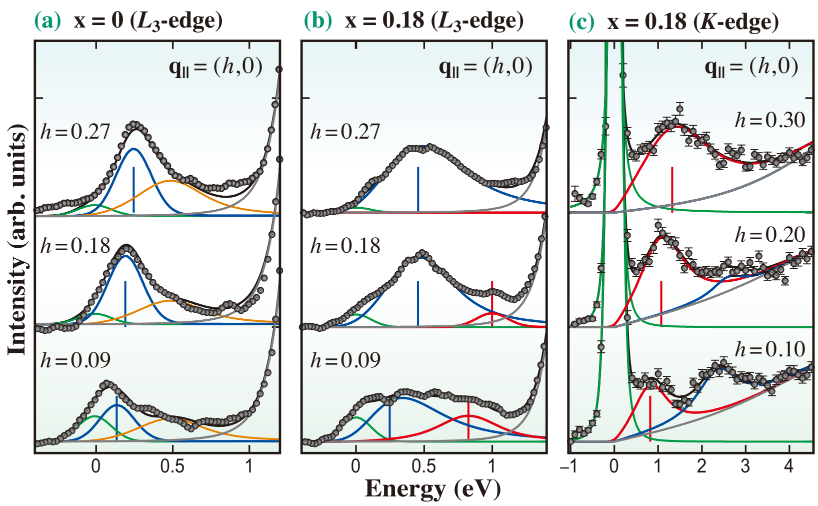

Fig. 1. RIXS spectra of $Nd_{2-x}Ce_xCuO_4$. Solid circles and solid lines denote the experimental data and the fitting results, respectively. Blue and red vertical bars indicate the peak positions of spin excitations (single-magnon) and intraband charge excitations, respectively.

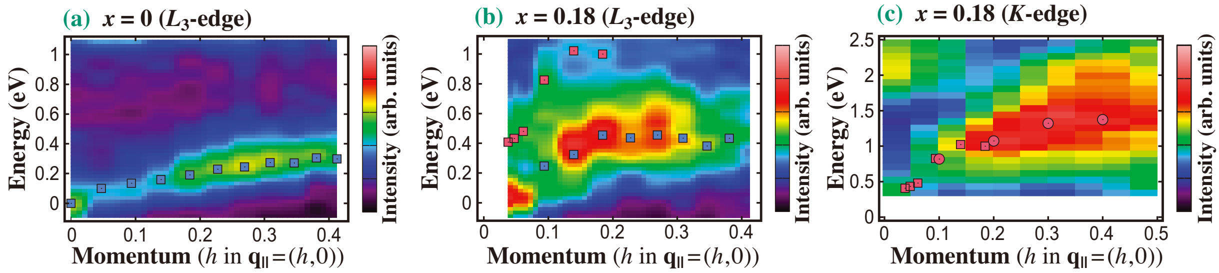

Fig. 2. RIXS intensity map of $Nd_{2-x}Ce_xCuO_4$. Blue and red squares indicate the peak positions of the spin and charge excitations obtained by fitting the Cu L_3-edge RIXS spectra, respectively. Red circles are the peak positions of the intraband charge excitations in the Cu K-edge RIXS spectra.

(light blue), which are prominent near the Brillouin zone center, and dispersive intraband excitations in the upper Hubbard band (red). In the parent Mott insulator, the latter is missing and charge excitations appear above 2 eV. The momentum dependence of the two excitations is clear in the RIXS intensity map in Fig. 2(c) and is consistent with our previous work for x = 0.15 [3]. The red circles on the map are the peak positions of the intraband excitations. Due to the improved energy resolution compared to the previous work, the lower-q region, could be accessed, allowing the additional spectral weight to be identified in the Cu L_3-edge RIXS spectra. We also plotted the peak of the additional spectral weight in the map (red squares). It is clear that the peak positions are smoothly connected to the dispersion of the intraband excitations in the Cu K-edge RIXS. Thus, the additional spectral weight in the Cu L_3-edge RIXS is attributed to the same charge origin. The energy scale of the charge excitations is on the order of transfer energy ($t \sim 0.4$ eV), and should be higher than the spin excitations, which is on energy scale of the exchange interaction ($J \sim 0.1$ eV). It is consistent with our assignment in the Cu L_3-edge RIXS spectra.

From RIXS at the Cu L_3- and K-edges, which is complemented by INS in the principal publication, we clarified the electron-doping evolution of the spin and charge excitations in cuprate superconductors. Figure 3, schematically summarizes the doping evolution of the spin and charge excitations, where characteristics of the elementary excitations in the cuprate superconductors are displayed. The high-energy shift of the spin excitations and their mixture with the charge excitations indicate that the electron dynamics of the electron-doped cuprates has a highly itinerant character in the sub-eV energy scale. In contrast, dispersion of the spin excitation in the hole-doped cuprates follows the spin-wave excitations of the parent antiferromagnetic insulator, which means that a more localized picture is suitable. Our findings impose constraints on theoretical models and show that a comprehensive description of the electronic excitations in the electron- and hole-doped cuprates is a prerequisite to completely understand the superconductivity. For instance, a recent theoretical study demonstrated that the t-J model, which is often used to describe the electronic properties of copper oxides, does not reproduce the high-energy shift of the spin excitations observed in this study [4]. Incidentally, our work is the first to demonstrate that the complementary use of X-rays and neutrons is very effective in inelastic scattering.

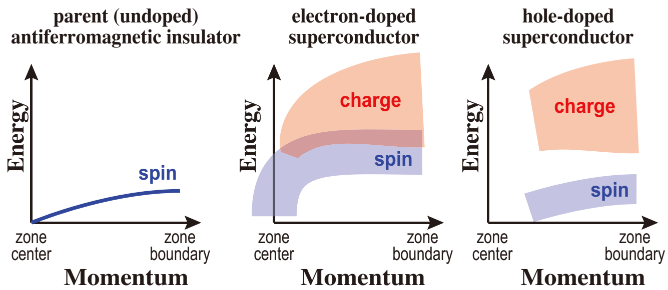

Fig. 3. Doping evolution of spin and charge excitations in the cuprate superconductors. In the present work, the parent antiferromagnetic insulator and an electron-doped superconductor are studied. Excitations in hole-doped superconductors have been reported in Refs. 2,5.

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