ARPES study on electronic evolution in $Ca_{2-x}Na_xCuO_2Cl_2$

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Abstract

Electronic evolution from an antiferromagnetic insulator to a high-T_c superconductor is revealed by ARPES experiments on tetragonal $Ca_{2-x}Na_xCuO_2Cl_2$ single crystals, which were grown for the first time under high pressures ($\leq$ 5.5 GPa). In an underdoped superconductor ($x = 0.1$), we found clear fingerprints of the parent magnetic insulator: a shadow band and a large gap-like structure. The results are most likely described by a "chemical potential shift", which contrasts clearly with the "pinned chemical potential" reported for the prototype $La_{2-x}Sr_xCuO_4$, demonstrating that the route to a high-T_c superconductor is not unique.

Key words: ARPES; $Ca_{2-x}Na_xCuO_2Cl_2$; electronic evolution; rigid band shift

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1. Introduction

All cuprate high-T_c superconductors are realized by doping carriers into their parent antiferromagnetic insulators (AFI). How the electronic structure evolves across the magnetic insulator to the high-T_c superconductor (metal) is, therefore, absolutely fundamental to a proper understanding of the mechanism of high-T_c superconductivity. The continuous improvement in the energy and momentum resolution of angle-resolved photoemission spectroscopy (APRES) has made it possible to discuss the details of the electronic structure, such as the band dispersion, energy gaps, etc, in the CuO_2 plane of high-T_c superconductors. Thanks to its excellent cleavage plane and the resulting fresh surface, the best ARPES data for the AFI have been obtained from $A_2CuO_2Cl_2$ (A = Ca, Sr) [1]. Another attractive feature in these oxychloride compounds is that they have an ideal (undistorted) square-lattice CuO_2 plane. Recently, we have succeeded in growing sizable single crystals ($\sim 1.5 \times 1.5 \times 0.2$ mm^3) of superconducting, Na (thus carrier) doped $Ca_2CuO_2Cl_2$ by a flux method under high pressures up to 5.5 GPa [2]. The obtained crystals were easily cleaved as the parent material, enabling us to examine the metallic side of the insulator-metal transition by ARPES experiments. The electronic evolution from the magnetic insulator to the high-T_c superconductor in $Ca_{2-x}Na_xCuO_2Cl_2$ appears as a "rigid band shift" rather than a "creation of in-gap states"" reported for the prototype $La_{2-x}Sr_xCuO_4$ [3].

2. Experimental

Single crystals of $Ca_{1.9}Na_{0.1}CuO_2Cl_2$, which are carrier underdoped and have T_c = 13 K, were prepared by the flux growth at 4 GPa using a powder mixture of $Ca_2CuO_2Cl_2$, NaCl, and $NaClO_4$. The details of the crystal growth and characterization of the obtained crystals can be found in Ref. [2]. The ARPES measurements were performed at beamline 5-4 of SSRL, Stanford University. The ARPES data were collected using 25.5 eV photons at 20 K by a SES-200 analyzer (ΔE = 16 meV and $\Delta\theta = 0.25°$).

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3. Results and Discussion

Figure 1 shows the energy distribution curves (EDCs) for $Ca_{1.9}Na_{0.1}CuO_2Cl_2$ along the high symmetry directions. From (0,0) to (π,π), a peak in the EDC disperses to the Fermi energy E_F. Even after the band has crossed E_F around ($\pi/2$,$\pi/2$), a broad feature still disperses backwards as indicated by tick-marks. This appears to be what is expected for shadow bands due to a band-folding caused by the unit-cell doubling. The low-energy electron diffraction at 20 K taken right after the ARPES measurements verified that the shadow band in this material did not derive from either a change in the crystal-structure or a surface reconstruction, suggesting antiferromagnetic correlations as the source of the band-folding.

In addition to the magnetic shadow band, a large gap-like feature is observed in the dispersion from (0,0) to (π,0) to (π,π). Although a peak in the EDC approaches E_F toward (π,0) to (π,π), no clear Fermi crossing is identified; instead, the structure in the EDC simply fades away on its approach to (π, π). The band near (π,0) is located ~ 200 meV below E_F, an extremely large value to be identified as a superconducting or pseudo gap.

By taking the second derivative of the EDCs, the band dispersion in $Ca_{1.9}Na_{0.1}CuO_2Cl_2$ is summarized in Fig. 2(a), where that for Na-free $Ca_2CuO_2Cl_2$ is also drawn for comparison. It is found that the dispersion between the two compounds is strikingly similar but differs only in the energy position, indicating that the valence bands of the parent insulator shifts rather rigidly toward E_F by carrier-doping. This is in striking contrast with what has been reported for the prototype $La_{2-x}Sr_xCuO_4$, where the parent bands remain the same with respect to E_F, while new states are created around E_F [3]. It clearly demonstrates that the route to a high-T_c superconductor is not unique.

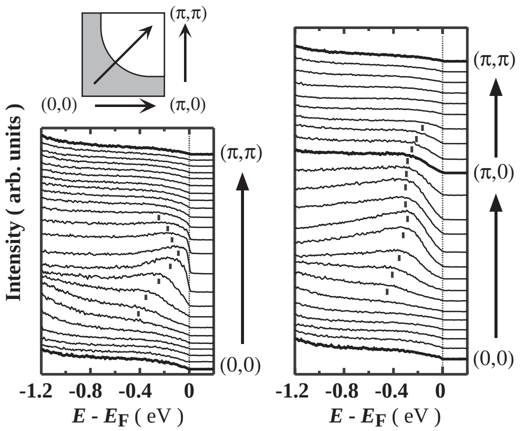

Fig. 1. ARPES spectra for $Ca_{1.9}Na_{0.1}CuO_2Cl_2$.

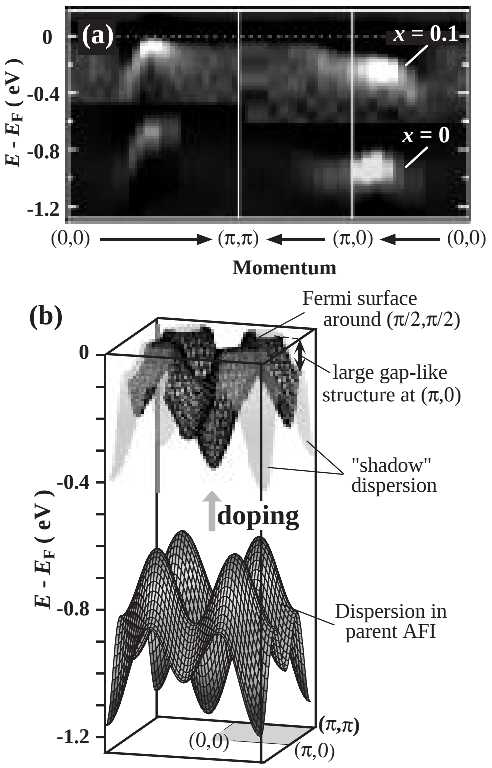

Fig. 2. (a) Band dispersion for $Ca_{1.9}Na_{0.1}CuO_2Cl_2$ and $Ca_2CuO_2Cl_2$. (b) Schematic representation of the "rigid band shift" electronic evolution.

In summary, this "rigid band shift" scenario in $Ca_{2-x}Na_xCuO_2Cl_2$ is illustrated in Fig. 2(b). As a result of the band-shift due to carrier-doping, the dispersion closest to E_F in the parent insulator [i.e., near ($\pi/2$,$\pi/2$)] touches E_F to form the Fermi surface, switching its state into the metal (superconductor). The energy difference between ~($\pi/2$,$\pi/2$) and (π,0) becomes the extremely large gap-like structure around (π,0). Finally, as a remnant of the antiferromagnetic parent material, the magnetic shadow band is observed near ($\pi/2$,$\pi/2$).

Acknowledgements

This work was supported by SORST-JST, by JSPS, and by the U.S. DOE.

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