Anomalies of conductivity behavior near the paramagnetic-antiferromagnetic transition in single-crystals La$_2$CuO$_{4+\delta}$

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Abstract

The temperature dependences of resistance, $R(T)$, of two single-crystals La$_2$CuO$_{4+\delta}$ samples have been studied with the aim to detect a possible change in the $R(T)$ behavior induced by paramagnetic-antiferromagnetic (PM-AFM) transition. One of the samples with $\delta \lesssim 0.01$, was fairly homogeneous in oxygen distribution (not phase-separated) with Néel temperature $T_N \approx 266$ K. Conductivity of this sample has been determined by Mott’s variable-range hopping below $T_N$. The other, far less resistive, sample with $\delta \approx 0.05$, was inhomogeneous (phase-separated) showing both PM-AFM ($T_N \approx 205$ K) and superconducting ($T_c \approx 25$ K) transitions. It is found that for the homogeneous sample the resistivity decreases above $T_N$ far faster with temperature than below it (for both directions of measuring current, parallel and perpendicular to basal CuO$_2$ planes). A similar behavior of conductivity near PM-AFM transition is also found for the phase-separated and less resistive sample. In this case a clear kink in $R(T)$ curve near $T_N \approx 205$ K can be seen. Furthermore, a transition to metallic ($dR/dT > 0$) behavior occurs far enough above $T_N$. The observed behavior of the samples studied is related to increased delocalization of charge carriers above $T_N$. This is in accordance with decrease in the AFM correlation length and corresponding enhancement of the hole mobility above $T_N$ known for low-doped lanthanum cuprates.

Key words: cuprates, paramagnetic-antiferromagnetic transition, phase separation, superconductivity
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1 Introduction

Research on cuprate high-$T_c$ superconductors is one of the topical problems of physics of solids. The nature of superconductivity in these compounds (discovered more than 20 years ago) is still not entirely clear. It is however evident that their magnetic and superconducting properties are closely related. In the normal state, the conducting properties can also be considerably dependent on the magnetic state of the system. For example, in single crystal La$_2$CuO$_4$ the metamagnetic antiferromagnetic (AFM) - weak ferromagnetic transition causes a sharp increase in the conductivity [12]. Investigations of the interaction between the charge carriers and the magnetic subsystem are important not only for high-$T_c$ superconductors but generally for a wide range of other magnetic conductors and semiconductors as well.

The above consideration has stimulated this study of transport and magnetic properties of single-crystal La$_2$CuO$_{4+\delta}$ cuprates near the Néel temperature $T_N$. The objective was to detect possible correlations of these properties. Non-doped La$_2$CuO$_4$ is an AFM insulator ($T_N \approx 320$ K) [3]. Its doping with excess oxygen ($\delta \neq 0$) gives rise to charge carriers (holes) and suppresses the AFM order (decreases $T_N$). The properties of a single crystal La$_2$CuO$_4$ are to a great extent determined by its crystal and magnetic structures [3,4]. In the temperature region of our interest (below 430 K), La$_2$CuO$_4$ has an orthorhombic perovskite-type lattice consisting of alternating CuO$_2$ and La$_2$O$_2$ layers. In the $Bmab$ space group the CuO$_2$ layers are perpendicular to the $c$-axis and parallel to the $ab$-plane [3]. The charge carriers (holes) are believed [5] to be mainly of oxygen character. The excess oxygen (when $\delta \neq 0$) is located in the La$_2$O$_{2+\delta}$ layers between the adjacent CuO$_2$ planes [5]. This provides hole delocalization from the CuO$_2$ planes and thus leads to the three-dimensional (3D) character of conductivity. The magnetic structure is formed by the copper ions $d^9$Cu$^{2+}$ with spin $S = 0.5$. The spins at the neighboring Cu$^{2+}$ sites are oppositely directed and aligned along the orthorhombic $b$-axis (Cu-Cu direction) in the $bc$-plane. The spins are canted ($\approx 0.17^\circ$) with respect to the $b$-axis. Each CuO$_2$ layer has a weak ferromagnetic moment perpendicular to the layer. Below $T_N$, the directions of the moments in neighboring CuO$_2$ planes are opposite, so that the system as a whole is a 3D AFM [1].

It is well known that in metallic conductors the transition from paramagnetic (PM) into AFM state usually implies a decrease in magnetic part of electric resistance that causes a kink in temperature dependence of resistance $R(T)$ [6] or even an appreciable drop of the resistance on transition to the AFM state [7]. In magnetic semiconductors, the influence of spin ordering at $T < T_N$ on conductivity is still not understood sufficiently well. It is, however, known [8,9] that conductivity of these compounds demonstrates often a weak kink in the $R(T)$ at $T \approx T_N$. It can be expected that similar effect takes place in
conductivity of La$_2$CuO$_{4+\delta}$ as well.

The effect of PM-AFM transition on temperature dependence of resistance has been noted in studies of some underdoped high-$T_c$ cuprates. For example, for single-crystal YBa$_2$Cu$_3$O$_{6+x}$ a clear feature near $T_N$ has been found in the out-of-plane resistivity $\rho_c(T)$ (that is when current is parallel to the c-axis), whereas, the in-plane $\rho_{ab}(T)$ (measured with current parallel to CuO$_2$ layers) shows no feature at $T = T_N$ [10]. The same behavior was found for lightly doped La$_{2-x}$Sr$_x$CuO$_4$ [11]. In both cases this reflects strong anisotropy of quasi-two-dimensional (2D) conductivity of these layered compounds.

The behavior of conductivity in non- or low-doped La$_2$CuO$_{4+\delta}$ near $T_N$ was approached previously [12,13]. In Ref. [12] the temperature dependence of electric resistance $R(T)$ shows an anomaly near or not very far from $T_N$ in the samples with $T_N$ in the range 250–310 K, but these anomalies appeared to be determined by the phase separation [14,15,16,17,18,19] upon cooling into oxygen-poor ($\delta \approx 0$) and oxygen-rich ($\delta > 0$) phases. In study of Ref. [13] anomalous behavior of resistance was searched for at $T \approx T_N$ by examining the temperature behavior of the activation energy of hopping conduction. It was found clearly in a sample with $T_N \approx 300$ K, but for other samples studied (with $T_N$ equal to 318 K and 275 K) it is hardly can be spoken about some definite resistance anomaly near $T_N$. The previous investigations thus, on the one hand, suggest a possibility of anomalous $R(T)$ behavior near $T_N$ in La$_2$CuO$_{4+\delta}$ crystals low-doped with excess oxygen, but, on the other hand, some crucial questions concerning this effect remained unanswered. For example, it is important to know how universal the anomaly is and what factors are responsible for it. We believe that the results of this study can be helpful in answering these questions.

2 Samples and experimental details

Two samples of single crystals La$_2$CuO$_{4+\delta}$ (both grown at the Institute of Solid State and Semiconductor Physics, Minsk, Belorussia by S. N. Barilo group) were used in this study. First of them (No. 1) was with dimensions about $4 \times 4 \times 3.3$ mm$^3$. The temperature of AFM transition ($T_N = 266$ K) was determined from the position of the peak in the temperature dependence of dc magnetic susceptibility $\chi(T)$ (Fig. 1) in the magnetic field directed along the c-axis. The measurements were made in a Faraday-type magnetometer. According to the known phase diagram for La$_2$CuO$_{4+\delta}$ [15,17,18,19] this value of $T_N$ corresponds to $\delta \lesssim 0.01$. At this low content of excess oxygen the La$_2$CuO$_{4+\delta}$ sample is expected to be in a rather homogeneous non-metallic phase state (no mixed-state or substantial phase separation effect) [15,17,18,19].
The second sample (No. 2) (with the size about $1.5 \times 2 \times 2$ mm$^3$) was studied previously in Ref. [20] where $\delta \approx 0.05$ was estimated. The sample was shown [20] to be inhomogeneous, consisted of AFM and superconducting phases. Temperature dependence of the dc magnetic susceptibility $\chi(T)$ (Fig. 2), measured in this study, shows both the AFM ($T_N = 205 \pm 2$ K) and superconducting ($T_c \approx 25$ K) transitions, reflecting its phase separated state. The values of $T_N$ and $T_c$ correspond well to those measured in Ref. [20].

The temperature dependences of resistivity, $\rho(T)$, were measured with a direct measuring current using the four-probe method in zero magnetic field ($H = 0$). To produce contacts for the current and potential leads, silver was deposited onto the contact pads and then thin gold wires were glued with silver paste. Current was passed both along and across the CuO$_2$ planes. The measuring current was selected (using the measured current-voltage characteristics) so that the Ohm’s law could be obeyed in the whole $T$-range of the measurement. The measuring current $J$ for sample No. 1 ($T_N \approx 266$ K) was varied within 3–10 $\mu$A. The less resistive sample No. 2 ($T_N \approx 205$ K) was measured at higher current $J \leq 100$ $\mu$A.

3 Results and discussion

3.1 Sample La$_2$CuO$_{4+\delta}$ with $\delta \lesssim 0.01$ and $T_N \approx 266$ K

The obtained dependences $\rho(T)$ (presented as log $\rho$ vs. $T^{-1/4}$) for the in-plane (current $J$ parallel to the CuO$_2$ planes) and out-of-plane ($J \parallel c$) transport are shown in Fig. 3. As expected for layered cuprates, the out-of-plane resistivity is far higher than the in-plane one. Both curves have a clear kink (or turn) near $T_N \approx 266$ K. Below $T_N$, the curves follow the Mott’s law for variable-range hopping

$$
\rho \approx \rho_0 \exp \left( \frac{T_0}{T} \right)^{1/4},
$$

which agrees with the previous data for La$_2$CuO$_{4+\delta}$ [22,21,22]. The value of exponent (1/4) corresponds with that expected for a 3D system [23]. Below $T_N$, the characteristic temperatures $T_0$ [see Eq. (1)] for the curves $\rho(T)$ in Fig. 3 are $3.3 \times 10^5$ K and $7.7 \times 10^4$ K for the in-plane and out-of-plane currents, respectively. The localization length $L_c$ can be estimated from the expression $kT_0 \approx 16/[N(E_F)L_c^2]$ [23], where $N(E_F)$ is the density of charge-carrier states at the Fermi level. Using $N(E_F) = 2.8 \times 10^{46}$ J$^{-1}$m$^{-3}$ [24], the values $L_c \approx 0.5$ nm and $L_c \approx 0.84$ nm can be obtained for the in-plane and out-of-plane directions of the current, respectively, for $T \leq 266$ K. This
corresponds to previous estimates of the value of $L_c$ in La$_2$CuO$_4$ below 200 K [21,22]. Above $T_N$, the investigated $T$-interval is not wide enough (on the inverse temperature scale) to determine precisely functional dependence $\rho(T)$ of an exponential type like Eq. (1). If, nevertheless, to apply dependence (1) formally for comparison with $\rho(T)$ above $T_N$, the calculated values of $L_c$ turn out to be too small (less than interatomic O-O or Cu-Cu distances) to believe in variable-range hopping in this temperature range. At the same time the simple exponential dependence $\rho(T) \propto \exp(E_a/kT)$ is obviously inconsistent with the experimental results in the whole temperature interval studied.

In Fig. 4 temperature behavior of resistivity is presented as log $\rho$ vs. $T$ that shows more evidently the change in $\rho(T)$ behavior near $T_N$. It is seen that below $T_N$ the $\rho(T)$ follows the Mott’s law (as indicated above) but above $T_N$ the resistivity decreases with temperature far faster than below it. This is in contrast with $\rho(T)$ behavior near $T_N$ in metallic conductors where resistivity increases when going from AFM to PM state [6,7,8]. At the same time, in some magnetic semiconductors a kink in $\rho(T)$ near $T_N$ similar to that shown in Fig. 4 is observed [9]. An increasing in activation energy of conducting electrons at AFM-PM transition was proposed as a possible explanation but it was not considered as a general rule [9]. For the sample studied no some definite activation energy exists below or above $T_N$, so we cannot apply this explanation.

Magnetoresistance (MR) was measured in this study using a rotating Kapitza electromagnet with the highest field 1.7 T. In the interval 170–400 K in fields up to 1.7 T, the MR was very low ($|\Delta R(H)/R(0)| < 10^{-3}$). This does not permit to judge about an anomaly in the MR behavior near $T_N$. It can not be excluded, however, that in rather strong fields MR of La$_2$CuO$_4$+$\delta$ could reveal some anomalous behavior near $T_N$. The possibility of such anomaly caused by the spin disorder influencing the mobility of charge carriers was discussed in [25]. An anomaly in the out-of-plane MR ($J \parallel c$) upon crossing $T_N$ with changing in temperature was found actually in single-crystal YBa$_2$Cu$_3$O$_{6+x}$ for both the in-plane and out-of-plane orientations of the field [10]. A rather high magnetic field, $H = 16$ T, was needed, however, in this case to increase MR magnitude up to about 1% below $T_N$ and to reveal clearly enough this effect [10].

The effect of magnetic ordering on the hopping conduction of magnetic semiconductors should be analyzed taking into account the type of magnetic order and the origin of charge carriers. The nature of holes in La$_2$CuO$_{4+\delta}$ is not completely clear so far (see the Discussion in [2]). Although it is commonly supposed that the holes in La$_2$CuO$_{4+\delta}$ are mainly of oxygen character [3], the overlapping of the $d$- and $p$-orbitals and the $d$- and $p$-band hybridization [26] can also have an appreciable effect on the nature and motion of holes. It is known that in high-$T_c$ cuprates the AFM order and the charge carriers (holes)
are antagonistic to each other \cite{27}. On the one hand, at $T < T_N$ holes of any type, in addition to lattice distortion and the related polaron effects, can cause considerable distortion of the AFM order in cuprates \cite{28}. The hole can have a spin and its motion can add to the disturbance of the AFM order (frustration effect) \cite{28}. On the other hand, the AFM order impedes the motion of the holes.

Above $T_N$, the long-range 3D AFM order starts to decay quickly \cite{27} though the 2D AFM correlations can persist in the CuO$_2$ planes even far above $T_N$ \cite{3}. It can be suggested, therefore, that in cuprates with weak coupling between the CuO$_2$ layers (quasi-2D conductors with strong conductivity anisotropy) some feature at $T \approx T_N$ is expected only for the out-of-plane conductivity, when the interlayer exchange coupling is destroyed above $T_N$. For the in-plane conductivity nothing special is to expect since 2D intralayer AFM correlation still exists even far above $T_N$. This type of behavior was really seen in quasi-2D cuprates like YBa$_2$Cu$_3$O$_{6+\delta}$ \cite{10} and lightly-doped La$_{2-x}$Sr$_x$CuO$_4$ \cite{11}, where features at $T \approx T_N$ were found only in the out-of-plane resistivity but not in the in-plane one. By contrast, the observed variations in conductivity near $T_N$ in the sample studied manifest that the destruction of the 3D AFM order by thermal fluctuations impacts on conductivity in both the in-plane and out-of-plane directions (Figs. 3 and 4). This is not surprising due to specific influence of oxygen holes which leads to 3D character of hopping conductivity in La$_2$CuO$_{4+\delta}$ with excess oxygen \cite{25} as it is mentioned above.

In low-doped La$_{2-x}$Sr$_x$CuO$_4$ the hole mobility increases as the AFM correlation length $\xi_{AF}$ decreases, i.e. as the AFM order grows weaker \cite{29}. At the same time, it is known \cite{3,27} that in La$_2$CuO$_{4+\delta}$ and low-doped La$_{2-x}$Sr$_x$CuO$_4$ the length $\xi_{AF}$ is practically independent on temperature below $T \approx 300$ K. However, above $T_N$, it decreases drastically with rising temperature \cite{3,27}. Thus, the change in the behavior of conductivity at temperature exceeding $T_N$ (Figs. 3 and 4) can be related to the decrease in the AFM correlation length and the corresponding enhancement of the hole mobility in this temperature region.

It should be noted that some of the authors of this article have observed more than once the $R(T)$ of La$_2$CuO$_{4+\delta}$ (with $T_N$ in the range 180–280 K) behaving near $T_N$ as in Fig. 3 (e.g., see \cite{2}). The study of Ref. \cite{2} was done, however, only up to 300 K, so that this effect does not appear quite clearly. In this study the temperature interval was extended to 430 K. This made possible to see the change in behavior of $R(T)$ near $T_N$ more clearly and allowed us to relate it to the decrease in the AFM correlation length above $T_N$.

It is significant as well that behavior of $R(T)$ near Néel temperature quite similar to that shown in Fig. 3 was found in other magnetic oxides like manganites (La$_{0.25}$Ca$_{0.75}$MnO$_3$ \cite{30}) or cuprates with variable-range hopping transport (Y$_{0.37}$Pr$_{0.63}$Ba$_2$Cu$_3$O$_7$ \cite{31}, YBa$_2$Cu$_3$O$_{6+\delta}$ \cite{10}, La$_{2-x}$Sr$_x$CuO$_4$ \cite{11}). It seems
therefore that particular features of the change in temperature behavior of hopping conduction of La$_2$CuO$_4$ near $T_N$ found in this study (variable-range hopping below $T_N$ and progressive decreasing in resistivity with temperature above $T_N$) is perhaps a common property of magnetic semiconducting oxides with perovskite-like lattice.

3.2 Sample $La_2CuO_{4+\delta}$ with $\delta \approx 0.05$ and $T_N \approx 205$ K

As it is mentioned above, this sample is inhomogeneous consisting of a mixture of AFM and superconducting phases, which is reflected in the temperature behavior of the dc susceptibility $\chi(T)$ (Fig. 2). Let us consider at first the temperature dependence $\rho(T)$ recorded in the in-plane current direction (Fig. 5) which is found to be in complete concordance with $\chi(T)$, showing resistive superconducting transition with $T_c \approx 25.5$ K and clear feature (kink) near $T_N \approx 205$ K.

On the whole, this sample (No. 2) is much ($\approx 10^3$) less resistive than the sample No. 1 with $T_N \approx 266$ K (compare Figs. 3 and 5). Nevertheless, the variable-range hopping [$\ln \rho(T) \propto T^{-1/4}$] is also revealed in this sample in the range between $T_c$ and $T_N$ (Fig. 5). In the same way as for the sample No. 1, an increased drop in resistivity with temperature is seen above $T_N$; furthermore, far enough above $T_N$ (at $T_{min} \approx 350$ K, where resistance minimum shows up) a transition from non-metallic ($d\rho/dT < 0$) to metallic ($d\rho/dT > 0$) temperature behavior of $\rho(T)$ takes place.

Resistive superconducting transition in the in-plane direction is fairly sharp (Fig. 5) with $T_c \approx 25.5$ K defined as the temperature of the inflection point of $\rho(T)$ curve (which was found using derivative). In low-temperature range of the resistive transition (at $T \leq 25$ K) some clear “shoulder” in $\rho(T)$ can be seen, as that commonly observed in granular (low- and high-$T_c$) superconductors with a rather weak connection between grains [32,33]. Thus it can be thought that superconducting regions in this inhomogeneous sample make some sufficiently continuous chain to show the rather sharp resistive superconducting transition. On the other hand, however, some links of this chain are weak which determines the shoulder in low-temperature part of the resistive transition curve [32,33]. The indicated features of the resistive transition (the "shoulder" together with non-zero resistance even at lowest temperature of this study) reflect unambiguously two-phase (phase-separated) state of the sample, in which some of the superconducting "grains" (clusters) are isolated within AFM matrix.

The resistivity in the out-of-plane ($J||c$) direction is (same as for sample No. 1) much larger than that for the in-plane one (Fig. 6). Nevertheless, the temper-
ature dependence of resistivity has main features similar to those in the in-plane current direction. Superconductivity shows itself as a considerable drop of resistance when approaching the temperature $T_c \approx 25.5$ K from above. The resistance, however, does not come down to zero, which is an evidence of higher phase inhomogeneity in this direction, so that a continuous superconducting chain is not formed. Change in $\rho(T)$ behavior after elevation of temperature above $T_N$ is identical to that for the in-plane current direction. Even a transition from nonmetallic to metallic $\rho(T)$ behavior takes place (resistance minimum at $T_{min} \approx 385$ K).

3.3 Summary

The results obtained demonstrate clear interrelation between the magnetic and conducting properties of low-doped $\text{La}_2\text{CuO}_{4+\delta}$. Both samples, although significantly different in resistivity and $T_N$, show the same characteristic changing in transport properties above $T_N$: system becomes less resistive and more metallic. In sample No. 2 even a nonmetal-metal transition takes place far enough above $T_N$. This conforms with known theoretical concepts and some experiments [27,28,34]. According to these, the AFM order enhances hole localization while thermal destruction of the AFM order induces delocalization of charge carriers, so that above $T_N$ a system can approach metallic state with increasing temperature. At $T \approx T_N$ only interplane AFM order disappears; whereas, the in-plane AFM can survive even rather far above $T_N$. At the same time, the AFM correlation length $\xi_{AF}$ decreases drastically with temperature above $T_N$ [31,27], and this should be accompanied with enhancement of the hole mobility [29]. Strong increase in conductivity above $T_N$ can lead in some cases to nonmetal-metal transition as it is observed in this study. These effects are seen in both the in-plane and out-of-plane directions which is determined by 3D character of hopping conduction in $\text{La}_2\text{CuO}_{4+\delta}$.

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Figures

Figure 1. Temperature dependence of the dc magnetic susceptibility $\chi$ in the field $H = 0.543$ T parallel to the crystallographic axis $c$ of single crystal La$_2$CuO$_{4+\delta}$ (sample No. 1, $\delta \lesssim 0.01$).

Figure 2. Temperature dependence of the dc magnetic susceptibility $\chi$ in the field $H = 0.83$ T parallel ($\parallel$) and perpendicular ($\perp$) to the crystallographic axis $c$ of single crystal La$_2$CuO$_{4+\delta}$ (sample No. 2, $\delta \approx 0.05$). The inset shows diamagnetic response below the superconducting transition temperature $T_c \approx 25$ K.

Figure 3. Temperature dependence of resistivity (presented as log $\rho$ vs. $T^{-1/4}$) of single crystal La$_2$CuO$_{4+\delta}$ (sample No. 1, $T_N = 266$ K) for the in-plane (current $J$ parallel to the CuO$_2$ planes) and out-of-plane ($J\parallel c$) directions of the measuring current. In both cases, solid lines present Mott’s law (Eq. 1) which is obeyed below $T_N$.

Figure 4. (Color online) Temperature dependence of resistivity of sample No. 1 (presented as log $\rho$ vs. $T$) for measuring current parallel to the c-axis. The dashed line presents Mott’s law (Eq. 1). It is seen that above $T_N$ progressive deviation from this law sets in.

Figure 5. Temperature dependence of resistivity of single crystal La$_2$CuO$_{4+\delta}$ (sample No. 2, $\delta \approx 0.05$, $T_N \approx 205$ K) for the in-plane (current $J$ parallel to the CuO$_2$ planes) direction of the measuring current. The inset presents this dependence as ln $\rho$ vs $T^{-1/4}$ to show hopping conductivity in the range between $T_c$ and $T_N$. The value of $T_c \approx 25.5$ K is defined from the position of a peak in the temperature behaviour of the derivative $d\rho/dT$ in the region of superconducting transition.

Figure 6. Temperature dependence of resistivity of single crystal La$_2$CuO$_{4+\delta}$ (sample No. 2, $\delta \approx 0.05$, $T_N \approx 205$ K) for the out-of-plane ($J\parallel c$) direction of the measuring current. The inset presents this dependence as ln $\rho$ vs $T^{-1/4}$. Positions of $T_N$ and $T_c$ are shown by arrows.
Figure 1 to paper Belevtsev et al.

\[ \chi (10^{-6} \text{emu/g}) \]

\[ T_N = 266 \text{ K} \]

\[ H \parallel c \]
Figure 2 to paper Belevtsev et al.

\[ \chi(10^6 \text{ emu/g}) \]

\[ T_c \approx 25 \text{ K} \]

\[ H=0.83 \text{ T} \]

\[ H || c \]

\[ T_N = 205 \pm 2 \text{ K} \]

\[ H \perp c \]

\[ H=15 \text{ mT} \]
Figure 3 to paper Belevtsev et al.
Figure 4 to paper Belevtsev et al.
Figure 5 to paper Belevtsev et al.
Figure 6 to paper Belevtsev et al.