Soft X-ray Angle-resolved Photoemission Spectroscopy of Strongly Correlated Bulk Electronic Structures of $SrCuO_2$ and Sr_2RuO_4

Angle-resolved photoemission spectroscopy (ARPES) with high resolutions in energy and momentum (or wave number k) realized at low photon energies (hν) between ~20 and ~100 eV has been extensively applied to correlated electron systems such as high-T_c cuprates to study their electronic structures [1]. However, it is known that such a low-hν photoemission spectroscopy (PES) is surface sensitive and often provides spectral shapes which are not consistent with bulk electronic structures in several transition metal and rare earth compounds [2,3]. Therefore, caution is required to suppress surface effects sensitively probed by the short photoelectron mean free path (λ). For this purpose, the bulk-sensitive ARPES above several hundred eV with λ > 10 Å has been strongly desired. Pioneering ARPES with high-hν X-ray was performed many years ago. However, the unsatisfactory angular (±2°) and energy (0.35 - 0.85 eV) resolutions made it impractical for band mapping and Fermiology.

By virtue of recent instrumental developments at beamline **BL25SU**, soft X-ray ARPES is successfully performed for the valence bands of quasi-one-dimensional (quasi-1D) $SrCuO_2$ and quasi-two-dimensional (quasi-2D) Sr_2RuO_4. Above several hundred eV, the photoionization cross section of the Cu 3*d* or Ru 4*d* states is much higher than that of the O 2*p* states. Clear differences from the results of low-hν ARPES have been observed for both materials.

In contrast to various misconceptions, the large photon momentum q (0.36 $Å^{-1}$ at hν = 700 eV) is simply transferred to the photoelectron momentum. Then $q_{//}$ = 0.25 $Å^{-1}$ is transferred to the photoelectron momentum parallel to the surface ($k_{//}$). For a lattice constant of c = 3.9 Å ($SrCuO_2$), for example, $k_{B//}$ at the Brillouin zone edge is π/c = 0.80 A^{-1} and the instrumental angular resolution of 0.25° corresponds to the sufficiently high $k_{//}$ resolution of ~0.06 A^{-1}.

It was proposed in quasi-1D $SrCuO_2$ by low-hν ARPES that the so-called "spin-charge separation" might be taking place. We have performed Cu 3*d*-sensitive and bulk-sensitive ARPES on a cleaved surface at room temperature and found a clear dispersion in the valence band region (Fig. 1(a)). In the so-called O 2*p* bands in the range of 2 - 4 eV, the Cu 3d and O 2*p* states are strongly hybridized. The band near 1 eV has a strong Cu 3*d* component. As shown in the density plot in Fig. 1(b), the V-shaped dispersion up to 1 eV is very prominent in k = 0 (or 2π) ± π/2. Dispersion is much less prominent in the region of π ± π/2. Although both spinon and holon branches are expected in the region of 0 (or 2π) ± π/2 according to the spin-charge separation scenario, there is no trace of the spinon branch irrespective of the present Cu 3*d* sensitivity. The larger dispersion due to the holon is also not observed in the region of π ± π/2. These results demonstrate that the spin and charge are not fully separated in the bulk of $SrCuO_2$ due to the finite magnitude of U/t = 7.5 (U is the Coulomb repulsive energy and t is the transfer energy) [4].

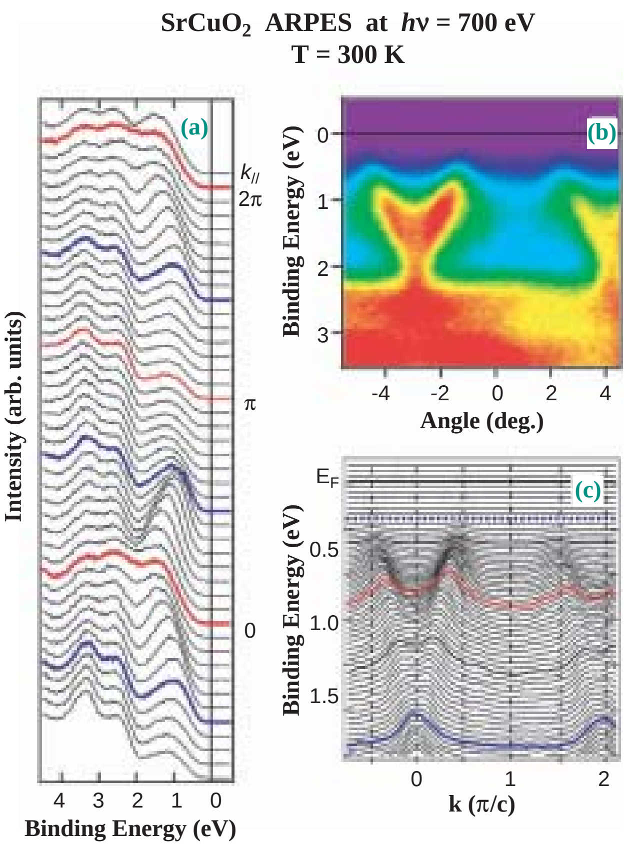

Fig. 1. Soft X-ray (*h*ν=700 eV) ARPES along the c-axis of 1D $SrCuO_2$ at 300 K. **(a)** Energy distribution curves. **(b)** Density plot of the measured photoemission intensity. **(c)** Momentum distribution curves.

Fermi surfaces (FSs) are key issues in understanding the physical properties of new functional materials. Soft X-ray ARPES is applied to the “triplet” superconductor Sr_2RuO_4. Quantum oscillation measurements and band-structure calculations suggest one holelike FS sheet centered at (π,π) (α sheet) and two electron-like FS sheets centered at (0,0) (β and γ sheets). On the other hand, Yokoya *et al*. have first concluded two holelike and one electron-like FSs from low-hν ARPES. A following low-hν ARPES on degraded surface suggests that the earlier finding originates from surface states and surface reconstruction, and that the bulk FSs are qualitatively similar to the result of band calculation.

ARPES spectra are here measured at hν = 700 eV along the (π,0)-(π,π) direction, demonstrating a holelike FS sheet α. Then ARPES spectra are measured along the (0,0)-(π,0) cut. They are complicated because the a sheet band is located at ~0.5 eV, while the β and γ sheet bands show dispersion crossing E_F. The behavior of the E_F crossing of the β and γ branches is also confirmed by the momentum distribution curves (MDCs). Thus the genuine bulk FSs are revealed in Figs. 2(a) and 2(b) without serious distortion owing to the bulk-sensitivity of the soft X-ray ARPES. This technique is further applied to high-T_c cuprates and other strongly correlated electron systems.

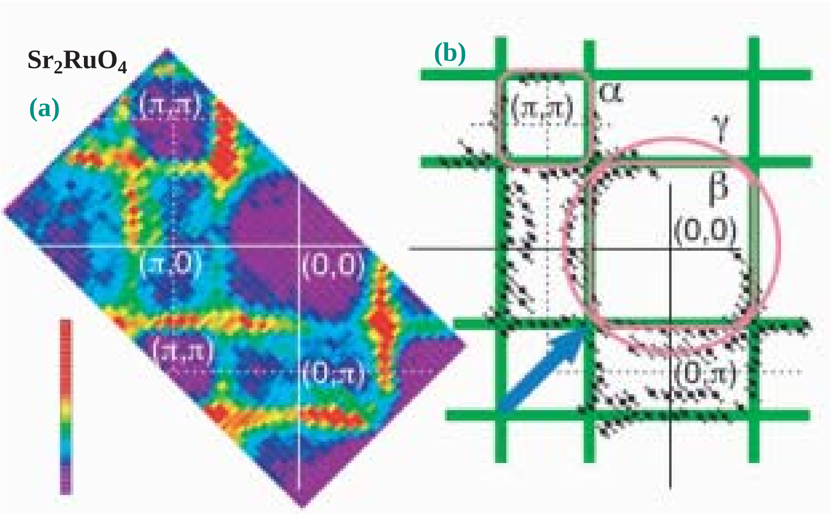

Fig. 2. Bulk Fermi surfaces of Sr_2RuO_4 probed by soft X-ray ARPES at 700 eV and 20 K. **(a)** Intensity integrated from E_F to –0.1 eV. **(b)** Estimated k_F and schematic drawing of FSs.

Shigemasa Suga* and Akira Sekiyama

Graduate School of Engineering Science, Osaka University

*E-mail: suga@mp.es.osaka-u.ac.jp

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