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Effect of oxygen deficit on temperature dependence of excess conductivity in $Y_1Ba_2Cu_3O_{7-\delta}$ single crystals

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Evolution of the excess conductivity in $Y_1Ba_2Cu_3O_{7-\delta}$ single crystals has been studied under stepwise oxygen removal by heat treatment. The concentration dependence of the coherence length along **c** axis $\xi_c(0)$ on the oxygen content has been obtained. The oxygen content reduction from $\delta \le 0.1$ to $\delta \approx 0.23$ has been shown to result in a substantial broadening of the temperature interval where the excess conductivity takes place. The $\xi_c(0)$ value increases more than twice and the 2D-3D crossover point is shifted in temperature.

Исследована эволюция избыточной проводимости монокристаллов $Y_1Ba_2Cu_3O_{7-\delta}$ при поэтапном удалении кислорода путем термообработки. Получена концентрационная зависимость длины когерентности в направлении оси **c** $\xi_c(0)$ от содержания кислорода. Показано, что понижение содержания кислорода от $\delta \le 0.1$ до $\delta \approx 0.23$ приводит к значительному расширению температурного интервала существования избыточной проводимости. При этом величина $\xi_c(0)$ увеличивается более чем в два раза и смещается по температуре точка 2D-3D кроссовера.

It is known that as the oxygen content in YBaCuO compounds decreases, the electric resistance increases and the critical temperature (T_c) is lowered [1, 2]. The defect ensemble composition and topology as well as the extent and character of the labile oxygen subsystem are of great importance in this case [2]. It is to note that, in spite of very comprehensive experimental material accumulated to date, the effect of small oxygen deficit on various conductance mode and, in particular, on the fluctuation conductance (FC) of those compounds remains still unclear. Perhaps this is due in part to the fact that the available experimental data were obtained mainly using texturized and ceramic samples containing numerous intergrain bonds as well as films deposited onto various substrates by wide variety of technologies [3–5]. Therefore, it is expedient to study the evolution of various FC modes in high-perfection YBaCuO single crystals in the course of oxygen content reduction.

$Y_1Ba_2Cu_3O_{7-\delta}$ single crystals were grown by solution-melt technique in a gold crucible at 850–970°C as described in [2, 6]. The typical sample size was $2.0 \times 0.3 \times 0.02$ mm^3, the smallest crystal dimension corresponding to **c** axis. To obtain the samples with optimum oxygen content of $\delta \le 0.1$, the selected crystals were annealed in oxygen stream at 400°C for 5 days. To reduce oxygen content, the samples were annealed in oxygen stream for 3–5 days at higher temperatures. The parameters of the samples studied are presented in the Table below.

The electric contacts were prepared by connecting silver wires to the crystal surface using silver paste. The electric resis-

Table

$7-\delta$	T_a, K	T_c, K	$R(300)$, Ω	T^*, K	α_1	α_2	ε_0	d, Å	$\xi_c(0)$, Å
6.92	670	91.738	0.253	143	−0.499	−1.012	0.064	11.69	1.48
6.87	720	90.845	0.303	171	−0.496	−1.032	0.104	11.7	1.89
6.83	760	88.712	0.314	192	−0.500	−1.005	0.145	11.71	2.23
6.81	790	88.394	0.353	215	−0.499	−1.010	0.206	11.71	2.65
6.77	810	78.515	0.397	232	−0.505	−1.031	0.292	11.72	3.17

tance was measured by the standard four-contact technique at 1 mA DC at two opposite current directions in zero magnetic field. In the experimental geometry, the transport current vector was directed at 45° with respect to the twin boundary (TB) plane. The temperature was measured by a copper-constantan thermocouple, the voltage across the sample and across the reference resistor, by V2-38 nanovoltmeters. The voltmeter signals were transferred to a PS through an interface. The measurements were done in temperature drift mode, the drift being about 0.1 K/min near T_c and about 5 K/min at $T > T_c$. The critical temperature was determined from the maximum position in temperature dependences of electric resistance derivative (in the ab plane) with respect to temperature, $dR_{ab}/dT(T)$, in the superconducting transition region. All the measurements were done three days after the annealing was over, thus providing the equilibrium distribution of oxygen over the sample volume at room temperature [2].

In Fig. 1, is shown the temperature dependences of electric resistivity in the ab plane measured after the sample annealing in oxygen stream at various temperatures. In the inset (a), the same dependences are presented as normalized to the resistivity value at room temperature, $\rho_{ab}(300)$. The conductivity as a function of temperature is seen to behave qualitatively as a quasi-metal one. As the annealing temperature increases, the absolute electric conductivity value increases while the linear region of $\rho_{ab}(T)$ becomes narrowed substantially at high temperatures. It is seen from the Table that the critical temperature drops as the annealing temperature increases. The initial narrow superconducting transition region ($\Delta T_c \leq 0.3$ K) is enlarged considerably and the transition itself takes a stepwise shape. Perhaps this evidences the appearance of several phases in the sample that have various critical temperatures of superconducting transition.

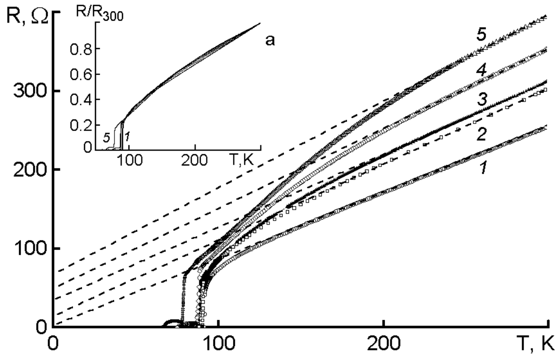

Fig. 1. $R(T)$ dependences for crystals annealed at 670, 720, 760, 790, and 810 K (curves *1–5*, respectively). Inset: the same dependences for electric resistance normalized to R_{300}. The curve numbers in the inset are the same as in main figure.

It is seen from Fig. 1 that below a certain characteristic temperature T^*, the $\rho_{ab}(T)$ dependences become "smoothed", that may be due to appearance of an excess conductance. The temperature dependence of the latter can be defined as

$$\Delta\sigma = \sigma - \sigma_0, \qquad (1)$$

where $\sigma_0 = \rho_0^{-1} = (A + BT)^{-1}$ is the conductivity determined by interpolating the linear section observed in the high-temperature region to zero temperature; $\sigma = \rho^{-1}$, the experimental conductivity value at $T < T^*$. The excess conductance near T_c is known to be due to fluctuating coupling of the current carriers and can be described by a power dependence obtaine in the theoretical Lawrence-Doniach model [7] that supposes a very smooth crossover from 2D to 3D mode of the fluctuating conductance as the sample temperature decreases:

$$\Delta\sigma = \left[\frac{e^2}{16\hbar d}\right]\varepsilon^{-1}\left\{1 + J\varepsilon^{-1}\right\}^{-1/2}, \qquad (2)$$

where $\varepsilon = (T - T^{mf}_c)/\ T^{mf}_c$ is the reduced temperature; T^{mf}_c, the critical temperature of the mean field approximation; $J = 2\xi_c(0)/d)^2$, the interplanar coupling constant; ξ_c, the coherence length along the **c** axis; and d, the 2D layer thickness. In the limiting cases (near T_c at $\xi_c >> d$, the interaction between the fluctuating Cooper pairs being realized in the whole superconductor volume, that is, in the 3D mode; or far from T_c at $\xi_c << \mathrm{d}$, the interaction being possible only in the planes of conducting layers, i.e., in the 2D mode), the expression (2) is transformed into the known relationships for the 3D and 2D cases, respectively, of the Aslamazov-Larkin theory [8]:

$$\Delta\sigma_{2D} = \frac{e^2}{16\hbar d}\varepsilon^{-1}, \tag{3}$$

$$\Delta\sigma_{3D} = \frac{e^2}{32\hbar\xi_c(0)}\varepsilon^{(-1/2)}. \tag{4}$$

When making a comparison with experimental data, it is of importance to determine accurately the $T^{mf}{}_c$ value influencing substantially the slope of $\Delta\sigma(\varepsilon)$ dependences. When comparing with experimental data, $\xi_c(0)$, d, and T_c are usually the fitting parameters in Eqs.(2)–(4) [9]. However, the use of such procedure results as a rule in considerable quantitative discrepancies between the theory and experiments. This, in turn, makes it necessary to use a scaling factor, so-called C factor, as an additional fitting parameter that enables the fitting of the experimental data to calculated ones, thus taking into account the possible inhomogeneous spreading of transport current for each specific sample [4]. In our case, it is T_c that was taken to be $T^{mf}{}_c$. The T_c was determined in the maximum points of $dR_{ab}/dT(T)$ dependences in the superconducting transition region, as has been proposed in [3].

Fig. 2 presents the temperature dependences $\Delta\sigma(T)$ in $\ln\Delta\sigma(\ln\varepsilon)$ coordinates. In the temperature region between T_c and 1.1–1.25 T_c, these dependences are approximated satisfactorily by straight lines with the slope α_1 about −0.5 that corresponds to the power index of −1/2 in Eq.(4), thus evidencing the 3D character of the fluctuating superconductivity within that temperature range. A further temperature elevation results in a considerable reduction of the $\Delta\sigma$ decrease rate (α_2 about −1) that, in turn, could be considered to be associated with the FC dimensionality change. As follows from (3) and (4), in the point of 2D-3D crossover,

$$\varepsilon_0 = 4\left[\xi_c(0)/d\right]^2. \tag{5}$$

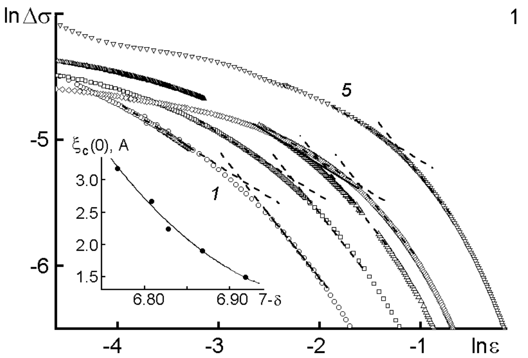

Fig. 2. $\Delta\sigma(T)$ dependences in $\ln\Delta\sigma(\ln\varepsilon)$ coordinates. The curve numbers are the same as in Fig. 1. Inset: $\xi_c(0)$ as a function of oxygen content ($7-\delta$).

In this case, having determined the ε_0 value and using literature data on T_c and interplanar spacing dependences on δ [11], $\xi_c(0)$ can be calculated. As seen in the Table, the $\xi_c(0)$ calculated using (5) is increased from 1.48 up to 3.17 Å as T_c drops. This agrees qualitatively with the $\xi_c(0) = f(\delta)$ dependence obtained for film YBaCuO samples [4]; but the absolute $\xi_c(0)$ values exceed considerably those found in [4] at oxygen deficit. It is to note also that the result obtained from the samples with the highest T_c agrees satisfactorily with the values $\xi_c = 2.3 \pm 0.5$ Å determined from the magnetic susceptibility measurements [6] for YBaCuO single crystals grown by a similar technique at optimum oxygen doping.

As is noted above, the oxygen content reduction is accompanied by a considerable widening of the excess conductance region towards higher temperatures ($T > 1.5T_c$) that cannot be obviously explained within the frame of the existing fluctuation theories. According to the modern concepts (see, e.g., [10, 11]), such a behavior of $\rho_{ab}(T)$ dependences may be due to transition into so-called pseudogap state typical of "underdoped" HTSC compositions. At the other hand, it seems that it is cannot be excluded that the mentioned feature may be caused in part also by the additive contributions to the conductance from impurity phases having higher T_c. For example, in earlier works aimed at FC studies in HTSC compounds [5], it has been shown that superconducting phases with $T_c > 140$ K, although being structurally unstable at normal conditions, may exist as impurity phases in doped and multiphase samples.

Thus, the oxygen content reduction results in a considerable broadening of the region where excess conductance exists, the coherence length being increased more than twice.

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Вплив кисневого дефіциту на температурну залежність надлишкової провідності монокристалів $Y_1Ba_2Cu_3O_{7-\delta}$

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Досліджено еволюцію надлишкової провідності монокристалів $Y_1Ba_2Cu_3O_{7-\delta}$ при поетапному видаленні кисню шляхом термообробки. Отримано концентраційну залежність довжини когерентності у напрямку осі **c** $\xi_c(0)$ від вмісту кисню. Показано, що зменшення вмісту кисню від $\delta \leq 0{,}1$ до $\delta \approx 0{,}23$ зумовлює значне розширення температурного інтервалу існування надлишкової провідності. При цьому величина $\xi_c(0)$ збільшується більш ніж у два рази та зміщується за температурою точка 2D-3D кросоверу.