Normal-State Resistivity Anisotropy in Underdoped $R\mathrm{Ba_2Cu_3O_{6+x}}$ Crystals

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We have revealed new features in the out-of-plane resistivity ρ_c of heavily underdoped $R\mathrm{Ba_2Cu_3O_{6+x}}$ (R = Tm, Lu) single crystals, which give evidence for two distinct mechanisms contributing to the c-axis transport. We have observed a crossover towards "metal-like" ($\partial\rho_c/\partial T > 0$) behavior at the temperature T_m which quickly *increases* with decreasing doping. The metal-like conductivity contribution dominates at $T < T_m$ and provides a saturation of the resistivity anisotropy, ρ_c/ρ_{ab}. The antiferromagnetic ordering is found to block this metal-like part of the c-axis conductivity and complete decoupling of $\mathrm{CuO_2}$ planes, which may be the reason of superconductivity disappearance. [S0031-9007(98)07984-8]

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The primary indication of an unusual normal state in high-T_c cuprates is the contrasting behavior of the in-plane (ρ_{ab}) and out-of-plane (ρ_c) resistivity. In most cuprates, for instance, $\mathrm{La_{2-x}Sr_xCuO_4}$, underdoped $\mathrm{YBa_2Cu_3O_{6+x}}$, $\mathrm{Bi_2Sr_2CuO_y}$, the "metal-like" electron transport along $\mathrm{CuO_2}$ planes coexists with a nonmetallic conductivity between planes, and the resistivity anisotropy, ρ_c/ρ_{ab}, diverges with decreasing temperature till the superconducting transition interrupts this tendency [1–3]. This behavior violating the conventional concept of band electron transport has brought into being many theories which imply blocking the c-axis coherent transport and charge confinement within the $\mathrm{CuO_2}$ planes [4–6]. The salient consequence of charge confinement is a possibility of superconductivity owing to interlayer pair tunneling [4,5]. The two-dimensional behavior is considered thus as a key quality of that unusual normal state giving rise to high-T_c superconductivity.

Important exceptions from this straightforward picture were however found, and the best known one is $\mathrm{YBa_2Cu_3O_7}$ (Y-123) which is a 90-K superconductor, but possesses a metallic out-of-plane conductivity [7]. A crossover towards the coherent c-axis electron transport with decreasing temperature was recently found in $\mathrm{YBa_2Cu_4O_8}$ (Y-124) [8,9]. This peculiar behavior of Y-123 and Y-124 systems was attributed to the metallic conductivity of their Cu-O chains. In contrast to expectations, a temperature crossover in $\rho_c(T)$ resembling that in Y-124 was observed also in heavily underdoped $R\mathrm{Ba_2Cu_3O_{6+x}}$ (R = Y, rare earth) crystals in which, obviously, the Cu-O chains were destroyed [10].

Analyzing experimental data for highly anisotropic high-T_c cuprates, one should take into account crystal perfection problems. Stacking faults can well block the c-axis conductivity and give rise to insulating $\rho_c(T)$, while an apparent metallic behavior and crossovers can originate from numerous screw dislocations [11] short-circuiting the whole set of $\mathrm{CuO_2}$ planes. Recently [12] we have found that R-123 crystals can grow not only as conventional thin or thick plates, but also like whiskers along the b axis. We succeeded in growing whiskerlike Tm-123 crystals which had a shape of thin, wide bars with the shiny bc faces being the *largest* ones. These unique crystals are very attractive for studying the resistivity anisotropy $\rho_c(T)/\rho_{ab}(T)$. While their shape is suitable for measuring *both* resistivity components, the growth mechanism being distinct from that of platelets implies the absence of screw dislocations along the c axis, i.e., in the direction transverse to the crystal growth one.

In the present work, using mainly these whiskerlike crystals, we demonstrate that the out-of-plane conductivity in $R\mathrm{Ba_2Cu_3O_{6+x}}$ *inherently* contains two distinct contributions associated presumably with two types of charge carriers. The first contribution is temperature activated and provides the familiar contrast between $\rho_c(T)$ and $\rho_{ab}(T)$. The second one roughly follows the in-plane conductivity σ_{ab} though reduced by 4 orders of magnitude. This contribution dominates the low-temperature c-axis transport, induces a crossover in $\rho_c(T)$, and prevents the resistivity anisotropy from diverging at low T. In contrast to the Y-124 system [9] this metal-like conduction cannot be associated with Cu-O chains which are destroyed in our underdoped crystals.

Both the plate- and whiskerlike $\mathrm{(Tm,Lu)Ba_2Cu_3O_{6+x}}$ crystals were grown by the flux method [12] and their oxygen stoichiometry was varied by subsequent high-temperature annealing [10]. Measurements of $\rho_c(T)$ and $\rho_{ab}(T)$ were performed by the four-probe method on two samples cut always from the same single crystal. Further description will concentrate mainly on whiskerlike Tm-123 crystals, since their shape and absence of screw c-axis dislocations allow a straightforward analysis of ρ_c data. For example, the sample in Fig. 1 was $45 \times 400 \times 550\ \mu\mathrm{m}^3$ with the largest dimension along the c axis. Mapping this crystal on an isotropic model, one obtains a thin, long wire ($\approx 0.01 \times 0.1 \times 10\ \mathrm{mm}^3$), for which evaluating ρ_c from raw data holds, obviously, no problems. For ρ_{ab} measurements, in their turn, a narrow bar of $45 \times 800 \times 140\ \mu\mathrm{m}^3$ was cut from the same whiskerlike crystal.

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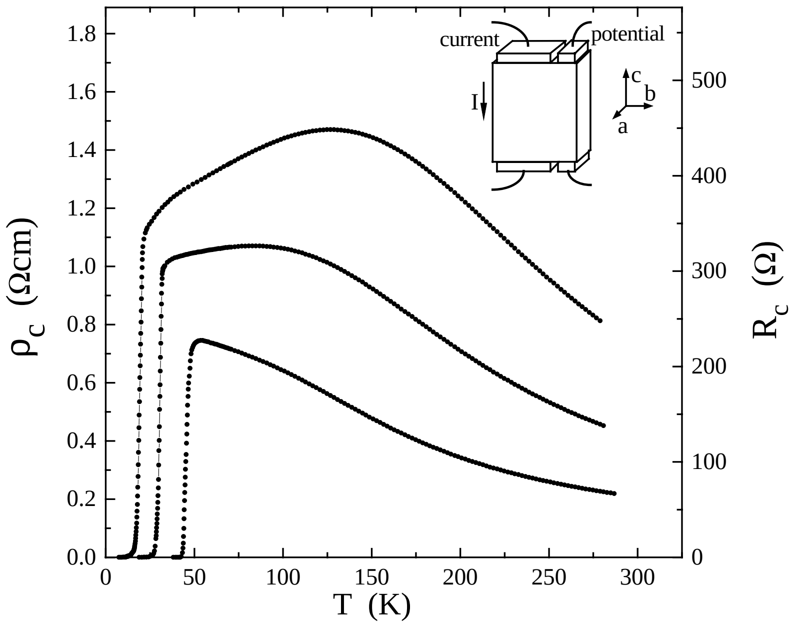

FIG. 1. The out-of-plane resistivity, $\rho_c(T)$, of the $TmBa_2Cu_3O_{6+x}$ "whiskerlike" single crystal at various oxygen contents, $x \approx 0.41$, 0.44, and 0.49 (top to bottom). Inset: The contact configuration.

In the $RBa_2Cu_3O_{6+x}$ system, superconductivity mostly hides the low-T region and the normal-state resistivity can be measured down to fairly low temperatures only in a narrow doping range in the vicinity of the antiferromagnetic-superconducting (AFM-SC) phase boundary. A selection of $\rho_c(T)$ curves obtained within this heavily underdoped region is presented in Fig. 1. In contrast to what one could expect the c-axis resistivity does not grow sharply with decreasing temperature but passes through a maximum at T_m and begins to drop. For the sample with $x \approx 0.41$, ρ_c monotonically decreases within a wide temperature range from 127 K down to $T_c \approx 19$ K. A tendency of ρ_c to saturate is apparent for $x \approx 0.49$ as well, but for such and higher doping levels the ρ_c crossover is masked, because T_m quickly decreases with doping, while superconductivity in its turn hides the larger temperature range. This is why other studies of $YBa_2Cu_3O_{6+x}$ dealing mainly with $x \geq 0.6$ [2] could not reveal this ρ_c peculiarity. The crossover observed gives evidence for the change of the dominating conductivity mechanism and implies most likely that the out-of-plane conductivity contains two contributions, the balance of which determines the shape of $\rho_c(T)$ curves.

The crossover temperatures T_m determined from 34 resistivity curves measured on nine plate- and whiskerlike Tm-123 and Lu-123 crystals with different oxygen contents are collected in Fig. 2. The data are plotted on the phase diagram in which both the Néel temperature T_N and the superconducting transition temperature T_c are determined from resistivity measurements [10] and presented as a function of the in-plane conductivity σ_{ab}, which is roughly proportional to the hole density in the CuO_2 planes. This diagram presentation looks similar to the usual T-x one but qualitatively accounts for the influence of oxygen ordering on the hole density [10]. As can be seen T_m quickly decreases with increasing doping (increasing x) and somewhere at the 60-K plateau the crossover line gets under the SC region. The Cu-O chains, when perfectly ordered, possess the metallic conductivity [7,9], and one could expect the range of metallic behavior to extend with increasing oxygen content. Obviously the phenomenon we are dealing with is of a different nature. The metal-like conductivity component dominates just in the region where the Cu-O chains are destroyed. The heavily underdoped crystals have no

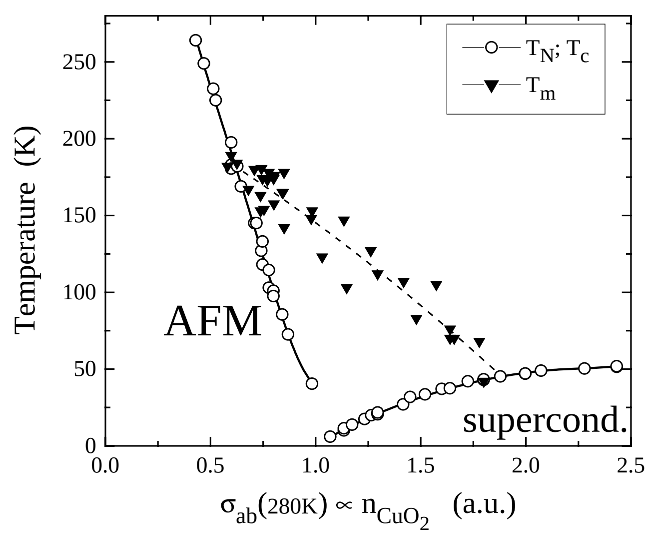

FIG. 2. A cumulative diagram for $TmBa_2Cu_3O_{6+x}$ and $LuBa_2Cu_3O_{6+x}$ crystals. The crossover temperature T_m together with the AFM and SC transition temperatures, T_N and T_c, are presented as a function of the in-plane conductivity σ_{ab}.

other conducting subsystem besides CuO_2 planes, and one hence has no choice but to attribute the metal-like conduction to the direct interplane charge transport.

Each conductivity contribution can be analyzed separately at a distance from the crossover line, i.e., in the range where it dominates. The metal-like contribution roughly tracks the in-plane conductivity behavior, while the activated one can be fitted by exponential expressions, the simplest of which is that of variable range hopping. The crossover behavior can be thus described as $\sigma_c(T) = K\sigma_{ab}(T) + C\exp(-B/T^{1/4})$, with the conductivity anisotropy approaching a constant value at low T.

To obtain additional information on the conductivity contributions we had available two simple approaches. First, we could move the crystal from the SC to AFM doping region, see Fig. 2, and analyze how the long-range magnetic order influences ρ_c. To test the nature of the metal-like conductivity one has to place the Néel temperature in that temperature region where this contribution dominates; i.e., T_N should be <100 K. The second possibility is to use the well studied phenomenon of chain-layer oxygen ordering, see Ref. [10], and references therein, as a convenient way of tuning the hole density in CuO_2 planes. The hole-doping level can be reduced by $\approx 20\%$ by heating the crystal to $\approx 120\,^{\circ}$C with subsequent quenching, and it can be gradually restored simply by room-temperature aging. The advantage of this procedure is that both the stoichiometry and the contact configuration remain exactly unchanged.

Figure 3 combines both approaches and presents the $\rho_c(T)$ and $\rho_{ab}(T)$ data obtained for nonsuperconducting crystals. One can see that at $T > 20$ K [13] ρ_{ab} has almost parabolic temperature dependence and roughly scales with hole doping. The interplane resistivity retains the pronounced crossover, but an anomaly associated with the AFM ordering [10] has appeared instead of SC transition; compare with Fig. 1. A steplike increase of ρ_c occurs upon cooling below the Néel temperature. We notice that T_N is essentially below the crossover point, and the ρ_c anomaly is located in the region where the metal-like conductivity component undoubtedly dominates, but at the same time no peculiarity is observed on $\rho_{ab}(T)$ curves. This is probably the most visual evidence that the ρ_c crossover cannot be associated with some admixture of the in-plane conductivity.

Figure 4 presents the $(\rho_c/\rho_{ab})(T)$ curve obtained from the data shown by solid circles in Fig. 3. Above T_N the resistivity anisotropy can be well fitted by the empirical expression implying that σ_c contains two contributions. One can see that in the absence of antiferromagnetic ordering, the anisotropy would saturate at a value of several thousand. The long-range AFM ordering obviously blocks the metal-like conductivity contribution and completes decoupling of CuO_2 planes. For comparison, in the right inset we show the anisotropy data obtained for a highly homogeneous platelike Lu-123 crystal. The only apparent difference is a sharper AFM transition, which makes the regular behavior of the resistivity anisotropy and, particularly, its tendency to saturation more spectacular. The same anisotropy saturation is obviously characteristic of SC samples as well, since $\rho_c(T)$ curves for crystals with $T_c \approx 19$ K and $T_N \approx 65$ K, Figs. 1 and 3, differ not much above $T_c(T_N)$.

The left inset in Fig. 4 compares fits of regular ($T > T_N$) $(\rho_c/\rho_{ab})(T)$ dependences for two hole-doping levels of the same crystal and illustrates one more surprising result. The resistivity anisotropy at high T considerably decreases with increasing density of carriers, but its extrapolated low-temperature value, in contrast to what

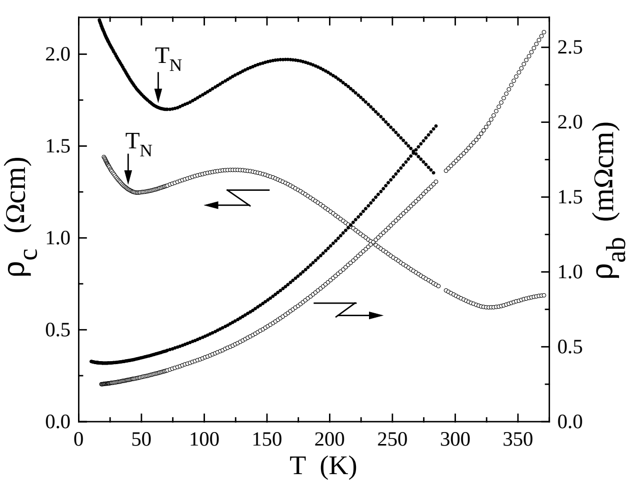

FIG. 3. In-plane, $\rho_{ab}(T)$, and out-of-plane, $\rho_c(T)$, resistivity of $TmBa_2Cu_3O_{6+x}$ ($x \approx 0.37$) crystals. Measurements were performed immediately after quenching (solid circles) and after five days aging at room temperature (open circles).

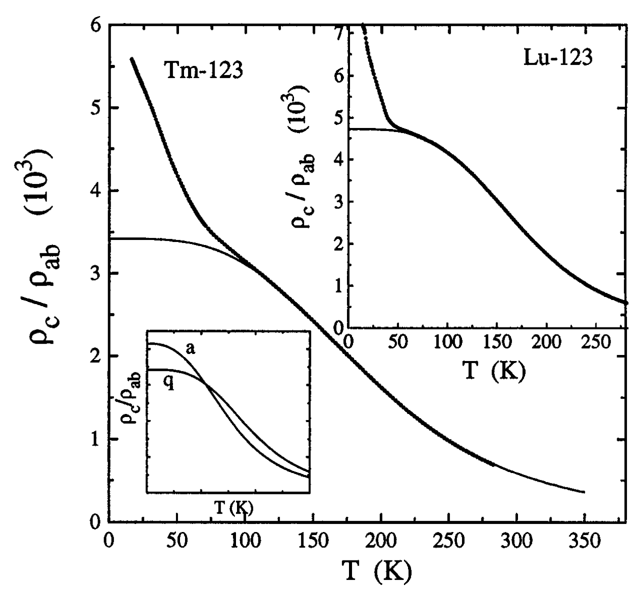

FIG. 4. Main graph: Resistivity anisotropy ρ_c/ρ_{ab} for whiskerlike $TmBa_2Cu_3O_{6.37}$; solid line: a fit of the regular temperature dependence ($T > T_N$). Left inset: Fits of the resistivity anisotropy for quenched and aged states of the same crystal. Right inset: Resistivity anisotropy for a "platelike" $LuBa_2Cu_3O_{6.34}$ crystal.

one could expect, *increases* with increasing doping. The metal-like conductivity contribution under discussion has therefore nothing to do with the metallic conductivity in fully oxygenated $YBa_2Cu_3O_7$. Instead, it is the temperature-activated contribution which should gradually acquire metallic features with $x \to 1$ and begin to dominate over the whole temperature range.

Few theories consider a possibility of crossover towards metal-like c-axis conductivity at low temperatures in underdoped cuprates [6,14]. The general problem which emerges upon constructing a picture based on multiband conduction, Kondo scattering, etc., is that ρ_c at any temperature appears to be too large to fit the concept of coherent conduction. To ascribe the metal-like conductivity observed to the coherent transport one has either to suppose that only a small fraction of carriers participates in the c transport or to introduce new heavy quasiparticles [14]. On the other hand, the incoherent c-axis conductivity can also track the behavior of σ_{ab}, if just the strong in-plane scattering blocks the interplane transitions [5].

The most attractive approach for the temperature-activated conductivity contribution is to attribute it to Cu-O chains. Really, if perfect Cu-O chains in Y-123 and Y-124 systems possess the metallic conductivity [7,9], fragmented ones in underdoped crystals should naturally provide the hopping electron transport. This assumption would explain the observed strong dependence of $\rho_c(T)$ on both the oxygen content and oxygen ordering.

However, we probably should search for a more general explanation for the puzzling c transport rather than that based on structural peculiarities of R-123. Actually, apart from the resistivity scale, similar behavior ($\rho_{ab} \propto T^2$ and a crossover in ρ_c at $\approx$120 K) was observed in the noncuprate layered system Sr_2RuO_4 [15], and the anisotropy saturation at low T was reported for $La_{2-x}Sr_xCuO_4$ [3]. Of course, approaches not assuming any role of Cu-O chains could be suggested. The concept of charge confinement, for instance, implies blocking of the interplane single-particle tunneling but retains a possibility of coherent transport for pairs [4,5]. The two types of carriers, namely, bosons and thermally excited fermions, could thus be responsible for distinct conductivity contributions observed. The pair formation not necessarily results in superconductivity and these are preformed pairs which are often considered as a cause of pseudogap effects in cuprates [14,16]. Just a glance is enough to find a similarity between the crossover temperature T_m in Fig. 2 and the pseudogap crossover temperature T^* in popular phase diagrams suggested for cuprates [17]. Some difference existing in the scales, but not in the doping dependences, is not valuable, since both T_m and T^* correspond to arbitrary determined crossover points.

The blocking phenomenon due to AFM ordering holds a problem for all cited above models and remains yet to be explained. If ρ_c was controlled by interplane scattering [5], it would not change at T_N, since the in-plane scattering obviously does not undergo considerable variation. On the other hand, if the metal-like conductivity component originates from preformed pairs [4,5,14], one necessarily faces the question how spinless carriers interact with the magnetic order. An interesting consequence within the preformed-pair model is that blocking of interplane pair transitions could explain why antiferromagnetism and superconductivity hardly coexist in cuprates.

In summary, we have found new features in the out-of-plane electron transport of heavily underdoped $RBa_2Cu_3O_{6+x}$ single crystals. We have shown that the c-axis conductivity intrinsically contains two contributions. The first one is the familiar semiconductorlike conductivity usually observed in moderately underdoped samples. The other looks metal-like and dominates the interplane transport at low temperatures and low doping, where the Cu-O chains are destroyed. Because of this contribution the resistivity anisotropy saturates at low T instead of diverging. The finding possibly having implication for the nature or high-T_c superconductivity is that in nonsuperconducting samples the metal-like part of σ_c is blocked by antiferromagnetic ordering.

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