Normal-state resistivity anisotropy in underdoped RBa$_2$Cu$_3$O$_{6+x}$ crystals

A. N. Lavrov$^{1}$, M. Yu. Kameneva, L. P. Kozeeva

Institute of Inorganic Chemistry, Siberian Branch of RAS, Lavrentyeva-3, 630090 Novosibirsk, Russia

(October 8, 2018)

We have revealed new features in the out-of-plane resistivity $\rho_c$ of heavily underdoped RBa$_2$Cu$_3$O$_{6+x}$ (R=Tm,Lu) single crystals, which give evidence for two distinct mechanisms contributing the c-axis transport. We have observed a crossover towards “metal-like” ($\partial \rho_c/\partial T > 0$) behavior at the temperature $T_n$ which quickly increases with decreasing doping. The “metal-like” conductivity contribution dominates at $T < T_n$ and provides a saturation of the resistivity anisotropy, $\rho_c/\rho_{ab}$. The antiferromagnetic ordering is found to block this “metal-like” part of the c-axis conductivity and complete decoupling of CuO$_2$ planes, which may be the reason of superconductivity disappearance.

74.25.Fy, 74.62.Dh, 74.72.Bk

The primary indication of an unusual normal state in high-$T_c$ cuprates is the contrasting behavior of the in-plane ($\rho_{ab}$) and out-of-plane ($\rho_c$) resistivity. In most cuprates, for instance La$_{2-x}$Sr$_x$CuO$_4$, underdoped YBa$_2$Cu$_3$O$_{6+\delta}$, Bi$_2$Sr$_2$CuO$_y$, the metal-like electron transport along CuO$_2$ planes coexists with a non-metallic conductivity between planes, and the resistivity anisotropy, $\rho_c/\rho_{ab}$, diverges with decreasing temperature till the superconducting transition interrupts this tendency. 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 CuO$_2$ planes. The salient consequence of charge confinement is a possibility of superconductivity owing to interlayer pair tunneling. 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 YBa$_2$Cu$_3$O$_7$ (Y-123) which is a 90-K superconductor, but possesses a metallic out-of-plane conductivity. A crossover towards the coherent c-axis electron transport with decreasing temperature was recently found in YBa$_2$Cu$_4$O$_8$ (Y-124). 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 RBa$_2$Cu$_3$O$_{6+x}$ (R=Y, rare earth) crystals in which, obviously, the Cu-O chains were destroyed.

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 short-circuiting the whole set of CuO$_2$ planes. Recently 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 whisker-like 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 whisker-like crystals, we demonstrate that the out-of-plane conductivity in RBa$_2$Cu$_3$O$_{6+x}$ 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 $\sigma_{ab}$ 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, this metal-like conduction cannot be associated with Cu-O chains which are destroyed in our underdoped crystals.

Both the plate- and whisker-like (Tm,Lu)Ba$_2$Cu$_3$O$_{6+x}$ crystals were grown by the flux method and their oxygen stoichiometry was varied by subsequent high-temperature annealing. 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 whisker-like Tm-123 crystals, since their shape and absence of screw c-axis dislocations allow a straightforward analysis of $\rho_c$ data. For example, the sample in Fig. 1 was $45 \times 400 \times 550 \mu m^3$ with the largest dimension along the c-axis. Mapping this crystal on an isotropic model, one obtains a thin, long wire ($\approx 0.01 \times 0.1 \times 10 mm^3$), for which evaluating $\rho_c$ from raw data holds, obviously, no problems. For $\rho_{ab}$ measurements, in their turn, a narrow bar of $45 \times 800 \times 140 \mu m^3$
In the RBa$_2$Cu$_3$O$_{6+x}$ 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 AF-SC phase boundary. A selection of $\rho_c(T)$ curves obtained within this heavily underdoped region is presented in Fig. 2. In contrast to what one could expect the $T$-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 $x \approx 0.41$, $\rho_c$ monotonically decreases within a wide temperature range from 127 K down to $T_c \approx 19$ K. A tendency of $\rho_c$ to saturate is apparent for $x \approx 0.49$ as well, but for such and higher doping levels the $\rho_c$ 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$_2$Cu$_3$O$_{6+x}$ dealing mainly with $x \geq 0.6$ could not reveal this $\rho_c$ 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 9 plate- and whisker-like 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 Neel 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 $\sigma_{ab}$, 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 [8], 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 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. Firstly, we could move the crystal from the SC to AF doping region, see Fig. 2 and analyze how the long-range magnetic order influences $\rho_c$. To test the nature of the...
metal-like conductivity one has to place the Neel 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 $\approx 120$°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.

Fig. 3 combines both approaches and presents the $\rho_c(T)$ and $\rho_{ab}(T)$ data obtained for non-superconducting crystals. One can see that at $T > 20$ K $\rho_{ab}$ has almost parabolic temperature dependence and roughly scales with hole doping. The interplane resistivity retains the pronounced crossover, but an anomaly associated with the AF ordering has appeared instead of SC transition, compare with Fig. 1. A step-like increase of $\rho_c$ occurs upon cooling below the Neel temperature. We notice that $T_N$ is essentially below the crossover point, and the $\rho_c$ 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 $\rho_c$ crossover cannot be associated with some admixture of the in-plane conductivity.

Fig. 3 presents the $(\rho_c/\rho_{ab})(T)$ curve obtained from the data shown by solid circles in Fig. 2. Above $T_N$ the resistivity anisotropy can be well fitted by the empirical expression implying that $\sigma_c$ 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 AF 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 plate-like LuBa$_2$Cu$_3$O$_{6.34}$ crystal. The only apparent difference is a sharper AF 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. [10], not much differ above $T_c(T_N)$.

The left inset in Fig. 3 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 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$_2$Cu$_3$O$_7$. 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 [1,4]. The general problem which emerges upon constructing a picture based on multi-band conduction, Kondo-scattering, etc., is that $\rho_c$ 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 $\sigma_{ab}$, if just the strong in-plane scattering blocks the interplane transitions [1].

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 [6,14], 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 $\rho_c$ at $\approx 120$ K) was observed in non-cuprate layered system Sr$_2$RuO$_4$ [3], and the anisotropy saturation at low $T$ was reported for La$_{2-\delta}$Sr$_\delta$CuO$_4$ [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 [3,4]. 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 [4,15]. Just a glance is enough to find 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 AF ordering holds a problem for all cited above models and remains yet to be explained. If $\rho_c$ was controlled by interplane scattering [4], 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 [4,14,15], 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$_2$Cu$_3$O$_{6+x}$ single crystals. We have shown that the $c$-axis conductivity intrinsically contains two contributions. The first one is the familiar semiconductor-like 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 non-superconducting samples the metal-like part of $\sigma_c$ is blocked by antiferromagnetic ordering.

ACKNOWLEDGMENTS

ANL is grateful to Prof. V. F. Gantmakher for many fruitful discussions.

[1] Y. Nakamura and S. Uchida, Phys. Rev. B 47, 8369 (1993).
[2] K. Takenaka, K. Mizuhashi, H. Takagi, and S. Uchida, Phys. Rev. B 50, 6534 (1994).
[3] Y. Ando et al., Phys. Rev. Lett. 75, 4662 (1995); 77, 2065 (1996).
[4] S. Chakravarty, A. Sudbo, P. W. Anderson, and S. Strong, Science 261, 337 (1993); P. W. Anderson, cond-mat/9801267.
[5] N. Kumar, T. P. Pareek and A. M. Jayannavar, Phys. Rev. B 57, 13399 (1998).
[6] H. C. Lee and P. B. Wiegmann, Phys. Rev. B 53, 11817 (1996).
[7] T. Ito et al., Nature 350, 596 (1991).
[8] J.-S. Zhou, J. B. Goodenough, B. Dabrowski, and K. Rogacki, Phys. Rev. Lett. 77, 4253 (1996).
[9] N. E. Hussey et al., Phys. Rev. Lett. 80, 2909 (1998).
[10] A. N. Lavrov and L. P. Kozeeva, Physica C 248, 365 (1995); 253, 313 (1995); JETP Lett. 62, 580 (1995).
[11] C. T. Lin, J. Crystal Growth 143, 110 (1994).
[12] L. P. Kozeeva et al., J. Crystal Growth, submitted.
[13] for discussion of localization effects at low temperatures see V. F. Gantmakher et al., JETP Lett. 65, 870 (1997).
[14] A. S. Alexandrov, V. V. Kabanov and N. F. Mott, Phys. Rev. Lett. 77, 4796 (1996).
[15] F. Lichtenberg, A. Catana, J. Mannhart, and D. G. Schlom, Appl. Phys. Lett. 60, 1138 (1992).
[16] V. B. Geshkenbein, L. B. Ioffe, and A. I. Larkin, Phys. Rev. B 55, 3173 (1997).
[17] B. Batlogg, V. J. Emery, Nature 382, 20 (1996).