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Aging-effect in optimal doped $YBa_2Cu_3O_{7-\delta}$ single crystals

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We investigate the conducting properties of the optimally oxygen doped $YBa_2Cu_3O_{7-\delta}$ single crystals in the basal *ab*-plane before and after a long time exposure (aging) at air atmosphere. Prolonged exposure leads to increase of amount 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 (T_c) is well described within the Aslamazov-Larkin theoretical model. Here we show that the prolonged exposure leads to a great extention of temperature range of the pseudogap state implementation and to narrowing the linear section of the dependence $\rho_{ab}(T)$.

Исследована проводимость в базисной плоскости оптимально допированных монокристаллов YBaCuO до и после длительной выдержки (aging) в атмосфере воздуха. Показано, что длительная выдержка приводит к возрастанию числа эффективных центров рассеяния нормальных носителей. Избыточная проводимость исследованных образцов в широком интервале температур подчиняется экспоненциальной температурной зависимости, а вблизи T_c удовлетворительно описывается теоретической моделью Асламазова-Ларкина. При этом длительная выдержка способствует значительному расширению температурного интервала реализации псевдощелевого состояния и существенному сужению температурного интервала линейной зависимости $\rho_{ab}(T)$.

Aging-effect в оптимально допованих монокристалах $YBa_2Cu_3O_{7-\delta}$. *З.Ф.Назиров, А.В.Попова, Р.В.Вовк.*

Досліджено провідність в базисній площині оптимально допованих монокристалів YBaCuO до і після тривалої витримки (aging) в атмосфері повітря. Показано, що тривала витримка призводить до зростання числа ефективних центрів розсіювання нормальних носіїв. Надлишкова провідність досліджених зразків у широкому інтервалі температур підкоряється експоненціальній температурній залежності, поблизу T_c задовільно описується теоретичною моделлю Асламазова-Ларкина. При цьому тривала витримка сприяє значному розширенню температурного інтервалу реалізації псевдощілинного стану та суттєвому звуженню температурного інтервалу лінійної залежності $\rho_{ab}(T)$.

1. Introduction

An important feature of technological use of so called 1-2-3 system, $ReBa_2Cu_3O_{7-\delta}$ (Re = Y, or other rare-earth element) compounds, is the oxygen subsystem stability. As is known in this compounds with non-stoichiometric oxygen composition the non-equilibrium state can be induced using temperature [1, 2] or high pressure [3, 4]. This is accompanied by redistribution of the labile oxygen (structural relaxation), which, in turn has a significant impact on the critical and electro-transport parameters of the superconductor [1–4].

It is assumed that in optimally oxygen doped $ReBa_2Cu_3O_{7-\delta}$ ($\delta \leq 0.15$) compounds such relaxation processes do not occur and that their electrical properties are practically unchanged under impact of external extreme influences [5]. Despite a large number of works [6, 7] devoted to the study of non-equilibrium conditions and the structural relaxation in the 1-2-3 system, the question about impact of the long-term effects of external factors (such as atmosphere) on structural parameters and electrotransport in such compounds is still open. The studies devoted to aging itself are rather limited and confined to ceramic [8], films [9] or textured [10] samples for very different technological applications. As a consequence, the experimental data are often highly controversial. For example, in [11] reported about significant increasing the superconductive volume fraction of the sample, the intragranular critical current density, and the pinning force density in the process of long-term aging. At the same time, other studies [8–10] revealed a significant degradation of the above mentioned properties under long time exposure in air. There is also a substantial variation in the superconducting state parameters.

Additionally, $ReBa_2Cu_3O_{7-\delta}$ compounds practically always have planar defects — twin boundaries (TB) [12], which complicate the study of electric transport properties under the external forces influence. It is important to clarify these issues and this requires the study of pure and perfect single-crystal samples. In particular, study on the longtime atmosphere influence on conditions and regimes of fluctuation paraconductivity (FC) [13, 14] and on so-called pseudogap anomaly (PG) existence in these compounds [15, 16] could be important not only for understanding the nature of high-temperature superconductivity (HTSC) but also for determination of the empirical ways to enhance their critical parameters. In the present study we investigate the impact of prolonged exposure at air on various regimes of conduction in the basal *ab*-plane of oxygen optimally doped $YBa_2Cu_3O_{7-\delta}$ single crystals with a high critical temperature $T_c \approx 90$ K.

2. Experimental

$YBa_2Cu_3O_{7-\delta}$ single crystals were grown in a gold crucible by solution-melt technology described in details in the previous study [12]. Electrical resistance in the *ab*-plane was measured by standard 4-point probes method in direct current up 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 ($\delta \le 0.1$). After these measurements, the crystal was stored in a glass container until re-measurements, which were carried out after 17 years. The resistivity measurements were made as re-testing of the sample immediately prior to studies of the magnetic flux dynamics in the samples, the results of which will be presented in the future study.

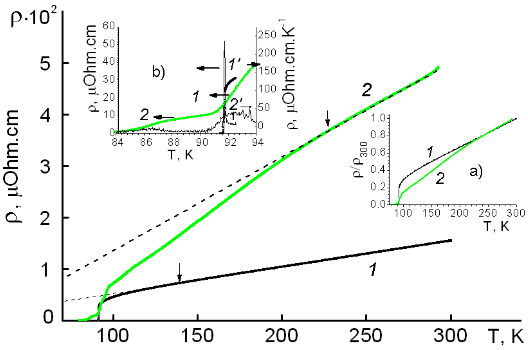

Fig. 1. $\rho_{ab}(T)$ Dependence of $YBa_2Cu_3O_{7-\delta}$ single crystal before and after long aging at air, curves *1* and *2*, respectively. Arrows show the mean field transition temperature to the pseudogap regime, T^*. Inset (a) shows the same dependence in $\rho_{ab}/\rho_{ab}(300)$ versus T coordinates, inset (b) shows transitions to the superconducting state in ρ_{ab} versus T and $d\rho_{ab}/dT$ versus T coordinates for the same samples. Numbering of the curves in the inset corresponds to the numbering in the figure.

3. Results and discussion

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. The resistive transitions to the superconducting state in coordinates of ρ_{ab} versus T and $d\rho_{ab}/dT$ versus T are shown in inset (a) of Fig. 1. It is seen that before and after exposure the fairly extended linear section remains on the dependences $\rho_{ab}(T)$, however, the temperature deviation from the linear dependence of T^* in the process of long-term exposure at air significantly shifted toward the higher temperatures. At the same time, electrical resistivity in the *ab*-plane at the room temperature increased from 155 to 491 μOhm·cm, and width of the resistive transition to the superconducting state ΔT_c increased approximately threefold (from 0.3 to ≈10 K). The transition has acquired a stepped form that testifies to appearance of

Table

Sample	T_c, K		ρ_{ab}(300), μΩ·cm	T^*, K	Δ^*_{ab}, meV	ε_0	α_{3D}	α2D	ξ_c(0), Å
Before aging	92.1		153	147	89	0.064	−0.502	−1.128	1.49
After aging	T_{c1}	86.4	491	228	42	0.035	−0.517	−1.060	1.09
	T_{c2}	92				0.172	−0.493	−1.031	2.43

the phase separation in their volume [2, 4]. This last comment is also supported by presence of the clearly pronounced additional peak in the dependence $d\rho_{ab}/dT$ versus T. In accordance to the previous studies [1–4], these peaks correspond to T_c of different phases within the volume of the crystal. In our case it was possible to identify at least three of these phases with different T_c. Parameters of the samples are given in Table. Using previously published data on the T_c dependence from oxygen concentration [17], we can conclude that its oxygen content with aging is slightly decreased (by 1–4 %) and remains within the limits $\delta \leq 0.2$.

As shown by metallurgical studies on optical microscope in polarized light the TB structure in the both samples did not change with aging. Therefore the electrical resistance increase cannot be due to influence of the twin boundaries. Therefore, the observed increase of ρ_{ab} is likely caused by decrease of the carriers density or by emergence of effective scattering centers. This is consistent with increasing the number of vacancies forming during the long time exposure at air and the increase of the degree of oxygen's nonstoichiometry connection.

As it can be observed in Fig. 1, when 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 transition to pseudogap regime (PG) [15, 16]. Currently there are two main scenarios of the pseudogap anomalies in HTSC systems. According to the first scenario, the occurrence of the PG is associated with short-range order fluctuations of "dielectric" type, taking place in underdoped compounds (see for example [18]). The second scenario assumes the formation of the Cooper pairs at temperatures significantly above the critical, $T^* >> T_c$, followed by establishment of its phase coherence at $T < T_c$ [19, 20].

Notably, from Table and Fig. 1, prolonged annealing leads to a significant narrowing of the field of linear dependence $\rho_{ab}(T)$ comparing to the original sample, and the temperature T^* is shifted to higher temperatures by about 80 K, indicating corresponding increase of temperature interval of the excess conductivity existence.

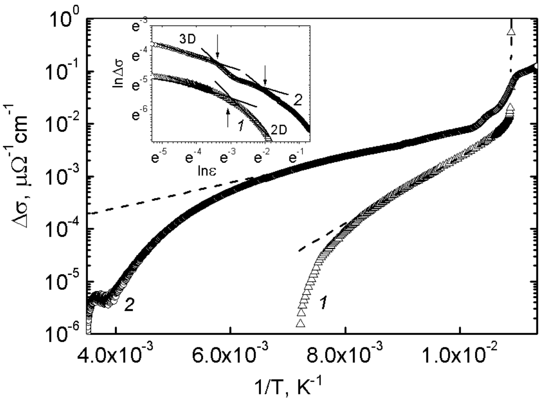

Fig. 2. Temperature dependences of the excess conductivity in the *ab*-plane for $YBa_2Cu_3O_{7-\delta}$ single crystal in lnΔσ — $1/T$ versus lnΔσ – lnε coordinates (inset). Names (notation) of the curves are relevant to Fig. 1. Dashed lines in Fig. 2 show the approximation of equation (2), and in the inset, straight lines — the approximation with slope $tg\alpha_1 \approx -0.5$ (3D regime) and $tg\alpha_2 \approx -1.0$ (2D regime). Arrows show the point of 2D–3D crossover.

Temperature dependence of the excess conductivity can be determined by the following equation:

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

where $\sigma_0 = \rho_0^{-1} = (A + BT)^{-1}$, is conductivity, determined by extrapolating the linear part to zero temperature, and $\sigma = \rho^{-1}$, is the experimentally determined value of conductivity in the normal state. The experimental dependences 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 description of the exponential dependence by the formula:

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

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

As it was observed previously in [20], the approximation of the experimental data can be greatly enhanced by introduction of the factor $(1 - T/T^*)$. In this case, the excess conductivity is proportional to density of the 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 $T_f < T < T^*$, in which the pseudogap state exists, is determined by the rigidity of the order parameter phase that in its turn depends on the oxygen deficiency or the doping element concentration. Specific mechanisms of quasiparticle scattering can play some role. The mechanisms are conditioned by the presence of structural and kinematic anisotropy in the system [21–24]. The value of Δ^*, obtained from Eq. 2 for our experimental samples is shown in Table. It is evident that the long aging leads to significant reduction in the absolute value of the pseudogap $\Delta^*_{K1}/\Delta^*_{K2} \approx 2.07$.

It can be inferred from Fig. 2 that by approaching the T_c the sharp increase in the value of $\Delta\sigma$ takes place. From the theory [25] it is known that near the T_c, the excess conductivity is caused due to fluctuation pairing of the carriers' processes, whose contribution to the conductivity at $T > T_c$ for 2D and 3D cases is determined by the degree dependences determined by the formulas:

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

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

where $\varepsilon = (T - T_c)/T_c$, e is the electron charge, $\xi_c(0)$, is the coherence length along the **c** axis at $T \to 0$ and d, is the characteristic size of a two-dimensional layer. In our case, T_c is defined by the maximum on the $d\rho_{ab}(T)/dT$ dependences in the superconducting transition region (Fig. 1(a)).

The inset in Fig. 2 shows the temperature dependence of $\Delta\sigma(T)$ in $\ln\Delta\sigma$ versus $\ln\varepsilon$ coordinates. Near T_c these relationships are satisfactorily approximated by straight lines with slope $\mathrm{tg}\alpha_1 \approx -0.5$ corresponding to the exponent $-1/2$ in (4), which clearly shows the three-dimensional nature of the fluctuation superconductivity in this temperature interval. At higher temperatures the rate of $\Delta\sigma$ decrease significantly increases ($\mathrm{tg}\alpha_2 \approx -1$), which, in turn, could be seen as an indication of the change in the fluctuation conductivity dimension. It should be also noted that the dependence, received after the prolonged annealing, is characterized by double crossover 2D–3D, corresponding to the various phases of T_c. As follows from (3) and (4), at 2D–3D crossover:

$$\xi_c(0)\varepsilon_0^{-1/2} = d/2. \qquad (5)$$

In this case, determining the value of ε_0 and using the bibliography data regarding dependence of the inter-plane distance from δ [12] ($d \approx 11.7$ Å), we could calculate the value of $\xi_c(0)$. The calculations have shown that in the process of aging the change in the coherence length value from $\xi_c(0) = 1.49$ Å up to $\xi_{c1}(0) = 1.09$ Å and $\xi_{c2}(0) = 2.43$ Å for fazes 1 and 2. Finally, the 2D–3D crossover point is significantly shifted by temperature (Table and inset in Fig. 2).

4. Conclusion

In conclusions the long time exposure of the optimally oxygen doped $YBa_2Cu_3O_{7-\delta}$ single crystals at air leads to partial degradation of the conducting properties and emergence of the effective scattering centers of the carriers. The excess conductivity, $\Delta\sigma(T)$ of $YBa_2Cu_3O_{7-\delta}$ single crystals, in a wide temperature range $T_f < T < T^*$ has exponential temperature dependence and in the case of approximation to T_c, is well described within the Aslamazov-Larkin theoretical model. Prolonged annealing of $YBa_2Cu_3O_{7-\delta}$ single crystals facilitates to great extent of the temperature range of implementation of the pseudogap state in the *ab*-plane, thus narrowing the linear portion of the dependence $\rho_{ab}(T)$. In the aging process it is also showing signs of the phase separation in the experimental samples volume, which are shown in the presence of additional peaks in the curves $d\rho_{ab}(T)/dT$ in the superconducting transition region.

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