

## Anisotropic resistivities of single-crystal $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ with different oxygen content

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The in-plane resistivity  $\rho_{ab}(T)$  and the out-of-plane  $\rho_c(T)$  have been extensively measured for the pure single-crystal  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi2212) annealed at different oxygen pressure. The  $\rho_c(T)$  and anisotropy [ $\rho_c(T)/\rho_{ab}(T)$ ] decreases rapidly with increasing carrier concentration. It is found that the out-of-plane resistivity decreases linearly with temperature down to about 120 K for the overdoped sample; its resistivity anisotropy is a weak temperature dependence. In the  $ab$  plane, the anisotropy is very weak and nearly independent of temperature. The data of  $\rho_c(T)$  and  $\rho_c(T)/\rho_{ab}(T)$  are well fitted by the bipolaron theory proposed by Alexandrov and Mott. [S0163-1829(98)05945-1]

A distinctive property for the high- $T_c$  layered cuprates is their extremely anisotropic resistivity at temperatures above the superconducting transition temperature  $T_c$ . In the underdoped regime, the  $c$ -axis resistivity has a “nonmetallic” temperature dependence while the in-plane resistivity is metallic in behavior.<sup>1-3</sup> The resistivity anisotropy  $\rho_c/\rho_{ab}$  and  $c$ -axis resistivity  $\rho_c$  decrease systematically with increasing carrier concentration.<sup>1,4</sup> In the overdoped regime, the  $c$ -axis resistivity  $\rho_c(T)$  also shows a metallic behavior,<sup>5,6</sup> especially the  $c$ -axis resistivity of highly oxygenated  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  ( $x > 0.93$ ) decreases linearly with temperature down to the superconducting transition temperature.<sup>5,7</sup> The resistivity anisotropy in overdoped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  ( $x \sim 0.34$ ) is temperature independent as expected of an anisotropic three-dimensional metal.<sup>4,6</sup> These are strong evidence that the transition into the “overdoped” regime involves a crossover from two-dimensional superconducting to three-dimensional metallic behavior. In  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , (Bi2212) however, the resistivity anisotropy  $\rho_c/\rho_{ab}$  is as high as  $10^5$ ,<sup>3,8</sup> which is significantly larger than those of other layered cuprates. The insulating trend in  $\rho_c(T)$  is observable as high as 200 K, even in the overdoped regime.<sup>9,10</sup> An important issue is whether the metallic behavior or  $T$ -linear dependence of the out-of-plane resistivity takes place in the overdoped sample of Bi2212. A further issue concerns the intrinsic behavior of the out-of-plane resistivity for the layered cuprates. Many models for the out-of-plane conduction have been proposed, however no consensus has been achieved so far.<sup>11</sup> Recently, a temperature linear dependence of the  $c$ -axis resistivity has been observed in the overdoped sample of Pb-doped Bi2212, and its magnitude is reduced by four orders and is nearly the same magnitude as that of in-plane resistivity.<sup>12</sup> It is anomalous that the anisotropy is too small, even much less than that

of highly oxygenated  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  and overdoped  $\text{La}_{1.66}\text{Sr}_{0.34}\text{CuO}_4$ . However, less work is made for the  $c$ -axis resistivity of the overdoped Bi2212 sample. It is because the heavy overdoped sample is difficult to obtain.

To clarify further the  $c$ -axis conduction, we have measured in detail the in-plane and the out-of-plane resistivity for more than twenty crystals of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  with a different oxygen content using the Montgomery method.<sup>13,14</sup> We will demonstrate that the in-plane resistivity of Bi2212 is nearly linear with temperature at high temperatures, but the slight deviation from linearity occurred at about 190 K, which is believed to arise from the pseudogap opening for the underdoped crystal. It is observed that the out-of-plane resistivity is metallic above 120 K, and the anisotropy is weakly temperature dependent for the overdoped sample. The out-of-plane resistivity and the anisotropy is well explained by the bipolaron proposed by Alexandrov and Mott.

High-purity single crystals of Bi2212 were grown by self-flux using CuO as flux. The crystals used to measure resistivity had a size of  $1.5 \text{ mm} \times 0.6 \text{ mm} \times 5 \mu\text{m}$  with the shortest dimension along the  $c$  axis. The thickness of the crystal was determined by a scanning electron microscope. To obtain the crystals with the different oxygen content, the as-grown single crystals were annealed at  $500^\circ\text{C}$  and the different oxygen partial pressures of  $10^{-7} \sim 10^2$  atm. Electrical contacts of less than  $2 \Omega$  resistance were established by soldering the copper leads onto the sample surface on which pure silver was evaporated. The Montgomery contact configuration is shown in the inset of Fig. 1 and Fig. 2. Instead of the point contact in the original Montgomery method, we extended the contacts along the edges of the samples to minimize the distortion effect of finite contact size along the  $c$  axis. The obtained crystals were characterized by x-ray dif-

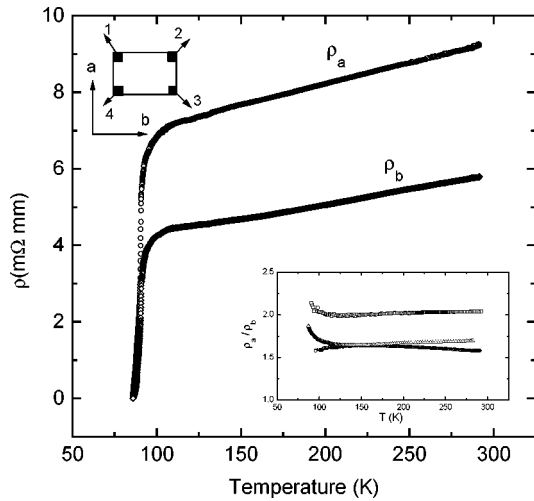


FIG. 1. The temperature dependence of anisotropic in-plane resistivity for the as-grown single crystal, and the resistivity ratio  $\rho_a(T)/\rho_b(T)$  (inset) for the as-grown single crystal (circles) and crystals annealed at the pressure of 100 atm (up triangles) and  $10^{-7}$  atm (squares), respectively. Inset: schematic of the electric contact configuration.

fraction with Cu  $K\alpha$  radiation. Lattice parameters for single crystals were refined using the (0,0,1) Bragg peaks over the  $\theta$  range.

X-ray diffraction shows that the crystals were of the proper Bi2212 phase, and grown preferentially along the  $c$  axis. There is no indication of the presence of the secondary phase. It is found that the  $c$ -axis parameter of crystal in-

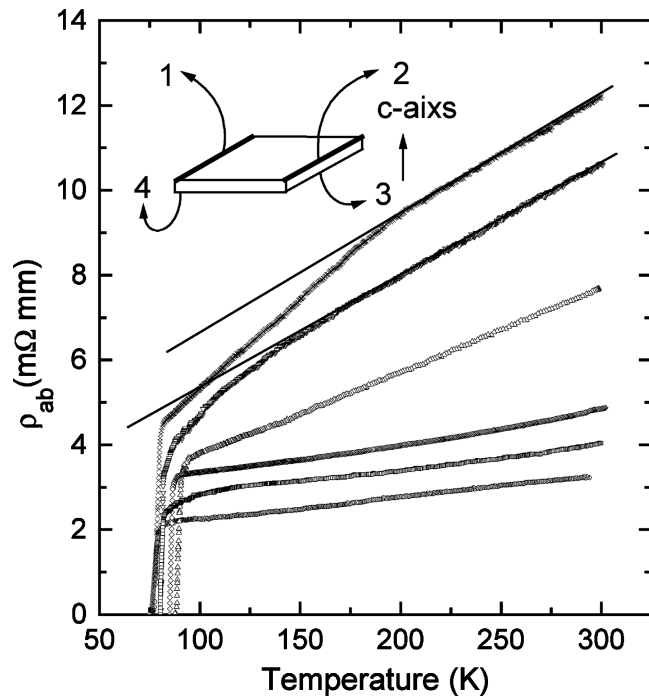


FIG. 2. The temperature dependence of in-plane resistivity  $\rho_{ab}(T)$  for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals annealed at different oxygen pressure; the crystals annealed at oxygen pressure of  $10^{-7}$  atm: crosses;  $10^{-5}$  atm: down triangles;  $10^{-3}$  atm: up triangles; 1 atm: diamonds; 10 atm: squares; 100 atm: circles. Inset: schematic of the electric contact configuration.

creases with decreasing annealing oxygen pressure, as is listed in Table I. It arises from the increase of excess oxygen incorporated between the  $\text{Bi}_2\text{O}_2$  double layers, being consistent with the previous reports.<sup>15,16</sup> To a certain extent, the change of the  $c$  axis is believed to be a token of the change in carrier concentration because the  $c$ -axis parameter is sensitive to oxygen content.

Figure 1 shows the temperature dependence of the anisotropic in-plane resistivity for the as-grown single crystal. A schematic of the Montgomery contact configuration is shown in the inset of Fig. 1. It is found that the normal state resistivity behavior along the  $a$ -axis and  $b$ -axis directions is nearly the same. The in-plane anisotropy [ $\rho_a(T)/\rho_b(T)$ ] is also shown in the inset of Fig. 1. The resistivity  $\rho_a(T)/\rho_b(T)$  is about 1.5–2.0 and temperature independent for the as-grown crystal and the crystals annealed at pressures of 100 and  $10^{-7}$  atm, respectively, which is consistent with the previous report,<sup>17</sup> suggesting a common scattering mechanism for  $a$ -axis and  $b$ -axis transport. Therefore, we will consider the transport along the  $a$  axis and the  $b$  axis to be isotropic for the entire sample. It is worth pointing out that the transport along the  $a$  and  $b$  directions shows a characteristic deviation from linearity at the same temperature in underdoped samples.

Figure 2 shows the temperature dependence of in-plane resistivity  $\rho_{ab}(T)$  for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals annealed at different oxygen pressure.  $T_c$  changes systematically with the annealing oxygen pressure. Here, we do not determine the value of the oxygen content because it is very difficult to accurately determine it for single crystal. However, the change of the  $c$ -axis parameter may be considered as a token of the change of oxygen content. It is noted that the decrease of  $c$ -axis parameter corresponds to the change of  $T_c$  from the underdoped to overdoped regime. The superconducting transition temperature changes from 76 K of the underdoped crystal to 78 K of the overdoped crystal. With increasing annealing oxygen pressure, the value of  $\rho_{ab}(T)$  and the slope  $d\rho_{ab}/dT$  decrease. It indicates that the carrier concentration in the crystals increases with increasing annealing oxygen pressure, corresponding to the decrease of the  $c$ -axis parameter. For the single crystals annealed under oxygen pressures of  $10^{-7}$  and  $10^{-5}$  atm, the resistivity  $\rho_{ab}$  deviates from high-temperature  $T$ -linear behavior at a characteristic temperature  $T^*$  far above  $T_c$ , where  $T^*$  is 190 and 160 K, respectively. This is believed to be evidence for the pseudogap (or spin gap) in the underdoped sample, which has been explained to be due to a decrease in spin scattering caused by the opening of the spin gap.<sup>18</sup> For the single crystals as-grown and annealed under  $10^{-3}$  atm, a typical  $T$ -linear behavior is observed and the  $T_c$  is about 90 K, which is transport characteristic for the optimally doped sample. A  $T^2$  component is observed in the single crystals annealed under oxygen pressures of 10 and 100 atm, indicating the transport behavior of the overdoped sample.

The temperature dependence of out-of-plane resistivity  $\rho_c(T)$  for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals annealed at different oxygen pressure is shown in Fig. 3. A systematic evolution of  $\rho_c(T)$  with reduction of the annealing oxygen pressure in these crystals is observed. That is, the magnitude of  $\rho_c$  increases rapidly. For the optimally doped and underdoped crystals, the resistivity temperature dependence  $d\rho_c(T)/dT$  is negative in the measuring temperature range between the superconducting transition temperature ( $T_c$ ) and

TABLE I. The fitting parameters  $B$ ,  $\sigma_b$ ,  $c$ ,  $\Delta_1$ , and  $Ax^{1/2}$  of Eq. (3) to the  $\rho_c(T)$  data and  $Ax^{1/2}$ , and  $\Delta_2$  of Eq. (1) to the  $\rho_c(T)/\rho_{ab}(T)$  data. The first line gives the annealing oxygen pressure. The second line gives the  $c$ -axis parameters.

Samples	$T_c$ (K)	$c$ axis ( $\text{\AA}$ )	$B$	$\sigma_b(\text{K}^{-1})$	$c(\text{K}^{-1})$	$\Delta_1$ (K)	$Ax^{1/2}$	$\Delta_2$ (K)
100 atm	78	30.58	0.112	0.1443	4.463	310.7	4100.6	152.4
10 atm	80.3	30.61	0.615	0.0026	0.768	409.6	3510.4	416.2
1 atm	85	30.72	0.054	0.0203	0.161	638.0	3097.1	580.4
$10^{-3}$ atm	89	30.92	0.111	0.0007	0.080	501.4	3798.2	507.7
$10^{-5}$ atm	81	30.98	0.027	0.0018	0.008	540.2	3320.6	571.3
$10^{-7}$ atm	76	31.09	0.029	0.0016	0.009	619.4	2809.4	646.7

300 K. For the overdoped crystals, the  $d\rho_c(T)/dT$  is positive at high temperature, while negative at low temperature. In between,  $\rho_c(T)$  has a minimum. The temperature  $T_{\min}$  corresponding to the minimum  $\rho_c$  is about 120 K for the crystal annealed at the oxygen pressure of 100 atm. This feature is shown rather clearly for the crystal annealed at the oxygen pressure of 100 atm by replotting the  $\rho_c$  vs  $T$  curve in the inset of Fig. 3.

Figure 4 shows the temperature dependence of the anisotropy  $\rho_c/\rho_{ab}$  for the same crystals as in Fig. 2. The anisotropy is rather larger, which is consistent with the previous report.<sup>3</sup> The anisotropy increases as the annealing oxygen pressure decreases. This suggests that the mechanisms governing the transport property along and perpendicular to the  $\text{CuO}_2$  plane are different. The temperature dependence of anisotropy becomes weak with increasing annealing oxygen pressure. This is consistent with what is expected since there is strong evidence that a crossover from 2D superconducting to 3D metallic behavior occurs when  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  is doped into the nonsuperconducting overdoped regime ( $x > 0.25$ ).<sup>4,6</sup>

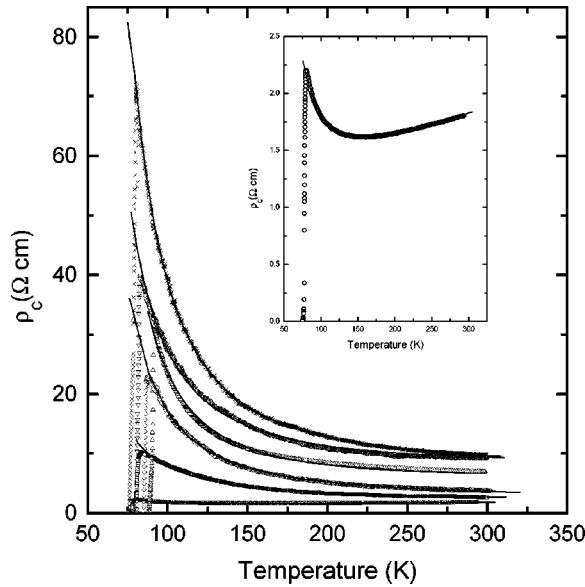


FIG. 3. The temperature dependence of out-of-plane resistivity  $\rho_{ab}(T)$  for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals annealed at different oxygen pressure; the crystals annealed at oxygen pressure of  $10^{-7}$  atm: crosses;  $10^{-5}$  atm: down triangles;  $10^{-3}$  atm: up triangles; 1 atm: diamonds; 10 atm: squares; 100 atm: circles. The solid curves are fits of  $\rho_c(T)$  data to Eq. (3). The  $\rho_c(T)$  vs  $T$  curve for the crystal annealed at the oxygen pressure of 100 atm is replotted in the inset to show more clearly the temperature behavior.

There have been several theoretical models aiming to elucidate the temperature dependence of  $c$ -axis transport  $\rho_c(T)$ ,<sup>11</sup> for example interlayer scattering or phonon (impurity) assisted tunneling, the ‘‘confinement’’ theory proposed by Anderson.<sup>19</sup> In Ref. 9, Watanabe *et al.* have discussed the theoretical models mentioned above. Here, we will interpret our experimental data using the bipolaron theory proposed by Alexandrov and Mott.<sup>20,21</sup> In their theory, polarons dominate in the  $c$ -axis transport at intermediate and high temperatures because they are much lighter in the  $c$  direction than bipolarons. At the same time, the polaron contribution to the in-plane transport is small at any temperature due to their low density compared with the bipolaron one. The normal state out-of-plane resistivity  $\rho_c(T)$  is given by the formula<sup>20,21</sup>

$$\frac{\rho_c(T,x)}{\rho_{ab}(T,x)} = Ax^{1/2} e^{\frac{\Delta(T,x)}{2T}}, \quad (1)$$

where  $A$  is constant,  $x$  is the doping level, and  $\Delta$  is the bipolaron binding energy.

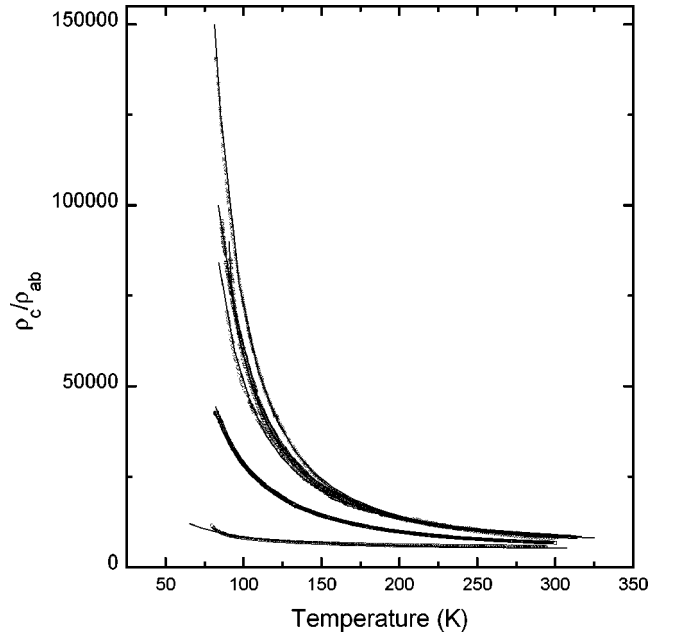


FIG. 4. The temperature dependence of the anisotropy  $\rho_c/\rho_{ab}$  for the same crystals as in Fig. 2 and Fig. 3; the crystals annealed at oxygen pressure of  $10^{-7}$  atm: crosses;  $10^{-5}$  atm: down triangles;  $10^{-3}$  atm: up triangles; 1 atm: diamonds; 10 atm: squares; 100 atm: circles. The solid curves are fits of  $\rho_c(T)/\rho_{ab}(T)$  to Eq. (1).

$$\rho_{ab}(T) = \frac{m^2 C v_0}{4e^2} \frac{T + \sigma_b T^2}{n - n_L + b n_L T}, \quad (2)$$

where  $C = C_{ac} + n_L C_{im}$  and  $\sigma_b = \alpha e^2 b n_L / m^2 C$  is the relative boson-boson scattering cross section,  $n_L$ ,  $n$  are the total number of localized bosons and bosons per unit cell, respectively. Therefore, we can obtain the expression of  $\rho_c(T)$ ,

$$\rho_c(T) = B e^{\Delta(T,x)/2T} T + \sigma_b T^2 / 1 + c T, \quad (3)$$

where  $B = m^2 C v_0 / 4e^2 (n - n_L)$  and  $c = b n_L / (n - n_L)$ .

We fit the experimental data in Fig. 3 and Fig. 4 by using the above expressions of  $\rho_c(T)$  and  $\rho_c(T)/\rho_{ab}(T)$ , respectively. In the fitting,  $\Delta$  is considered to be temperature independent. The fitting curves are shown in Fig. 3 and Fig. 4 as solid lines, and the parameters obtained are listed in Table I. It is found that the experimental results can be well described by the bipolaron theory. The  $\Delta$  obtained by fitting anisotropy [ $\rho_c(T)/\rho_{ab}(T)$ ] data using Eq. (1) is nearly the same as that obtained by fitting the  $c$ -axis resistivity  $\rho(T)$  using Eq. (3), but the  $\Delta$  obtained from Eq. (1) is two times more than that obtained from Eq. (3) for the overdoped sample annealed at the oxygen pressure of 100 atm. It should be pointed out that the anisotropy data of the overdoped samples cannot be well reproduced by Eq. (1). It indicates that our experimental result is consistent with that predicted by the bipolaron theory for the underdoped and optimal samples. From Table I, it is found that the fitting parameters to the experimental data of the sample annealed at the oxygen pressure of 1 atm seem to be anomalous compared with that for the other samples. The  $Ax^{1/2}$  increases with increasing annealing oxygen pressure. It indicates that the carrier concentration increases with the annealing oxygen pressure, which is expected. The  $\Delta$  increases with decreasing annealing oxygen pressure; that is, the bipolaron binding energy shifts to high temperature with the decrease of carrier concentration. Alexandrov *et al.* expected a strong dependence of the binding energy,  $\Delta \equiv \Delta(T, x)$ , on doping because of the screening.<sup>21</sup> The change of the bipolaron binding energy with doping is consistent with that of

the ‘‘spin gap’’ temperature. In Ref. 21, the  $\Delta$  has been believed to be normal-state gap (or the ‘‘spin gap’’ in the uniform spin susceptibility), but the obtained  $\Delta$  is not consistent with the ‘‘spin gap’’ temperature in Fig. 2. It suggests that the bipolaron binding energy might not be considered as the ‘‘spin gap.’’ But the bipolaron theory successfully accounts for the deviation from linearity in in-plane resistivity due to the opening of the pseudogap.<sup>20</sup> One can see from Table I that the parameter  $c$  increases with doping, different from the case of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  in which both the number of localized bosons and the number of extended bosons increase with doping, but  $b n_L / (n - n_L)$  decreases with doping.<sup>20</sup>

It has been reported that numerical fits were carried out using the functional form  $\rho_c(T) = (a/T) \exp(\Delta/T) + bT + c$ , where  $a$ ,  $b$ ,  $c$ , and  $\Delta$  are fitting constant.<sup>9,10</sup> It is found that the experimental results of the underdoped and optimally doped samples can be well reproduced with the above formula and the parameter  $b$  was set to zero. But the experimental data of the overdoped samples cannot be fitted by setting  $b=0$ . This is consistent with the previous report,<sup>10</sup> suggesting that a  $T$ -linear contribution to  $\rho_c(T)$  has to be added. While its  $\rho_c(T)$  can be well fitted by the bipolaron theory for a whole doping level, the anisotropy data of the overdoped samples cannot be well reproduced by Eq. (1).

In summary, an intensive study of the anisotropy ( $\rho_{ab}$  and  $\rho_c$ ) on the single crystals  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  annealed at the different oxygen pressure was made. The spin gap development with carrier concentration has been observed in in-plane resistivity. The  $\rho_c(T)$  and the anisotropy [ $\rho_c(T)/\rho_{ab}(T)$ ] can be well reproduced by the bipolaron theory proposed by Alexandrov and Mott. The  $\rho_c(T)$  and the anisotropy [ $\rho_c(T)/\rho_{ab}(T)$ ] decrease with increasing carrier concentration. There exists a minimum out-of-plane resistivity for the overdoped samples;  $\rho_c(T)$  has a positive slope at the high temperature. The anisotropy [ $\rho_c(T)/\rho_{ab}(T)$ ] has a weak temperature dependence for the overdoped samples, which is consistent with that predicted by the bipolaron theory.

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