

**LOW-TEMPERATURE RESISTIVITY OF SINGLE CRYSTALS  
YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> IN THE NORMAL STATE**

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A scan of the superconductor - nonsuperconductor transformation in single crystals YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> ( $x \approx 0.37$ ) was done in two alternative ways, namely, by applying the magnetic field and by reducing the hole concentration through the oxygen rearrangement. The in-plane normal-state resistivity  $\rho_{ab}$  obtained in both cases was quite similar; its temperature dependence can be fitted by logarithmic law in the temperature range of almost two decades. However, a different representation of the  $\sigma_{ab} = 1/\rho_{ab}$  by a power law typical for a 3D-material near a metal - insulator transition is also plausible. The vertical conductivity  $\sigma_c = 1/\rho_c$  followed the power law and neither  $\sigma_c(T)$ , nor  $\rho_c(T)$  could be fitted by  $\log T$ . It follows from the  $\rho_c$  measurements that the transformation at  $T = 0$  is split into two transitions: superconductor - normal-metal and normal-metal - insulator. In our samples, they are distanced in the oxygen content by  $\Delta x \approx 0.025$ .

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In this paper, we intend to apply the scaling theory of localization [1] to the underdoped system YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub>. The theory [1] allows to classify temperature dependence of the conductivity in the close vicinity of the metal - insulator transition in a 3D-material. There is a critical region near the transition where the conductivity  $\sigma(T)$  follows a power law

$$\sigma = \alpha + \beta T^m. \quad (1)$$

When the main inelastic processes in the critical region are controlled by the electron-electron interaction, the exponent  $m = 1/3$  [2, 3]. The constant  $\alpha$  in the relation (1) is negative,  $\alpha < 0$ , and the critical region there is restricted from below by a crossover temperature  $T^*$  on the insulating side of the transition. Below  $T^*$ , the conductivity falls off exponentially [4, 5]

$$\sigma \propto \exp[-(T_0/T)^n], \quad n = 1, 1/2, \text{ or } 1/4. \quad (2)$$

The crossover temperature  $T^*$  and the constant  $\alpha$ , both become zero in the transition point so that  $\sigma(T)$  in this point is proportional to  $T^{1/3}$  [2, 3]:

$$\sigma = \beta T^{1/3}. \quad (3)$$

This relation can be used for detecting the transition point.

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A low-temperature crossover line exists in the critical region of the transition on the metallic side too. The conductivity here is described by dimensionless equation [6]

$$s^{3/2} = s^{1/2} + t^{1/2}, \quad s = \sigma(T)/\sigma(0), \quad t = T/T^*. \quad (4)$$

The function  $s(t)$  transforms into power laws (1) in the opposite temperature limits, with  $\alpha = \sigma(0)$  and  $m = 1/2$  at low temperatures, when  $t \ll 1$ , and with  $\alpha = \frac{2}{3}\sigma(0)$  and  $m = 1/3$  when  $t \gg 1$ . It follows from the Eq.(4) that at the crossover temperature, when  $t = 1$ , the conductivity is  $\sigma(T^*) = 1.75\sigma(0)$ .

According to the theory [1], normal-metal - insulator transition does not exist in 2D-materials: any film is expected to become insulating at  $T = 0$ . With decreasing the temperature, the localization starts by the so called quantum corrections to the classical conductivity  $\sigma_0$

$$\sigma = \sigma_0 + \Delta\sigma = \sigma_0 + \gamma_\sigma \log T, \quad \Delta\sigma \ll \sigma_0. \quad (5)$$

Relation  $\Delta\sigma < \sigma_0$  cannot be violated because the conductivity  $\sigma(T)$  is always positive. When  $\sigma_0$  and  $\Delta\sigma$  become comparable the weak localization turns into the strong localization and logarithmic behavior (5) transforms into exponential one (2): at low enough temperature  $\sigma(T)$  should fall off exponentially.

When the metal is superconducting the pattern of the transition into insulating state changes. In 2D, the transition superconductor - insulator has been observed experimentally [7]. In 3D, it is not clear whether such transition can take place as a single transition or it would come through intermediate state of normal metal. In order to study low-temperature behavior of a superconductor, we can bring it to the transition, suppress the superconductivity by magnetic field  $H$ , and then investigate the transition and its vicinity keeping the field constant. It was assumed that material with suppressed superconductivity would behave at finite temperatures as an usual metal. However, since 1980, experimental indications repeatedly appeared that for a superconducting material there is an intermediate region in the vicinity of the transition in which the normal resistivity varies logarithmically with the temperature [8]:

$$\rho(T) = \rho_0 + \Delta\rho = \rho_0 - \gamma_\rho \log T. \quad (6)$$

The temperature dependence (6) was found in granular aluminum [8] and granular niobium nitride films [9], in percolating lead films [10], in  $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-y}$  ceramics [11]. In all these cases, the resistance changes by several times in the range of the logarithmic temperature dependence, the logarithmic term in (6) becoming the leading one at low temperature:

$$\Delta\rho \gg \rho_0. \quad (7)$$

Hence, the relation (6) cannot be converted into (5) despite formal resemblance between them.

The interest to this problem renewed after publications by Ando, Boeinger, et al. [12, 13]. They revealed the  $\log T$ -term in the resistivity of the underdoped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  in pulsed magnetic fields of 60 T. This interest has several aspects. (i) As the volume of experimental data is rather poor one cannot be confident and must check whether the  $\log T$ -term really exists - it is not simple and even

not always possible to distinguish between  $\log T$  and a power-law dependence (1). (ii) What are the specific properties of those materials where this term appears. If these were the high- $T_c$  superconductors only [11–14], it would point to a specific role of strong correlations in electron systems; on the other hand, it may occur that it is granularity which is of the primary importance [8–11, 14].

Motivated by these goals, we present below the measurements of the temperature dependence of the in-plane  $\rho_{ab}$  and vertical  $\rho_c$  resistance of the single crystals  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ . By decreasing the doping level in the high- $T_c$  superconductor systems, one can suppress the superconducting transition temperature  $T_c$  and bring the system to the border of the superconducting region. In  $\text{YBaCuO}$ -system, this can be done by decreasing the oxygen content or, in a limited range, by oxygen rearrangement in the planes of  $\text{CuO}$ -chains [15, 16].

Single crystals  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  were grown by the flux method in alumina crucibles [17]. Being oxygenated at  $500^\circ\text{C}$  in flowing oxygen they had  $T_c$  of about  $90 - 92$  K and fairly narrow resistive transition  $\Delta T_c < 1$  K. To bring samples to the border of the superconducting region, the oxygen content was reduced by high-temperature ( $770 \div 820^\circ\text{C}$ ) annealing in air with subsequent quenching into liquid nitrogen [16, 17]. To rearrange the chain-layer oxygen subsystem, the  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  crystal was heated up to  $120 \div 140^\circ\text{C}$  and quenched into liquid nitrogen. This procedure reduces the mean length of  $\text{Cu-O}$  chains; hence, the hole doping of  $\text{CuO}_2$ -planes decreases [18, 19]. The equilibrium state with larger hole density can be restored simply by room-temperature aging. Dosing the aging time, one can also obtain intermediate states. So, the quenching–aging procedure allows to vary gradually the charge carrier density and to tune the sample state through the border of the superconducting region.

A special care was paid to measure reliably the separate resistivity components. The in-plane resistivity was measured on thin,  $20 - 40 \mu\text{m}$ , plate-like crystals by the four-probe method with current contacts covering two opposite side-surfaces of the crystal [17]. The contacts were painted by silver paste and were fixed by annealing. To measure the vertical resistivity, the circular current electrodes were painted on the opposite sides of the plate with potential probes in the middle of the circles. The resistivity was measured by the standard low-frequency (23 Hz) lock-in technique in the temperature range  $0.37 - 300$  K, the measuring current being low enough to avoid any sign of sample overheating even at the lowest temperature.

Below we present the temperature dependence of the in-plane resistivity obtained for the "aged" and "quenched" states of one of the  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  ( $x \approx 0.37$ ) crystals. For the both states, the resistivity passed through a minimum near  $50$  K. The ratio  $r$  of the resistance at room temperature to that at minimum was  $\approx 3$ . (In terms of classical metal physics this means that the crystal is not perfect: crystals with  $r \approx 10$  do exist.) In the quenched state, no signs of the superconducting transition were observed on the  $\rho(T)$  curve down to the lowest temperature. In the aged state, the resistivity growth at low temperatures was interrupted by the superconducting transition. Owing to low  $T_c < 10$  K, the superconducting transition could be suppressed almost completely by the available magnetic field  $H||c$ .

Attempts to fit the  $\rho_{ab}(T)$  data by an exponential law (2) were unsuccessful. On the contrary, we succeeded in fitting the data by the logarithmic law (5), see Fig.1. The quenched and the intermediate states, both without superconductivity,

demonstrate the resistivity which increased logarithmically with decreasing  $T$  over almost two decades of temperature. The magnetoresistance of the quenched state was below 1%; so, the perfect fit demonstrated in Fig.1 is valid with and without magnetic field, as well. It can be seen that the  $\rho_{ab}(T)$  curves for the aged state step by step approach with increasing magnetic field the straight line. Apparently, the deviations from the logarithmic law (5) indicate only that the largest applied field 7.7 T was not strong enough. So, the representation of the data given in Fig.1 agrees with that of Refs.[11–14].

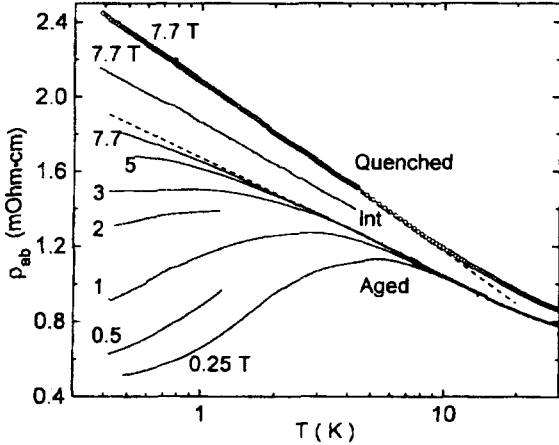


Fig.1. In-plane resistivity versus  $\log T$  for a single crystal YBaCuO with a fixed content of oxygen but with different its arrangements (quenched, intermediate and aged states). Only aged state is superconducting and the set of curves demonstrates how the field  $H||c$  destroys the superconductivity. Dashed lines are extrapolation of the linear dependence  $\rho_{ab}(\log T)$ . Experimental points are plotted only on one curve

However, this is not the only possible interpretation. Assuming that our sample is a 3D-material, we can analyse the data by the help of Eqs.(1) and (4). According to Fig.2, the data for the quenched state of the sample replotted as  $\sigma$  vs.  $T^{1/3}$  approach the straight line at large  $T$  and satisfy the equation (4) with the values of the parameters  $\sigma(0) = 0.23 \text{ mOhm}^{-1} \cdot \text{cm}^{-1}$  and  $T^* = 0.37 \text{ K}$ . Those for the intermediate state are  $\sigma(0) = 0.32 \text{ mOhm}^{-1} \cdot \text{cm}^{-1}$  and  $T^* = 1.25 \text{ K}$ . So, this approach is self-consistent: the aging of the sample increases the hole density and thus leads to the increase of the conductance  $\sigma(0)$  and of the crossover temperature  $T^*$ .

So, at this stage we cannot choose between  $\log T$ - and  $T^m$ -representations, i.e. between representations (5) and (1), (4). We have made experiments with a crystal with  $\tau \approx 10$  too but were left with the same uncertainty. However, in any case, the superconducting state is alternated not by an insulator; representation (5) would indicate that it converts into some specific strongly correlated metallic state, representation (1) would point to a normal-metal state. The conclusion that the transformation in YBaCuO consists of two stages: into normal metal first and into insulator after further decrease of the hole density, was made previously [20] basing on extrapolation of the transport data from high temperatures (from above the resistance minimum). Here the extrapolation edge is far less – 0.4 K only.

The transport properties of YBaCuO-crystals near the border of the superconducting region are highly anisotropic the ratio  $\rho_c/\rho_{ab}$  exceeding  $10^3$  [21]. The crystals can be considered as a stack of weakly bound conducting  $\text{CuO}_2$ -planes. This brings some uncertainty into the question whether the in-plane transport should be considered to be of 2D or of 3D type. At the same time, the vertical

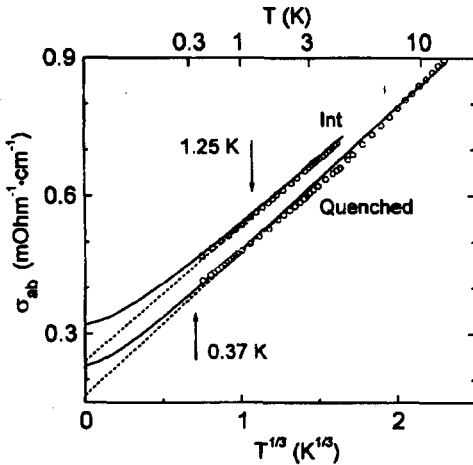


Fig.2. The data for quenched and intermediate states from Fig.1 replotted as  $\sigma_{ab}$  vs  $T^{1/3}$ . Solid lines - fits by Eq.(3) with the  $T^*$  values indicated by arrows, dashed lines - asymptotes in the  $t > 1$  region

transport is certainly 3D and its temperature dependence is of special interest. We have measured several crystals and present below examples of typical behavior.

In Fig.3, the  $\rho_c$  data for two crystals annealed in air at  $800^\circ\text{C}$ , c1 and c2, are presented, each in two states, quenched (curves  $c1_q$  and  $c2_q$ ) and aged ( $c1_a$  and  $c2_a$ ). They certainly do not follow  $\log T$ , but perfectly fit the  $T^{1/3}$ -representation (1) with  $\alpha$  of different signs - see inset. Hence, these aged and quenched states should be placed to different sides of the metal - insulator transition. All the presented data were obtained with magnetic field of 7.7 T but the magnetoresistance was small and did not affect the representation.

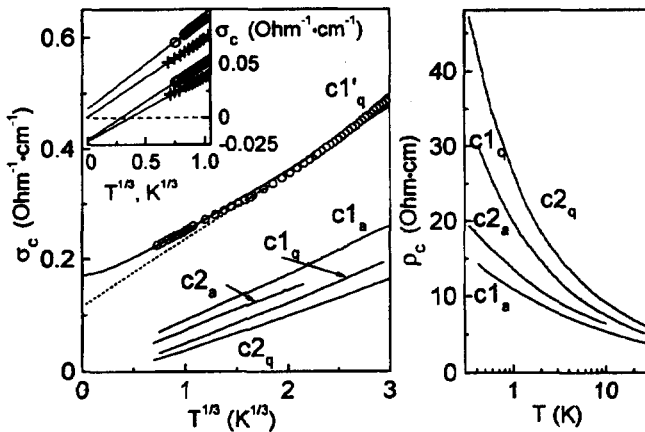


Fig.3. Vertical resistivity data for two crystals, c1 and c2, plotted as  $\sigma_c$  vs  $T^{1/3}$  (left) and as  $\rho_c$  vs  $\log T$  (right). Experimental points are removed from several curves. Inset is enlarged part of the main plot

In the left part of Fig.3, we present in addition the curve  $c1'_q$  obtained for the crystal c1 annealed at  $780^\circ\text{C}$  and quenched; the curve is fitted by eq.(4) with  $\sigma_0 = 1.5\alpha = 0.18 \text{ Ohm}^{-1}\text{cm}^{-1}$ . The aged state of the crystal with this oxygen content reveals symptoms of the superconductivity, such as onset kink and positive magnetoresistance at lower temperatures. Hence, these aged and quenched states should be placed to different sides of the superconductor - normal-metal transition on the phase diagram on Figs.5 and 7 of ref.[16]. According to these diagrams, in the  $800^\circ\text{C}$  range of the annealing temperature, the  $20^\circ$  change results in

$\Delta x \approx 0.025$  difference of the oxygen content and in  $\Delta n/n_c \approx 0.07$  difference of the hole concentration  $n$  normalized to the critical value  $n_c$ . Precise positions of both transitions may depend on the degree of disorder in the crystals. But the obtained numbers can be regarded as estimates for the distance between the superconductor - normal-metal and the metal - insulator transitions along the abscissa axis of the phase diagram.

In conclusion, the low-temperature  $\rho_c(T)$  curves of  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  ( $x \approx 0.37$ ) single crystals follow scaling temperature dependence in the vicinity of the metal - insulator transition and allow to specify the transition point. The difference in the oxygen concentrations  $x$  between this point and those of the normal-metal - superconductor transition is approximately  $\Delta x \approx 0.025$ . It remains still unclear whether the representation of the in-plane resistivity  $\rho_{ab}(T)$  in the region between these transitions in the  $\log T$  scale is meaningful or the description by the functions (1) and (4) is more adequate.

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