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# Effect of long aging on the resistivity properties of optimally doped $YBa_2Cu_3O_{7-\delta}$ single crystals



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#### ABSTRACT

We investigate the conducting properties in the basal ab-plane before and after a long time exposure in air atmosphere of the optimally oxygen doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals. Prolonged exposure leads to an increase of the effective scattering centers of the normal carriers. The excess conductivity in a wide temperature range has exponential temperature dependence and near the critical temperature is well described within the Aslamazov–Larkin theoretical model. The prolonged exposure increases to a great extent the temperature range of the implementation of the pseudogap state, narrowing the linear section of the temperature dependence of the resistivity in the ab-plane,  $\rho_{ab}(T)$ .

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#### 1. Introduction

YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> based superconductors have been intensively studied due to their unique properties and their incorporation in superconducting electronics [1]. An important feature of the technological use of the so called 1-2-3 system, ReBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (Re=Y, or other rare-earth element) compounds, is the stability of the oxygen subsystem. In ReBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> with a nonstoichiometric oxygen composition, using temperature [2,3] or high pressure, [4,5] a non-equilibrium state can be induced. This is accompanied by a redistribution of the labile oxygen (structural relaxation) that in turn has a significant impact on the critical and electrotransport parameters of the superconductor [6].

It is assumed that in  $ReBa_2Cu_3O_{7-\delta}$  ( $\delta \leq 0.15$ ) compounds, such relaxation processes does not occur and that their electrical properties are practically unchanged to the impact of external influence [7]. In spite of numerous studies [2–6,8,9], investigating the impact of non-equilibrium conditions on the structural relaxation in the 1-2-3 system, the impact of the long-term effects of external factors (such as the atmosphere) on the structural parameters and the electrotransport in such compounds is not determined. Studies devoted on aging itself are rather limited and confined to ceramic [10], films [11], or textured [12] samples for very different technological applications. As a consequence, the experimental data is often highly controversial. For example,

Qing-Rong Feng et al. [13] reported a significant increase of the superconductive volume fraction, the intragranular critical current density, and the pinning force density in the process during long-term aging. Other studies [10–12] revealed a significant degradation of the above mentioned properties under long time exposure in air. There is also a substantial variation in the parameters of the superconducting state.

Additionally, the  $ReBa_2Cu_3O_{7-\delta}$  compounds practically always have planar defects – twin boundaries (TB) [5,14], which complicate the study of electric transport properties under the influence of external forces. It is important to clarify these issues and this requires the study of pure and perfect single-crystal samples. In particular, a study on the longtime atmosphere influence to the conditions and regimes of fluctuation paraconductivity (FC) and to the so-called pseudogap anomaly (PG) existence in these compounds [15] could be important not only for understanding the nature of high-temperature superconductivity (HTSC) but also for determining the empirical ways to enhance their critical parameters. In the present study we investigate the impact of prolonged exposure in air on various regimes of conduction in the basal ab-plane of the oxygen optimally doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystals with a high critical temperature  $T_c$ .

### 2. Methodology

The  $YBa_2Cu_3O_{7-\delta}$  single crystals were grown in a gold crucible with the solution-melt technology, described in detail in a previous study [15]. The electrical resistance in the ab-plane was measured by a standard 4-contacted method in direct current up

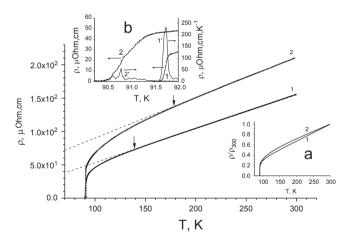
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to 10 mA. The sample temperature was measured with a platinum resistance thermometer. The first measurements of the electrical resistivity in the basal ab-plane were performed immediately after removing the crystal from the melt and their oxygen saturation to the optimum value ( $\delta$ <0.1). After these measurements, the crystal was stored in a glass container until the re-measurements, which were carried out after 6 years. The resistivity measurements were made as re-testing of the sample immediately prior to the studies of the magnetic flux dynamics in the samples, the results of which will be presented in a future study.

#### 3. Results and discussion

The temperature dependence of the resistivity in the ab-plane  $\rho_{ab}(T)$ , measured before and after long time exposure in air atmosphere, is shown in Fig. 1 and inset (a). The resistive transitions to the superconducting state in the  $\rho_{ab}$  versus T and  $d\rho_{ab}/dT$  versus T are shown in the inset (b) of Fig. 1. In both cases (prior and after aging), the dependence is quasi-metallic, but the ratio  $\rho_{ab}(300 \text{ K})/\rho_{ab}(0 \text{ K})$ , before and after the long aging in air is significantly decreased from 40 to 8. In this case, the value  $\rho_{ab}(0 \text{ K})$  was determined by extrapolation of the linear section of the  $\rho_{ab}(T)$  dependence, as is shown in Fig. 1.

At the same time, the electrical resistivity in the ab-plane at room temperature, increased from 155 to 209  $\mu\Omega$  cm, and the critical temperature dropped from 92 to 90.8 K. The width of the resistive transition to the superconducting state  $\Delta T_{\rm c}$ , increased approximately threefold (from 0.3 to $\approx$ 1 K) and the transition has acquired a stepped form. The parameters of the samples are given in Table 1.



**Fig. 1.** The  $\rho_{ab}(T)$  dependence of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> single crystal, before and after long aging in air, curves 1 and 2, respectively. The arrows show the mean field transition temperature to the pseudogap regime,  $T^*$ . The inset (a) shows the same dependence in  $\rho_{ab}/\rho_{ab}(3\ 0\ 0)$  versus T coordinates, the inset (b), shows the transitions to the superconducting state in  $\rho_{ab}$  versus T and  $d\rho_{ab}/dT$  versus T coordinates for the same samples. The numbering of the curves in the inset corresponds to the numbering in the figure.

 Table 1

 Resistivity parameters before and after aging of the samples.

Sample		$ \rho_{ab}(300), $ (μ $\Omega$ cm)		Δ* <sub>ab</sub> , (meV)	$\varepsilon_0$	$\alpha_{\mathrm{3D}}$	$\alpha_{\mathrm{2D}}$	$\xi_{c}(0)$ (Å)
Before aging	92	155	143	87	0.065	-0.509	- 1.124	$1.49\pm0.05$
After aging	90.8	209	172	65	0.034	-0.512	-0.992	$1.98 \pm 0.05$

Using previously published data on the  $T_c$  dependence from the oxygen concentration [16], we can conclude that its oxygen content with aging is slightly decreased (by 1.2%) and remains within the limits ( $\delta \le 0.15$ ). The increase in the width of the resistive transition suggests a significant reduction in the degree of homogeneity of the samples [15]. Additionally, it is consistent with the step-like form, which is observed after aging, showing signs indicating the appearance of phase separation in their volume [3,5]. This is also supported by the presence of a clearly pronounced additional peak in the dependence  $\mathrm{d}\rho_{ab}/\mathrm{d}T$  versus T. In accordance to previous studies [3,5], these peaks correspond to the  $T_c$  of different phases within the volume in the crystal.

As shown by metallurgical studies on the optical microscope in polarized light the TB structure in both samples has not changed with aging. Therefore the increase in electrical resistance cannot be due to the influence of the TB. Therefore, the observed increase  $\rho_{\rm ab}$  is likely caused by a decrease of the density of the carriers or the emergence of effective scattering centers, as it is also evident from the change in the ratio  $\rho_{\rm ab}(300~{\rm K})/\rho_{\rm ab}(0{\rm K})$ . This is consistent with increasing the number of vacancies forming during the long time exposure in air and the increase of the oxygen's hypostoichiometry.

As it can be observed from Fig. 1, when the temperature falls below a certain characteristic value  $T^*$ , a deviation of the  $\rho_{ab}(T)$  from the linear dependence occurs indicating the appearance of excess conductivity, which can be due to the transition to the pseudogap regime [15–19]. There are two main scenarios of the pseudogap anomalies in HTSC systems. According to the first scenario, the occurrence of PG is associated with short-range order fluctuations of "dielectric" type, taking place in hypostoichiometric compounds (see for example Ref. [18]). The second scenario assumes the formation of Cooper pairs at temperatures significantly above the critical,  $T^* \gg T_c$ , followed by the establishment of its phase coherence at  $T < T_c$  [17,19].

Notably, from Table 1 and Fig. 1, prolonged aging leads to a significant narrowing of the field of linear dependence  $\rho_{\rm ab}$  (T) comparing to the original sample, and the temperature  $T^*$  is shifted to higher temperatures by about 30 K, indicating a corresponding increase of temperature interval of the existence of excess conductivity.

The temperature dependence of the excess conductivity can be determined by the equation:

$$\Delta \sigma = \sigma - \sigma_0 \tag{1}$$

where  $\sigma_0 = \rho_0^{-1} = (A+BT)^{-1}$  is the conductivity, determined by extrapolating the linear part to zero temperature, and  $\sigma = \rho^{-1}$  is the experimentally determined value of the conductivity in the normal state. The experimental dependencies of  $\Delta\sigma(T)$  are shown in Fig. 2 in  $\ln \Delta\sigma$  versus 1/T coordinates. For a wide temperature range, these curves are straight lines corresponding to the description of the exponential dependence by the formula:

$$\Delta\sigma \sim \exp(\Delta^*_{ab}/T) \tag{2}$$

where  $\Delta_{ab}^*$  is the value which defines a certain thermal activation process through the energy gap, the "pseudogap".

As it was observed previously [17], the approximation of the experimental data can be greatly enhanced by the introduction of the factor  $(1-T/T^*)$ . In this case, the excess conductivity is proportional to the density of superconducting carriers  $n_s \sim (1-T/T^*)$ , and inversely proportional to the number of pairs  $\sim \exp(-\Delta^*/kT)$ , destroyed by the thermal motion. In this case,  $T^*$  is considered as the mean field transition temperature and the temperature range  $T_f < T < T^*$  (where  $T_f$  is the temperature of transition in the fluctuation conductivity (FC) regime determined as a point of a deviation of value  $\Delta \sigma$  upwards from a linear dependence  $\ln \Delta \sigma(1/T)$  when lowering the temperature), in which the pseudogap state exists, is determined by the rigidity of the order parameter phase that in its

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