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Phase separation in oxygen deficient $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals: effect of high pressure and twin boundaries

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Abstract

We investigate the influence of high hydrostatic pressure on the electrical resistance in the ab-plane in $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals with oxygen deficiency. It is determined that the high-pressure induced redistribution of the labile oxygen enhances the phase separation, which is accompanied by structural relaxation and ascending diffusion within the volume of the sample. This results in a significant displacement of the temperature intervals that correspond to metal-to-dielectric type transitions and the realization of a pseudo-gap anomaly regime. It is determined that the formation of the low-temperature phase can occur at the twin boundaries.

Key words: *HoBaCuO single crystals, hydrostatic pressure, labile oxygen, pseudo-gap anomaly, metal-to-dielectric transition, twin boundaries.*

Introduction

An important characteristic of high temperature superconducting compounds (HTSC) of the system $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (Re = Y or rare earth ion) is the ability to realize a non-equilibrium state under a specific oxygen non-stoichiometry [1-5]. This can be initiated by external means, such as temperature [1] or high pressure [5]. The non-equilibrium state can be accompanied by a redistribution process of the labile oxygen and structural relaxation, which in turn can affect substantially the electro-transport parameters of the system [1,2,5]. Importantly, the conductivity characteristics and the crucial parameters can be modified by substituting yttrium (Y) with rare-earth elements. When Y is substituted with holmium (Ho) that has a large magnetic momentum (more than $10\mu_B$), $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ is paramagnetic in the normal state [3,6]. Nevertheless, as with other rare earth elements, the substitution of Y with the paramagnetic ions Ho or dysprosium (Dy), the superconducting properties in stoichiometric $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds (with $\delta \leq 0.1$) do not change substantially [6]. This can be explained by the localization of Ho or Dy away from the superconducting planes, which in turn interfere with the formation of distant magnetic order. In oxygen deficient HTSC compounds of the 1-2-3 system the rare earth ion is sensitive to the local symmetry of its environment and to the charge density distribution. Their change affects the crystalline field, forming the electronic structure of this ion [7]. A characteristic peculiarity of the oxygen deficit samples (with $\delta \geq 0.3$) is the broadening of the resistivity transfers under pressure [5,8,9]. The reason of such behavior has not been completely determined. In spite of a number of studies on the relaxation processes in the 1-2-3 system under high pressure, many aspects such as the charge transfer and the nature of the redistribution of the vacancy subsystem are still undetermined. Notably, a substantial part of the experimental material was ceramic and polycrystalline samples with a high content of grain boundaries (intergrain bonds) [4,8,9]. In the case of single crystals the existence of twin boundaries (TB) [5] influences the transport properties in the normal state. This aspect is not yet sufficiently studied, due to experimental difficulties in determining the contribution of these defects. In the present study we investigate the effect of hydrostatic pressure of up to 5 kbar on the electro-transport characteristics and the structural relaxation in the *ab*-plane of oxygen deficient $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals. Two different kinds of transport current geometry were investigated: (a) parallel ($\mathbf{I} \parallel \text{TB}$) when the impact of the TB in the carriers scattering processes is minimized and (b) at an angle of $\alpha = 45^\circ$ between \mathbf{I} and TB.

Experimental techniques

HoBa₂Cu₃O_{7- δ} single crystals were grown from the flux in a gold crucible using similar technology as for the growth of YBa₂Cu₃O_{7- δ} single [1,5]. To obtain samples with oxygen concentrations having $\delta \leq 0.15$, the crystals were annealed in an oxygen flow at a temperature of 370-410°C for five days [2]. This procedure is accompanied by the formation of TB system, which effectively minimizes the elastic energy of the crystal lattice in the tetragonal-orthogonal transition [5]. For the resistance measurements the single crystals were selected from the same batch and had dimensions: 1.7 mm X 1.2 mm X 0.2 mm (sample K1) and 1.9 mm X 1.5 mm X 0.3 mm (sample K2). In these samples the *c*-axis was oriented along the shorter dimension, which had singly-directed TB areas (see Fig. 1) [2,3]. The experimental geometry was selected so that the transport current vector in the *ab*-plane, was either parallel, $I \parallel TB$, (sample S1), or at an angle of $\alpha = 45^\circ$ (sample S2) to the twin boundaries, as it is shown in the insets (a) and (b) of Fig. 2. To produce hypostoichiometric samples, the crystals were annealed in an oxygen flow at higher temperature range for three to five days (see Table1) [2]. Electric contacts were formed with the standard four-contact scheme by applying silver paste onto the crystal surface and the connection of silver conductor. The resistance in the *a-b* plane was measured using the standard method for two opposite directions of a direct current up to 10 mA as it was described in detail previously [1]. As in previous work [2,5], the hydrostatic pressure was applied in an autonomous chamber (piston–cylinder type) and was measured using a manganin manometer. The temperature measurements were performed using a copper-constantan thermocouple mounted at sample level on the outside of the chamber [2]. To determine the influence of the structural relaxation, the measurements were made a few days after the pressure application and removal as it is anticipated that the relaxation processes were then completed [2].

Results and discussion

Fig. 2 (a) and (b) report the temperature dependence of the resistivity in the *ab* plane, $\rho_{ab}(T)$, for the S1 and S2 single crystals measured after the high hydrostatic pressure application and removal process. Part of the curves in this and the following figures is not coerced for clarity. The insets (a) and (b) of Fig. 2 show the resistivity transitions to the superconducting state in $\rho_{ab}(T)$ and $d\rho_{ab}(T)/dT$ coordinates, respectively. The analysis of the experimental curves 1, measured before the application of high pressure, show that with the reduction of the oxygen concentration the critical temperature of the sample decreases. Furthermore, this leads to the expansion of the temperature width of the resistivity transition into the

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superconducting state compared to the initial sample (expansion by more than 10 times from $\Delta T_c \leq 0.3\text{K}$ to $\Delta T_c \approx 3.5\text{K}$). Additionally, the superconducting transition acquires an expressed stepwise form. This indicates that there are at least two phases, having different critical temperatures (T_{c1} and T_{c2}) to the superconducting transition [1,5]. The reduction of the oxygen concentration in both crystals leads in the transition from a quasi-metallic behaviour of $\rho_{ab}(T)$ [1,5] to the dependence with the characteristic thermally activated sagging [2]. This will be discussed in more detail in what follows. The application of pressure, leads to a decrease of resistance and to an increase of T_c (see Table 1), with a rate $dT_c/dP \approx 0.7 \text{ K.kbar}^{-1}$. This value is in consistent with previous studies [5,8,9], concerning oxygen deficient $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ samples. Interestingly, the electrical resistivity is decreasing not only as a consequence of the high pressure, but also in the isobar process of retaining the sample at RT following the application of high pressure [2]. For example, in Fig.2 (a) and (b) curves 2 and 3 show the dependence for the samples S1 and S2 respectively, measured directly after the application of pressure (4.8 kbar), as well as after the isobar process of retaining the sample at room temperature for five days following the application of high pressure. This exposure leads to an additional decrease in electrical resistance from 4-5%. Similar behaviour of $\rho_{ab}(T)$ was also observed after the removal of high pressure. Thus, in Fig. 2 curves 1 and 4 correspond to the dependence, measured before the application and immediately after the removal of pressure. Comparison of these curves indicates that the results are affected on the time that the sample is at room temperature. Immediately after the pressure removal, the value of the electrical resistance of the sample at room temperature, was approximately 4% less than the value measured prior the application of the pressure and consequently relaxed for three days to its equilibrium value. After this, the $\rho_{ab}(290\text{K})$ value saturates and the $\rho_{ab}(T)$ dependence for both crystals almost completely coincides with the original curves, obtained before the prior the application of pressure. This confirms the reversibility of the process.

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As it was mentioned previously, the reduction of the oxygen content, apart from the decrease of the T_c (from 92 to 63 K), leads to the transformation of the $\rho_{ab}(T)$ dependence, which is expressed by a transition from a quasi-metallic behaviour to that with a characteristic thermo activated bending, characteristic of the stoichiometric sample [5].

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Fig. 3 presents these dependencies in $\ln[\rho_{ab}/T] - 1/T$ coordinates. The dependence is determined to be linear in a sufficient wide interval of temperatures ($125\text{K} \leq T \leq 240\text{K}$). This behaviour is described by:

$$\rho = AT \exp(\Delta/T), \quad (1)$$

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where A is a constant and Δ is the activation energy. Under further temperature reduction, we observed a more steep increase in $\rho_{ab}(T)$, which in conjunction to the Mott principles [10],

provides an indication of the realization of an “Anderson” type metal-to-dielectric (MD) transition in the system. The analysis of our experimental data (refer to Fig. 4), in the temperature interval where there is systematic deviation from linearity (in $\ln[\rho_{ab}(T)] - 1/T$ coordinates) indicates that our curves are well described by the means of the asymptotical expression:

$$1/\rho \propto T^{1/3}, \quad (2)$$

Such a behaviour in the $\rho(T)$ dependence, was previously observed in experimental work of the amorphous alloys Gd-Sn [11]. According to Gantmakher *et al.* [11] the dependence of Eq. (2) results from the scaling description in the vicinity of the metal–dielectric transition when the system is in a “critical” regime in which the conductivity has a quantum character [11]. An additional argument in supporting this supposition may be the $\rho_{ab}(T)$ curves cross in the interval of the resistance transition to the superconducting state, that was observed during exposure of the experimental samples at fixed pressure conditions (see inset in Figure 2).

The detailed investigation of these effects needs to be addressed in future work.

It is established that the Anderson transition can also occur in materials that are not amorphous, but which have a degree of disorder [10]. In the compounds of 1-2-3 such disordering may be traced to the labile components [1-5,12]. This is consistent to the presence of residual displacement in the temperature interval of the implementation of the MD-transition. Comparing curves 1 and 2 in Figure 3 (obtained at zero pressure prior the application and immediately after the pressure removal), shows that in curve 2 the temperature that the MD-transition commences is shifted by about 5-6 K compared to curve 1. We should mention again that, after holding the sample at room temperature for three days, the $\rho_{ab}(T)$ dependence is identical with the corresponding dependence measured before the pressure application, indicating the reversibility of the process.

From Fig. 5 (transition to the superconducting phase in $d\rho_{ab}/dT$ - T coordinates), it is determined that the retention of the sample at room temperature in the application-removal of high pressure process has a significant influence on the width and shape of the superconducting transition. A characteristic feature of the influence of retaining the samples at room temperature for 3-5 days, is the significant peak displacement, up to $\Delta T_c \approx 0.5$ - 1.5 K, in the $d\rho_{ab}(T)/dT$ dependence (according to [13] the temperature corresponding to this maximum is T_c), upwards and downwards in temperature, which indicates the change of the transport current flow paths. This in turn can occur in the case of strengthening the phase separation of the non-stoichiometric oxygen samples. The latter requires oxygen transport between phases with different T_c .

In Fig. 5 curve 1 was measured prior to the application of pressure, curve 2 - immediately after the application of pressure 4.8 kbar, curve 3- after retaining the sample at room temperature

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2 under pressure 4.8 kbar for five days, curve 4 is measured immediately after the pressure
3 removal and curve 5 after retaining the sample at room pressure for three days. As it can be
4 observed from the figure, holding the sample at room temperature during the pressure
5 application – removal process, except from the absolute change of the T_c value, leads to
6 significant qualitative changes of the width and the form of the transition to the superconducting
7 state. A comparison of 2 and 3 demonstrates that after retaining the sample under pressure for 7
8 days, the absolute value $d\rho_{ab}(T)/dT$ is reduced by about 1.5 times. Additionally, there is a
9 significant spreading in the transfer (refer to [2] for the case of YBCO). The dependence
10 measured directly after the pressure removal (Fig. 5, curve 4) there is change only in the absolute
11 value of T_c , while the transfer width and value $d\rho_{ab}(T)/dT$ both remain practically unaltered. The
12 comparison between curves 4 and 5, demonstrates that after the sample experienced atmospheric
13 pressure for 3 days at room temperature, the width and the transfer regained their initial values.
14 Therefore, when the pressure increases a part of the oxygen of the lower T_c phase, migrates to a
15 higher critical temperature phase and when the pressure is reduced, the reverse process takes
16 place. This effect can be possible in the case of the realisation of an ascending diffusion process
17 [2,14], in which there is an inhomogeneous field of mechanical stresses (eg. due to external
18 compression). In a compressive environment large impurity atoms (with respect to the lattice
19 atoms) will experience larger force compared to the lattice atoms. Atoms with large ionic radius
20 are likely to diffuse to tensile stress regions, whereas atoms with smaller volume diffuse in the
21 region of compression. These concepts of ascending diffusion may also govern point defects [14]
22 as in the present work oxygen vacancies. The reversibility of this effect is also an important
23 feature. For example, when removing the external stress the reverse ordering of the
24 concentration of the point defects in the sample will occur.

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43 Fig. 5 (a) and (b) show the temporary dependence of the isobar relaxation $T_c(t)$ of the
44 critical temperature for high T_c and low T_c phases (dark and light symbols respectively) for both
45 samples, obtained during the step by step annealing at room temperature after the application of
46 pressure of 4.8 kbar. The solid lines are presented the results by the expression proposed by
47 Jorgencen *et al.* [15]:

$$48 \quad T_c(t) = T_c(\infty) + [T_c(0) - T_c(\infty)] \exp[-(t/\tau)^{1/2}], \quad (3)$$

49 where $T_c(\infty)$ and $T_c(0)$ are the equilibrium and the initial value of the critical temperature,
50 respectively, and τ is a characteristic time for the relaxation process [1].

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Calculations using Eq. (3) demonstrate that the equilibrium value $T_c(\infty)$ in gradual
annealing is accelerated for the low temperature phases in comparison to the high temperature
phases: $\tau(T_{c1})/\tau(T_{c2}) \approx 1.32$ [1]. Apparently, this phenomenon is due to a difference of the
diffusion path during the high-pressure-induced redistribution of labile components. It appears

that, for the same reason, the reverse process (alignment of the labile oxygen's concentrations) is occurring, after the pressure removal (refer to Table1). In the second case, the oxygen atoms have to overcome smaller distances to leave the high-temperature (oxygen richer) phase than in the first case when the atoms of the labile oxygen have to overcome greater distances in order to find a vacancy in the low-temperature (oxygen depleted) phase.

Using the values of τ , obtained by Eq. (3) using our experimental data, we can determine the activation energy of the relaxation process in our samples [10] using the Arrhenius law:

$$\tau = \tau_0 \exp\left(\frac{E_A}{k_B T}\right), \quad (4)$$

where $\tau_0 = 1.4 \cdot 10^{-12}$ s is the characteristic period [4], which, according to [12], is independent of the pressure. The activation energy value obtained from our data $E_A \approx 0.94$ eV is slightly lower than the typical values for the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds with reduced oxygen concentration [16]. It should also be noted that all the characteristic shape changes in the temperature dependence and absolute values of the resistivity parameters that were observed in the isobaric annealing process at room temperatures, in $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ were more pronounced compared to $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. In $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ the Ho ion (which has larger ionic radius than Y) plays an important role in the structural order of the system affecting the oxygen ions interactions in the CuO planes. Previous studies confirmed that when we substitute Y to rare-earth elements with larger ions significant qualitative changes are evident regarding the $T_c(\delta)$ dependence in $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [7]. Herewith, the characteristic for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ dependence $T_c(\delta)$ with two plateaus at 60 K and 90 K degenerates in a more sharp monotonic dependence. Additionally, the Ortho-II structure is not realized at all [7]. It can be assumed that oxygen hypostoichiometric $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds have a more extended disordered oxygen hyperstructure than the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds.

In previous works [2,17,18], the value of critical temperature in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ was correlated with the number of holes in the CuO_2 layers *via*:

$$T_c = T_c^{\max} \left[1 - 82.6(n - n_{\text{opt}})^2\right], \quad (5)$$

where T_c^{\max} is the maximum critical temperature and $n_{\text{opt}} = 0.16$ is the optimal number of holes in the surface (layer) for this compound. According to Eq. 4 when we increase the pressure at room temperature, the number of carriers for the low-temperature phase is reduced by about 2-3%, whereas at the same time in the high-temperature phase the reverse process is taking place as the number of holes increases. This is in agreement with the concept that the application of high pressure leads to a redistribution (by diffusion) of the labile components from the phase of lower T_c to HTSC phase

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2 [2,5]. It is an important confirmation of the interrelation between structural and electro-transport
3 anomalies in the system.
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5 In our case, one of the possible reasons of the phase separation could be the origination of
6 the low T_c in the TB boundaries. Indeed, experiments on the vortex structure [19,20] have shown
7 that the density of vortices in TB increases compared with their density in the volume of the
8 superconductor, indicating the suppression of T_c in the TB. This can be due to the low oxygen
9 concentration in the TB plane, which effectively implies the high concentration of oxygen
10 vacancies [21], as a consequence of mechanical tensions. Vacancies create strong attractive
11 fields (i.e, repulsion field for the oxygen atoms). This assumption can be justified by the different
12 forms of superconducting transitions obtained with the different experimental geometries (here
13 $\mathbf{I} \parallel \text{TB}$ and an angle between \mathbf{I} and the TB of 45°). Figure 5 shows that for $\mathbf{I} \parallel \text{TB}$ the height of the
14 peak of the $d\rho_{ab}(T)/dT$ dependency of the low-temperature phase is almost 3 times lower than the
15 height of the peak corresponding to the high-temperature phase. Together with this, the more
16 pronounced maximum, corresponding to high-temperature phase. In the second experiment
17 geometry (angle between \mathbf{I} and the TB of 45°) we observed an inverse $d\rho_{ab}(T)/dT$ dependence
18 with a more pronounced maximum, corresponding to low-temperature phase. Since in the first
19 case, the TB is parallel to the transport current there is a high probability of percolation paths
20 mediating the current flow in the high-temperature phase. When the TB is oriented at an angle
21 of 45° relative to the transport current vector (measurement of electrical resistance of the single
22 crystal K2), the percolation paths of current flow in the high-temperature superconducting phase
23 are missing. Therefore, the intensity of current carriers scattering should be minimal when the
24 experiment geometry is $\mathbf{I} \parallel \text{TB}$, which is reflected in the transformation of the shape in the
25 corresponding $d\rho_{ab}(T)/dT$ dependence. The latter proposition is indirectly confirmed by the
26 difference of the absolute value of resistivity at room temperature, which is less than 7% in the
27 case of the experiment geometry $\mathbf{I} \parallel \text{TB}$, in comparison with the case of the experiment geometry
28 when the angle between \mathbf{I} and TB is 45° . Here, however, it should be noted that an additional
29 possible reason regarding the differences in the behaviour of the temperature dependencies of the
30 resistance under different orientations of the transport current in relative to TB planes could
31 serve the anisotropy of the resistance parameters in the nodal and anti-nodal directions.
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53 It should be noted that the final conclusion about the nature of the impact of TB on the
54 phase separation in $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals, requires additional experimental studies. A
55 future direction should be the investigation of superconducting transitions in untwining and
56 twinning single crystals with a specific distribution of TB planes. In one segment of the crystal
57 the transport current vector has to be oriented in parallel, and in another perpendicular to the TB
58 plane. In the latter case, $\mathbf{I} \perp \text{TB}$, the transport current will cross over the TB plane in one of the
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2 segments of the single crystal. As it can flow within the volume of the superconductor it passes
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4 by the TB plane in the other segment of the crystal (when $\mathbf{I} \parallel \text{TB}$). The measurements in the
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6 untwined crystals provide data regarding the existence (or lack) of the step-like form of the
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8 transition to the superconducting phase, as well as the difference ($T_{c1}-T_{c2}$) in the absence of
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10 plane defects in the sample.

11 12 13 **Conclusions**

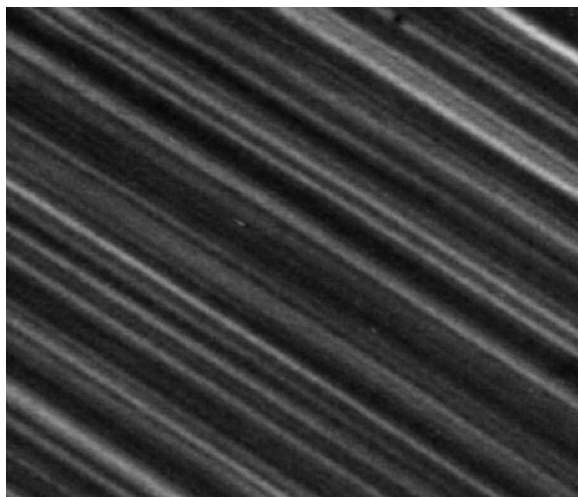
14 To conclude, twin boundaries are effective scattering centres of normal carriers in $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$
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16 compounds. Reducing the oxygen stoichiometry of $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals, results to an
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18 uneven distribution of oxygen in the volume of the crystal and the formation of phases with
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20 different critical temperatures. Herewith, the substitution of yttrium with holmium significantly
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22 affects the charge distribution and the effective interaction in the CuO-planes, thereby
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24 stimulating the disordering in the oxygen subsystem. Induced by high pressure the redistribution
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26 of labile oxygen is enhances the phase separation in the volume of oxygen deficient
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28 $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals. Additionally, it stimulates ascending diffusion processes between
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30 the superconducting phases with different oxygen stoichiometry. This results in a significant
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32 displacement of the temperature intervals that correspond to MD type transitions.
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Table 1. Sample data and material parameters under different pressures

Sample	S1 ($I_{ab} TB$)					S2 ($\alpha_{I,TB} = 45^\circ$)				
	P (kbar (under RT))	ρ_{300} ($\mu\text{Ohm}\cdot\text{cm}$)	T_{c1} (K)	T_{c2} (K)	τ_1 (hr)	τ_2 (hr)	ρ_{300} $\mu\text{Ohm}\cdot\text{cm}$	T_{c1} (K)	T_{c2} (K)	τ_1 (hr)
0	752.8	62.28	-	-	-	807.5	62.56	61.88	-	-
4.8 (immediately after the application of pressure)	661.4	65.35	64.86	11.03	8.4	714	65.7	64.7	10.55	8
4.8 (five days after the application of pressure)	637.1	65.56	64.34	-	-	682.9	66.02	63.67	-	-
0 (immediately after pressure removal)	721.9	62.49	61.67	7.05	5.9	780.4	62.9	60.89	6.8	5.3
0 (three days after pressure removal)	751	62.33	-	-	-	806.2	62.53	61.82	-	-



H
1 μm

Fig.1. Photograph of the singly –oriented TB areas, in polarized light.

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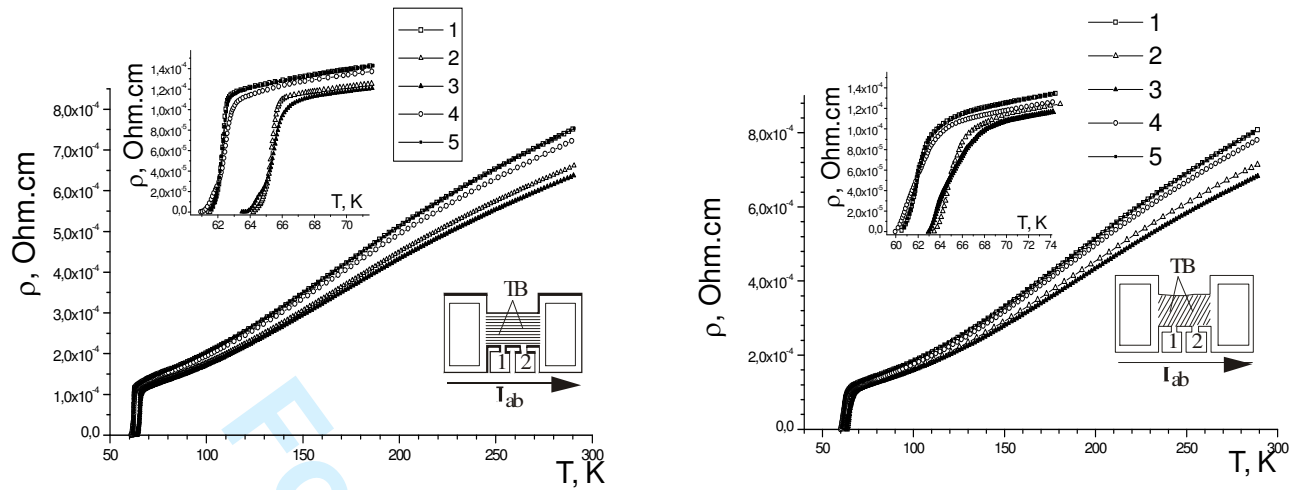


Fig. 2 (a) and (b.) Temperature dependence of the resistivity $\rho_{ab}(T)$ for samples at different pressures. Curve 1 was obtained prior the application of pressure; curve 2 – was obtained immediately after the application of pressure 4.8 kbar; curve 3 – was measured after keeping the sample in room temperature under pressure 4.8 kbar within a week; curve 4 – was obtained immediately after the removal of pressure and curve 5 – was measured immediately after keeping the sample for three days under zero pressure. The insets of Fig. 2 show the geometry of the experiment and the resistivity transitions to the superconducting phase.

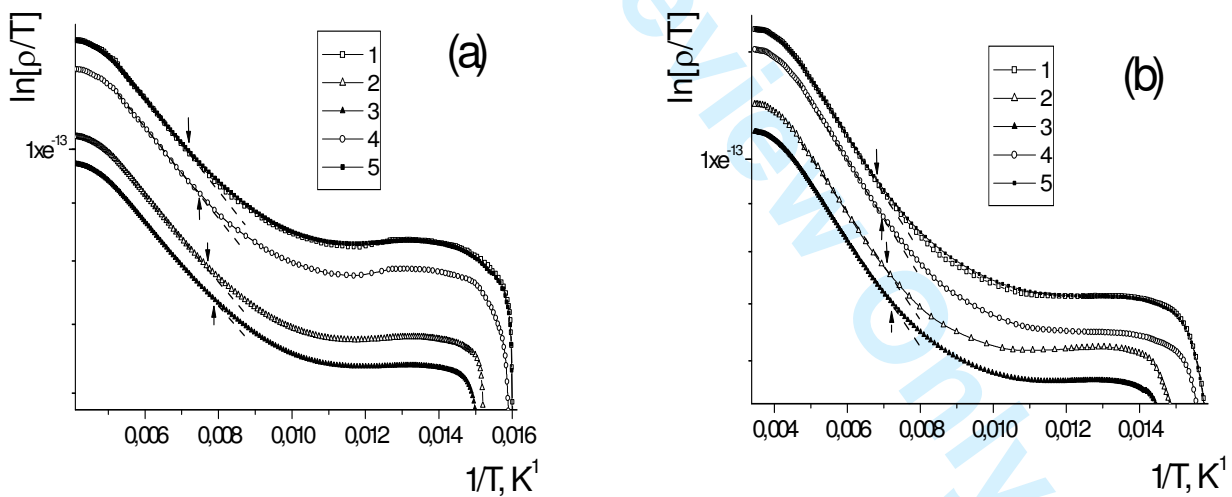


Fig. 3. The $\ln(\rho_{ab}/T)$ versus $1/T$ dependence in different pressures. The numbering of the curves corresponds to the numbers in Fig. 2. Arrows show the points that the systematic deviation from linearity commences.

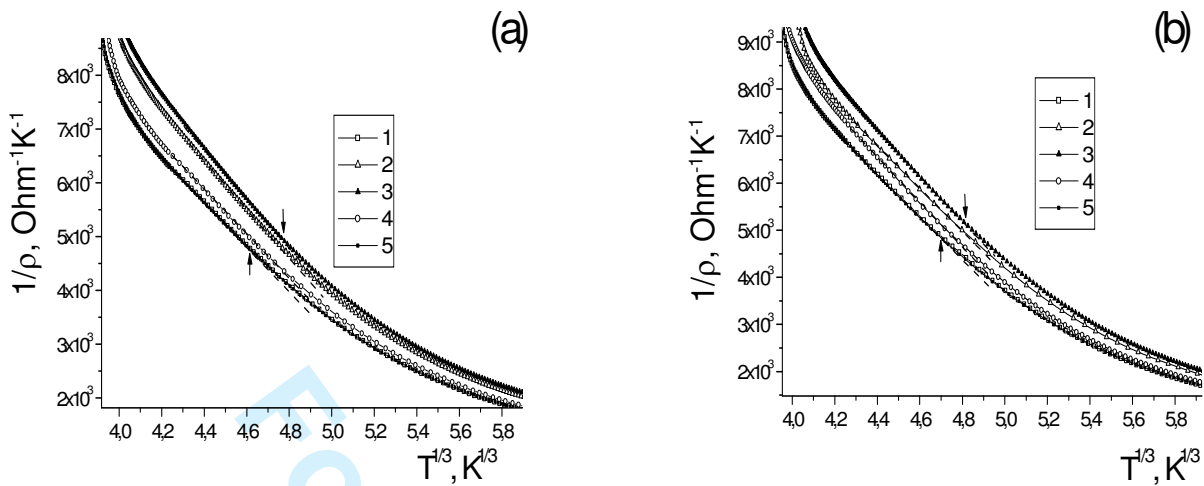


Fig. 4. Plot of $1/\rho_{ab} - T^{1/3}$ in different pressure. The numbering of the curves corresponds to the numbers in Fig. 2. Arrows show the points that the systematic deviation from linearity commences. .

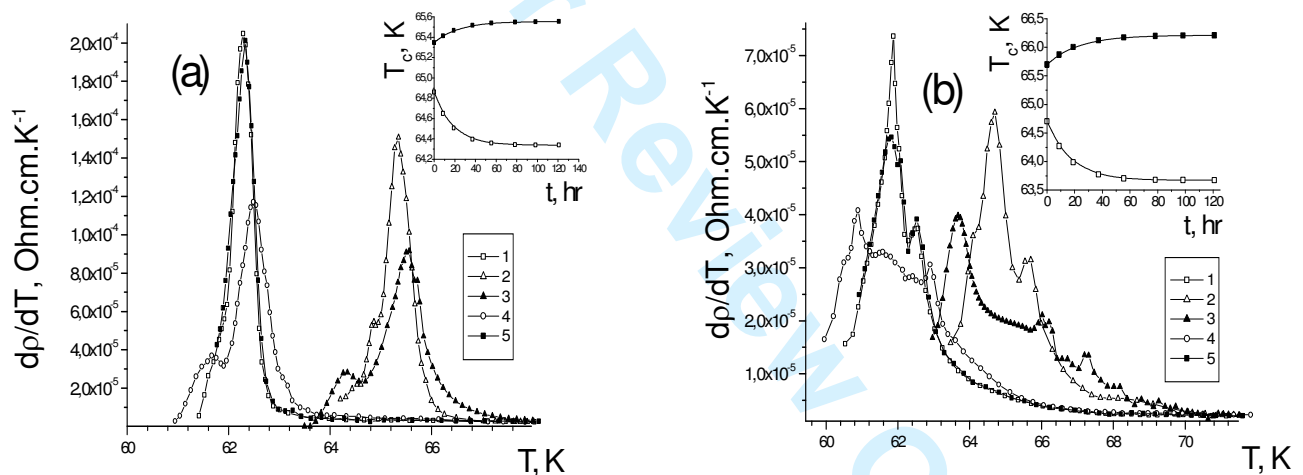


Fig. 5. The temperature dependence $d\rho_{ab}(T)/dT$ in the interval of the transition to the superconducting phase. The numbering of the curves corresponds to the numbers in Fig. 2. The insets (a) and (b) in show the temporary dependence of the isobar relaxation $T_c(t)$ of the critical temperature for high T_c and low T_c phases (dark and light symbols respectively) for both samples, obtained during the step by step annealing at room temperature after the application of pressure 4.8 kbar.