Negative differential resistance in single crystal La$_2$CuO$_4$ at low temperature

B. I. Belevtsen and N. V. Dalakova

B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Sciences, pr. Lenina 47, Kharkov 61103, Ukraine

A current-controlled negative differential resistance has been revealed in the $I$-$V$ characteristics of single crystal La$_2$CuO$_{4+\delta}$ in the low temperature region. The non-linear behavior of conductivity is accompanied by a transition from positive to negative magnetoresistance when the current is growing. Possible reasons for the effect observed are discussed.

La$_2$CuO$_{4+\delta}$ is a mother compound for one of the family of high-$T_c$ superconductors (HTSC). The stoichiometric La$_2$CuO$_4$ ($\delta = 0$) is an antiferromagnetic (AFM) Mott insulator with the Néel temperature $T_N \approx 320$ K. On doping it with oxygen ($\delta \neq 0$), charge carriers (oxygen holes) appear in the system, which leads to destruction of the AFM order and brings the system into the metallic (superconducting) state. The excess oxygen resides between the LaO planes and thus determines the three-dimensional (3D) character of conductivity in La$_2$CuO$_{4+\delta}$. Because of its high mobility, the excess oxygen forms a favorable condition for chemical (impurity-induced) phase separation. Indeed, as neutron-diffraction data show, below 320 K crystalline La$_2$CuO$_{4+\delta}$ separates into two phases which are crystallographically close to each other. One of the phases has stoichiometry similar to that of La$_2$CuO$_4$. The other is rich in oxygen and becomes superconducting below $T_c \approx 38$ K. More evidence supporting phase separation in this material was obtained by quadrupole and nuclear magnetic resonance techniques. The phase separation and its investigation are topical problems of HTSC physics. The structural and stoichiometric inhomogeneities caused by phase separation can affect significantly the behavior of the transport properties of copper oxides.

This study is concerned with the effect of current upon the conductive and magnetoresistive properties of single crystal La$_2$CuO$_{4+\delta}$ ($T_N = 182$ K). The dc resistivity in the direction parallel to the CuO$_2$ planes was measured using the Montgomery method at different pre-assigned current. The magnetic field was parallel to the tetragonal $\vec{c}$-axis of the crystal and perpendicular to the transport current.

The temperature dependencies of the resistivity $\rho_\parallel$ measured along the $\vec{c}$-axis for the different amplitudes of the measuring currents $J$ are shown in Fig. 1. It is seen that at $J \leq 1 \mu A$ the resistance is only slightly dependent on current in the whole interval of temperatures (4.2–300 K). The Mott’s law for variable-range hopping (VRH) is well obeyed in the region 20–200 K for $J \leq 1 \mu A$:

$$R \approx R_0 \exp \left( \frac{T_0}{T} \right)^{1/4}, \quad (1)$$

In this region the Ohm’s law is obeyed well. The exponent (1/4) in Eq. (1) corresponds to the behavior of a

$$\text{FIG. 1: The dependencies of } \lg \rho_\parallel \text{ vs. } T^{-1/4} \text{ at different transport currents } (J \parallel \vec{a}).$$

For $T < 10$ K and $J \leq 1 \mu A$ the resistance grows with lowering temperature more rapidly than it is predicted by Eq. (1). This behavior is typical for crystal La$_2$CuO$_{4+\delta}$ in the low temperature region. The effect may be produced by the isolated superconducting inclusions that appear in the dielectric matrix on phase separation when the volume fraction of the superconducting phase is much smaller than the percolation threshold.

At $J > 1 \mu A$ there is a significant deviation of $\rho_\parallel(T)$ from the Mott’s law in low temperature region. When the current increases, the resistance drops drastically, and the temperature at which $\rho_\parallel(T)$ starts to deviate from Eq. (1) shifts towards higher temperatures (Fig. 1). This behavior accounts for the non-linear effects in the conductivity. The non-linear $I$-$V$ curves are illustrated in Fig. 2. At $T < 8$ K some regions with negative differential resistance (NDR) can be seen where $dV/dI < 0$. Earlier, a voltage-controlled NDR effect at low temperatures ($T < 10$ K) was observed in single crystal La$_2$CuO$_4$ with inhomogeneous distribution of oxygen. Here we report for the first time a current-controlled NDR in single-crystal La$_2$CuO$_4$ with more homogeneous distribution of
According to Ref. 8, the influence of electric field on resistance under the VRH condition is described by

\[ R(T, E) = R_0(T) \exp\left(-\frac{eE r_h \gamma}{kT}\right), \]  

(2)

where \( R_0(T) \) is the resistance for \( E \to 0 \) described by Eq. 1, \( r_h \) is the mean hopping distance, \( \gamma \) is a factor of the order of unity. It is evident from Eq. (2) that in rather low fields (\( E \ll kT/er_h\gamma \)), resistance is field independent, i.e. the Ohm’s law is obeyed. As follows from estimation, this is true for the sample studied even in the highest fields of the experiment. In this context the non-linear behavior of the \( I-V \) curves (Fig. 2) can hardly be related to the influence of the electric field on hopping conduction.

There may be another reason for the non-linearity, namely, electron overheating with rather high currents. If the charge carriers do not have enough time to give up quickly the energy received from the field to the lattice, their temperature rises and exceeds that of the phonons. The overheating affects the mobility of the carriers and leads to violation of the Ohm’s law. The theory of “hot” electrons was applied successfully to explain the violation of the Ohm’s law in experiments on doped semiconductors. In Ref. 10 the non-linearity of experimental \( I-V \) characteristics of doped Ge with hopping conduction was described quantitatively taking into account electron overheating and the “thermal model” of electron-phonon energy transfer. It was assumed that the resistance of the sample was determined only by the electron temperature \( T_e \) irrespective of the value of current. In this case the nonlinearity of \( I-V \) curves was due to a decrease in the sample resistance \( R(T_e) \) caused by the heating of the charge carriers to \( T_e \). As a result, the voltage over the sample \( V = IR(T_e) \) can decrease when the current increases. Below a certain critical temperature \( T_x \) an extreme point \( dV/dI = 0 \) appears in the \( I-V \) curves, which is followed by a NDR region. This is a region of instability, current and resistance oscillations, and nonequilibrium transitions. The known theories attribute NDR, among other things, to a non-uniform distribution of impurities and defects over the crystal, which produce regions with electric fields of different intensities. In the sample studied, NDR can be caused by phase separation into superconducting and dielectric regions.

Qualitatively, the \( I-V \) curves in Fig. 2 correspond to those calculated in Ref. 10 taking into account the overheating effect. For the sample studied critical temperature transition to NDR is about 6 K (Fig. 2); whereas estimations made in the frame of the “thermal model”\( ^{11} \) give the value close to 1 K. This discrepancy may be attributed with phase separation into superconducting and dielectric regions. The model in Refs. 10,11 was developed for semiconductors and did not allow for superconducting inclusions as factors of inhomogeneities. Nevertheless, the basic concepts of the model\( ^{10,11} \) account on the whole for the results obtained. The observed current-controlled NDR effect can be interpreted as NDR typical for percolation systems\( ^{12} \) in which increasing electric fields (currents) lead to elongation of the existing high-conductivity percolation paths or even to the formation of new ones. However, the results obtained are not sufficient to analyze comprehensively or to draw conclusions about particular mechanisms of this effect in the investigated sample.

The behavior of magnetoresistance (MR) in the single...
The effect of current is particularly evident in the low temperature region (Fig. 3). For rather low currents $J \leq 1 \mu A$ (with conductivity close to the Ohmic one), MR is positive in the low temperature region ($T \leq 10 K$) (Fig. 4). We can attribute this positive MR to the influence of superconducting inclusions, like in $La_2CuO_{4+\delta}$ sample with much higher $T_N$. An increase in the current produces the Joule heating and corresponding pair-breaking effect. As a result, the positive MR disappears. When the current reaches $J \approx 10 \mu A$, MR becomes negative.

The possible sources of the negative MR in $La_2CuO_{4+\delta}$ at $T > 10 K$ was considered in details in Ref. 13. In fields above $\approx 5 T$, MR is to a large extent determined by the metamagnetic AFM - weak FM transition. The competition of two different MR mechanisms and the transition from positive to negative MR under electron overheating are illustrated in Fig. 3. This corresponds to the temperature behavior of MR at low currents (Fig. 4).

The results of the MR investigation thus attest to the effect of electron overheating, which in turn stimulates NDR at high currents in the low temperature region. The latter effect evolves from the inhomogeneous composition of the sample: because of phase separation typical for this system, superconducting inclusions are produced in the dielectric matrix at low temperatures.

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**FIG. 4:** Temperature dependencies of magnetoresistance for $J \parallel \vec{a}$ taken at two magnitudes of magnetic field $H \parallel \vec{c}$.

Crystal $La_2CuO_{4+\delta}$ studied is also strongly dependent on transport current and sensitive to electron overheating.

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* Electronic address: belevtsev@ilt.kharkov.ua

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