Angle-resolved photoemission study of Zn-doped PrBa₂Cu₄O₈: Possible observation of single-particle spectral function for a Tomonaga-Luttinger liquid

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We have studied the electronic structure of metallic Cu-O double chains in Zn-doped PrBa₂Cu₄O₈ (Pr124) using angle-resolved photoemission spectroscopy. The single-particle spectral function of Zn-doped Pr124 is compatible with a power-law spectral function expected for a Tomonaga-Luttinger liquid. This is in contrast to the fact that the photoemission spectrum of pure Pr124 shows a Fermi step and can be interpreted as a Fermi liquid realized in coupled metallic chains. The exponent α is estimated to be 0.6 ± 0.1 that is one of the smallest values among those obtained for various one-dimensional metals.

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Low-energy excitations in one-dimensional (1D) metals described by the Tomonaga-Luttinger (TL)-liquid scheme show power-law behaviors in the spectral functions of various physical quantities.[1–4] The exponents of the spectral functions are determined by the magnitude of coupling constant K_ρ that characterizes each 1D metallic system. Angle-resolved photoemission spectroscopy (ARPES) is a powerful tool to observe the single-particle spectral function $A(k,\omega)$ as a function of momentum k and energy ω relative to the Fermi level E_F. For the TL-liquid state, $A(k,\omega)$ is expected to show a power-law singularity near E_F rather than the δ function peak of the Fermi liquid and the momentum-integrated single-particle spectral function $\rho(\omega) = \Sigma_k A(k,\omega)$ is expected to show the power-law behavior of $|\omega|^\alpha$ for small $|\omega|$. There have been considerable efforts to observe the TL spectral function using photoemission spectroscopy in 1D organic conductors,[5,6] Au chains on Si(111) surface,[7] and Mo oxides.[8,9] In these systems, the spectral weight near E_F is considerably suppressed and the anomalous exponent α is close to or larger than 1.[5] The relatively large α commonly observed in these 1D metallic systems suggests that these 1D systems may commonly have strong fluctuations of charge ordering or charge-density wave coupled with lattice degrees of freedom.[1,2] On the other hand, the interpretation of the ARPES data still remains highly controversial. It has been pointed out that the apparent TL behaviors of the organic conductors and of the Mo oxides are due to the bad surface quality[6] and the limited momentum resolution,[9] respectively.

The Cu-O chain is one of the most idealistic systems to study the single-particle spectral function of the 1D metals. The hole concentration or the filling of the Cu-O chain can be varied. In addition, the coupling between the chains can be controlled by chemical substitution. Also, it is very interesting to compare the spectral function of the hole-doped Cu-O chain with that of the hole-doped CuO₂ plane, which shows the high T_c superconductivity and may be another class of non-Fermi liquid. It was found that ARPES of the undoped Cu-O single chains in Sr₂CuO₃ and the double chains in SrCuO₂ have spinon and holon dispersions that are manifestation of the spin-charge separation in 1D Mott insulators.[10,11] An ARPES study of PrBa₂Cu₃O₇ (Pr123) has shown that the hole-doped Cu-O single chain in Pr123 has a band gap opening at E_F due to charge ordering and also shows spinon and holon dispersions.[12] On the other hand, in PrBa₂Cu₄O₈ (Pr124), the photoemission spectrum of the hole-doped Cu-O double chain shows a Fermi step at E_F and does not behave like a TL liquid.[13] This indicates that the chain-chain coupling is substantial in Pr124 to realize a Fermi-liquid state. This is consistent with the fact that the conductivity of Pr124 shows a two-dimensional (2D) behavior at low temperatures within the a-b plane.[14]

Very recently, Nakada *et al.* found that, when Zn is doped into Pr124, the resistivity along the a-axis direction shows a nonmetallic temperature dependence down to at least 2 K while the b-axis direction preserves its metallic behavior.[15] Here, the b axis is in the Cu-O double chain direction as shown in Fig. 1. It is believed that the Zn ions are mainly substituted for the Cu ions of the CuO₂ plane. Actually, the band dispersion from the Cu-O double chain reported in the present work is as clear as that of pure Pr124, indicating that the Zn ions do not disturb the hole-doped Cu-O chain. Therefore, Zn-doped Pr124 is more suitable for studying the 1D nature of the Cu-O double chain than pure Pr124. In this work, we report ARPES data of the Zn-doped Pr124 that show a TL-liquid spectral function in contrast to the Fermi-liquid spectral function of pure Pr124. The exponent α estimated from the ARPES data of Zn-doped Pr124 is 0.6 ± 0.1. This is the smallest value among those reported for various 1D metals.
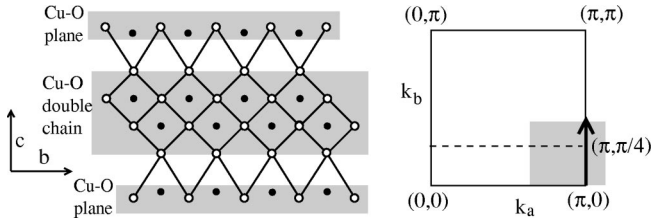
FIG. 1. Left panel: A schematic drawing of the Cu-O double chain and the CuO_2 plane in Zn-doped Pr124. The open and closed circles represent oxygen and Cu ions, respectively. The Cu-O chain is along the b axis. The cleaved surface is perpendicular to the c axis. Right panel: The first Brillouin zone of the Cu-O chain and the CuO_2 plane. The dashed line indicates an expected Fermi surface for the 1/4-filled Cu-O chain.

Naturally untwinned single crystals of Zn-doped Pr124 were grown by a flux method as reported in the literature.[15,16] The amount of Zn doping is nominally 5%. The ARPES measurements of Pr124 were performed at beam line 5-4 equipped with a Scienta SES 200 electron analyzer, at Stanford Synchrotron Radiation Laboratory (SSRL). The chamber pressure during the measurements was less than 5 $\times 10^{-11}$ Torr. The samples were cooled to 10 K and cleaved *in situ*. The cleaved surfaces were the a-b plane and the CuO_4 square units of the Cu-O chain are perpendicular to the cleaved surface (see Fig. 1). Orientation of the single crystals was done by Laue diffraction before the measurement. We have cleaved two samples and checked the reproducibility. The observation of the dispersive feature near E_F indicates that the surface quality is comparable to or better than that of Pr123.[12] The position of E_F was calibrated with the gold spectra. The experimental uncertainty in the energy calibration was ± 1 meV. The incident photons were linearly polarized and had an energy of 22.4 eV. The polarization vector of the incident photons had a component parallel to the Cu-O chain direction. The total-energy resolution including the monochromator and the analyzer was approximately 20 meV. The angular resolution was $\pm 0.28°$ that gives the momentum resolution of $\pm 0.01\pi$ in units of $1/a$ or $1/b$ ($a = 3.88$ Å and $b = 3.90$ Å).

Figure 2(a) shows a set of ARPES data of Zn-doped Pr124 taken along the Cu-O chain direction, which is indicated by the arrow in the right panel of Fig. 1. 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 going from $k_b/\pi = 0.0$ to 0.25, a broad dispersive feature from the Cu-O chain moves to E_F. The dispersive feature reaches E_F around $k_b/\pi = 0.25$ and loses its intensity for $k_b/\pi > 0.25$, indicating that Fermi momentum k_F is $\sim \pi/4$. This dispersive behavior does not depend on k_a although its intensity depends on k_a due to a transition-matrix element effect.

The 1D behavior is clearly seen in the spectral weight mapping of the ARPES data shown in Fig. 3. The spectral weight is integrated from -0.05 eV to E_F and is plotted as a function of k_a/π and k_b/π. The mapped region is indicated by the shaded area in the right panel of Fig. 1. One can see a horizontal red belt where the dispersive structure is
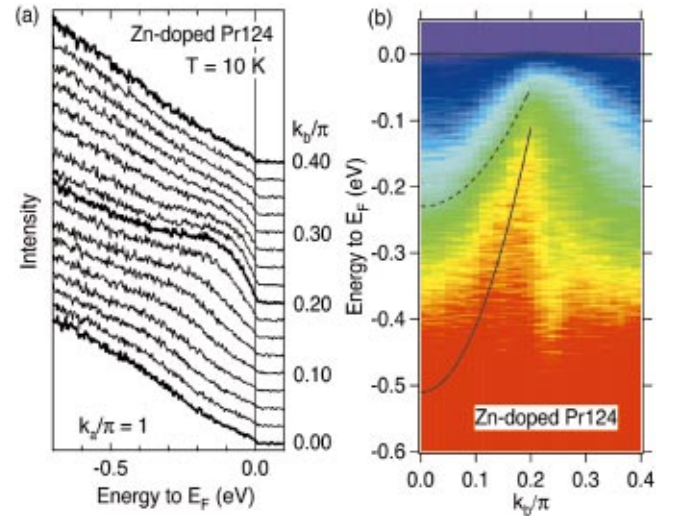

FIG. 2. (Color) (a) ARPES spectra and (b) density plot along the Cu-O chain in Zn-doped Pr124. k_a and k_b are the momentum perpendicular to the chain and that along the chain, respectively. The solid and dashed curves roughly indicate the dispersion of the broad feature and the contour on the high-energy side, respectively.

located in the window from -0.05 eV to E_F. The upper edge of this red region indicated by the thick dashed line is approximately given by $k_b/\pi \sim 0.25$ and does not depend on k_a/π. Because of the transition-matrix element effect, the spectral weight is enhanced for $k_a/\pi \sim 1$, which is consistent with the recent model calculation.[17] Although the band dispersion and the Fermi surface is almost 1D, the almost 1D band structure can be compatible with a Fermi liquid as well as a TL liquid. In order to clarify whether the system is a quasi-1D Fermi liquid or a TL liquid, the line shape of the spectral function should be analyzed.

In the ARPES data shown in Fig. 2(a), the centroid of the broad dispersive feature has the dispersion of ~ 0.5 eV,
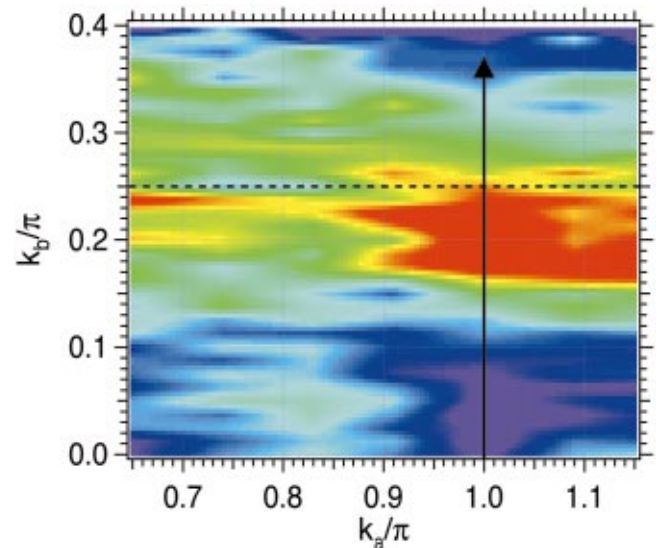

FIG. 3. (Color) Spectral weight mapping for Zn-doped Pr124. The spectral weight is integrated from -0.05 eV to E_F.

which approximately agrees with the magnitude of t in the t-J model for the cuprates and can be assigned to the holon dispersion.[18] On the other hand, the spinon dispersion is not clearly separated from the holon dispersion. However, as shown in the following paragraphs, Zn-doped Pr124 behaves as a TL liquid with anomalous exponent α larger than 0.5. In this case, since the TL model predicts that the spinon feature appears as a cusp instead of a power-law divergence,[1] the spinon feature is expected to be obscured and may not separately be observed from the holon feature. Actually, in Zn-doped Pr124, the holon dispersion from $k_b/\pi = 0.0$ to 0.25 has a tail on the high-energy side that could be assigned to the obscured spinon feature. The momentum dependence of the tail feature can clearly be seen in the density plot [Fig. 2(b)]. The tail on the high-energy side of the holon dispersion gives a clear parabolic contour in going from $k_b/\pi = 0.0$ to 0.25 as indicated by the dashed line in Fig. 2(b). It is possible to assign this parabolic contour to the cusp of the spinon feature predicted for a TL liquid with $\alpha > 0.5$.[1] This parabolic contour has the dispersion of ~ 0.2 eV that approximately agrees with the magnitude of J in the t-J model for the cuprates.[18] Interestingly, this parabolic contour becomes hampered in pure Pr124,[13] probably because the cusp of the spinon feature is destroyed by the chain-chain coupling.[19]

Although α was deduced from the ARPES data near k_F in some earlier works,[7,8] Xue *et al.* pointed out that determining α in this way would be difficult if the momentum resolution is not good enough.[9] On the other hand, it is possible to estimate α if one can construct an angle-integrated photoemission spectrum from the ARPES data. Since the 1D band of the Cu-O double chain is occupied for $0 < k_b/\pi < 0.25$, it is reasonable to construct the angle-integrated photoemission spectrum by summing the ARPES data from $(\pi,0)$ to $(\pi,0.4\pi)$. Here, we used the ARPES data normalized at -0.9 eV. We have confirmed that the normalization procedure does not affect the conclusion. For example, the estimated α is not changed when the ARPES data are normalized using the integrated intensity up to -0.9 eV. Figure 4 shows the angle-integrated photoemission spectrum that can be viewed as a momentum-integrated spectral function $\rho(\omega)$. We have estimated α by fitting the angle-integrated photoemission spectrum above -0.2 eV to a power-law function $A\omega^\alpha$ and a linear background. The linear background is expected, e.g., from the $k_b = \pi/4$ spectrum and is probably derived from the CuO_2 plane or the Pr-O layer. Here, the power-law function is convoluted with a Gaussian to take into account the energy resolution. In Fig. 4, a typical fitted result is shown by the solid curve that reproduces the experimental result. We have estimated the error bar for α by fitting the spectrum to the power-law function without the linear background as well as by changing the energy range down to -0.1 eV. The estimated exponent α falls in the range of 0.6 ± 0.1, which is much smaller than those deduced for the organic conductors,[5] $Li_{0.9}Mo_6O_{17}$,[8] and Au chains on Si(111).[7] Since α is given by $(K_\rho + K_\rho^{-1} - 2)/4$, α of 0.6 ± 0.1 gives K_ρ of 0.24 ± 0.04 that is in good agreement with the value ($K_\rho = 0.24$) obtained from the optical study by Tak-
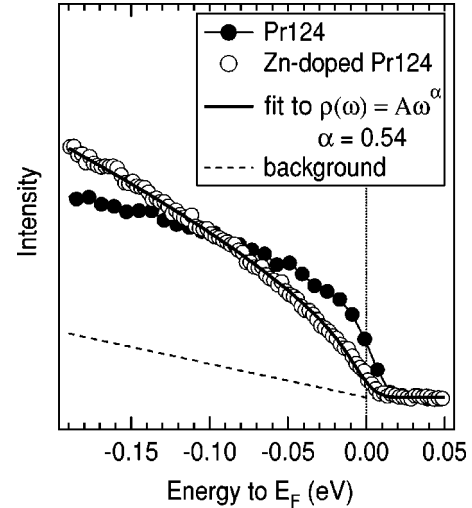

FIG. 4. Photoemission spectra of Zn-doped Pr124 and pure Pr124 obtained by summing the ARPES spectra from $(\pi,0)$ to $(\pi,0.4\pi)$. The solid curve indicates the fitted result and the dashed line represents a linear background for the fitting.

enaka *et al.*[20] It has recently been suggested that the charge ordering in the 1/4-filled Cu-O chain is frustrated in the double chain system.[21] Therefore, even when the chain-chain coupling is reduced by Zn doping, the metallic Cu-O double chain in Pr124 can avoid charge ordering and exist as a TL liquid. Since the fluctuation of charge ordering tends to be small compared to other 1D systems, the Cu-O double chain can have relatively small α. Another possibility is that the boundary effect[4] that can enhance α is less important in Pr124 than other 1D systems.

In Fig. 4, the angle-integrated spectrum of pure Pr124 is compared with that of Zn-doped Pr124. The spectral weight at E_F is substantial for Pr124 indicating that the spectral function of Pr124 cannot be fitted to a power-law function and can be fitted to a step function broadened due to the energy resolution. On the other hand, since the spectral weight at E_F is almost zero for Zn-doped Pr124, it is possible to fit the spectral function of Zn-doped Pr124 to a power-law function as shown in Fig. 4. Therefore, while Pr124 can be interpreted as a Fermi liquid, the spectral function of Zn-doped Pr124 shows the TL behavior within the limitation of the finite-energy resolution.[22] Probably, in Pr124, the chain-chain coupling is strong enough to give the Fermi-liquid behavior. This is consistent with the fact that pure Pr124 has a large Hall coefficient at low temperatures and behaves as a 2D system.[14] Since holes are sitting in the $x^2 - y^2$ orbital of the CuO_4 square units in the Cu-O chain, the direct hopping term between the neighboring Cu-O chains is expected to be small. On the other hand, since the CuO_4 square plane in the Cu-O chain sharing the apical oxygen with the CuO_5 pyramid of the CuO_2 plane below it (see Fig. 1), the hopping term via the CuO_2 plane is important compared to the direct hopping term and mainly gives the coupling between the Cu-O chains. When the CuO_2 plane is disturbed by Zn doping, the chain-chain coupling is reduced and, consequently, the conductivity perpendicular to the Cu-O chain would become nonmetallic.[15]

In conclusion, we have studied the hole-doped Cu-O double chains in Zn-doped Pr124 using ARPES. The ARPES data show that the Cu-O double chain in Pr124 is approximately 1/4 filled. The spectral function near E_F can be fitted to a power-law spectral function, suggesting that a TL-liquid state is realized in the Cu-O double chain of Zn-doped Pr124. The exponent α estimated from the photoemission spectra is 0.6 ± 0.1. This value is one of the smallest among those obtained for various 1D metals investigated by photoemission spectroscopy. With α larger than 0.5, the TL scheme predicts that the holon component has a singularity while the spinon part has only a cusplike structure. This prediction reasonably agrees with the ARPES data of Zn-doped Pr124. In future, ARPES of Zn-doped Pr124 should be performed with higher energy and momentum resolution in order to exclude the possibility of a Fermi liquid. Also further experimental and theoretical studies are required to explain the relatively small α in the Cu-O double chain and to systematically understand the magnitude of α obtained for various 1D metals.

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