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The systematic study of the normal-state transport properties of Bi-2212 crystals

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Abstract. The temperature dependences of the in-plane resistivity $\rho_{ab}(T)$ and out-of-plane resistivity $\rho_c(T)$ in Bi-2212 crystals covering the region from the underdoped to the overdoped regime have been measured. In the underdoped regime, $\rho_{ab}(T)$ deviates from a linear temperature dependence below a characteristic temperature T^* , well above T_c , whose value decreases with increasing hole concentration. For underdoped crystals, $\rho_{ab}(T)$ shows a typical S-shaped temperature dependence and $\rho_{ab} = \rho_0^* + \beta \exp(-\Delta/T)$ is satisfactorily obeyed over a much wider temperature range from slightly above T_c up to T^* . Near the optimal region, the T-linear dependence of $\rho_{ab}(T)$ is maintained over a wide temperature interval. In contrast, a power law $\rho_{ab} \sim T^n$ (n = 1.5-1.8) is followed in the overdoped regime. As regards the out-ofplane resistivity, on the other hand, $\rho_c(T)$ for the underdoped Bi₂Sr₂CaCu₂O_v crystals shows a semiconductive behaviour, which is well described by the formula $\rho_c = (C_1/T) \exp(C_2/T) +$ $C_3T + C_4$. The difference between the temperature dependences of $\rho_c(T)$ in the overdoped $Bi_2Sr_2CaCu_2O_y$ and $Bi_{1.85}Pb_{0.15}Sr_2CaCu_2O_{8+\delta}$ crystals, with basically the same values of T_c and nearly the same power-law temperature dependences of $\rho_{ab}(T)$ ($\rho_{ab} \sim T^{1.4}$), reveals that the inter-plane disorder in the form of oxygen vacancies and substituted cations acting as an extra blocking layer plays an important role in out-of-plane transport.

1. Introduction

It is well recognized that the understanding of the normal-state transport properties in layered cuprates is important if one is to elucidate the mechanism of high- T_c superconductivity. First, the in-plane resistivity ρ_{ab} evolving as a function of the doped carrier concentration offers an opportunity to characterize the high- T_c electronic state. Previous studies on charge transport have been carried out on $La_{2-x}Sr_xCuO_4$ at different doping levels [1, 2]. In the underdoped regime, the in-plane resistivity ρ_{ab} deviates from a linear T-dependence below a characteristic temperature T^* , well above T_c , whose value decreases with increasing hole concentration. Near the optimal region, the T-linear dependence of the in-plane resistivity is maintained over a wide temperature interval. In contrast, the power law $\rho_{ab} \sim T^n$ shows a gradual change of n from n = 1 towards n = 2 upon increase of the hole concentration in the overdoped regime. Similar studies on underdoped YBa₂Cu₃O_{7-x} [3] and slightly overdoped YBa₂Cu₃O_y [4] have also been reported. Although some transport properties of Bi-2212 crystals [5, 6] have been reported, there have been fewer systematic studies of Bi-2212 crystals reported. On the other hand, the anisotropic resistivity of the layered cuprates, which reveals unusual features for the normal state, has attracted extensive attention. In the normal state, a semiconductive out-ofplane resistivity $(d\rho_c/dT < 0)$ coexists with a metallic in-plane resistivity ρ_{ab} over a wide

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temperature and carrier concentration range [7–9]. As regards the origin of the semiconductive behaviour of the *c*-axis resistivity ρ_c , many ideas have been advanced [10]. However, there is no consensus. Bi₂Sr₂CaCu₂O_y is one of the most anisotropic HTSC and is appropriate for consideration in studies of the 2D nature inherent to HTSC.

In this paper, we have measured the in-plane and out-of-plane resistivities for a series of $Bi_2Sr_2CaCu_2O_y$, $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_8$ and $Bi_xPb_{2-x}Sr_2CaCu_2O_y$ crystals, which cover the range from underdoped to overdoped levels. Our work mainly concentrates on the anomalous transport properties, which arise in connection with the evolution of the pseudogap and $\rho_c(T)$ from showing a semiconductive behaviour to showing a metallic behaviour.

2. Experimental procedures

A series of Bi₂Sr₂CaCuO_y, Bi₂Sr₂Ca_{1-x}Y_xCu₂O₈ and Bi_xPb_{2-x}Sr₂CaCu₂O_y crystals were grown from a Bi_2O_3 -rich melt in Al_2O_3 crucibles. The as-grown $Bi_2Sr_2CaCuO_y$ crystals were annealed at 450 °C and various oxygen partial pressures in the range 10^{-7} - 10^{2} atm so as to obtain crystals covering from the range from underdoped to overdoped levels. The samples are so thin that the oxygen diffusion in the samples is very rapid, and thus all samples have a homogeneous, reproducible oxygen content after undergoing annealing treatment. All crystals for use in the measurements were verified to be single phase by means of x-ray diffraction (XRD). The typical dimensions of these crystals are 1.5 mm \times 0.8 mm \times (6–10) μ m, with the shortest dimension along the c-axis. Thicknesses were measured with a scanning electron microscope. The in-plane resistivities $\rho_{ab}(T)$ of the Bi₂Sr₂Ca_{1-x}Y_xCu₂O₈ crystals were measured by a direct four-probe method, while the anisotropic resistivities of the other crystals were measured using a generalization of the Montgomery method [11, 12]. To ensure low contact resistance, the electrical pads were made by soldering copper leads on evaporated silver electrodes. The degrees of Y and Pb doping, as well as the different oxygen contents, determine the carrier concentrations of Bi-2212 crystals. However, it is not straightforward to compare the doping levels of these samples. The doping levels can be deduced from the following:

- (a) the behaviour of temperature dependences of ρ_{ab} and ρ_c ,
- (b) the superconducting temperature T_c , and
- (c) the magnitudes of ρ_{ab} and ρ_c .

3. Experimental results and analyses

Figure 1 shows the temperature dependences of the in-plane resistivities ρ_{ab} for Bi₂Sr₂CaCuO_y single crystals. Crystals A, B, C, D, E and F were respectively annealed under oxygen pressures of 10⁻⁷, 10⁻⁵, 10⁻³, 1, 10 and 10² atm. T_c and the resistivities ρ_{ab} for the six crystals alter systematically. A typical *T*-linear behaviour of the resistivity and the highest T_c (90 K) are observed for the optimally doped C crystal. Although the oxygen contents of the crystals have not been directly determined, it can certainly be deduced that crystals A and B are both in the underdoped regime, while crystals D, E and F are in the overdoped regime. For the overdoped D, E and F crystals, a slightly upward curve of ρ_{ab} is observed and $\rho_{ab} = \rho_0 + \alpha T^n$ (n = 1.5-1.8) is satisfactorily obeyed for the above crystals. This result is similar to that for overdoped La_{2-x}Sr_xCuO₄ and YBa₂Cu₃O_y. For the underdoped crystals A and B, ρ_{ab} deviates from the high-temperature *T*-linear behaviour at a characteristic temperature T^* (shown by the arrows), far above T_c . This behaviour resembles that observed for underdoped YBa₂Cu₃O_y (y = 6.85, 6.75) and is not an effect of a superconductive fluctuation. However, the curve for



Figure 1. The temperature dependences of the in-plane resistivities ρ_{ab} for Bi₂Sr₂CaCuO_y single crystals. Crystals A, B, C, D, E and F were respectively annealed under oxygen pressures of 10^{-7} , 10^{-5} , 10^{-3} , 1, 10 and 10^2 atm.

 $\rho_{ab}(T)$ is not a typical S-shaped temperature dependence. In fact, crystals A and B are both slightly underdoped, and this is confirmed by their higher values of T_c .

In an attempt to obtain samples that were more underdoped, the as-grown crystals were sealed in the quartz vacuum tube and annealed. Unfortunately, the optically smooth and shiny samples became dull and changed their properties upon annealing, so more heavily underdoped crystals were not successfully obtained by further decreasing the oxygen content. However, more heavily underdoped crystals were obtained by substitution of Y³⁺ for Ca²⁺ in Bi₂Sr₂Ca_{1-x}Y_xCu₂O₈. Bi₂Sr₂Ca_{1-x}Y_xCu₂O₈ crystals (with nominally x = 0.0, 0.03, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.05, 0.0(0.08, 0.11) were grown, but the actual x-value was not directly determined. There has been one investigation [13] focusing on the characteristics of single crystals in this system that were grown, and determining the relationship between the nominal and actual Y concentrations in these crystals. From a comparison with these previously obtained results, we established roughly that the Y content is 0.1 for the compound with nominally x = 0.03, is 0.2 for that with x = 0.05, is 0.3 for that with x = 0.08 and is 0.4 for that with x = 0.11. Figure 2 shows the temperature dependences of the in-plane resistivities ρ_{ab} for Bi₂Sr₂Ca_{1-x}Y_xCu₂O₈ (x = 0.0, 0.03, 0.05, 0.08, 0.11) single crystals. It can be observed that the $\rho_{ab}(T)$ for $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_8$ (x = 0.03, 0.05, 0.08, 0.11) crystals deviate from the high-temperature T-linear behaviour at a characteristic temperature T^* (shown by the arrows), far above T_c , and that the characteristic temperature T^* increases with the increase of the level of Y doping of the crystals, i.e., the reduction of the carrier concentration. $\rho_{ab}(T)$ for the x = 0.11 crystal shows a typical S-shaped temperature dependence, and the upturn near T_c arises from the onset of localization or superconducting fluctuation. In the temperature range from around T_c up to about 150 K, ρ_{ab} can be described as $\rho_{ab} = \rho_0 + \alpha T^n$ with $\rho_0 = 0.76498$ m Ω cm, $\alpha = 0.00004 \text{ m}\Omega \text{ cm K}^{-1}$ and n = 1.98, as shown in figure 2. This is in agreement with a previous report [3]. However, over a much wider temperature range from slightly above T_c up to T^* , ρ_{ab} can be described as $\rho_{ab} = \rho_0^* + \beta \exp(-\Delta/T)$ with a temperature-independent parameter Δ ; here $\rho_0^* = 0.8946 \text{ m}\Omega \text{ cm}, \beta = 5.868 \text{ m}\Omega \text{ cm}, \Delta = 290.27 \text{ K}$. The residual



Figure 2. The temperature dependences of the in-plane resistivities ρ_{ab} for Bi₂Sr₂Ca_{1-x}Y_xCu₂O₈ (x = 0.0, 0.03, 0.05, 0.08, 0.11) single crystals.



Figure 3. The temperature dependences of the out-of-plane resistivities ρ_c for crystals A, B, C, D, E and F.

resistivity ρ_0^* is ~17% larger than the residual resistivity ρ_0 . A similar fit was also found for A15 superconductors even over the extremely wide temperature range [14].

Figure 3 shows the temperature dependences of the out-of-plane resistivities $\rho_c(T)$ for crystals A, B, C, D, E and F. With increasing level of underdoping, the overall magnitude

of ρ_c , as well as its semiconductive temperature dependence, increases. The equation $\rho_c = (C_1/T) \exp(C_2/T) + C_3T + C_4$ was used to fit our resistivity data; this equation was suggested by Ong and co-workers [15]. The parameters C_1 , C_2 , C_3 and C_4 are constants, and the parameter C_2 is related to a pseudogap. The fitted data are shown in figure 3 as a solid curve and the parameters C_1 , C_2 , C_3 and C_4 are listed in table 1. Although the data for the overdoped samples D, E and F can be fitted by the above equation, the parameters C_3 and C_4 give unphysical negative values. This indicates that the $\rho_c(T)$ for overdoped D, E and F do not fit the above equation, and seems to be consistent with pseudogaps only being observed in underdoped superconductors [16, 17]. However, the semiconductive gap C_2 always opens at our measurement temperatures for crystals A, B and C, but the pseudogap is only observed below T^* [16, 17]. Furthermore, the semiconductive behaviour of the slightly overdoped crystal casts doubt on the origin of the semiconductive behaviour being a pseudogap. It is worth pointing out that there is always an activation-type component included in the semiconductor behaviour of $\rho_c(T)$ even for the slightly overdoped Bi-2212 crystal.

Table 1. The parameters C_1 , C_2 , C_3 and C_4 for the underdoped Bi₂Sr₂CaCuO_y crystals.

	$C_1 (\Omega \text{ cm K})$	<i>C</i> ₂ (K)	$C_3 (\mathrm{m}\Omega \mathrm{cm} \mathrm{K})$	$C_4 (\Omega \text{ cm})$
A	504.5 ± 10.1	187.1 ± 1.5	2.71 ± 0.66	5.75 ± 0.22
В	878 ± 17	105.4 ± 1.26	15.1 ± 0.6	0.517 ± 0.216
С	273 ± 20	198.5 ± 5.8	12.0 ± 1.3	1.958 ± 0.432

In order to make the different temperature behaviour of ρ_c for the overdoped Bi-2212 crystals clear, we have measured in detail the anisotropic resistivities of more than twenty crystals of the overdoped Bi₂Sr₂CaCuO_y and Bi_xPb_{2-x}Sr₂CaCu₂O_y systems. Some of the results are shown in figure 4. It shows the temperature dependences of $\rho_{ab}(T)$ and $\rho_c(T)$ for both Bi₂Sr₂CaCuO_y and Bi_{1.85}Pb_{0.15}Sr₂CaCu₂O_y crystals. In figure 4, we see that the temperature dependence of $\rho_{ab}(T)$ shows the basically same T_c and nearly the same power-law temperature dependence ($\rho_{ab} \sim T^{1.4}$) as would be expected for a slightly overdoped form for both crystals. In contrast, the temperature dependences of $\rho_c(T)$ for the two crystals reveal very different behaviours. The facts that T_c is the same and the temperature dependence behaviours of $\rho_{ab}(T)$ are similar seems to indicate that the two crystals have the same carrier concentration. Therefore, the different temperature dependence behaviour of $\rho_c(T)$ indicates that it is not completely ascribed to the carrier concentration.

Transport properties are easily obscured by many factors. There are many more complications which mainly stem from the extreme sensitivity of the properties to the compositions (stoichiometry) which control the carrier concentration in the CuO₂ plane and the inter-plane disorder. However, even the dimension size of the crystals used in the measurements affects the behaviour of the temperature dependence of the resistivity. Forro [18] reported that very thin Bi₂Sr₂CaCuO_y crystals (sample thickness $d < 0.4 \ \mu$ m) usually show a metallic-like temperature dependence of $\rho_c \ (d\rho_c/dT > 0)$. Although we have not prepared such thin samples, all of the crystals on which measurements were made in this paper are verified to be of good quality by means of rocking curves. Recently, Hussey *et al* [19] studied the anisotropic resistivity of YBa₂Cu₄O₈ crystals (among the cleanest high- T_c cuprates, naturally stoichiometric and having the least inter-plane disorder) and suggested that inter-plane disorder plays a critical role in determining the behaviour of $\rho_c(T)$. Bi-2212 crystal is one of the most anisotropic high- T_c cuprates; an extra blocking layer due to (i) inter-plane disorder in the form of substituted cations and (ii) oxygen vacancies reduces the mean free path along the *c*-axis, and possibly strongly affects the out-of-plane transport. From the data of figure 4, the *c*-axis



Figure 4. The temperature dependences of the in-plane resistivities ρ_{ab} and out-of-plane resistivities ρ_c of both Bi₂Sr₂CaCuO_y and Bi_{1.85}Pb_{0.15}Sr₂CaCuO_y crystals. \bullet : Bi₂Sr₂CaCuO_y; \blacktriangle : Bi_{1.85}Pb_{0.15}Sr₂CaCuO_y.

conductivities in both crystals are well into the insulating side of the Mott limit, although a positive slope $(d\rho_c/dT > 0)$ is observed below room temperature for one crystal. According to the Mott theory, the minimum conductivity is $\sigma_{min} = 0.03e^2/(hd_c/2\pi)$, where d_c is the lattice parameter along the *c*-axis. In the case of Bi-2212, d_c is about 15 Å and σ_{min} is about 47 Ω^{-1} cm⁻¹. Therefore, the $\rho_c(T)$ data are far beyond the boundary of the Mott minimum conductivity. Note that, even for basically the same magnitude of $\rho_c(T)$, the temperature dependence is completely different. In fact, the two-dimensional nature of high- T_c cuprates is characterized not by the magnitude of the out-of-plane resistivity, but by the anisotropic scattering times [4]. In the case of the above result, it could be suggested that (i) an extra blocking layer due to Pb substituting for Bi in a Bi₂O₂ layer and (ii) oxygen vacancies have different effects on the *c*-axis transport.

4. Conclusions

The temperature dependences of the in-plane resistivities $\rho_{ab}(T)$ and out-of-plane resistivities $\rho_c(T)$ have been measured for Bi-2212 crystals covering the range from the underdoped to the overdoped regime. Near the optimal region, the T-linear dependence of $\rho_{ab}(T)$ is maintained over a wide temperature interval. In the overdoped regime, a power law, $\rho_{ab} \sim T^n$ (n = 1.5–1.8), is obeyed. In the underdoped regime, $\rho_{ab}(T)$ deviates from a linear temperature dependence below a characteristic temperature T^* , well above T_c , whose value decreases with increasing hole concentration. These results are similar to those obtained for $La_{2-x}Sr_xCuO_4$ [1, 2] and YBa₂Cu₃O_{7-x} [3, 4]. However, the temperature dependence of $\rho_{ab}(T)$, which shows a typical S-shaped curve, seems to be more satisfactorily fitted by the formula $\rho_{ab} \sim \beta \exp(-\Delta/T)$ than by the reported formula $\rho_{ab} \sim \alpha T^n$ [3] over a much wider temperature range, from slightly above T_c up to T^* . Recently, ARPES experiments [20] have revealed that the Fermi surface in underdoped Bi-2212 shows an unusual behaviour developing on cooling the sample, when a pseudogap opens up. We deduce that the temperature dependence of $\rho_{ab} \sim \beta \exp(-\Delta/T)$ could be related to the above feature. In order to make this clearer, more careful studies of transport properties, including magnetotransport, must be made over a certain underdoped composition region (the localization can obscure $\rho_{ab}(T)$ below T^* in more heavily underdoped samples and the superconducting fluctuation can obscure $\rho_{ab}(T)$ below T^* in slightly underdoped samples). The out-of-plane resistivities $\rho_c(T)$ for underdoped Bi₂Sr₂CaCu₂O_y crystals are well described by the formula $\rho_c = (C_1/T) \exp(C_2/T) + C_3T + C_4$ as reported in [15]. However, the completely different temperature dependences of $\rho_c(T)$ for the overdoped $Bi_2Sr_2CaCu_2O_y$ and $Bi_xPb_{2-x}Sr_2CaCu_2O_y$ crystals, for basically the same T_c and nearly the same power-law temperature dependence of $\rho_{ab}(T)$ ($\rho_{ab} \sim T^{1.4}$), reveal that the inter-plane disorder, in the form of oxygen vacancies and substituted cations acting as an extra blocking layer, plays an important role in out-of-plane transport. Therefore, the model for the *c*-axis resistivity should pay considerable attention to inter-plane disorder at least in the Bi-2212 system.

Acknowledgments

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