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Angle-resolved photoemission study of Sr_2CuO_3

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We report angle-resolved photoemission spectroscopy (ARPES) on a single Cu-O chain compound Sr_2CuO_3 with Ne I photons. In contrast with previous ARPES with higher-energy photons, the present result clearly shows two well-separated branches of dispersions near E_F ascribable to spinon and holon excitations, respectively. This indicates that spin and charge degrees of freedom are separated in one-dimensional Cu-O chains, supporting the theoretical prediction as well as the previous ARPES interpretation. Comparison with previous ARPES results indicates that surface, photoionization cross section, and finite-temperature effects substantially modify ARPES spectra. [S0163-1829(99)04508-7]

I. INTRODUCTION

Low-dimensional correlated electron systems have attracted much attention because of their interesting properties originating in the low dimensionality and quantum fluctuation. In particular, the one-dimensional (1D) system has been intensively studied theoretically for its simplicity and abnormality. It has been theoretically established[1] that the spin and charge degrees of freedom are separated in the 1D correlated electron system, and behave as independent elementary excitations called ''spinon'' and ''holon'', respectively.[2,3] Many experimental efforts have also been made to synthesize and discover ideal 1D metallic materials to check the theoretical prediction. However, it has been gradually recognized that almost all 1D materials discovered so far are insulating, and carrier doping has been hardly succeeded. This fact causes difficulty for the experimental confirmation of the ''spin-charge separation'' using conventional experimental techniques.

Photoemission spectroscopy (PES) is a unique experimental technique to probe the electronic structure of materials directly using the external photoelectric effect. In the final state of photoemission process, a hole created by a photon (photohole) is left in the system. This provides a unique unconventional doping method (photodoping) in otherwise strongly insulating 1D materials, and the photoelectron ejected from material should reflect the electronic structure of the photodoped system through energy- and momentum-conservation laws. In particular, angle-resolved photoemission spectroscopy (ARPES) is the most direct experimental check of the existence of two independent quasiparticles (spinon and holon) created by spin-charge separation, since it has been theoretically predicted that they should show different energy dispersions in momentum (k) space.[3] Actually many ARPES measurements have been performed on various 1D materials to study the anomalous electronic structure near the Fermi level (E_F).[4–11] The first ARPES observation of spin-charge separation was reported by Kim *et al.*[9] on $SrCuO_2$ which consists of corner-sharing double Cu-O chains (Fig. 1). The experimental result shows an asymmetric energy dispersion near E_F with respect to $k=\pi/2$, which they interpreted as an evidence of spin-charge separation. Almost the same experimental result was later reported by Fujisawa *et al.*[11] on a simpler 1D Cu-O chain compound Sr_2CuO_3 which has a single Cu-O chain (Fig. 1). Although both experimental results agree qualitatively with the theoretical predictions,[3,9] there have remained several quantitative disagreements between the experiment and the calculation, such as the intensity ratio between the spinon and holon bands. While it has been suggested that some of the discrepancies may be removed by taking into account of the photoionization cross section and/or the surface effect,[10] no experimental confirmation has been presented so far.

In this paper, we report ARPES results on a 1D single chain compound Sr_2CuO_3 with lower-energy photon (Ne I, 16.8 eV), which is more bulk sensitive than those used in the previous ARPES measurements.[9–11] The photoionization cross-section ratio between Cu $3d$ and O $2p$ atomic orbitals is also substantially different from the previous ones. We compare the present experimental result with the previous reports, and discuss possible origins for the discrepancies between the experiment and the calculation.

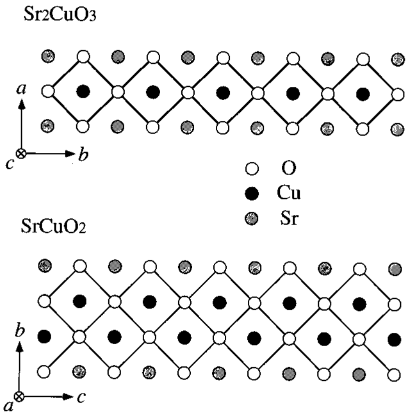

FIG. 1. Crystal structure of Sr_2CuO_3 consisting of single Cu-O chains, compared with that of $SrCuO_2$ having double Cu-O chains.

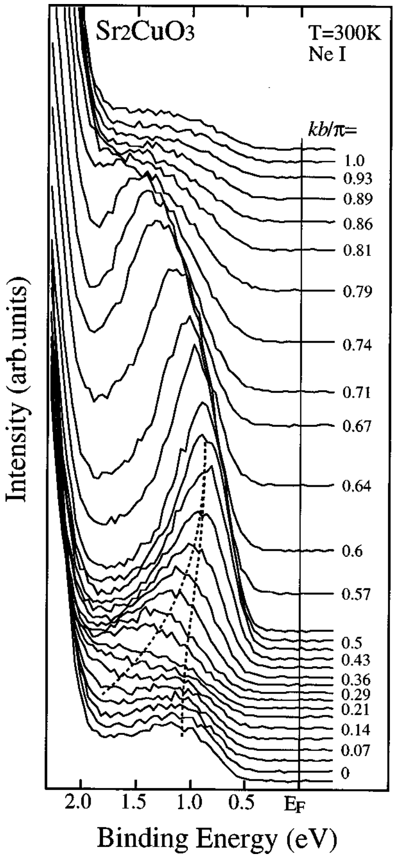

FIG. 2. ARPES spectra of Sr_2CuO_3 near E_F, measured along the b (chain) direction using Ne I photons. The approximate k point in the Brillouin zone is indicated on each spectrum. Broken lines show possible energy dispersions in the first half of the Brillouin zone.

II. EXPERIMENT

Sr_2CuO_3 single crystals were grown by a traveling-solvent-floating-zone method with a CuO solvent. The crystals obtained were characterized by x-ray powder diffraction as well as the Laue method. A photoemission measurement was performed using a home-built ARPES spectrometer with unpolarized Ne I resonance line (16.8 eV) from a discharging lamp. The base pressure of the spectrometer was 5×10^{-11} Torr, and the overall energy resolution was about 100 meV. In order to avoid the charging-up effect of the sample, the photon flux was tuned to be as small as possible, and the sample was kept at room temperature. We have confirmed that charging up did not take place by changing the photon flux and/or the sample temperature. The single crystal was cleaved *in situ* along the bc plane just before ARPES measurement, and all the spectra were recorded within 10 h after cleaving. The Fermi level (E_F) of the sample is referenced to that of a gold film evaporated on the sample substrate. We have measured ARPES spectra for four different samples, and confirmed the reproducibility of data.

III. RESULTS AND DISCUSSION

Figure 2 shows ARPES spectra near E_F measured along the chain direction (b axis) from $kb/\pi=0$ to 1.0 in the Brillouin zone. The intensity of each spectrum is normalized with the incident photon flux. The ARPES spectrum at the Γ point ($k=0$) has a very distinct peak at 1.2 eV below E_F, which is followed by a strong main peak located at about 2.6-eV binding energy (not shown in Fig. 2). On increasing the wave vector (k) from the Γ point, an additional small structure appears around 1.5–2.0 eV, which is evident from the gradual filling of a dip between the 1.2-eV peak and the main peak at 2.6 eV. On further increasing k, this new structure gradually grows up with approaching E_F, merging with the 1.2-eV peak around $kb/\pi=0.29-0.36$, then finally forms a prominent peak at 0.8 eV at $kb/\pi=0.5$. This spectral change suggests the existence of two bands near E_F in the first half of the Brillouin zone, which have different energy dispersions but with a common maximum closest to E_F at $kb/\pi=0.5$. One of new findings by Ne I measurement is that the ARPES spectrum at Γ point ($k=0$) has a distinct peak at 1.2 eV in contrast to the previous reports with higher-energy photons,[9–11] where only a steplike structure has been observed in the same energy region. This new finding indicates that there is a well-defined peak (band) around 1.2 eV at Γ point. In the second half of the Brillouin zone ($kb/\pi=0.5-1.0$), on the other hand, the prominent peak at 0.8 eV at $kb/\pi=0.5$ rapidly moves toward high binding energy as kb/π is further increased to 1.0. At $kb/\pi=1.0$ this prominent peak disappears into the higher-binding-energy region, and a weak broad structure is left around 1.0–1.5 eV. We find that the intensity of this broad structure around $kb/\pi=1.0$ is much smaller than that of a distinct peak at Γ point ($k=0$). The origin of this weak structure will be discussed later.

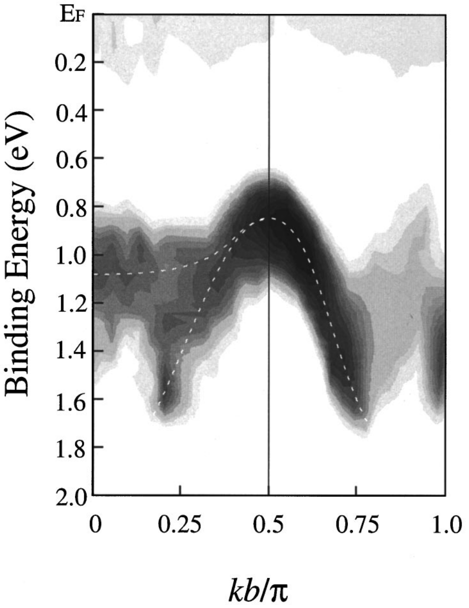

FIG. 3. ''Band dispersion'' near E_F of Sr_2CuO_3 obtained from the present ARPES measurement (Fig. 2). Dark parts correspond to ''bands.'' White broken lines are guides for the eye.

Figure 3 shows the ''band dispersion'' near E_F of Sr_2CuO_3 derived from the present ARPES measurements. In mapping out the ''band dispersion,'' we took the second derivative of ARPES spectra after moderate smoothing, and plotted the intensity with gradual shading as a function of the wave vector and the binding energy; dark parts correspond to ''bands.'' We set the gray-scale image in Fig. 3 so as to have the apparent bandwidth in the gray-scale image almost equal to the full width at half maximum of the corresponding peak in Fig. 2. All the characteristic features of band dispersions observed in the raw ARPES spectra are more clearly visible in Fig. 3. We find again that there are two well-separated dispersive bands in the first half of Brillouin zone ($kb/\pi=0$ to 0.5), and they have a common maximum closest to E_F at $kb/\pi=0.5$. It is also clear in Fig. 3 that one of the two bands with a larger energy dispersion is symmetric with respect to $kb/\pi=0.5$, but has a much stronger intensity in the second half of the Brillouin zone. We observe a weak structure at 1.4 eV around $kb/\pi=1.0$. This structure corresponds to a weak broad structure at 1.0–1.5 eV around $kb/\pi=1.0$ in the raw ARPES spectra (Fig. 2). It is noted that this weak structure in Fig. 3 is isolated from other dispersive bands.

Next we discuss the origin of this weak structure by comparing the present results by Ne I light (16.8 eV) with previous reports with higher-energy photons of He I (21.2 eV) or synchrotron radiation (22.4 eV).[9–11] The surface sensitivity is suppressed in Ne I measurement in comparison with He I and/or 22.4-eV photon measurements, since the escape depth of photoelectrons is longer in the Ne I case.[12] We find that the intensity of the broad structure around $kb/\pi=1.0$ is considerably smaller in the Ne I measurement in comparison with those with higher-energy photons,[9–11] suggesting that the broad structure is substantially of surface origin. Actually we observed that the spectral intensity gradually increases with time. Also as found in Fig. 3, the weak structure located at 1.4 eV around $kb/\pi=1.0$ is isolated from other dispersive bands. All these experimental facts suggest that the weak structure is extrinsic, and may be due to surface degradation.

According to the 1D Hubbard model[3,13,14] at $U\rightarrow\infty$, the spectral function $A(k,\omega)$ shows two divergences due to the spinon and holon excitations; the former corresponds to a flatband located at $\omega=2t$ appearing only in the first half of Brillouin zone ($k=0$ to $\pi/2$), while the latter is a symmetric dispersive band with respect to $k=\pi/2$. These characteristic features indicative of the spin-charge separation are clearly observed in the present experiment (Fig. 3). On the other hand, the t-J model calculation has shown that the dispersions of spinon and holon bands have widths of $\pi J/2$ and $2t$, respectively.[15] Since the spinon and holon bands in Fig. 3 show a bandwidth of about 0.25 and 1.1 eV, respectively, we obtain $J\sim0.16\,\text{eV}$ and $t\sim0.55\,\text{eV}$. These values are almost the same as those obtained with He I photons,[11] and also consistent with the result from the magnetic-susceptibility measurement.[17–19]

In spite of a good qualitative agreement between the experiment and the calculation, we find some quantitative discrepancies as observed in the previous measurements.[9–11] The calculations[3,13,14] have predicted that the spectral intensity of holon band is almost symmetric with respect to $k=\pi/2$, while the experimental holon band shows a much stronger intensity in the second half of the Brillouin zone than in the first one (see Fig. 2). Further, while the intensity of the spinon band is comparable to that of the holon band in the calculations, we observed a relatively smaller spectral weight for the spinon band, as shown in Fig. 2. These discrepancies may be accounted for in terms of the photoionization cross section, since the photoionization cross section of Cu $3d$ is much smaller than that of O $2p$ in the present photon-energy range,[20] and the t-J calculation[10] has predicted a larger spectral weight from Cu $3d$ near $k=0$. If it is the case, the observed well-resolved spinon branch near $k=0$ in the Ne I measurement (Fig. 2) suggests that the spinon branch near $k=0$ has a relatively larger O $2p$ weight compared with the holon branch, since the photoionization cross section of Cu $3d$ is further suppressed in the Ne I measurement.[20] As for an alternative explanation for the discrepancy in the spectral intensity, a finite-temperature effect may be important since our measurement was done at room temperature to avoid the charging-up effect. The spectral function of the 1D t-J model at finite temperatures[16] shows that there takes place a spectral weight transfer from the spinon to the holon band at finite temperatures. In fact, in a recent ARPES study of NaV_2O_5[21] temperature-induced spectral weight transfer has been reported, although the spinon and holon band are not well resolved due to the smaller J value in NaV_2O_5. Thus the finite-temperature effect may be a cause for the relatively smaller spectral weight of the spinon band in the first half of the Brillouin zone, but cannot explain the intensity difference of the holon band between the first and the second half of the Brillouin zone.

IV. CONCLUSION

We have performed ARPES measurements on Sr_2CuO_3, and compared the results with previous reports with higher-energy photons to study the surface and photoionization cross-section effects in spin-charge separation in a 1D Cu-O chain. In contrast to the previous reports, a series of ARPES spectra measured with Ne I photons shows two well-resolved branches of band dispersions near E_F ascribable to the spinon and holon excitations, respectively. The intensity profiles of the spinon and holon bands are different from the theoretical predictions. These discrepancies are partially removed by taking into account surface, photoionization cross-section, and finite-temperature effects.

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