Fermi Surface and Band Dispersion in La$_{2-x}$Sr$_x$CuO$_4$

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Using angle-resolved photoemission spectroscopy (ARPES), we observe the band structure, the Fermi surface and their doping dependences in La$_{2-x}$Sr$_x$CuO$_4$. The results reveal that the Fermi surface undergoes a dramatic change: it is holelike and centered at $(\pi,\pi)$ in underdoped ($x=0.1$) and optimally doped ($x=0.15$) samples as in other cuprates, while it is electronlike and centered at $(0,0)$ in heavily overdoped ($x=0.3$) ones. The peak in the ARPES spectra near $(\pi/2,\pi/2)$ is broad and weak unlike that in other cuprates. In the underdoped and optimally doped samples, a superconducting gap ($\Delta = 10 - 15$ meV) is observed near $(\pi,0)$.

KEYWORDS: La$_{2-x}$Sr$_x$CuO$_4$, ARPES, Fermi surface, band dispersion, superconducting gap, doping dependence

Among the family of high-$T_c$ cuprate superconductors, La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) provides unique opportunities to study the systematic evolution of the electronic structure with hole doping. First, LSCO has a simple crystal structure with single CuO$_2$ layers. It has neither Cu-O chains as in YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) nor complicated structural modulation of the block layers as in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212). Second, the hole concentration in the CuO$_2$ plane can be controlled over a wide range and uniquely determined by the Sr concentration $x$ (and the small oxygen non-stoichiometry). One can therefore investigate the doping dependence of the electronic structure continuously from the heavily overdoped limit ($x \sim 0.35$) to the undoped insulator ($x = 0$) in the same system. This information would be highly useful to critically check existing theories of electron correlations and superconductivity in the CuO$_2$ plane. Indeed, the doping dependences of thermodynamic and transport properties have been extensively studied for the LSCO system.$^{1-5}$

Angle-resolved photoemission spectroscopy (ARPES) is a powerful method to probe the electronic structure of low-dimensional systems. In particular, band structures, Fermi surfaces$^{6-12}$ and superconducting and normal-state gaps in the high-$T_c$ cuprates$^{13-19}$ have been observed by ARPES. However, most of the ARPES experiments have focused on the Bi2212 system and its family compounds; ARPES studies of LSCO have been hindered probably because LSCO is difficult to cleave and its surface is not as stable as that of Bi2212 under an ultrahigh vacuum.

We have recently focused on the LSCO system and carried out a series of photoemission studies.$^{20,21}$ The angle-integrated photoemission (AIPES) spectra of LSCO show a broad feature (at $\sim -100$ meV) and a suppression of the density of states at the Fermi level ($E_F$) in the underdoped region.$^{21}$ In the present study, we have overcome the experimental difficulties in the ARPES of LSCO using high-quality single crystals, and have measured ARPES spectra of underdoped ($x = 0.1$), optimally doped ($x = 0.15$) and heavily overdoped ($x = 0.3$) samples in order to investigate the doping dependence of the electronic structure of the CuO$_2$ plane, in particular, the shape of the Fermi surface and the band dispersions.

Single crystals of LSCO ($x = 0.1, 0.15$ and 0.3) were grown by the traveling-solvent floating-zone method. ARPES measurements were carried out at beamline 5-3 of Stanford Synchrotron Radiation Laboratory (SSRL), using incident photons with energies of 22.4 eV and 29 eV. The wave vector and the electric vector of the incident photons and the sample surface normal were kept in the same plane and the angle of incidence was $45^\circ$. The total energy resolution was approximately 45 meV and the angular resolution was $\pm 1$ degree. The spectrometer was kept in an ultrahigh vacuum better than $5 \times 10^{-11}$ Torr during the measurements and the samples were cleaved in situ. Since the surface degraded rapidly at high temperatures, the measurements were done only at low temperatures ($T \sim 15$ K). The cleanliness of the surface was confirmed by the absence of a hump at energy $\sim -9.5$ eV and a shoulder in the valence band at $\sim -5$ eV. All the spectra presented here were taken within 10 hours and mostly five hours after cleaving. The position of the Fermi level was calibrated with gold spectra for every measurement and the experimental uncertainty of the energy calibration was about $\pm 2$ meV.

ARPES spectra for underdoped ($x = 0.1$) and optimally doped ($x = 0.15$) LSCO are shown in Fig. 1. Although the dispersive features are broad, an angular dependence is identified. As one goes from $(0,0)$ to $(\pi,0)$ in...
the first Brillouin zone (BZ) or from (2\pi,0) to (\pi,0) in the second BZ [Figs. 1(a) and 1(b)], a broad “quasiparticle” (QP) peak emerges from lower energies and then stays somewhat below \(E_F\) around the (\pi,0) point, never crossing the Fermi surface. The midpoint of the leading edge is below \(E_F\) (\(\sim -20\) meV) at \(\sim -(\pi,0)\). As for Fig. 1(b), the peak intensity decreases in going from (1.2\pi,0) to (\pi,0). This is probably due to a matrix-element effect peculiar to the incident photon energy of \(h\nu = 22.4\) eV because such an intensity modulation is not seen in the spectra taken at another photon energy \(h\nu = 29\) eV, as shown in Fig. 1(a). The dispersion of the QP peak is very weak around the (\pi,0) point, meaning that there is a “flat band”, namely, an extended saddle point around (\pi,0), similar to that in other cuprates.9) On the other hand, as one goes from (\pi,0) towards (\pi,\pi) [Fig. 1(d)], the broad peak and its leading edge further approach \(E_F\) and then the peak disappears around \(\sim (\pi,0.2\pi)\). Since a superconducting gap is opened on the Fermi surface at this temperature (\(T \sim 15\) K < \(T_c\)), the leading-edge midpoint stays below \(E_F\) (\(\sim -8\) meV) even for the peak closest to \(E_F\). It has been indeed reported that the minimum-gap locus in the superconducting state coincides with the Fermi surface in the normal state.22) Therefore, we may conclude that the band crosses an underlying Fermi surface around \(\sim (\pi,0.2\pi)\). (If the energy gap of underdoped LSCO remains opened in the normal state as indicated for the underdoped Bi2212,15–17) no real Fermi surface exists in the underdoped LSCO.) As for the (0,0)→(\pi,\pi) cut [Fig. 1(c)], no QP peak is identified in the present spectra. However, the fact that the QP band is below \(E_F\) at (\pi,0) implies that the Fermi surface underlying the superconducting gap is holelike and centered at (\pi,\pi) for \(x = 0.1\), as in other cuprates studied previously such as Bi2212, Bi2201, YBCO and Nd2-xCe_xCuO4.7–12)

Fig. 1. ARPES spectra of underdoped (\(x = 0.1\)) and optimally doped (\(x = 0.15\)) \(La_{2-x}Sr_xCuO_4\) near the Fermi level. Insets show measured points in the momentum space and the polarization of incident photons (arrows).

Fig. 2. ARPES spectra of heavily overdoped (\(x = 0.3\)) \(La_{2-x}Sr_xCuO_4\) near the Fermi level. Insets show measured points in the momentum space and the polarization of incident photons (arrows).

The leading-edge midpoint reaches about -3 meV at the peak moves upwards, reaches \(E_F\) around \(\sim (0.8\pi,0)\) or \(\sim (1.2\pi,0)\), then decreases in its intensity and almost disappears at (\pi,\pi). Here, the leading-edge midpoint also reaches \(\sim +6\) meV above \(E_F\) at \(\sim (1.2\pi,0)\), suggesting that the QP band has indeed reached the Fermi level. That should be contrasted with the spectra observed for

\[ E_F = 22.4 \text{ eV} \]

\[ F_T = 29 \text{ eV} \]

\[ h\nu \]

\[ k_x = 0 \]

\[ k_y = 0 \]

\[ k_x = 0 \]

\[ k_y = 0 \]

\[ k_x = 0 \]

\[ k_y = 0 \]
Bi2212, where the leading-edge midpoint is always below $E_F$ in the $(0,0)\rightarrow(\pi,0)$ cut $\textsuperscript{7,15}$. The intensity decrease at $\sim(0.8\pi,0)$ occurs in both the first and second BZs and in the spectra taken at both $h\nu = 29$ eV and $h\nu = 22.4$ eV. Hence, it is difficult to explain all the observed intensity decreases unless a Fermi-surface crossing occurs there besides the effect of matrix elements, because the matrix-element effect is dependent on the energy of incident photons and is different between the first and second BZs. As for the spectra taken at $h\nu = 29$ eV, the intensity decrease at $\sim(0.8\pi,0)$ and $\sim(1.2\pi,0)$ observed for $x = 0.3$ [Fig. 2(a)] is not found for $x = 0.1$ [Fig. 1(a)], distinguishing the two compositions. Furthermore, in the spectra taken at $h\nu = 22.4$ eV in the first BZ, the QP peak seen at $(0.75\pi,0)$ almost disappears at $(0.92\pi,0)$ for $x = 0.3$ [Fig. 2(b)], in contrast to the spectra for $x = 0.15$ [Fig. 1(e)]. Therefore, we have concluded that the QP band crosses the Fermi surface around $(0.8\pi,0)$ for $x = 0.3$. Close inspection of the spectra [Figs. 2(b) and 2(c)] reveals that a small part of the QP peak weight appears to remain below $E_F$ at $\sim(\pi,0)$, suggesting that the “flat band” stays only slightly above $E_F$ at $(\pi,0)$. In the presence of strong electron correlation, the QP peak is no more a single peak and part of the spectral weight is distributed on the other side of $E_F$. This also explains the intensity drop in going from $(\pi,0)$ to $(\pi,\pi)$ as seen in Fig. 2(c). Along the $(0,0)\rightarrow(\pi,\pi)$ cut [Fig. 2(d)], although the peak is not clearly identified, the edge intensity slightly increases at $\sim(0.38\pi,0.38\pi)$ and then drops at $\sim(0.44\pi,0.44\pi)$, a behavior indicative of Fermi-surface crossing. This intensity variation was reproduced in several measurements. The QP peak near $(\pi/2,\pi/2)$ is broad and weak compared to the peak near $(\pi,0)$, in contrast to other cuprates showing clear QP peaks around $(\pi/2,\pi/2)$ $\textsuperscript{7,8,14,15,23}$. To summarize, the observations indicate that the Fermi surface is electronlike and centered at the $(0,0)$ point for $x = 0.3$, unlike the other cuprates studied so far.

Thus the Fermi surface undergoes a dramatic change from holelike to electronlike between $x = 0.15$ and $x = 0.3$, as the flat band around $(\pi,0)$ moves from below $E_F$ to above $E_F$. The change in the Fermi-surface topology may be related to the observation that the sign of the Hall coefficient changes from positive to negative around $x = 0.25$ in LSCO $\textsuperscript{24,25}$ and has the same tendency as that expected from the local-density-approximation (LDA) band-structure calculation of La$_2$CuO$_4$ by shifting the Fermi level as in the rigid band model. $\textsuperscript{26,27}$ The behavior of the $(\pi,0)$ flat band upon doping is consistent with numerical studies of the Hubbard model $\textsuperscript{28,29}$ Figure 3 shows the Fermi surfaces suggested from the present experiments for $x = 0.1$ and 0.3. The area enclosed by the Fermi surface is $71 \pm 3\%$ of the half BZ area for $x = 0.3$, which agrees well with the number of electrons $1-x$ as expected from the Luttinger sum rule, indicating a “large Fermi surface”. As for $x = 0.1$, the Fermi surface near $(\pi/2,\pi/2)$ (dashed curve) has been tentatively drawn in Fig. 3 so that the enclosed area is consistent with the Luttinger sum rule. According to Fig. 3, the Fermi surface for $x = 0.3$ seems to be almost square and have a large straight portion around $(\pi/2,\pi/2)$. Recently, Fermi-surface nesting $\textsuperscript{30}$ and short-range stripe order $\textsuperscript{31}$ have been proposed as the origins of the incommensurate peaks in the dynamical magnetic structure factor $S(q,\omega)$ observed by inelastic neutron scattering. However, the Fermi-surface nesting would not explain why the incommensurate peaks are smeared out in the overdoped region ($x > 0.25$), as recently observed $\textsuperscript{32}$ in the presence of the straight Fermi surface for $x = 0.3$.

The width of the QP peak around $(\pi,0)$ for LSCO is almost the same as that for Bi2212 with corresponding hole doping $\textsuperscript{7,8,14,15,23}$ even though the dispersive features are weaker for LSCO than for Bi2212. The QP peak becomes broader as $x$ decreases from the overdoped to the underdoped regions. The broad peak on the flat band around $(\pi,0)$ may be related to the broad feature in the AIPES spectra, where the spectral intensity starts to decrease towards $E_F$ in the underdoped LSCO $\textsuperscript{21}$ that is, what is called a high-energy pseudogap or a weak pseudogap $\textsuperscript{33}$ on an energy scale ($\sim 100$ meV) much larger than that of the superconducting gap and the “normal-state gap” ($\lesssim 20$ meV). $\textsuperscript{15-19}$ Both the broad $(\pi,0)$ peak in the ARPES spectra and the high-energy pseudogap in the AIPES spectra have nearly the same energy scales, that is, the order of the superexchange energy $J \sim 100$ meV, and their energies also increase as $x$ decreases, following the development of antiferromagnetic correlations. $\textsuperscript{21}$ The QP peak line shape and its doping dependence in the $(0,0)\rightarrow(\pi,0)$ cut of LSCO are also consistent with the trend seen among Bi2212, Sr$_2$CuO$_2$Cl$_2$ and the $t-t'-t''-J$ model calculation $\textsuperscript{23,34}$ These observations suggest that the broad line shape of the ARPES spectra around $(\pi,0)$ originates from antiferromagnetic correlations.

The magnitude of the superconducting gap may be roughly estimated from the shift of the leading edge on the Fermi surface (Fig. 4) $\textsuperscript{17,18}$ For the non-
superconducting sample \((x = 0.3)\), the leading-edge midpoint is pushed above \(E_F\) \((\sim +6 \text{ meV})\) at the Fermi-surface crossing \((1.2\pi, 0)\) due to the finite instrumental resolution \((\sim 45 \text{ meV})\). On the other hand, the leading-edge midpoint is below \(E_F\), \(\sim -8\) and \(\sim -3 \text{ meV}\) for \(x = 0.1\) and 0.15, respectively, on the Fermi surface near \((\pi, 0)\). Accordingly, the relative shift of the leading edge is about 10 – 15 meV on the Fermi surface near \((\pi, 0)\) for \(x = 0.1\) and 0.15, and is smaller than the typical value \(\sim 25 \text{ meV}\) for Bi2212 and YBCO, \(\sim 4 \text{ Letters}\) doped LSCO. We found that the holelike underlying dispersions and Fermi surfaces in overdoped and underdoped LSCO. We found that the holelike underlying dispersions and Fermi surfaces in overdoped and underdoped LSCO. We found that the holelike underlying dispersions and Fermi surfaces in overdoped and underdoped LSCO. We found that the holelike underlying dispersions and Fermi surfaces in overdoped and underdoped LSCO.

In conclusion, we have successfully observed band dispersions and Fermi surfaces in overdoped and underdoped LSCO. We found that the holelike underlying Fermi surface centered at \((\pi, \pi)\) for \(x = 0.1\) and 0.15 is converted into the electronlike Fermi surface centered at \((0, 0)\) for \(x = 0.3\) (heavily overdoped). The peak in the ARPES spectra near \((\pi, \pi/2)\) is quite broad and weak compared to that of other cuprates. In the underdoped and optimally doped samples, a superconducting gap \((\Delta = 10 – 15 \text{ meV})\) is observed on the Fermi surface at \(\sim (\pi, 0.2\pi)\). More studies are necessary to identify the anisotropy, doping dependence and temperature dependence of the superconducting gap.

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Fig. 4. ARPES spectra for momenta on the Fermi surface (minimum-gap locus) near \((\pi, 0)\) shown by open circles in the inset. For each composition \(x\), the spectrum at \((0,0)\) has been subtracted as the angle-independent background.
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