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Systematics of transport anisotropy in single-domain $La_{2-x}Sr_xCuO_4$ films

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The ability to control the tilt angle of CuO_2 planes with respect to the film surface has led to successful synthesis of single-domain, epitaxial $La_{2-x}Sr_xCuO_4$ films of (103) and (109) orientations corresponding to tilt angles of 21° and 49°, respectively. The systematics of the *c*-axis resistivity and transport anisotropy ρ_c/ρ_{ab} have been studied as a function of Sr doping varying from 0.04 to 0.34 over $2 \leq T \leq 800$ K. The temperature dependence of the *c*-axis resistivity evolves from semiconductinglike behavior in the lightly doped and optimally (superconducting) doped regions ($x \leq 0.15$) to metallic behavior in the heavily doped region ($x \geq 0.25$), displaying a notable discontinuity of the slope at the tetragonal-orthorhombic phase transformation. The crossover from two-dimensional metallic transport to strongly anisotropic three-dimensional metallic transport was observed near the superconducting phase boundary ($x \sim 0.22$). The evolution of transport anisotropy ρ_c/ρ_{ab} closely resembles the evolution of the electronic orbital character of doping-induced holes, hence suggesting a connection between the two-dimensional electronic structure and superconductivity.

The mechanism for high-temperature superconductivity in the cuprates has been widely recognized to be closely associated with anisotropic normal-state properties. Despite systematic convergence in the description of CuO_2-plane properties,[1,2] the nature of out-of-plane charge conduction and its relevance to superconductivity remained elusive. Progress on experimental investigations of the out-of-plane properties has been hampered by the lack of samples with useful geometry and reliable quality. Marked inconsistency was found among earlier reports on the *c*-axis resistivity of crystals, and illustrated a sensitive dependence on doping content and defect distribution within the sample.[3] On a theoretical ground, profound disagreement exists among different formalisms. Band-structure calculations using local density approximation indicated an anisotropic three-dimensional metal for high-T_c cuprates, where the Fermi surface is characterized as an array of square cylinders exhibiting low dispersion in the *c*-axis direction.[4] Alternatively, two-dimensional Luttinger-liquid theory based on one-band Hubbard model emphasized the confinement of charge carriers in the CuO_2 planes, which in turn inhibits coherent transport across the planes.[5] Incoherent Giaver tunneling was proposed to be responsible for the *c*-axis transport displaying "semiconducting" behavior with a $1/T$ dependent resistivity.[5]

To shed light on this issue, we have undertaken extensive investigations of the full temperature dependence of the normal-state resistivity for both in-plane[6] and out-of-plane directions, as the carrier concentration is varied systematically by chemical doping. The simplicity of the crystal structure and the possibility of cation doping make the $La_{2-x}Sr_xCuO_4$ system ideally suited for this type of study. The measurement of *c*-axis resistivity over 800 K was enabled by our recent attainment of single-domain $La_{2-x}Sr_xCuO_4$ films of (103) and (109) orientations, of which the CuO_2 planes are uniaxially tilted off the substrate plane by 49° and 21°, respectively.[7] The values of ρ_c deduced from these two sets of films with different tilts of CuO_2 planes show a high degree of consistency, thus lending support to the method employed. A large magnitude and strong T dependence of resistivity anisotropy ρ_c/ρ_{ab} were observed in the lightly doped and optimally doped (superconducting) regions, indicative of two-dimensional transport. However, in the heavily doped region ρ_c/ρ_{ab} is nearly T independent with the magnitude approaching the prediction of band calculation,[4] implying anisotropic three-dimensional (3D) character. The crossover from 2D transport to anisotropic 3D transport occurs at compositions near the disappearance of bulk superconductivity. Furthermore, the systematics of transport anisotropy are correlated with the evolution of the electronic orbital character of doping-induced holes,[8] thus suggesting a connection between the 2D electronic structure and the occurrence of superconductivity.

The $La_{2-x}Sr_xCuO_4$ films of $0.04 \leq x \leq 0.34$ were prepared by *in situ* growth using 90° off-axis sputtering previously described.[9] Single-domain growth of (103) and (109) orientations employs vicinal (101) $SrTiO_3$ and (103) $SrTiO_3$ substrates, respectively. The structural analysis was done by x-ray diffraction on a four-circle diffractometer.[7,9] For resistivity measurements, the samples were mechanically cleaved to ~ 0.8 mm wide strips along the in-plane axes of [30$\bar{1}$] and [010] for (103) films, and [90$\bar{1}$] and [010] for (109) films, respectively. A conventional four-probe method was used with the electrodes formed by evaporating silver films. The high-temperature (> 300 K) measurements were conducted in a quartz tube furnace under clean O_2 flow.

Growth on vicinal (101) $SrTiO_3$ substrates with surface normal rotated about [010] by a few degrees results in single-domain (103) oriented films.[7] Deliberate miscut in this fashion exposes the (100) $SrTiO_3$ surface steps on which the (001) perovskite planes of $La_{2-x}Sr_xCuO_4$ nucleate favorably. The fact that the interplanar spacings in the *c*-axis direction of $La_{2-x}Sr_xCuO_4$ are incommen-

surate with the a,b lattice parameters has suppressed nucleations of degenerate antidomains to a concentration less than 10^{-4}. Following similar energetics, this model predicts single-domain growth of other orientations such as (109) films on (103) $SrTiO_3$. The high index plane (103) of $SrTiO_3$ has an asymmetric surface structure exposing the (100) surface steps naturally without intentional miscut. As illustrated in Fig. 1, nucleation of (001) perovskite plane of $La_{2-x}Sr_xCuO_4$ takes place on $SrTiO_3$ (100) steps, and leads to (109) oriented growth. Unwanted (101) domains are completely suppressed. The tilt angle of CuO_2 planes thus amounts to be the angle between the (100) step and the (103) face of $SrTiO_3$. X-ray diffraction employing longitudinal and ϕ scans confirmed that the (109) films are indeed of single variant with few impurities. The film morphology exhibits regular stepped terraces running along the film [90$\bar{1}$] with a typical dimension of $\sim$700 Å in width.

Based on two sets of single-domain samples with respective tilt angles of 21° and 46°, the c-axis resistivity can be deduced from the measured resistivities along the two in-plane axes according to the relation: $\rho(\theta)=\rho_{[010]}\cos^2\theta+\rho_c\sin^2\theta$, where $\rho_{[010]}$ and ρ_c are the resistivity along the CuO_2 plane and c axis, respectively, and θ is the angle between ρ and $\rho_{[100]}$. Because the in-plane contribution $\rho_{[010]}$ is negligible compared to ρ_c, the ratio of $\rho_{30\bar{1}}/\rho_{90\bar{1}}$ is approximately $\sin^2(46°)/\sin^2(21°)$ $\sim$4.4. Shown in Fig. 2 is the resistivity data along [30$\bar{1}$] and [90$\bar{1}$] from the (103) and (109) $La_{1.93}Sr_{0.07}CuO_4$ films, respectively. The $\rho_{30\bar{1}}/\rho_{90\bar{1}}$ data agree with the expected value within 10%. The minor discrepancy below 100 K is attributed to a sensitive dependence of ρ_c on the Sr content in the lightly doped region, and unavoidable compositional spreads in a solid solution system. Since ρ_c is prone to be affected by distributions of defects and dopants,[3] the consistency between (103) and (109) samples assures that our results do represent the intrinsic properties of the materials.

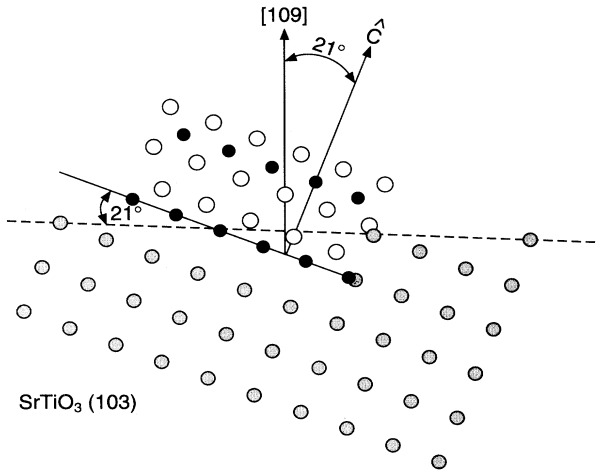

FIG. 1. Schematics illustrating single-domain growth of (109) films nucleating from (001) surface steps naturally exposed on a (103) $SrTiO_3$ substrate. The cation positions of film and substrate structure are indicated. The solid, open, and shaded circles stand for Cu, La (or Sr), and Ti (or Sr) atoms, respectively.

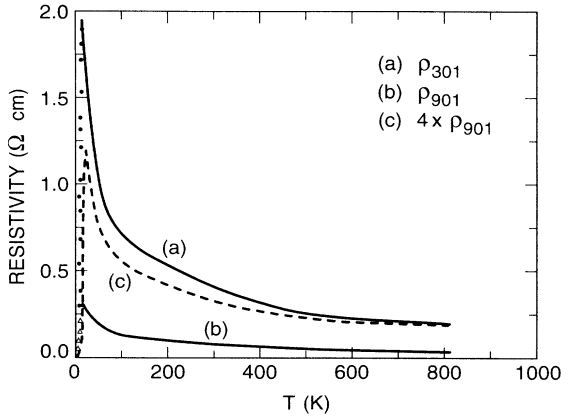

FIG. 2. Temperature dependence of the resistivity along [30$\bar{1}$] and [90$\bar{1}$] of (103) and (109) films of $x=0.07$, respectively.

Plotted in Fig. 3 is the systematic evolution of ρ_c up to 800 K with increasing Sr doping. In general, the ρ_c vs T data are characterized by a sum of $\rho_0+\rho_1(T)$. The first term is a T-independent "offset" ρ_0, which shows a rapid reduction by nearly four orders of magnitude with x increasing from 0.04 to 0.34. The second term ρ_1 is T dependent and shows semiconductinglike behavior in the lightly doped region ($x<0.15$), and evolves to metallic behavior in the heavily doped region ($x\geq 0.25$). The ρ_1 data of $x=0.04$ and 0.07 are consistent with an activated behavior showing approximately an $\exp(1/T)$ dependence, whereas the $x=0.34$ data follow a superlinear power law of $T^{1.3}$ for 20 K$\leq T\leq$800 K. In the opti-

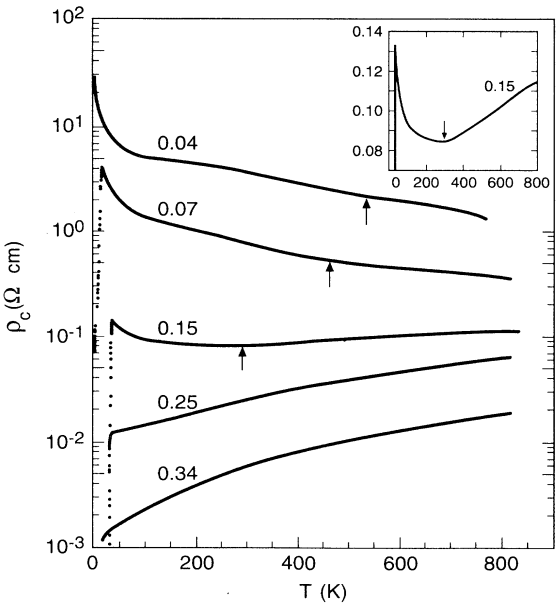

FIG. 3. Semilog plots of ρ_c vs T for $x=0.04$, 0.07, 0.15, 0.25, and 0.34. The inset shows the linear plot of ρ_c for $x=0.15$. The arrows denote the temperature of the orthorhombic-to-tetragonal transformation where a discontinuous change of $d\rho_c/dT$ appears.

mized superconducting region ($x \sim 0.15$) ρ_1 is weakly metallic showing a linear dependence above 300 K, and changes to a $1/T$ dependence below 300 K. Evidence for a discontinuous change of the resistivity slope, as marked by arrows in Fig. 3, is noted at 540, 470, and 300 K for $x = 0.04$, 0.07, and 0.15, respectively. A discontinuous change of the sign of $d\rho_c/dT$ was first reported in single-crystal data of $x = 0.15$, and attributed[10,11] to the structural transformation from a high-temperature tetragonal phase to a low-temperature orthorhombic phase at $T_{O,T}$. Our observation in the thin film data, though weaker, is consistent with such an interpretation.

The ρ_c data at the superconducting composition $x \sim 0.15$ clearly show nonmetallic behavior below 300 K, in contrast to the metallic T dependence of ρ_c in fully oxygenated $YBa_2Cu_3O_{7-x}$ crystals. In fact, for $x < 0.2$, the c-axis conductivity is smaller than the Mott minimal metallic conductivity, and displays a semiconducting T dependence. Here the Mott minimal metallic conductivity $\sigma_{\min}$ in the c-axis direction is about 10^2 S/cm, deduced from the extension of the Mott-Ioffe-Regel minimum metallic conductivity criterion for an anisotropic system.[3,12] The low conductivity in these cases implies that the c-axis mean free path is shorter than the unit cell dimension, and that c-axis momentum cannot be defined. Furthermore, conventional localization due to disorder cannot be responsible since localization is a coherent backscattering phenomenon in all three directions, and localization in solely one direction is not possible.[5]

A number of alternative mechanisms for the c-axis conduction have been suggested, yet presently there is a clear lack of agreement. The unusual $1/T$ dependence below 300 K for $x = 0.15$ was ascribed to incoherent Giaver tunneling between the CuO_2 planes by the Luttinger-liquid theory.[5] However, the origin of the significant contribution ρ_0 is not at all clear. Resonating tunneling through localized states in the insulating barrier between the CuO_2 planes was proposed to be responsible for this T-independent term.[13] At $T > 300$ K, it has also been suggested, by analogy to the 1D organic conductors, that the metallic behavior with $d\rho_c/dT > 0$ may come from the T-linear dependence of the in-plane scattering rate of charge carriers that control the tunneling rate to the neighboring plane.[14] Finally, a "dynamic dephasing" description proposed that the c-axis conduction has to do with scatterings from in-plane thermal fluctuations, and depends on the ratio of the interlayer hopping rate of CuO_2 sheets to the thermal energy.[15]

Shown in Fig. 4 is the transport anisotropy, expressed as the ratio ρ_c/ρ_{ab}. In general the anisotropy reduces drastically in magnitude with increasing Sr doping as well as with increasing T; however, markedly different T dependence is noted in different doping regimes. Following the generalized Drude model, ρ can be expressed as $(m^*/ne^2)\tau^{-1}$, where the scattering rate τ^{-1} is solely responsible for the observed T dependence of ρ according to optical conductivity measurements.[16] Relating the anisotropy data to the scattering rate leads to the following conclusions: in the lightly doped and superconducting regions, the large magnitude and the strong T dependence of the resistivity anisotropy suggest that the carriers are tightly confined to the planes, and that the in-plane and the out-of-plane conductions obey distinctly different scattering mechanisms, as a signature for 2D transport. On the other hand, as Sr doping exceeds 0.25, the resistivity anisotropy is much reduced, and approaching the band calculation[4] for an effective mass ratio of $m_c^*/m_{ab}^* \sim 25$. Further, the weak T dependence of transport anisotropy suggests that the scattering rates τ_c^{-1} and τ_{ab}^{-1} are comparable and follow a similar T dependence, consistent with a picture of anisotropic 3D transport within the framework of Fermi-liquid theory. The dimensional crossover of charge transport from 2D to anisotropic 3D occurs in a composition range where bulk superconductivity disappears.[17] Furthermore, the much weakened T dependence of ρ_c/ρ_{ab} at $T \geq T_{O,T}$ may suggest that the tetragonal phase is likely described by the anisotropic 3D picture, and that the orthorhombic phase displays the unusual 2D transport, along with the appearance of superconductivity.

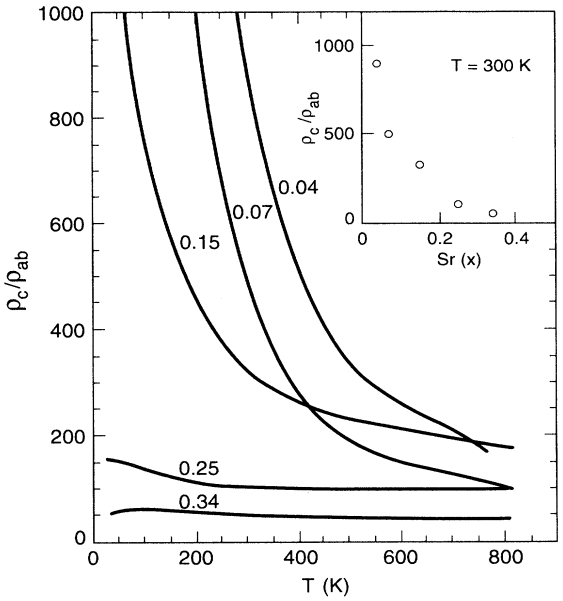

FIG. 4. Temperature dependence of transport anisotropy ρ_c/ρ_{ab} for $x = 0.04$, 0.07, 0.15, 0.25, and 0.34. Plotted in the inset is the ρ_c/ρ_{ab} ratio at 300 K vs Sr content.

Our transport anisotropy systematics share similar trends of compositional dependence and T dependence with previous results of single crystals measured below 300 K.[3,10,11,18] In particular, quantitative agreement is seen between our lightly doped films and the work by Kimura *et al.*,[11] where high-quality single crystals have been produced by the floating zone traveling solvent method. In the heavily doped region, our samples of $x = 0.34$ show an anisotropy about a factor 2 lower than bulk crystals.

Plotted in the inset of Fig. 4 is the resistivity anisotropy at 300 K vs Sr content. The systematic increase of conductivity anisotropy with doping suggests an enhanced interplanar coupling, and that may have to do, in part, with redistributions of the underlying electronic states of charge carries. As demonstrated recently in

polarization-dependent O K-, and Cu L-edge absorption spectroscopies on a similar set of single-domain $La_{2-x}Sr_xCuO_4$ films, the distribution of the hole-doped states evolves from being largely two-dimensional-like of predominantly O $2p_{x,y}$ character in the lightly and optimally doped regions ($x \leq 0.15$) to essentially three-dimensional-like displaying significant O $2p_z$ character in the heavily doped region ($x \geq 0.25$).[8] The consistency between these microscopically and the macroscopically determined results thus reveals a unified picture that the electronic structure crosses over from a 2D to anisotropic 3D when transversing the superconducting phase boundary.

In conclusion, the systematic evolution of anisotropic transport with carrier concentrations in the one-layered cuprate $La_{2-x}Sr_xCuO_4$ has been determined over 800 K. The single-domain epitaxial films with tilted CuO_2 planes enable the investigations of charge conduction across the planes, and demonstrated the striking crossover from 2D transport in the lightly doped and superconducting regions to anisotropic 3D transport in the heavily doped nonsuperconducting region. The c-axis transport property in the former case is consistent with a picture of incoherent tunneling between the planes. Further hole-doping into the latter region, the system eventually acquires the nature of an anisotropic metal with a superlinear T-dependent scattering rate in all three directions, and an effective mass anisotropy approaching the band calculation. Our present findings, together with the observation of the electronic orbital character of the doped hole evolving from 2D to 3D by soft x-ray spectroscopy, suggest a close link between the 2D electronic structure and the occurrence of superconductivity in $La_{2-x}Sr_xCuO_4$.

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