Angle-Resolved Photoemission Study of Insulating and Metallic Cu-O Chains in PrBa$_2$Cu$_3$O$_7$ and PrBa$_2$Cu$_4$O$_8$

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We compare the angle-resolved photoemission spectra of the hole-doped Cu-O chains in PrBa$_2$Cu$_3$O$_7$ (Pr123) and in PrBa$_2$Cu$_4$O$_8$ (Pr124). While, in Pr123, a dispersive feature from the chain takes a band maximum at k_b (momentum along the chain) $\sim \pi/4$ and loses its spectral weight around the Fermi level, it reaches the Fermi level at $k_b \sim \pi/4$ in Pr124. Although the chains in Pr123 and Pr124 are approximately $1/4$ filled, they show contrasting behaviors: While the chains in Pr123 have an instability to charge ordering, those in Pr124 avoid it and show an interesting spectral feature of a metallic coupled-chain system.

One-dimensional (1D) or quasi-1D compounds have attracted great interest because of their unique and interesting physical properties such as Peierls instability, spin-charge separation, and Tomonaga-Luttinger (TL) liquid behavior [1]. In order to realize the TL liquid state, the system should have good one dimensionality. On the other hand, a good 1D system is expected to have instability towards spin and/or charge ordering and tends to be insulating. In every 1D system, a metallic state, which is possibly a TL liquid, is competing with the instability to the insulating state. It is therefore very important and interesting to study the electronic structure of quasi-1D systems which are close to the boundary between the quasi-1D metal and insulator. This subject is also related to the electronic structure of the quasi-1D stripe phase in 2D systems and is important in light of the competition between various quantum mechanical ground states in 1D and 2D systems [2].

Angle-resolved photoemission spectroscopy (ARPES) is a powerful tool to study spin-charge separation as well as band gap opening due to charge ordering. It has been found that ARPES of both the Cu-O double chain in SrCuO$_2$ and the Cu-O single chain in Sr$_2$CuO$_3$ have two dispersive features which are successfully interpreted as spinon and holon dispersions of the undoped Cu-O chains and are a manifestation of the spin-charge separation in 1D Mott insulators [3]. In an ARPES study of PrBa$_2$Cu$_3$O$_7$ (Pr123), it has been reported that the hole-doped Cu-O single chain in Pr123 shows possible spinon and holon dispersions and also has a band gap opening at the Fermi level (E_F) proba-

bly due to charge ordering [4]. Recently, an ARPES study of a quasi-1D metal Li$_{0.9}$Mo$_6$O$_{17}$ has shown that spectral weight near E_F is considerably suppressed and can be interpreted as a TL liquid with large α which is the anomalous exponent of the momentum distribution function as well as of the single-particle density of states [5]. The large α suggests strong fluctuations of charge ordering or charge density wave (CDW) in Li$_{0.9}$Mo$_6$O$_{17}$ although the analysis of the ARPES data is still controversial [6].

In Pr123 and PrBa$_2$Cu$_4$O$_8$ (Pr124), the CuO$_2$ planes remain antiferromagnetic and insulating and do not have enough carriers to cause superconductivity [7–9]. On the other hand, the Cu-O single chains in Pr123 and the Cu-O double chains in Pr124 are heavily hole-doped and show semiconducting and metallic behaviors, respectively [7–9]. Therefore, Pr123 and Pr124 give us a unique opportunity to study the electronic structures of the hole-doped Cu-O chains which are close to the boundary between the quasi-1D metal and insulator. In this Letter, we report on a new set of ARPES data for Pr124 which has metallic Cu-O double chains. The ARPES data of the Cu-O double chain in Pr124 show a sharp contrast to those of the Cu-O single chain in Pr123. Since the structures of the Cu-O chains in Pr123 and Pr124, which are the same as the Cu-O chains in Sr$_2$CuO$_3$ and SrCuO$_2$, respectively, are simple compared to Li$_{0.9}$Mo$_6$O$_{17}$, the interpretation of the ARPES data is rather straightforward. By comparing the ARPES data of Pr123 and Pr124, we discuss the effect of interchain coupling and the instability towards charge ordering in the hole-doped Cu-O chains.

Naturally untwinned single crystals of Pr124 were grown by a flux method under oxygen pressure of 11 atm. The resistivity along the chain direction is metallic as reported in the literature [10,11]. The ARPES measurements of Pr124 were performed at beam line 5-4 equipped with a Scienta SES 200 electron analyzer, Stanford Synchrotron Radiation Laboratory (SSRL). The chamber pressure during the measurements was less than 5×10^{-11} Torr. The samples were cooled to 10 K and cleaved *in situ.* The cleaved surfaces were the *ab* plane, where the *b* axis is in the Cu-O chain direction. The cleanliness of the surfaces was checked by the absence of a hump at ~9.5 eV. The position of E_F was calibrated with gold spectra. The experimental uncertainty in the energy calibration was ± 1 meV. The ARPES data of Pr123 were taken at beam lines 5-3 of SSRL. The details of the measurements were described in a previous paper [4]. For the ARPES data of Pr123 and Pr124 shown in this paper, incident photons were linearly polarized and had an energy of 29 eV for Pr123 and 22.4 eV for Pr124. The total energy resolution including the monochromator and the analyzer was approximately 40 meV for Pr123 and 20 meV for Pr124. The angular resolution was $\pm 1°$ for Pr123 and $\pm 0.28°$ for Pr124, which gives the momentum resolution of $\pm 0.05\pi$ for Pr123 and $\pm 0.01\pi$ for Pr124 in units of $1/a$ or $1/b$. ($a = 3.87$ Å and $b = 3.93$ Å for Pr123; $a = 3.88$ Å and $b = 3.90$ Å for Pr124.)

The experimental arrangement is schematically shown in Fig. 1. The polarization vector of the incident photons had a component parallel to the Cu-O chain direction. The CuO$_4$ square planes of the Cu-O chains are perpendicular to the cleaved surface. In Fig. 2, the ARPES spectra of Pr124 along the Cu-O chain direction are shown for $k_a = \pi$ [12]. Here, k_a and k_b are the momentum perpendicular to the chain in units of $1/a$ and the momentum along the chain in units of $1/b$, respectively. In Fig. 2, one can see that a dispersive feature from the Cu-O chain moves to E_F in going from $k_b/\pi = 0.1$ to $k_b/\pi = 0.2$. For $0.2 < k_b/\pi < 0.25$, the dispersion becomes flatter and the dispersive feature gradually loses its intensity. This behavior is clearly seen in the right panel in Fig. 2, where the spectral weight integrated from -0.05 eV to E_F is plotted as a function of k_b/π. The integrated spectral weight decreases from $k_b/\pi = 0.20$ to 0.27, indicating that the

dispersive feature crosses E_F at $k_b/\pi = 0.23 \pm 0.03$ [13] and that the hole concentration of the Cu-O chain in Pr124 is 0.46 ± 0.06. This is consistent with the recent optical study which shows that the hole concentration of the Cu-O chain in Pr124 is ~0.4 [14].

The ARPES spectra of Pr123 and Pr124 along the Cu-O chain are compared in Fig. 3 for $k_a = \pi$. In Pr123, the dispersive feature from the Cu-O chain reaches the band maximum of -0.3 eV at $k_b/\pi = 0.24$ and stays there at $k_b/\pi = 0.29$. This feature gradually loses its weight for $k_b/\pi > 0.29$ without reaching E_F. On the other hand, in Pr124, the dispersive feature reaches E_F at $k_b/\pi \sim 0.23 \pm 0.03$. Although the Cu-O chains are approximately 1/4 filled both in Pr123 and in Pr124, the spectral feature near E_F of Pr124 is remarkably different from that of Pr123. As shown in Fig. 4, the dispersions of the Cu-O chain features become clear in the density plots of the raw ARPES spectra. While, in Pr124, the width of the Cu-O band below E_F is ~0.5 eV and the band dispersion is relatively large, the bandwidth is reduced to ~0.3 eV in Pr123 because of the band gap opening at E_F.

For Pr123, the band gap opening can be attributed to the charge ordering or CDW formation in the 1/4-filled Cu-O single chain [4]. Actually, charge instability in the Cu-O chain has been observed by nuclear magnetic resonance and nuclear quadrupole resonance measurements of Pr123 [15]. The CDW picture is also consistent with the experimental result that the gap opening at E_F reduces the band width of Pr123 compared to Pr124. On the other hand, the Cu-O double chain of Pr124 does not have charge ordering and is metallic. A small deviation from the 1/4 filling

FIG. 2. Left panel: ARPES spectra along the Cu-O chain in Pr124. k_a is the momentum perpendicular to the chain and k_b is the momentum along the chain. The dotted line outlines the band dispersion. Right panel: spectral weight integrated from -0.05 eV to E_F as a function of the momentum along the chain. Intensity is normalized to the peak height.
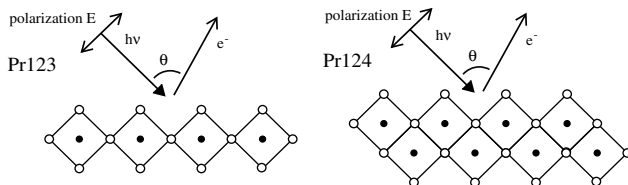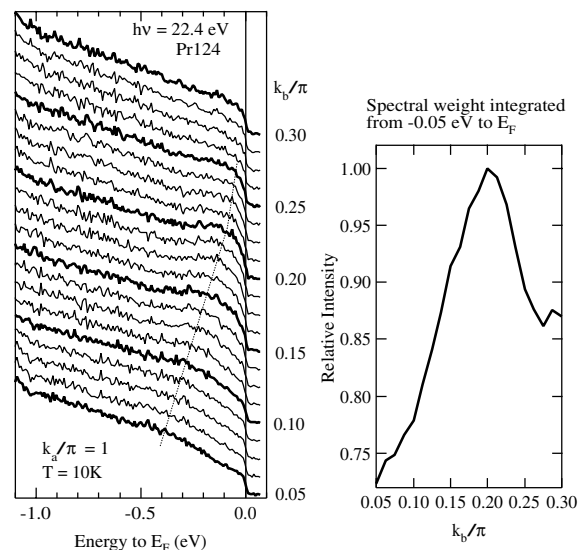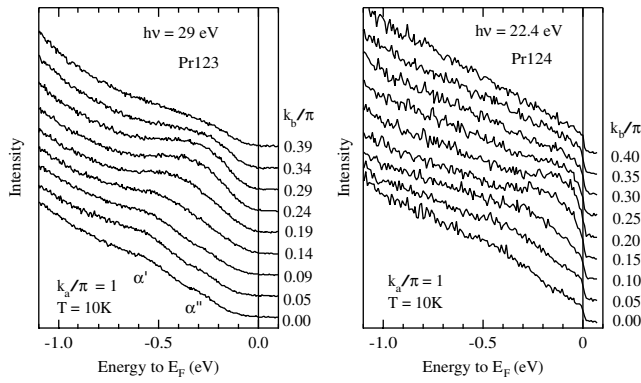

FIG. 1. Schematic drawings of the experimental arrangement and the Cu-O single and double chains at the cleaved surfaces of Pr123 and Pr124. The closed and open circles indicate Cu and oxygen ions, respectively.

FIG. 3.   ARPES spectra taken along the Cu-O chain in Pr123 and Pr124. k_a is the momentum perpendicular to the chain and k_b is the momentum along the chain.

would be responsible for this suppression of charge ordering in Pr124. It is also possible that, since each double chain of Pr124 consists of two single chains (see Fig. 1), charge ordering is unstable because of the weak interaction between the two single chains in each double chain [16].

The ARPES study of $Li_{0.9}Mo_6O_{17}$ shows that the spectral weight near E_F is considerably suppressed even in the metallic region and gives the anomalous exponent α of 0.9 [5], indicating that $Li_{0.9}Mo_6O_{17}$ is a TL liquid close to the CDW instability. On the other hand, the ARPES data of Pr124 have substantial spectral weight at E_F compared to $Li_{0.9}Mo_6O_{17}$ [5] and, in the low-energy region, Pr124 does not show the TL-liquid behavior. This is consistent with the fact that Pr124 has a large Hall coefficient at low temperature and behaves as a 2D system [10]. Probably, the deviation from the TL-liquid behavior is caused by the hopping between the double chains which becomes relevant in the low-energy region.
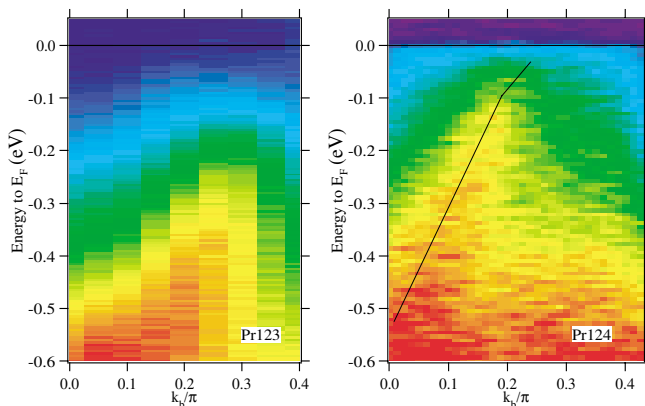
The hopping term between the double chains is estimated to be ~ 10 meV from the 1D-2D crossover observed in the transport measurements of Pr124 [10]. In the energy region higher than the hopping term, the 1D character is expected to manifest in the ARPES spectra. In order to show the dispersions more clearly, the second derivatives of the ARPES spectra are displayed in Fig. 5. In Pr123, two dispersive features labeled as α' and α'' are visible as two bright belts which can be attributed to holon and spinon dispersions, respectively, and are the manifestations of the spin-charge separation in 1D systems as predicted theoretically [17]. The holon and spinon dispersions have the width of ~ 0.5 and ~ 0.1 eV, respectively, which approximately agree with t and J in the t-J model for the cuprates [18]. On the other hand, Pr124 does not show separate spinon and holon features expected for the 1D t-J model [18]. One possible explanation is that, in the high-energy region, Pr124 behaves as a TL liquid with anomalous exponent α larger than 0.5 because of its proximity to charge ordering as proposed in the recent optical study on Pr124 [14]. In this case, since the TL model predicts that the spinon feature appears as a cusp instead of a power-law divergence, the spinon feature is expected to be broad and may not separately be observed from the holon feature [5,17].

As seen in Figs. 3 and 4, the ARPES spectra for $k_b/\pi <$ 0.20 have substantial spectral weight near E_F although the dispersive feature is located well below E_F (see Fig. 2). In the second derivative shown in Fig. 5, this spectral weight near E_F appears as a horizontal bright belt near E_F ranging from $k_b/\pi = 0.0$ to 0.4. In addition, for $0.20 < k_b/\pi <$ 0.25, the dispersion becomes flatter and it looks like a small pseudogap tends to open at E_F. These behaviors in the metallic Cu-O chains of Pr124 are similar to those observed along $(\pi, 0) \rightarrow (\pi, \pi)$ above T_c in the underdoped CuO_2 plane of the high-T_c cuprates [19], which in the stripe phase represent the dispersion along the 1/4-filled

FIG. 4 (color).   Density plot of the ARPES spectra for Pr123 and Pr124 along the chain direction. k_b is the momentum along the chain. Intensity increases in going from blue to red regions. The solid line outlines the Cu-O band dispersion in Pr124. While, in Pr124, the width of the dispersion below E_F is ~ 0.5 eV, the bandwidth is reduced to ~ 0.3 eV in Pr123 because of the band gap opening.
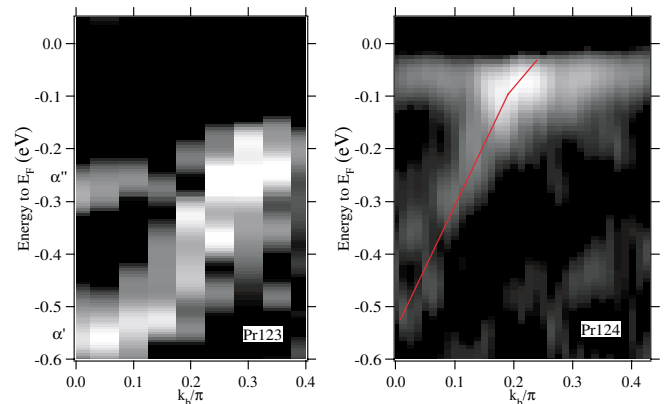

FIG. 5 (color).   Second derivatives of the ARPES spectra for Pr123 and Pr124 along the chain direction. k_b is the momentum along the chain. The red solid line outlines the Cu-O band dispersion in Pr124. In Pr123, two dispersive features are visible as two bright belts.

stripe [20]. The similarity between the coupled chains and the high-T_c cuprates has been argued by Kopietz, Meden, and Schönhammer in light of the crossover between a TL liquid and a 2D Fermi liquid [21]. It would be interesting to investigate whether the coupled-chain model can explain the present ARPES data of Pr124 as well as those of the high-T_c cuprates.

In conclusion, we have studied the hole-doped Cu-O chains in Pr123 and Pr124 using ARPES. The ARPES data show that the Cu-O single chain in Pr123 and the Cu-O double chain in Pr124 are approximately 1/4 filled. While, in Pr123, the 1D features from the Cu-O single chain lose their weight near E_F, the dispersive feature from the Cu-O double chain reaches E_F in Pr124. These facts indicate that the charge ordering occurs in the Cu-O single chain and is suppressed in the Cu-O double chain. In the low-energy region, the line shape of the ARPES spectra of Pr124 cannot be explained by the TL-liquid picture or by the Fermi-liquid picture and looks rather close to that of the 2D CuO$_2$ plane with stripes. It would be useful to study further the differences and similarities between the metallic Cu-O chains in Pr124 and the metallic stripe phase in the high-T_c cuprates.

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