Systematic Evolution of Temperature-Dependent Resistivity in $La_{2-x}Sr_xCuO_4$

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The in-plane resistivity (ρ_{ab}) of $La_{2-x}Sr_xCuO_4$ has been studied over a wide temperature (4–1000 K) and composition range ($0 < x < 0.35$). The much discussed T-linear resistivity is observed only in the narrow composition region associated with optimal superconductivity. In the underdoped range ($x < 0.1$), we observe first indications of resistivity saturation and analyze the resistivity as indicative of a "small" Fermi surface. In the overdoped range ($x > 0.2$), ρ_{ab} follows a novel power-law dependence, $\rho \propto T^{1.5}$, over the entire temperature range up to 1000 K.

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The unusual charge dynamics in the layered cuprates has been widely recognized to be a key for the understanding of high-temperature superconductivity and has promoted a number of theoretical models for 2D strongly correlated electron systems [1,2]. One of the hallmarks of the charge dynamics is the nearly T-linear in-plane resistivity (ρ_{ab}) over a remarkably wide temperature range, from just above T_c to near 1000 K, which is commonly observed in most cuprates showing the optimum superconductivity [1–4]. Closely related with this is the $1/\omega$ decay of the free carrier conductivity at optical frequencies, which reflects an anomalous frequency-dependent scattering rate proportional to ω at least up to 0.5 eV, instead of ω^2 as would be expected for a conventional Fermi liquid [5]. Lacking so far, however, is a reliable experimental study of ρ_{ab} over a wide temperature range on well characterized materials when the hole concentration is varied from far below to far above the optimum composition for superconductivity.

Here we report the electrical resistivity up to 1000 K on high-quality $La_{2-x}Sr_xCuO_4$ single-crystal thin films and polycrystalline materials, not only for the superconducting phase but also at lower and higher doping level. We find that the T-linear behavior for the resistivity over a wide temperature range is confined to a narrow composition range for optimum superconductivity. A well-defined decrease of the resistivity slope occurs above room temperature in the underdoped region, below $x \sim 0.1$. In the overdoped region above $x \sim 0.2$, a novel power-law dependence, $\rho = \rho_0 + AT^n$, $n \sim 1.5$, not T linear or T^2, dominates the resistivity over a wide temperature range up to ~ 1000 K. This is in contrast to earlier suggestions of conventional behavior in this composition range. These results not only indicate a close correlation between the T-linear scattering and superconductivity, but also provide a crucial test for theoretical models of the charge transport in CuO_2 sheets.

The c-axis-oriented single-crystal thin films with thicknesses of 3500–8000 Å were grown on $SrTiO_3(100)$ substrates [6]. The polycrystalline samples were prepared through solid-state reaction processing [7]. The resistivity measurements were performed using the conventional four-probe method or the Van der Pauw technique [8]. The high-temperature measurements above 300 K were conducted in a quartz tube furnace under O_2 flow. In order to check for possible O_2 desorption effects, we performed measurements under N_2/O_2 mixed gas flow ($P_{O_2} \sim 10^{-3}$ atm) for representative compositions. No difference was observed between the two runs up to 1000 K for $0.05 \lesssim x \lesssim 0.2$, and at least up to 800 K for $x > 0.2$, indicating no oxygen desorption up to these temperatures

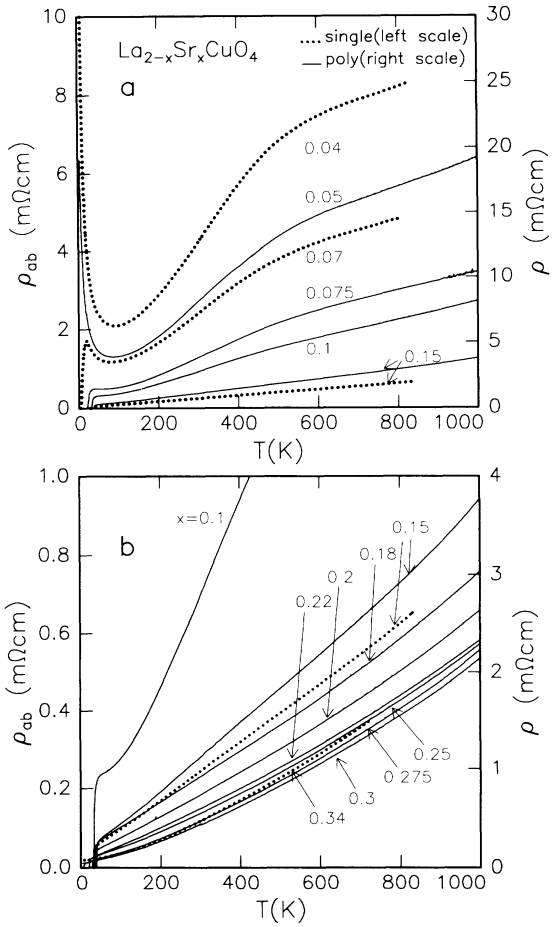

FIG. 1. The temperature dependence of the resistivity for $La_{2-x}Sr_xCuO_4$. (a) $0 < x \leq 0.15$, (b) $0.1 \leq x < 0.35$. Dotted lines, the in-plane resistivity (ρ_{ab}) of single-crystal films with (001) orientation; solid lines, the resistivity (ρ) of polycrystalline materials. Note, $\rho_M = (h/e^2)d = 1.7$ m Ω cm.

[9].

The evolution of the in-plane resistivity (ρ_{ab}) upon Sr doping is given in Fig. 1, showing ρ_{ab} of single-crystal thin films (dots). The high quality of these films is evident from the resistivity at room temperature, e.g., 240 $\mu\Omega$ cm for $x=0.15$, which is significantly lower than the values previously reported for thin films and single crystals [10], and a relatively high $T_c(R=0)$ of 35 K for $x=0.15$. The polycrystalline data are also displayed in Fig. 1 to examine the temperature dependence of resistivity with smaller x intervals. The temperature dependence of the resistivity in the polycrystalline materials is essentially the same as ρ_{ab} of the thin films and the magnitude is 3–4 times larger, as expected for this strongly anisotropic system.

The discussion of the data naturally focuses on three different ranges of carrier concentration. In Fig. 1, the T-linear resistivity can be seen over an extended temperature range only for a narrow composition range near $x=0.15$. This is the composition range where superconductivity with a substantial Meissner signal and specific-heat jump at T_c can be obtained [7].

In the underdoped region below $x\sim0.1$ [Fig. 1(a)], the resistivity shows a pronounced S-shaped temperature dependence. At low temperatures, the resistivity increases as $\Delta\sigma\sim(e^2/h)\ln T$, suggesting weak localization. At high temperatures, we observe a decrease in the slope. This feature becomes less pronounced upon Sr doping, however, it is still noticeable even for $x=0.1$. This becomes clearer when we take the temperature derivative of the resistivity, as shown in the top panel of Fig. 2. A maximum is seen near 300 K, growing in magnitude with decreasing Sr content.

The decrease of the slope at high temperatures is reminiscent of resistivity saturation [11]. It is widely accepted that the resistivity in a metal saturates at high temperatures when the mean free path l of the charge carriers becomes comparable to the interatomic distance, i.e., the minimum scattering length. The gross features of the resistivity curves in Fig. 1 certainly resemble those observed in $A15$ compounds [11].

A remarkable contrast between the present underdoped cuprate and $A15$ compounds, however, is that the resistivity here is more than 1 order of magnitude larger than those of $A15$ compounds. Through an analysis of the magnitude of resistivity based on Boltzmann transport theory, which provides an estimate of the Fermi momentum k_F, this contrast turns out to be related to the issue of whether the Fermi surface in this composition range is as large as that of a nearly half-filled band with $1-x$ electrons or as small as expected for x holes per Cu. (A review of this topic is given in Ref. [12].) In Boltzmann theory, the magnitude of 2D resistivity is given by k_F and l as $\rho=\rho_M/k_Fl$, where $\rho_M=(h/e^2)d=1.7$ mΩ cm (the resistivity at Ioffe-Regel limit, d is the interplane distance ~6.4 Å). Since l must be larger than the minimum scattering length, $a_0/2$ (CuO bond length, a_0 denotes in-

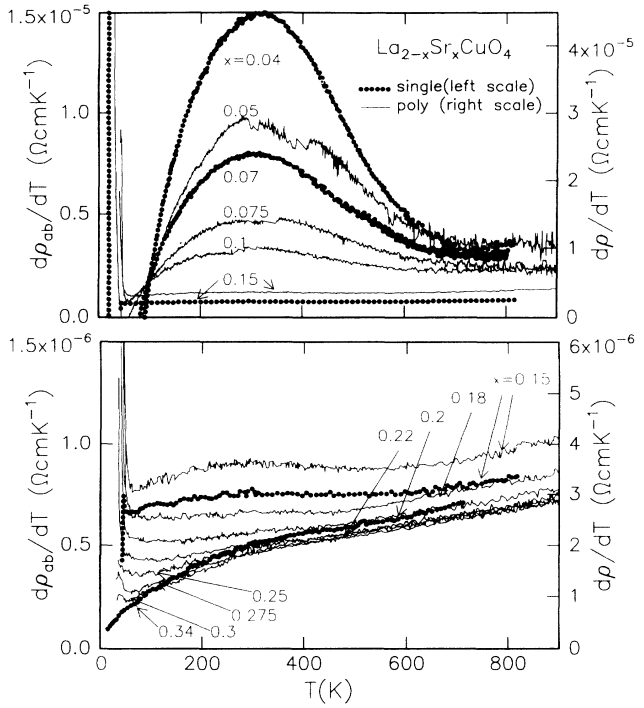

FIG. 2. The temperature derivative of the resistivity $d\rho/dT$ for the samples shown in Fig. 1. Dots, single-crystal thin films; solid line, polycrystalline materials.

plane lattice constant), the fact that the magnitude of the resistivity is even larger than $2\rho_m$ [Fig. 1(a)] implies $k_F\ll1/a$. This yields a substantially smaller k_F than would be expected from the large Fermi surface of a nearly half-filled band, where k_F is of the order of π/a. For a numerical estimate, we assume that the decrease of resistivity slope is indeed due to resistivity saturation. Since $\rho\propto(k_Fl)^{-1}$, and l in the saturation range is a constant $\sim a_0/2$, the systematic decrease of high-temperature resistivity implies an increase of k_F with doping. From the saturated resistivity ρ_s (roughly estimated as ~10 and 6 mΩ cm for $x=0.04$ and 0.07), we obtain k_F values of $0.34/a_0$ and $0.56/a_0$ for $x=0.04$ and 0.07, respectively. They are close to k_F values estimated for a "small" Fermi surface of assumed cylindrical shape, containing x holes ($\sim0.5/a_0$ and $0.66/a_0$ for $x=0.04$ and 0.07). This is the first experimental estimate of k_F in the underdoped region and is consistent with the observation of the large and positive Hall coefficient $\sim1/xe$ [13]. The discussion here therefore strongly suggests the breakdown of the large k_F picture or possibly the inadequacy of the Boltzmann transport model, in the lightly doped region below $x=0.1$.

In the optimum composition range for superconductivity near $x\sim0.15$, where we do not see any saturation up to 1000 K, the resistivity is approximately $(\frac{1}{2}-\frac{1}{3})\rho_M$, i.e., $k_Fl\sim2$–3, at 1000 K. This can be consistent with the "large" k_F picture, since l can be still larger than the minimum scattering length at least up to 1000 K. How-

ever, if that is the case, the compounds with x near 0.15 must be on the border of resistivity saturation at 1000 K.

We focus now on the heavily doped composition region shown in Fig. 1(b). We found an intriguing superlinear behavior of the resistivity up to an unexpectedly high temperature $\sim$1000 K. In the overdoped region above $x\sim0.2$, where superconductivity disappears at the structural phase boundary between orthorhombic and tetragonal [7], the resistivity shows a significant upturn, in contrast to dominant T-linear behavior near $x=0.15$. Similar superlinear behavior has been observed for overdoped $Tl_2Ba_2CuO_6$ up to room temperature [14]. Furthermore, the resistivity becomes almost insensitive to doping above $x\sim0.25$, again in contrast to the compositions with lower Sr content.

In Fig. 3, we plot the resistivity of the samples in the composition range where the resistivity does not strongly depend on doping, as $\log(\rho-\rho_0)$ vs $\log T$. One can see that the resistivity approximately follows a simple power law, $\rho=\rho_0+AT^n$ with $n=1.5\pm0.2$, over a wide temperature range from 4 to nearly 1000 K. By taking the temperature derivative in Fig. 2, we observe a crossover from T linear to $T^{1.5}$ as a function of x. Above $x=0.25$, $d\rho/dT$ is approximately in proportion to $\sqrt{T}$, reflecting the $T^{1.5}$ dependence. With reduced Sr content below $x=0.25$, a plateau region develops at low temperatures, corresponding to T-linear behavior.

It is worth pointing out a close correlation between the temperature dependence of $\rho_{ab}(T)\propto1/\tau_{tr}(T)$, and the frequency dependence of scattering rate, $1/\tau(\omega)$, obtained by fitting the optical conductivity $\sigma(\omega)$ with the generalized Drude formula. The correlation holds true not only in the superconducting phase but also in the overdoped region. In contrast to the ω-linear behavior in the superconducting phase, $x=0.15$, $1/\tau(\omega)$ shows significant superlinear behavior in the overdoped region [15]. We found $1/\tau(\omega)\propto\omega^{1.6}$ in the $x=0.34$ sample of Ref. [15], very close to the power law observed for the temperature-dependent resistivity with the same composition. This is an example of the remarkably general charge dynamics in layered cuprates: $1/\tau_{tr}(T)$ and $1/\tau(\omega)$ are sublinear in the "underdoped," very close to linear in the optimally doped, and superlinear in the overdoped regions of the phase diagram [16].

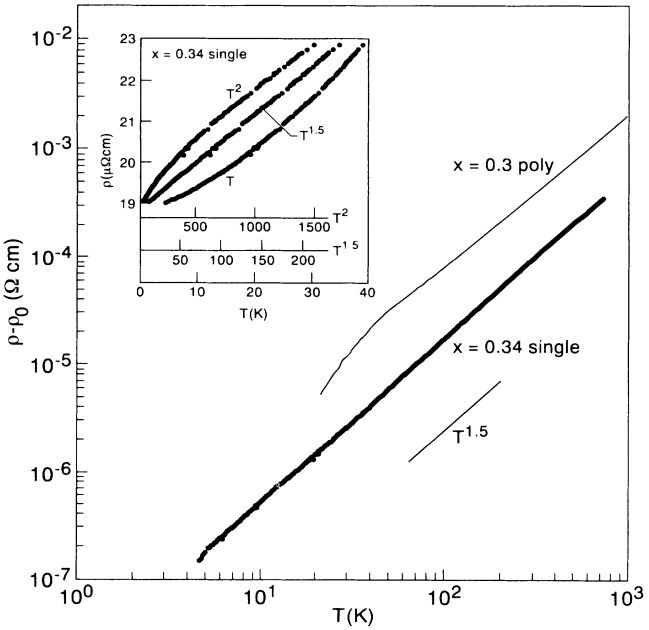

FIG. 3. The temperature dependence of the resistivity for overdoped $La_{2-x}Sr_xCuO_4$. Main panel: $\log(\rho-\rho_0)$ vs $\log T$ for the $x=0.34$ single-crystal thin film (solid line, $\rho_0=1.88\times10^{-5}$ Ω cm) and $x=0.3$ polycrystalline material (thin line, $\rho_0=5\times10^{-5}$ Ω cm). Inset: The resistivity ρ_{ab} vs T, $T^{1.5}$, and T^2 at low temperatures for the $x=0.34$ single-crystal thin film.

Although the overdoped region where superconductivity disappears has been frequently discussed as a conventional Fermi liquid, the unexpected result that the resistivity follows such a simple power law, $\sim T^{1.5}$, over more than 2 orders of magnitude in temperature, does not allow for such a straightforward interpretation. The absence of the T-linear term up to such a high temperature is obviously hard to explain in terms of the conventional electron-phonon coupling, which predicts T-linear behavior above $S\theta_D$ (θ_D denotes Debye temperature, $S=0.2$ - 0.3, see, for example, Ref. [17]).

One may argue that the observed resistivity behavior in the overdoped regime is due to dominant electron-electron scattering which gives rise to scattering in proportion to T^2. The observed behavior, in fact, seems distinctly different, at least from the T^2 resistivity which has been observed in the conventional three-dimensional metals. First of all, the experimentally obtained power is substantially lower than 2. If the obtained power was an artifact of the coexistence of electron-phonon and electron-electron scattering, then the resistivity would be dominated by the T^2 term at low temperatures where the electron-phonon contribution can be neglected. Even below 30 K, however, the temperature dependence is substantially weaker than T^2, as seen from the inset of Fig. 3. A combination of T^2 and T contributions, $\rho_0+BT+CT^2$, does not reproduce the present data. This is evident in Fig. 2, where $d\rho/dT$ is not linear in T. Second, the low-T limiting slope in a ρ vs T^2 plot, 5×10^{-3} $\mu\Omega\,\mathrm{cm\,K^{-2}}$, would be anomalously large in view of the moderate linear specific-heat coefficient, $\gamma\sim7$ mJ/mol K^2, obtained from specific-heat measurements [18]. Although this quantity depends on the details of scattering process, the prefactor for the T^2 term is more than 1 order of magnitude smaller in transition metals with similar γ values [19].

In a 2D Fermi liquid, it has been proposed that the electron-electron scattering is in proportion to $T^2\ln T/T_F$, since the quasiparticle lifetime $\tau(E)\propto1/(E^2\ln E/E_F)$ [20,21]. An attempt to fit the present data by such a modified form failed, giving unphysical T_F values of the order of 10-10^2 K. We note that power laws weaker but

closer to T^2 than the present result have been observed in $Tl_2Ba_2CuO_6$ [14] and $Nd_{2-x}Ce_xCuO_4$ [21]. Obviously, high-temperature studies of these systems would be very useful to clarify the origin of the anomalous superlinear behavior up to high temperatures.

In summary, our extensive study of the electrical resistivity in $La_{2-x}Sr_xCuO_4$, a prototype of the layered cuprates, has revealed an intriguing evolution of the charge transport in the CuO_2 layers as a function of doping. In the underdoped region, a well-defined decrease of the resistivity slope at high temperature, which is likely due to resistivity saturation, was observed for the first time. A simple analysis of the magnitude of resistivity, based on Boltzmann transport, yields a small Fermi surface rather than a large Fermi surface expected from a nearly half-filled band. The very wide temperature range with $\rho \propto T^{1.5}$ in the overdoped region, as well as the $\sim\omega^{1.6}$ dependence of the frequency-dependent scattering rate, illustrates the incomplete understanding of the charge transport, even in this composition region where the simple Fermi-liquid picture has been believed to be applicable. It is impractical here to make a close comparison with a number of pictures developed for the normal state charge transport (see, for example, Refs. [2,22–29]). However, the present data contribute significantly to the ongoing discussions on issues such as "Fermi liquid or non-Fermi-liquid," "large or small" Fermi surface, strong temperature and frequency dependence of relaxation time, and role of antiferromagnetic spin fluctuation.

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