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## EFFECT OF HIGH-PRESSURE-INDUCED STRUCTURAL RELAXATION ON EVOLUTION OF THE TEMPERATURE DEPENDENCE PSEUDOGAP IN $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$ SINGLE CRYSTALS

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In present work the influence of high pressure on the conductivity in the basal plane of the oxygen underdoped  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals is investigated. It is found that excess conductivity  $\Delta\sigma(T)$  in the  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals over a wide temperature range ( $T_c < T < T^*$ , where  $T_c$  is the critical temperature and  $T^*$  represents the mean field temperature of the superconducting transition) obeys an exponential temperature dependence. The description of the excess conductivity with  $\Delta\sigma \sim (1-T/T^*)\exp(\Delta^*_{ab}/T)$  can be interpreted in terms of mean field theory. The temperature dependence pseudogap is satisfactorily described within the framework of the BCS-BEC crossover.

**KEY WORDS:** excess conductivity, hydrostatic pressure,  $\text{HoBaCuO}$  single crystals, high-temperature superconductivity, crossover, pseudogap state.

### ВПЛИВ ІНДУКОВАНОЇ ВИСОКИМ ТИСКОМ СТРУКТУРНОЇ РЕЛАКСАЦІЇ НА ЕВОЛЮЦІЮ ТЕМПЕРАТУРНОЇ ЗАЛЕЖНОСТІ ПСЕВДОЩІЛИНИ МОНОКРИСТАЛІВ $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$

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В роботі досліджено вплив високого тиску на провідність в базисній площині ВТНП-монокристалів  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$  з нестачею кисню. Встановлено, що надлишкова провідність  $\Delta\sigma(T)$  монокристалів  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$  у широкому інтервалі температур  $T_c < T < T^*$  підкоряється експоненційній температурній залежності. При цьому описання надлишкової провідності за допомогою співвідношення  $\Delta\sigma \sim (1-T/T^*)\exp(\Delta^*_{ab}/T)$  може бути інтерпретовано в термінах теорії середнього поля, де  $T^*$  представлена, як середньопольова температура надпровідного переходу, а температурна залежність псевдощілини задовільно описується в рамках теорії кросовера БКШ-БЕК.

**КЛЮЧОВІ СЛОВА:** надлишкова провідність, гідростатичний тиск, монокристали  $\text{HoBaCuO}$ , високотемпературна надпровідність, кросовер, псевдощілинний стан.

### ВЛИЯНИЕ ИНДУЦИРОВАННОЙ ВЫСОКИМ ДАВЛЕНИЕМ СТРУКТУРНОЙ РЕЛАКСАЦИИ НА ЭВОЛЮЦИЮ ТЕМПЕРАТУРНОЙ ЗАВИСИМОСТИ ПСЕВДОЩЕЛИ МОНОКРИСТАЛЛОВ $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$

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В работе исследовано влияние высокого давления на проводимость в базисной плоскости ВТНП-монокристаллов  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$  с недостатком кислорода. Установлено, что избыточная проводимость  $\Delta\sigma(T)$  монокристаллов  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$  в широком интервале температур  $T_c < T < T^*$  подчиняется экспоненциальной температурной зависимости. При этом описание избыточной проводимости с помощью соотношения  $\Delta\sigma \sim (1-T/T^*)\exp(\Delta^*_{ab}/T)$  может быть интерпретировано в терминах теории среднего поля, где  $T^*$  представлена, как среднеполевая температура сверхпроводящего перехода, а температурная зависимость псевдощели удовлетворительно описывается в рамках теории кросовера БКШ-БЭК.

**КЛЮЧЕВЫЕ СЛОВА:** избыточная проводимость, гидростатическое давление, монокристаллы  $\text{HoBaCuO}$ , высокотемпературная сверхпроводимость, кросовер, псевдощелевое состояние.

Study of pseudogap anomalies (PG) - a significant decrease in the density of electronic states observed in the phase diagram of underdoped compounds, continues to be one of the most important areas of physics of high temperature superconductivity (HTSC). However, despite the large accumulated literary material, still remain unclear as the nature of origin of PG and questions about its role in the formation of the superconducting state in HTSC. Currently in the literature are discussed extensively two basic scenarios of pseudogap anomalies in HTSC systems. According to the first occurrence of PG associated with short-range order fluctuations of "dielectric" type, for example, antiferromagnetic fluctuations, waves of charge and spin density, etc. [1]. The second scenario allows for the formation of Cooper pairs at temperatures significantly above the critical value  $T^* \gg T$ , followed by the establishment of phase coherence at  $T < T_c$  [2,3].

Among the theoretical studies, which supporting the second view, we should note the theory of the crossover from the BCS mechanism to the mechanism of the Bose-Einstein condensation (BEC) [3], which were obtained by the temperature dependence of the pseudogap in the case of weak and strong coupling. In general, these relationships are described by the equation:

$$\Delta(T) = \Delta(0) - \Delta(0) \sqrt{\frac{\pi}{2}} \sqrt{\frac{T}{\Delta(0)}} \exp\left[-\frac{\Delta(0)}{T}\right] \left[1 + \operatorname{erf}\left(\sqrt{\frac{\sqrt{x_0^2 + 1} - 1}{T/\Delta(0)}}\right)\right], \quad (1)$$

where  $x_0 = \mu/\Delta(0)$  ( $\mu$  is the chemical potential of the carrier system and  $\Delta(0)$  is the value of energy gap at the zero temperature and  $\operatorname{erf}(x)$  is the error function. In the case that of  $x_0 \rightarrow \infty$  (weak coupling), Eq. (1) becomes:

$$\Delta(T) = \Delta(0) - \Delta(0) \sqrt{2\pi\Delta(0)T} \exp\left[-\frac{\Delta(0)}{T}\right], \quad (2)$$

that is well known in the BCS theory. In the limit of the strong interactions in the three-dimensional case ( $x_0 < -1$ ), Eq. (1) becomes:

$$\Delta(T) = \Delta(0) - \frac{8}{\sqrt{\pi}} \sqrt{-x_0} \left(\frac{\Delta(0)}{T}\right)^{3/2} \exp\left[-\frac{\sqrt{\mu^2 + \Delta^2(0)}}{T}\right]. \quad (3)$$

At the same time, as was shown in [2], at a sufficiently high accuracy values  $\Delta(T)$  of the pseudogap in a wide temperature range can be determined from the dependency  $\rho_{ab}(T)$  (resistivity in the basal plane) at temperatures below a characteristic value  $T^*$  (temperature the opening of the pseudogap).

The most promising for research on this issue is the connection  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$  ( $\text{Re} = \text{Y}$  or other rare-earth ion), which is associated with a wide range of variation of their conductive properties and critical parameters by varying the degree of oxygen nonstoichiometry [2] or replacement of components of the component [4]. An important feature of HTSC compounds  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is the ability to implement them in a nonequilibrium state with a certain degree of oxygen deficiency [5,6], which can be induced by external influences, such as temperature [5] or high pressure [6]. This condition is accompanied by processes of redistribution of labile oxygen and structural relaxation, which in turn has a significant impact on electrotransport parameters of the system [5,6]. An important role is played by the replacement of yttrium, rare earth isoelectronic analogamiami it. Of particular interest in this aspect, is replacing yttrium by holmium, which has a large (more than 10  $\mu_B$ ) magnetic moment [7], which provides connections paramagnetism in its normal state. However, as in the case of other rare earth elements in the implementation of the replacement of Y on paramagnetic ions  $\text{Re} = \text{Ho}, \text{Dy}$  doped superconducting properties of oxygen compounds  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$  with  $\delta \leq 0.1$ , do not change significantly [7]. Apparently this is due to the localization of these ions away from the superconducting planes, which, in turn, prevents the formation of long-range magnetic order. At the same time, we know that in samples of HTSC 1-2-3 system, the oxygen non-stoichiometric composition, rare-earth ion can serve as a sensor which is sensitive to local symmetry of its environment and the distribution of charge density, since they change affects the crystal field forming the electronic structure of the ion [5].

In the absence of a microscopic theory of high temperature superconductivity, of particular importance were the experimental methods to identify the parameters of superconductors, which greatly affect the whole of their physical characteristics in the normal and superconducting state. One of the most important methods in this respect is the use of high pressure [2,6,8]. Analysis of a very extensive study of literary material for PG in the 1-2-3 system shows that to date there are enough small number of papers in which the effect of pressure on the PG mainly in samples close to the stoichiometric composition [8]. However, with this in mind, the most informative is the study of this sample with low oxygen content, because it not only helps to clarify the role and influence of the structural features of the formation of super state, but also allows us to model the leading characteristics and the critical parameters of the superconductor.

The goal of this paper are investigation of the influence of high hydrostatic pressure up to 11 kbar on the different types of conductivity in  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals with oxygen deficient  $\delta < 0.35$  and the critical temperature  $T_c \approx 67$  K.

### EXPERIMENTAL TECHNIQUES

$\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals were grown from the flux in a gold crucible using similar technology as for the growth of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single [2,6]. 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. For the resistance measurements the single crystal were selected and had dimensions: 1.7 mm X 1.2 mm X 0.2 mm. In these sample the  $c$ -axis was oriented along the smallest dimension. To reduce the oxygen concentration, the crystals were annealed in an oxygen flow at higher temperature range for three to five days. 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  $ab$  plane was measured using the standard method for two opposite directions of a direct current up to 10 mA. The hydrostatic pressure was produced in an autonomous chamber of the piston–cylinder type [2,6] and was measured using a manganin manometer. The temperature measurements were performed using a copper-constantan thermocouple which was mounted at sample level on the outside surface of the chamber. To determine the influence of the structural relaxation, the measurements were made a few days after the pressure application and removal, when the relaxation processes were completed.

## RESULTS AND DISCUSSION

Fig. 1 presented the temperature dependence of resistivity measured at the orientation of the transport current  $\perp$  (  $\rho_{ab}(T)$ ) and different pressures. The application of pressure leads to a decrease in resistivity and an increase in  $T$  at  $dT/dP \approx 0.7$  K-kbar $^{-1}$ , in qualitative agreement with published data [2,6], obtained for samples of  $YBa_2Cu_3O_{7-\delta}$  with reduced oxygen content. It is important to note that the decrease in resistivity is not only a result of exposure to hydrostatic pressure, but also in the process of isobaric holding the sample at room temperature immediately after the application of high pressure. For example, in Fig. 1 curves 2 and 3 correspond to dependence, measured immediately after the application of pressure at 4.8 kbar, and also after isobaric sample extracts with the same pressure at room temperature for five days, respectively. It is evident that such exposure leads to an additional decrease in resistivity of about 5%.

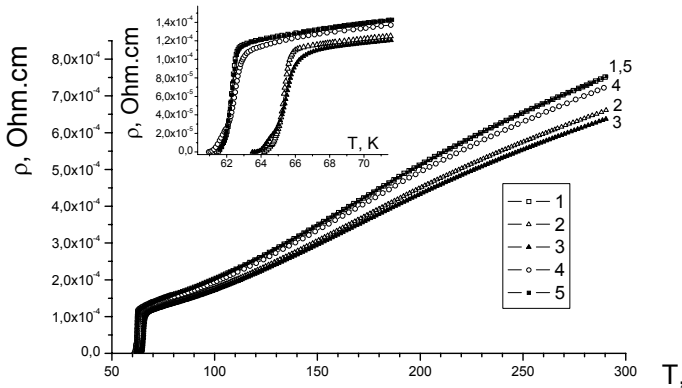


Fig. 1. Temperature dependence of the resistivity  $\rho_{ab}(T)$  for  $HoBa_2Cu_3O_{7-\delta}$  sample at different pressures.

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 show the resistivity transitions to the superconducting phase.

the systematic deviation of experimental points begins downwards from the linear dependence. According to previous work the  $T^*$  corresponds to the initiation temperature of the PG [1-6]. This will be discussed in more details in what follows.

The abrupt decrease of the  $\rho_{ab}(T)$  value, which is observed at temperatures  $T < T^*$ , indicates the appearance of the so-called excess conductivity  $\Delta\sigma$  in the crystal. The temperature dependence of excess conductivity is usually determined from the equation:

$$\Delta\sigma = \sigma - \sigma_0, \quad (4)$$

where  $\sigma_0 = \rho_0^{-1} = (A+BT)^{-1}$  is the conductivity obtained from the extrapolation of the linear part to the zero temperature, and  $\sigma = \rho^{-1}$  is the experimental value of the conductivity in the normal state. Analysis has shown that the experimental dependence  $\Delta\sigma(T)$  in the coordinates  $\ln\Delta\sigma - 1/T$  are straight lines in a wide temperature range, which corresponds to the description of the exponential dependence of the form:

$$\Delta\sigma \sim \exp(\Delta^*_{ab}/T), \quad (5)$$

where  $\Delta^*_{ab}$  the value that defines a thermal activated process across the energy gap - the pseudo-gap.

The exponential dependence of  $\Delta\sigma(T)$  was also observed earlier, in YBCO samples [2]. In [2] was demonstrated that the description of experimental data can be significantly expanded by introducing the factor  $(1-T/T^*)$ . The excess conductivity becomes proportional to the superconducting carrier density  $n_s \sim (1-T/T^*)$  and inversely proportional to the pairs  $\sim \exp(\Delta^*/kT)$ , that are destroyed by thermal motion:

$$\Delta\sigma \sim (1-T/T^*) \exp(\Delta^*_{ab}/T), \quad (6)$$

where the temperature interval  $T_c < T < T^*$ , in which the PG regime exists is defined by the phase of the order parameter that depends on either the oxygen hypostoichiometry or the dopant concentration. Therefore, by using the method [8] we can construct on the experimental curve  $\ln\Delta\sigma$  for the temperature dependence  $\Delta^*_{ab}(T)$  until the  $T^*$ .

Fig. 2 shows the temperature dependence of the PG in  $\Delta^*(T)/\Delta^*_{max}$  versus  $T/T^*$  coordinates, where  $\Delta^*_{max}$  is the  $\Delta^*$  value in the plateau, away from  $T^*$ , measured under different pressures. The  $\Delta^*(T)/\Delta^*(0)$  versus  $T/T^*$  dependence is calculated using Eqs (2), (3) in the mean-field approximation within the context of the BCS-BEC crossover theory [3].

Qualitatively similar behavior of curves  $\rho_{ab}(T)$  was observed after removal of high pressure. Thus, in the same figure curves 1 and 4 correspond to dependence, measured before application and immediately after the removal of pressure. Comparison of these curves shows that the measurement results depend significantly on the time of exposure of the sample at room temperature. Thus, immediately after the removal of pressure, the value of the resistivity of the sample at room temperature, the value was approximately 4% lower than measured before application of pressure to continue to relax for about three days to an equilibrium value. After that, the value  $\rho_{ab}(290K)$ , saturates, and depending  $\rho_{ab}(T)$  is almost completely identical (curve 5) to the original curves (curve 1), measured before the application of high pressure. This indicates that the reversibility of the process.

The application of pressure, also leads to a significant (up to 15 K) expansion of the linear interval of the  $\rho_{ab}(T)$  dependence in the region of the higher temperatures. The last one appears to reduce the value of the temperature  $T^*$ , at which

The values of the crossover parameters  $\mu/\Delta(0) = 10$  (limit of the BCS) and  $\mu/\Delta(0) = -2, -5, -10$  (limit of the BEC) are shown in Fig. 2 as the dotted lines. From this figure it is observed that by increasing the applied pressure there is a shifting of the experimental curves from Eq. (3) (strong coupling) to Eq. (2) (weak coupling). This behaviour is qualitatively similar to the effect of transformation temperature dependence of the pseudogap samples YBCO, observed during the reduction of oxygen non-stoichiometry [2,6]. It is obvious that these correlations in the behaviour of the curves  $\Delta^*(T)$  are not arbitrary. Indeed, as is well known from the literature (see eg. [6]), the application of high pressure to the samples of HTSC 1-2-3 system, as well as decreasing the oxygen content leads to improved conductive properties, which include the increases in the critical temperature and significant reductions of the electrical resistivity.

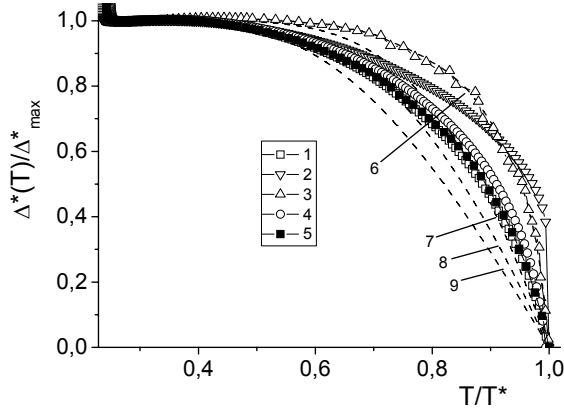


Fig. 2. The temperature dependence of the PG for the  $\text{YjBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystal, in  $\Delta^*(T)/\Delta_{\max}$  versus  $T/T^*$  coordinates, where  $\Delta_{\max}$  is the  $\Delta^*$  value in the plateau, away from  $T^*$ .

The numbering of the curves corresponds to the numbers in the Fig. 1. Dotted lines for the  $\Delta^*(T)/\Delta(0)$  versus  $T/T^*$  dependence were calculated according Eq. (3) for the values of the crossover parameters  $\mu/\Delta(0)=10$  (limit of the BCS, curve 6) and  $\mu/\Delta(0) = -2, -5, -10$  (limit of the BEC, curves 7-9).

behavior, the agreement between experiment and theory in this case can be considered quite satisfactory.

It should be noted that the structural relaxation is observed in the isobaric holding the sample at room temperature immediately before and after the application of high pressure has a significant influence on the qualitative behavior of the dependences  $\Delta^*(T)$ . This shows a comparison of curves 2 and 3 in Fig. 2, corresponding to the dependence of the measured immediately after the application of pressure at 4.8 kbar, and also after isobaric sample extracts with the same pressure at room temperature for five days, respectively. It is evident that the direct application of pressure leads only to a shift from dependence on the experimental curve of the form (3) (2), while retaining some qualitative differences in the behavior of the theoretical and experimental curves. At the same time, after isobaric aging the sample at room temperature, there is almost complete coincidence of these curves.

It is important to note once again that after removing the pressure and the completion of all the relaxation processes, depending  $\rho_{\text{ab}}(T)$  is almost completely identical to the original curves, measured before the application of high pressure, indicating the reversibility of the process. Thus, given a certain arbitrariness of determining the value of the opening of the pseudogap at  $T^*$  depending on reject  $\rho_{\text{ab}}(T)$  from linear behavior, the agreement between experiment and theory in this case can be considered quite satisfactory.

## CONCLUSIONS

In conclusion, we briefly sum up the main results obtained in this work. The application of high pressure to  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals ( $\delta \approx 0.35$ ) leads to a significant expansion of the interval linear dependence  $\rho_{\text{ab}}(T)$  and a narrowing of the temperature section of the implementation of the pseudogap regime. In this case excess conductivity obeys an exponential temperature dependence in wide temperature range and temperature dependence of the pseudogap - is satisfactorily described within the framework of the BCS-BEC crossover.

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