Two- to Three-Dimensional Crossover in the Electronic Structure of $(Bi, Pb)_2(Sr, La)_2CuO_{6+\delta}$ from Angle-Resolved Photoemission Spectroscopy

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The hole-concentration (x) dependence of the three-dimensional energy-momentum dispersion in $(Bi, Pb)_2(Sr, La)_2CuO_{6+\delta}$ has been investigated by angle-resolved photoemission spectroscopy. For a heavily overdoped sample of $T_c \leq 0.5$ K, an energy dispersion of $\sim$10 meV in width is observed in the vicinity of the $(\pi, 0)$ point with varying momentum along the c axis (k_z). This k_z dispersion is zero for underdoped, optimally doped, and slightly overdoped samples up to a doping level corresponding to $T_c = 22$ K. At higher doping levels we observe significant dispersion of the order of 10 meV (sample with $T_c \leq 0.5$ K). This is clear evidence that at a doping value corresponding to $T_c = 22$ K, a crossover from two- to three-dimensional electronic structure occurs.

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The high-T_c superconductors are widely known to exhibit two-dimensional CuO_2 layers, which are separated from each other by charge reservoir planes, such as the BiO-SrO layers in $Bi_2Sr_2Ca_{n-1}Cu_nO_{4+2n+\delta}$ ($n = 1, 2, 3$), (La,Sr)O layers in $(La, Sr)_2CuO_4$, and BaCuO layers in $YBa_2Cu_3O_{7-\delta}$. Since the electrical resistivity in the CuO_2 plane is, in general, 100 to $\sim$1000 times smaller than that along the direction normal to the CuO_2 plane (along the c axis), it is believed that the carriers are confined to the CuO_2 planes and that the electrical conduction along the c axis is caused by tunneling across the insulating charge reservoir planes. The superconductor-insulator-superconductor junction (Josephson junction) between two CuO_2 layers separated by insulating charge reservoir planes reported for optimally doped superconductors strongly proves the insulating nature of the charge reservoir planes [1–3]. This naturally constructed Josephson junction in the cuprate superconductors is known as an intrinsic Josephson junction.

Recently, however, Hussey *et al.* [4] reported the presence of a coherent three-dimensional Fermi surface in the overdoped $Tl_2Sr_2CuO_{6+\delta}$ (Tl2201) superconductor on the basis of magnetoresistance measurements in the normal state. The observation of an energy gap in tunneling measurements through the intrinsic Josephson junction and the observation of a three-dimensional Fermi surface, both of which are unambiguously determined experimental facts, are definitely contradictory to each other. Since the dimension of the electrons' momentum would be closely related to the high-T_c superconductivity, investigation into the mechanism leading to the these contradictory experimental facts is of great importance in order to gain a deeper insight into the nature of the high-T_c superconductors.

By measuring the electrical resistivity in the CuO_2 plane ($\rho_{a,b}$) and along the c axis (ρ_c), one may readily notice that both $\rho_{a,b}$ and ρ_c decrease with increasing hole concentration x [Figs. 1(a) and 1(b)]. This is most likely due to the reduction in the scattering probability inherent in the weakened electron correlation. Notably, the decrease of ρ_c with

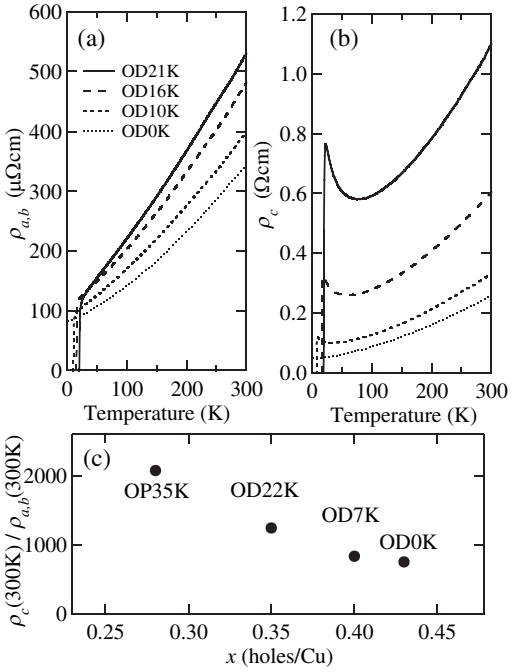

FIG. 1. (a) Electrical resistivity in the a, b plane ($\rho_{a,b}$) and (b) along the c axis (ρ_c) of $(Bi, Pb)_2(Sr, La)_2CuO_{6+\delta}$ at various hole concentrations. The anisotropy of the electrical resistivity $\rho_c/\rho_{a,b}$ at room temperature is shown in (c).

increasing x is obviously larger than that of $\rho_{a,b}$, and hence $\rho_c/\rho_{a,b}$ is drastically suppressed with increasing x as shown in Fig. 1(c). By considering the facts that a three-dimensional Fermi surface was reported for overdoped samples [4], while the insulating nature of the charge reservoir planes was reported in optimally doped conditions, [1–3], one can expect that this dramatic decrease of the $\rho_c/\rho_{a,b}$ ratio is caused by the development of a three-dimensional electronic structure and a coherent electrical conduction with increasing hole concentration. If this scenario is valid, the band that crosses the Fermi level ε_F should have finite dispersion along the k_z direction in the overdoped condition, and this ε-k_z dispersion would be reduced with decreasing x and disappear at a critical hole concentration p_c.

$(Bi, Pb)_2(Sr, La)_2CuO_{6+\delta}$ (Bi2201) is one of the best suited cuprate superconductors to investigate the hole-concentration dependence of dimensionality in the electronic structure, partly because its hole concentration can be controlled from heavily overdoped conditions to underdoped ones [5,6], and partly because it is easily employed for angle-resolved photoemission spectroscopy (ARPES) measurements [7–10]. Using synchrotron-based ARPES measurements we find clear evidence of a ε-k_z dispersion in heavily overdoped Bi2201 and its suppression with decreasing hole concentration. In this Letter, we present the experimental evidence for a three-dimensional dispersion in heavily overdoped Bi2201 and show how the system transforms into a two-dimensional one with decreasing hole concentration.

The Bi2201 single crystals were prepared by the conventional floating zone method and subsequent heat treatments. Details of the sample preparation conditions are reported elsewhere [7,8,11,12]. We employed a heavily overdoped sample with $T_c \leq 0.5$ K, overdoped samples with $T_c = 7$ and 22 K, and an optimally doped sample with $T_c = 35$ K. These are labeled as OD00K, OD7K, OD22K, and OP35K, respectively. The samples are reported to possess large hole concentrations (0.28, 0.35, 0.40, and 0.43 for the OP35K, OD22K, OD7K, and OD0K, respectively), which have been estimated from the momentum area surrounded by the holelike Fermi surface centered at the (π, π) point determined via ARPES measurements [7,8,11].

The ARPES spectra were acquired with the Scienta SES2002 and SES200 hemispherical analyzers at the port 011 undulator-NIM beam line of the Synchrotron Radiation Center, University of Wisconsin at Madison. The total energy resolution was about 20 meV, which was determined by the intensity reduction from 90% to 10% at the Fermi edge of a reference gold in electrical contact with the samples.

Figs. 2(a) and 2(b) represent the ARPES intensity of the OD0K sample acquired at 20 K with two different incident photon energies of 16 and 22 eV, respectively. The intensity images were measured along the cuts shown in

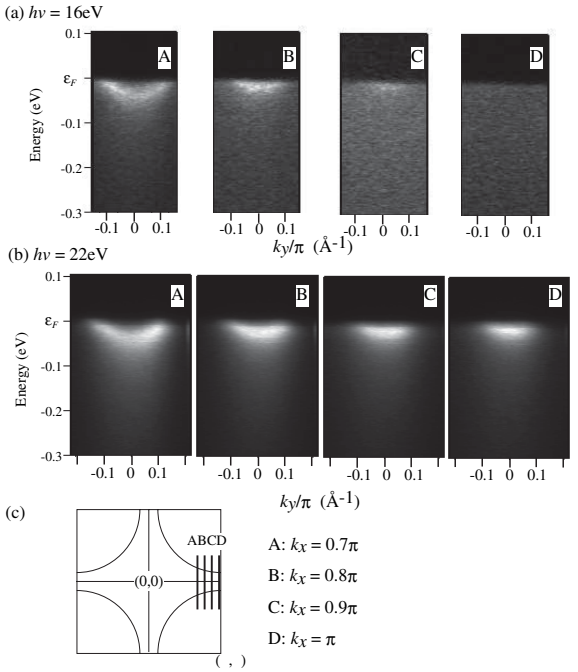

FIG. 2. ARPES intensity images for two different incident photon energies (a) $h\nu = 16$ eV and (b) 22 eV, as a function of energy and momentum obtained along various momentum cuts. These momentum cuts are shown in the two-dimensional Fermi surface in (c).

Fig. 2(c) If the conduction electrons are confined in the two-dimensionally extended CuO_2 planes, one should see no variation in the eigenvalues when the k_z component is varied by altering the photon energy. However, the variation in the eigenvalues with incident photon energy is clearly observed in the ARPES spectra. At $h\nu = 22$ eV, the eigenvalues in the vicinity of $(\pi, 0)$ point stay below E_F, while at $h\nu = 16$ eV the eigenvalues around the $(\pi, 0)$ point are completely lifted up above E_F and no states are observable below E_F. Thus, it is strongly argued that the wave function of the conduction electrons in a CuO_2 plane has finite overlap with that in the next CuO_2 plane, and that coherent Bloch states are extended three-dimensionally at least in the heavily overdoped condition.

The three-dimensional energy-momentum dispersion in the OD0K is more clearly confirmed by observing the shape of the two-dimensional Fermi surface (FS) at different values of the k_z component of the momenta, i.e., at different photon energies of $h\nu = 16$, 18, 22, and 28 eV. We determined the Fermi wave vectors (k_F's) from the peak momentum in the momentum distribution curves at E_F, as shown in Fig. 3(a). We then use them to construct the Fermi surface in the two-dimensional first Brillouin zone for the four photon energies shown in Fig. 3(b). Variation in the shape of the FS is observable especially near the $(\pi, 0)$ point; the holelike FS centered at the (π, π) point makes contact with the surrounding FS's at the $(\pi, 0)$ point when it is measured at $h\nu = 16$ eV, while at the other photon energies it never touches the other FS's. The varia-

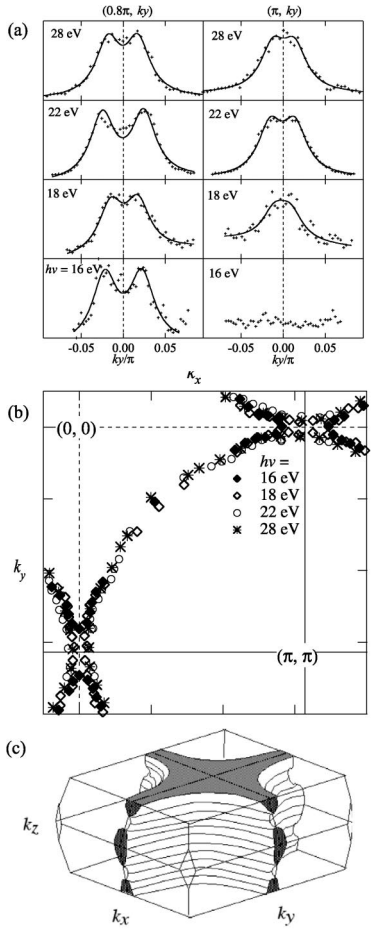

FIG. 3. (a) Momentum distribution curves at two momentum cuts, B and D, shown in Fig. 2(c). Solid lines indicate fitting result with a function consisting of two Lorentzian peaks. (b) Two-dimensional Fermi surfaces of the OD0K sample measured at four different incident energies of $h\nu = 16$, 18, 22, and 28 eV. (c) Schematic illustration of the three-dimensional Fermi surface of the OD0K. The Fermi surface has a cylindrical shape centered at the (π, π, k_z) line, and touches the surrounding ones at around the $(\pm\pi, 0, \pm\pi/2)$ and $(0, \pm\pi, \pm\pi/2)$ points.

tion in the FS is also pronounced near the node direction [$(0, 0)$-(π, π) direction]. The variation of the two-dimensional FS determined from this work is certainly different from that reported by Hussey *et al.* [4], where almost no variation of the two-dimensional FS near the $(\pi, 0)$ point and anti-node direction was observed.

By using the free-electron final state approximation and a roughly estimated 10 eV inner potential, we calculate k_z at the $(\pi, 0)$ point for different incident photon energies. The resulting k_z's for $h\nu = 16$, 18, 22, and 28 eV are $k_z \sim -0.3\pi$, 0.1π, 0.8π, and -0.1π, respectively. A schematic illustration of the three-dimensional FS constructed from the measured data is shown in the inset of Fig. 3(b). A cylindrical Fermi surface with its center at the (π, π, k_Z) momentum line makes contact with the neighboring ones at around $(\pm\pi, 0, \pm\pi/2)$ and $(0, \pm\pi, \pm\pi/2)$.

Within the framework of the Boltzmann transport theory, the direction dependence of the electrical conductivity can be roughly estimated from the area of the Fermi surface A_{FS}, obtained from a projection of the FS along the current-direction. A small but finite A_{FS} can be observed along the k_z direction for the OD0K sample. This strongly suggests the presence of a Boltzmann-type coherent electrical conduction along the c axis. The A_{FS} obtained in the direction perpendicular to the c axis clearly has a much larger value than that along the c axis. This large anisotropy in A_{FS} should be responsible for the large anisotropy in the electrical conductivity of the heavily overdoped samples of the three-dimensional coherent metallic system. One should note however that the contribution of the anisotropy of the A_{FS} is much weaker than that caused by the insulating nature of the charge reservoir planes in the underdoped and optimally doped samples, and therefore $\rho_c/\rho_{a,b}$ drastically increases with decreasing hole concentration.

To quantitatively investigate the hole-concentration dependence of the three-dimensional electronic structure, we determined the ε-$k_{x,y}$ dispersion in the vicinity of the $(\pi, 0)$ point along the anti-node direction at different photon energies. This momentum cut was selected because the in-plane momentum $k_{x,y}$ can be unambiguously determined by observing the folding point of the measured ε-k dispersion and the variation in the shape of FS for the OD0K sample was pronounced around $(\pi, 0)$ point. The ε-k dispersions determined from the ARPES data are shown in Fig. 4. The eigenvalue of the saddle point is located slightly above E_{F} in the OD0K and just a few meV below E_{F} in the OD7K. The values of the dispersion above the chemical potential were obtained from ARPES spectra measured at 200 K divided by the Fermi-Dirac distribution function. This method is very useful for revealing the state slightly above the chemical potential, that is chopped off by the Fermi-Dirac distribution function

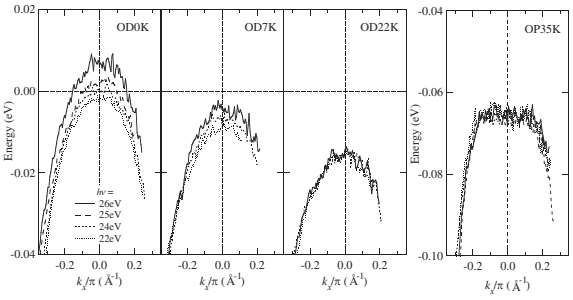

FIG. 4. Energy-momentum dispersions about the $(\pi, 0)$ point along the antinodal line measured with different incident photon energies. Each eigenvalue is determined by the peak energy of the energy distribution curves. Energy dispersion of 10 meV along the k_z direction was observable for OD0K, and this ε-k_z dispersion disappears with decreasing hole concentration. Only the dispersion curves of the OP35K show a wide flat portion about the $(\pi, 0)$ point. This is related to the enhanced electron correlation associated with the reduced hole concentration.

[13]. The ARPES spectra for OD22K and OP35K could be measured at low temperature (20 K), because the reduction in the hole concentration leads to a higher E_F and the saddle point moves sufficiently down below E_F where the intensity reduction due to both the Fermi-Dirac distribution function and superconducting gap ($\Delta \sim 8$ meV at OP35K) is negligible.

The width of the ε-k_z dispersion thus determined is about 10 meV for the OD0K, and that in the OD7K is reduced to $\sim$5 meV as can be seen in Fig. 4. Eventually, no variation with incident energies was observable in the ε-k_z dispersion of OD22K and OP35K. The hole-concentration dependence of the three-dimensional electronic structure suggests that the overlap of the wave functions between the CuO_2 planes is reduced with decreasing hole concentration and eventually the electronic structure becomes two dimensional for $x \sim$ values less than 0.35 holes/Cu.

We should comment on the results obtained by tunneling spectroscopy using the intrinsic Josephson junction. This is the best way to experimentally reveal the two-dimensionally confined electronic states in the CuO_2 planes. Recent experiments of the tunneling spectroscopy on our OD0K sample using a small mesa island on a clean surface definitely showed an absence of the intrinsic Josephson junction along the c axis in sharp contrast to its presence for the OP35K sample. [14] These observations lend strong support to our scenario of the x induced three- to two-dimensional crossover in the electronic structure.

Finally, we briefly discuss the mechanism leading to the hole concentration x-induced dimensional crossover in the electronic structure of Bi2201. The electronic structure in a two-dimensional square lattice is characterized by a van Hove singularity (vHs), which is manifest as a sharp peak in the electronic density of states $N(\varepsilon)$. The sharp peak in $N(\varepsilon)$ is produced by the saddle structure in the ε-k dispersion at the $(\pi, 0)$ point in the two-dimensional Brillouin zone. The Fermi level is pinned exactly at this peak energy (ε_{peak}) at the hole concentration with which the electronlike FS centered at the Γ point takes the place of the holelike one centered at (π, π). This situation, $\varepsilon_F \sim \varepsilon_{peak}$, must induce instability of the two-dimensional system because of the extremely large population of electrons at the highest kinetic energy. In such a case, the crystal structure would be slightly deformed as the two-dimensional electronic structure transforms into the three-dimensional one, because such a deformation greatly broadens the peak at $N(\varepsilon_F)$ and the total kinetic energy of the electrons is greatly reduced. Indeed the three-dimensional dispersion is most pronounced in the OD0K sample, which is the only one that shows the electronlike FS, in sharp contrast to the holelike ones in other samples with smaller x [7,8].

It is worthwhile to mention here that two-dimensional-like properties were theoretically predicted by Klemm *et al.* [15] for layered superconductors at temperatures below T_c even though the three-dimensional long range ordering in electronic structure is established from layer to layer. Dimensional crossover predicted by Klemm *et al.* was experimentally confirmed in some layered superconductors [16,17]. The observation of the two-dimensional nature in the optimal-doped high T_c superconductors [1–3] could be closely related to the Klemm's theory. In order to gain much deeper insight into the origin of the dimensional crossover experimentally revealed in the present work, further studies including careful consideration of the Klemm's theory are strongly required.

In summary, we find direct evidence for a three-dimensional energy-momentum dispersion in an overdoped Bi2201 high-T_c superconductor using ARPES with a synchrotron radiation photon source. The energy dispersion along the k_z direction in the heavily overdoped OD0K sample is $\sim$10 meV. With decreasing hole concentration this energy dispersion is gradually reduced and becomes zero at a hole concentration of $x = 0.35$. These experimental facts indicate that the x induces a dimensional crossover of the wave functions in the high T_c superconductor.

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