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Effect of pressure on paraconductivity in $HoBa_2Cu_3O_{7-\delta}$ single crystals with oxygen deficiency

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In this work, we have investigated the effect of high pressure on conductivity in basal plane of the high temperature super conducting single crystals $HoBa_2Cu_3O_{7-\delta}$ with oxygen deficiency. It has been determined that the excess conductivity $\Delta\sigma(T)$ of $HoBa_2Cu_3O_{7-\delta}$ single crystals in temperature interval near the critical temperature (T_c) is described within the framework of the Aslamazov-Larkin theoretical model. It has been shown that evolution of the transverse coherence length $\xi_c(0)$ in the case of application/removal of the high pressure is largely determined by the "relaxation" pressure effect during prolonged exposure of the sample under load at the room temperature.
Keywords: excess conductivity, hydrostatic pressure, YBaCuO single crystals, high-temperature superconductivity, crossover, pseudogap state.

Исследовано влияние высокого давления на проводимость в базисной плоскости ВТСП-монокристаллов $HoBa_2Cu_3O_{7-\delta}$ с недостатком кислорода. Установлено, что избыточная проводимость $\Delta\sigma(T)$ монокристаллов $HoBa_2Cu_3O_{7-\delta}$ в интервале температур вблизи критической T_c удовлетворительно описывается в рамках теоретической модели Асламазова-Ларкина. Показано, что эволюция поперечной длины когерентности $\xi_c(0)$ в случае приложения-снятия высокого давления в значительной степени определяется "релаксационным" эффектом давления в процессе длительной выдержки образца под нагрузкой при комнатной температуре.

Вплив тиску на парапровідность монокристалів $HoBa_2Cu_3O_{7-\delta}$ з нестачею кисню. *Е.В.Тютерева, Я.В.Довгополова, О.А.Чорновол-Ткаченко, І.Л.Іврій, Р.В.Вовк, С.І.Приходько.*

Досліджено вплив високого тиску на провідність у базисній площині ВТСП-монокристалів $HoBa_2Cu_3O_{7-\delta}$ з нестачею кисню. Встановлено, що надлишкова провідність $\Delta\sigma(T)$ монокристалів $HoBa_2Cu_3O_{7-\delta}$ в інтервалі температур поблизу критичної T_c задовільно описується у рамках теоретичної моделі Асламазова-Ларкіна. Показано, що еволюція поперечної довжини когерентності $\xi_c(0)$ у разі прикладання зняття високого тиску в значній мірі визначається "релаксаційним" ефектом тиску у процесі тривалої витримки зразка під навантаженням при кімнатній температурі.

1. Introduction

The question about influence of dimensionality to formation of the superconducting state of HTSC cuprates is still open [1–5]. In spite of 30 years of intense experimental and theoretical studies, microscopic nature of the high T_c superconductivity is

still unclear [6, 7]. The layered structure of HTSC compounds [8, 9], along with short coherence length [10, 11] and large depth of penetration [12], contributes to emergence of a wide temperature section of excess conductivity [13, 14]. It is established that near the critical temperature T_c, the excess conductivity is conditioned by fluctuation pairing of carriers (fluctuation conductivity — FC) [15], and at the higher temperatures $T > T_c$ by so-called pseudo-gap anomaly (PG) [16, 17]. According to the contemporary concepts [6], these unusual phenomena can serve as a key to understanding the nature of HTSC.

From experimental viewpoint, the most preferable compounds to conduct electrotransport studies is so-called 1-2-3 system $ReBa_2Cu_3O_{7-\delta}$ (Re=Y, or another rare earth ion) [18]. This is mainly due to the fact that composition of these compounds can be relatively easily varied by changing of oxygen content [19, 20] or by substituting elements [21, 22] and, thus, with a controlled manner to change their conductive [23, 24] and critical [25, 26] parameters. Notably, despite of huge number of works devoted to the study of FC [1–5, 27, 28] and PG [16, 17, 29] anomalies in HTSC, many aspects of these physical phenomena remain unclear until now, including the question of impact of structural ordering [30–32] in conducting Cu–O planes, in various mechanisms of conductivity in HTSC.

As is known, the presence of labile oxygen [32,33] in $ReBa_2Cu_3O_{7-\delta}$ (Re=Y, or another rare earth ion) compounds, can contribute to emergence of the non-equilibrium state, which can occur at high pressures [34, 35], "hopping" changes in temperature [36, 37] and long-term storage [38–40] and, in turn, may contribute to the process of phase separation [41, 42], ascending diffusion [43, 44] and emergence of various kinds of superstructures [45, 46]. All these processes have significant impact on the physical properties of HTSC in the normal and in superconducting states. This is pronounced by the oxygen hypostoichiometric composition of the samples [47, 48].

Substitution of yttrium by isovalent rare-earth atoms is important. Particularly, the substitution of yttrium by holmium, having sufficiently high (more than 10 m_B) magnetic moment [49] provides the compound paramagnetism in the normal state. Nevertheless, as in the case of other rare-earth elements, when implementing the substitution of Y by the paramagnetic ions Re = Ho, Dy, the superconducting properties of optimally oxygen doped $ReBa_2Cu_3O_{7-\delta}$ compounds with $\delta \le 0.1$, do not change significantly [49]. Apparently this is due to localization of these ions away from the superconducting planes, which, in turn, prevents the formation of long-range magnetic ordering. At the same time, it is known that in $ReBa_2Cu_3O_{7-\delta}$ the rare earth ion can serve as a sensor sensitive to the local symmetry of its environment and to the charge density distribution, since their change affects to the crystal field, that is forming the electronic structure of such ion [47]. Notably, the yttrium substitution by holmium can increase in intensity of the redistribution processes in the oxygen subsystem [3, 44].

In the absence of microscopic theory of the high-temperature superconductivity, experimental methods reveal the superconducting parameters, which essentially affect the physical characteristics in the normal and superconducting states. One of the most important methods in this aspect is the use of high pressure [50, 51], as it not only allows to clarify the role and influence of the structural features of the system on the superconducting state formation, but also gives the possibility of modeling the conductive characteristics and the critical parameters of the superconductor.

Taking the above under consideration, in the present study we investigated influence of the structural relaxation induced by the high hydrostatic pressure to FC on $HoBa_2Cu_3O_{7-\delta}$ single crystals with an oxygen deficiency $\delta \approx 0.35$ and critical temperature $T_c = 67$ K, with a given topology of plane effects (twin boundaries — TB). The use of single-crystal samples allowed us practically, to eliminate the influence on the conductive properties of these structural factors such as intergranular boundaries, dislocations, phase switching, etc, that are often characteristic for films, ceramic and textured samples. Further, the use of bridges with unidirectional TB with geometry I$\|$TB helped us to minimize the carriers scattering on the TB. Notably, TB are always present in $HoBa_2Cu_3O_{7-\delta}$ compounds under oxygen saturation [52–54] minimizing the elastic energy. Until now, the question regarding the impact of TB on the resistive properties remained open, because of the experimental difficulties encountered in determining their contribution to the electrotransport process.

2. Experimental

$HoBa_2Cu_3O_{7-\delta}$ single crystals were grown by the solution-melt method in a gold crucible, similar to the technology of $YBa_2Cu_3O_{7-\delta}$ single crystal growth [55]. For oxygen saturation of the crystals to the optimum concentrations $\delta \leq 0.15$, the single crystals were annealed in oxygen flow at temperature range of 370–410°C for five days. For resistive measurements, from the same batch of single crystals, we selected a single crystal with dimensions $1.7 \times 1.2 \times 0.2$ mm^3 (the smallest dimension is the direction along c axis). To reduce oxygen content, the samples were annealed for three to five days in the oxygen flow at the higher temperatures. Electrical contacts were made from silver wire and were connected to the crystals with silver paste. The resistivity in ab-plane was measured at constant current up to 10 mA at the two opposite current directions by the standard four-contact method. Hydrostatic pressure was created in a piston-cylinder autonomous chamber. The pressure value was measured by a manganin pressure gauge; the temperature, by a copper-constantan thermocouple mounted in the outer surface of the chamber on the sample position level. To determine the degree of influence of the structural relaxation, measurements were carried out after expiration of a few days after the application-removal pressure, upon completion of the relaxation processes.

3. Results and discussion

Figure 1 shows the temperature dependences of resistivity measured at transport current orientation $\mathbf{I} \perp c$ ($\rho_{ab}(T)$) and at different pressures. The application of pressure leads to decrease of the electrical resistance and to increase in T_c with $dT_c/dP \approx 0.7$ K·kbar^{-1}, which is in qualitative agreement with previous studies [34, 50] obtained for $YBa_2Cu_3O_{7-\delta}$-samples with low oxygen content.

Notably, the decrease in resistivity occurs not only as a result of the hydrostatic pressure, but also during isobaric exposure of the sample at the room temperature immediately after application of the high pressure. For example, in Fig. 1 curves *2* and *3* correspond to the dependences, measured immediately after application of pressure 4.8 kbar and after the isobaric exposure of the sample at the same pressure at the room temperature for a week, respectively. It is determined that such exposure leads to additional decrease of the resistivity by approximately 5 %.

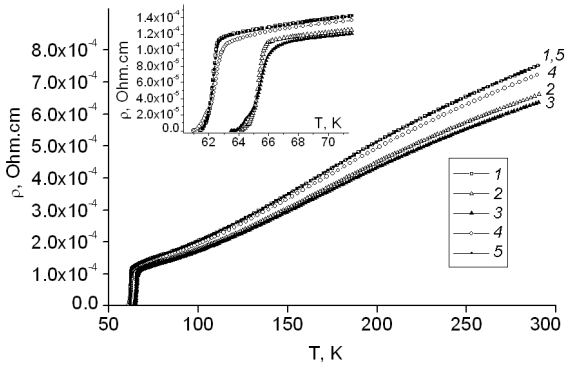

Fig. 1. Temperature dependences of resistivity $\rho_{ab}(T)$ for $HoBa_2Cu_3O_{7-\delta}$ single crystal in the process of application-removing of pressure (see text). The inset shows resistive transition to the superconducting state. Symbols in the inset correspond to the figure.

The qualitatively similar behavior of the curves $\rho_{ab}(T)$ was observed after removal of the high pressure. Thus, in Fig. 1 curves *1* and *4* show the dependences, measured before application and immediately after the pressure removal. Comparison of these curves shows that the results are dependent upon the exposure time of the sample at the room temperature. So, just immediately after the pressure removal electrical resistance of the sample at room temperature reached value of about 4 % lower than the value measured before the pressure application and further, it relaxed for about three days until the equilibrium value. After that, the value ρ_{ab}(290 K) saturates and the dependence $\rho_{ab}(T)$ (curve *5*) is almost identical to the original curve, measured prior to the high-pressure application. This demonstrates the process reversibility.

The pressure application also leads to substantial (up to 15 K) expansion of the linear section of $\rho_{ab}(T)$ dependence at high temperatures. The latter is displayed by reducing the value of so-called pseudogap temperature T^*, at which it begins systematic deviation of the experimental points downwards from the linear dependence. Faster than the linear, is decrease of the $\rho_{ab}(T)$ value, which is observed in the temperature range $T < T^*$, indicating the appearance of so-called excess conductivity $\Delta\sigma$ in the crystal. Temperature dependence of the excess conductivity is usually determined by the equation:

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

where $\sigma_0 = \rho_0^{-1} = (A + BT)^{-1}$, is conductivity determined by extrapolation of the linear section to zero temperature, and $\sigma = \rho^{-1}$ is the experimental value of conductivity in the normal state. According to the contemporary concepts, far away from the T_c appearance of $\Delta\sigma$ in HTSC compounds is often associated with appearance of so-called pseudogap anomaly [16, 17], which is analyzed in more detail in the previous work [56]. As is established [57], near the T_c, the excess conductivity is caused due to the processes of fluctuation pairing of charge carriers and can be described by the Lawrence-Doniach theoretical regime, implying the presence of a very smooth crossover from two-dimensional to three-dimensional regime of the fluctuation conductivity by reduction of the sample temperature:

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

where $\varepsilon = (T - T^{mf}_c)/T^{mf}_c$ is the reduced temperature; T^{mf}_c is the critical temperature in the mean-field approximation; $J = (2\xi_c(0)/d)^2$ is the interplane coupling constant; ξ_c is the coherence length along c axis and d is thickness of the two-dimensional layer. In extreme situations (near T_c, when $\xi_c >> d$, the interaction between the fluctuation Cooper pairs is implemented throughout the entire volume of the superconductor — 3D-regime, or away from T_c, when $\xi_c << d$, interaction is possible only in the plane of the conductive layers — 2D-regime), the expression (2) is converted into the well-known relations for the three- and two-dimensional cases from the theory Aslamazov-Larkin [15]:

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

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

In the case of comparison with the experimental data it is important to accurately define the value of T^{mf}_c, which significantly affects the angle of slope of $\Delta\sigma(\varepsilon)$. Typically, when comparing with the experimental data $\xi_c(0)$, d and T_c in Equations (2–4) are adjustable parameters [2]. However, when employing such a technique,

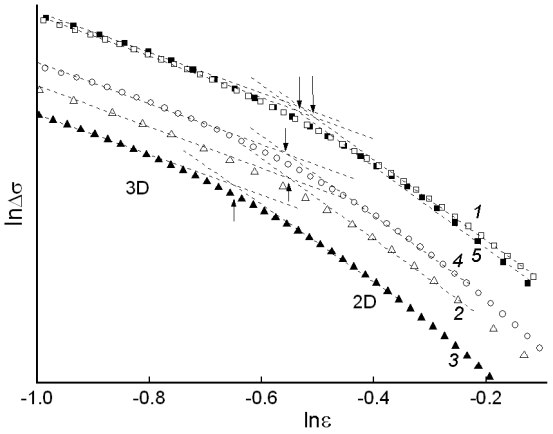

Fig. 2. Temperature dependences $\Delta\sigma(T)$ in $\ln\Delta\sigma(\ln\varepsilon)$ — coordinates for a series of pressures. The curve numbering corresponds to Fig. 1.

usually there are deviations between the theory and experiment. This, in turn, necessitates the use as an additional adjustable parameter the scaling factor, the so-called C-factor, which allows combining the experimental data with the calculated and thus taken into account the possible spreading of inhomogeneity of the transport current for each sample [2]. In our case, for T^{mf}_c it was taken T_c, defined as indicated above, at the point of maximum on $dR_{ab}/dT(T)$ dependences in the regime of the superconducting transition, as suggested in the previous work [10].

Figure 2 shows the temperature dependences $\Delta\sigma(T)$ in $\ln\Delta\sigma(\ln\varepsilon)$ coordinates. It is shown that near T_c, these dependences are satisfactorily approximated with the straight lines with the slope angle $\alpha_1 \approx -0.5$, corresponding to the exponent $-1/2$ in Eq. (4), indicating the three-dimensional nature of the fluctuating superconductivity in this temperature interval. At the higher temperatures the rate of the decrease of $\Delta\sigma$ increases substantially ($\alpha_2 \approx -1$), which can be considered as an indication of the change in the dimension of FC. As it follows from Equations (3) and (4), in the 2D–3D crossover point:

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

In this case, by specifying the value ε_0 and using data from the bibliography on the T_c dependence and interplanar distance from δ [8, 55], we can calculate the value of $\xi_c(0)$. As it is shown in Fig. 3, the value of $\xi_c(0)$, calculated in accordance with Eq. (5),

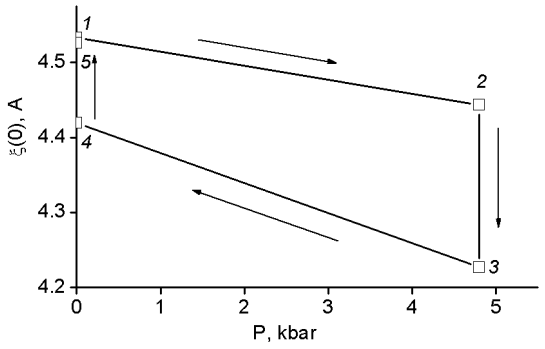

Fig. 3. Pressure dependences $\xi_c(0)$ in process of pressure removal-application.

decreases from 4.53 to 4.44 by the pressure increasing, that is in qualitative agreement with the behavior of the pressure dependences of $\xi_c(0)$ obtained for YBaCuO single crystals with the low oxygen content [58]. It should be noted that the value of the transverse coherence length changes not only in the case of the direct application of high pressure (so-called "true" pressure effect [58, 59]) — section 1–2, but also during the isobaric exposure of the sample at fixed pressure at the room temperature for a week — section 2–3 (the "relaxing" effect [34, 35]). Herewith, in the second case, the relative change of $\xi_c(0)$ is significantly larger (from 4.44 to 4.23). In the case of the pressure reset, the opposite pattern is observed. In the section 3–4, after the direct pressure reset, $\xi_c(0)$ increased from 4.23 to 4.42 , and subsequently, after further exposure of the sample at atmospheric pressure for three days at the room temperature (section 4–5), it recovered to almost the initial value of 4.525 . This asymmetry in the evolution of the value of $\xi_c(0)$ may be due to several factors.

Firstly, as is known from the literature [58], the structural relaxation, observed in the process of isobaric exposure of YBaCuO samples at the room temperature immediately after application-removal of the high pressure, can be accompanied by formation of various types of superstructures, such as the pressure-induced lengthening-shortening of oxygen chains in Cu–O planes or, the formation-decay processes of phase clusters with different degrees of oxygen nonstoichiometry [44, 58] and, accordingly, different superconducting characteristics. Such processes are often asymmetric due to the difference in the diffusion path in direct and reverse processes. Indeed, in order to find a vacant place in the chain, after application of the load to oxygen atom, it requires substantially more time than the decay time of such a chain after the load shedding [58]. As an indirect confirmation of this hypothesis, in [34] phenomena of asymmetry relaxation of the critical temperature T_c and of electrical resistance, the absolute value of which is directly related to the size of such clusters [31, 44].

Secondly, the presence in the system of TB, as it was above mentioned, can also lead to the phase separation in the system, since the TB, as defects of higher dimension, could act as powerful current sources of defects of the lower dimensions, for example, for the same oxygen vacancies [12, 60]. Thus, TB turning out to be the centers of nucleation of the depleted oxygen phase with the reduced critical characteristics [60].

As it was determined previously [42] the high pressure application to HTSC samples with a developed twin structure may facilitate the flow in the system of ascending diffusion processes, when the part of oxygen from phase with the lower T_c migrates into the phase with the higher critical temperature, and by decreasing pressure, occur the opposite redistribution. Indeed, as it is shown in the inset in Fig. 1, the resistive transitions of our sample are characterized by the clearly expressed stepwise form. This, apparently, indicates presence in the sample of at least two phases, with correspondingly, different critical temperature (T_{c1} and T_{c2}) of the transition to the superconducting state [42].

In this Figure, curve *1* corresponds to the dependence measured prior the pressure application; curve *2* was measured immediately after the application of pressure 4.8 kbar, curve *3*, after the sample was retained at the room temperature under a pressure of 4.8 kbar for a week; curve 4 was obtained immediately after the pressure removal and curve *5*, after holding the sample at the atmospheric pressure for three days. It can be deduced from this figure that the sample exposure at the room temperature in the process of the pressure application-removal, in addition to the absolute change in the value of T_c, leads to significant qualitative changes in width and in shape of the superconducting transition. Comparison between curves *2* and *3* shows that after exposure of the sample under the pressure for a week, the superconducting transition is broadened considerably (i.e. the difference between the steps correspond-

ing to the high and low temperature phases ($T_{c1} - T_{c2}$) increases). In the dependence, measured immediately after the pressure removal (curve *4*), changes only the absolute value of T_c, whereas the width of the transition remains practically unchanged. The comparison of the curves *4* and *5* shows that after the sample exposure at the atmospheric pressure for three days at the room temperature, the width and the shape of the transition is almost fully restored to their original values, indicating the process reversibility.

According to the previous work [23], the value of the critical temperature of compound $ReBa_2Cu_3O_{7-\delta}$ (Re=Y is related to the number of holes in CuO_2 plane, through the universal parabolic relationship:

$$T_c = T_c^{\max}\left[1 - 82.6(n - n_{opt})^2\right], \qquad (6)$$

where T_c^{max} is the maximum critical temperature and $n_{opt} = 0.25$ is the optimal content of the holes number on a plane for this compound. The calculations performed based on this relation, indicate that by increasing the pressure during exposure at the room temperature, the number of carriers for the low-temperature phase is reduced by about 3–4 %, while regarding the high temperature phase, the reverse process (i.e. increasing the number of holes) occurs.

Finally, the asymmetry in change of $\xi_c(0)$ in the case of pressure application-removal processes, may be partly due to the fact that the time of pressure rising in our experiment was substantially more than the pressure reset time. This is due to the specific of obtaining the high pressure in multipliers, piston-cylinder type [61], which is usually generated by a slow (about 30 minutes) indentation of the piston in the working channel by press. At the same time, the pressure reset until the atmospheric pressure is happening rather fast, just in a few seconds. Thus, it is logical to assume that under the relatively slow pressure increase, the structure is improved (for example, the concentration of vacancies decreases) and the pressure reset leads to destruction of the order with formation of the excess (non-equilibrium) defects, whose concentration at $P = 0$ relaxes to the initial equilibrium value [42]. Thus, in this case, a certain influence could have the specific mechanisms of quasiparticle scattering [62–64], due to the presence in the system kinetic and structural anisotropy.

Notably, in the case of our sample there is a clear correlation in the behavior of the pressure dependences $\xi_c(P)$ and $T_c(P)$. With the growth of $T_c(P)$, the value $\xi_c(P)$ decreases, and vice versa, which could indicate that the ratio between these values may be governed by the general theory of superconductivity [65], from which it follows that $\xi_0 \sim h\nu_F/k_BT_c$.

It is important to note once again that when the pressure is removed and all the relaxation processes are completed, the dependence $\rho_{ab}(T)$ practically coincides with the original curves, measured prior the high pressure application, which indicates the reversibility of the process. Thus, given some conventionality in the definition of 2D–3D crossover temperature ε_0, we can assume that evolution of the transverse coherence length $\xi_c(0)$ in the case of the high-pressure application-removal to the non-stoichiometric oxygen samples $HoBa_2Cu_3O_{7-\delta}$ is probably related not only to the change of the lattice parameters, the electron-phonon interaction, the connections between layers, etc. (so-called "true" pressure effect), but also, to the large extent, is determined by the "relaxation" pressure effect during the prolonged exposure of the sample under load at the room temperature. That in its turn is caused by the redistribution of the labile oxygen.

4. Conclusions

In conclusion, we briefly sum up the main results obtained in this study. Application of the high pressure to single crystals $HoBa_2Cu_3O_{7-\delta}$ ($\delta \le 0.35$), leads to significant expansion of the interval of the linear dependence $\rho_{ab}(T)$, which in turn, the corresponding narrowing of the temperature section of the pseudogap regime realization. Thus, the excess conductivity in the temperature range near the critical T_c is satisfactorily described within the framework of the Aslamazov-Larkin theoretical model. Under pressure the evolution of the FC regime of non-stoichiometric by oxygen $HoBa_2Cu_3O_{7-\delta}$ samples, is apparently determined by the two processes: a general trimerization of the system, due to changes in the relation between ξ_c and d, on the one hand, and the pressure "relaxation" effect conditioned by the redistribution of the labile oxygen, from the other. Herewith, in contrast to the undoped samples $YBa_2Cu_3O_{7-\delta}$, application of the high pressure leads to substantial increase in the value of the pressure derivatives dT_c/dP and $d\xi_c/dP$.

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