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Metal – dielectric transition and temperature dependence of the pseudo-gap in $YBa_2Cu_3O_{7-\delta}$ single crystals: effect of pressure and oxygen nonstoichiometry

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The effect of high hydrostatic pressure on the electric resistance in the *ab* plane of the oxygen-deficient $YBa_2Cu_3O_{7-\delta}$ single crystals has been investigated. It is shown that the pressure-induced redistribution of the labile oxygen results in an intensified phase layering accompanied by structural relaxation processes within the experimental sample volume. A significant displacement of the temperature ranges that correspond to metal-dielectric type transitions and the pseudo-gap realization regime. The description of the excess conductivity as $\Delta\sigma\sim(1 - T/T^*)\exp(\Delta^*_{ab}/T)$ can be interpreted in terms of mean field theory where T^* is the mean-field temperature of the superconducting transition and the temperature dependence of the pseudo-gap is well described in the framework of the BCS-BEC crossover theory. Increasing the applied pressure results in the narrowing of the temperature range where the PG regime is realized, thus extending the area of linear $\rho(T)$ dependence in the *ab* plane.

Исследовано влияние высокого гидростатического давления на электросопротивление в *ab*-плоскости монокристаллов $YBa_2Cu_3O_{7-\delta}$ с недостатком кислорода. Установлено, что индуцируемое высоким давлением перераспределение лабильного кислорода приводит к усилению фазового расслоения, которое сопровождается процессами структурной релаксации в объеме экспериментального образца. При этом происходит существенное смещение температурных участков, отвечающих переходам вида металл-диэлектрик и режима реализации псевдощелевой аномалии. Описание избыточной проводимости $\Delta\sigma$ при помощи 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 кроссовера БКШ-БЭК. Увеличение приложенного давления приводит к эффекту сужения температурного интервала реализации ПЩ-режима, тем самым расширяя область линейной зависимости $\rho(T)$ в *ab*-плоскости.

1. Introduction

The study of the resistivity and the structural characteristics of high-temperature superconducting cuprates (HTSC) [1] is an active research direction in condensed matter physics. The significant anisotropy, the rather complex crystal structure [2], the inhomogeneity of the defects distribution within the crystal, the clustered inclusions and the relaxation effects [3] are among specific features of those compounds that, in turn, cause a number of interesting effects. For example, among the electric transport properties of HTSC, to mention are a wide temperature area of excess para-conductivity in the ab plane [3], the non-coherent cross-wise electric transport [2], the metal-insulator type transitions [3] and the pseudo-gap (PG) anomaly. These phenomena are very important in solving a fundamental problem in solid state physics: the discovery of new functional materials with high current bearing capability. In this regard, it is important to use experimental methods to determine the factors affecting the physical characteristics the most and if the theoretical models describe the results adequately. The use of a high hydrostatic pressure in HTSC [4, 5] is among the methods of critical importance.

An appropriate system to investigate these issues is $YBa_2Cu_3O_{7-\delta}$ (1-2-3 system). This is due to the possibility to modify the conductivity characteristics and the crucial parameters by varying the oxygen stoichiometry and doping with substituent elements [4, 5]. It has been found that the pressure application to the oxygen-deficient $YBa_2Cu_3O_{7-\delta}$ results in a non-equilibrium state accompanied by the labile component redistribution [4]. This, in turn, will change the transport characteristics of the sample both in the normal and superconducting states as well as in the PG regime realization region [4, 5]. Although the PG anomaly in 1-2-3 system has been studied intensively, there are only few studies that address the influence of pressure upon the PG [5] in optimally oxygen doped samples. In this work, we have investigated the influence of hydrostatic pressure up to 11 kbar on the conductivity in the *ab* plane of the oxygen deficient $YBa_2Cu_3O_{7-\delta}$ single crystals.

2. Experimental

To grow the $YBa_2Cu_3O_{7-\delta}$ single crystals, we used the solution-melt technique in a gold crucible as described in [3, 4]. For the resistivity measurements, selected were rectangular single crystals of 3.0×0.5×0.03mm3 size with the *c* axis oriented along the smallest dimension. The crystals were annealed in oxygen flow at 400°C for five days in order to obtain stoichiometric samples. To produce oxygen deficient samples, the crystals were additionally annealed in air flow at 600°C for one day with a following hardening. The standard four-contact scheme was used to form the electric contacts. The silver paste was applied onto the crystal surface and gold conductors (0.05mm in diameter) were connected thereto followed by 3 hours annealing at 200°C in oxygen atmosphere. The technique provided a contact transition resistance of less than 1 Ω and made it possible to measure the resistivity at transport currents up to 10 mA in the *ab* plane. The hydrostatic pressure was produced in an autonomous chamber of the piston–cylinder type [4] and it was measured using a manganin manometer. A copper-constantan thermocouple mounted at the sample level on the outside surface of the chamber provided the temperature measurements. To ensure that the relaxation process was over when determining the influence of the oxygen redistribution, the measurements were made three to seven days after the pressure application and removal.

3. Experimental results and disscusion

The temperature dependence of the electric resistivity in the *ab*-plane, $R_{ab}(T)$, measured after the pressure application and removal is presented in Fig. 1. Some curves thereon and next two pictures are not shown to simplify the general picture. It is seen that as the applied pressure increases, the critical temperature (T_c) of the sample rises, the electric resistance decreases and the interval of the linear dependence $R_{ab}(T)$ broadens significantly in the area of higher temperatures. This is reflected in the lowered characteristic T^* value, from which on a systematic deviation of the experimental data commences downwards from the linear dependence. T^* is the temperature

of transition to a pseudo-gap mode [2, 3]. Notably, all the above changes occur not only as a result of the hydrostatic pressure action but also during the sample holding at room temperature under isobar conditions, immediately after the pressure application-removal procedure. For example, in Fig. 1, curves *1* and *4* show the dependences measured prior to the application and immediately after the pressure removal. Comparing the two curves, we observe that the results depend significantly on the holding time of the sample at room temperature. So, immediately after the pressure removal, the electric resistance value of the sample was about 5% less than that measured prior to the pressure application and relaxed further, until the equilibrium condition, within 3 days. Thereafter, the $\rho_{ab}(T)$ dependence matched with that measured before the pressure application. This testifies the reversibility of the process that we will consider in more detail below.

The decrease of the oxygen content results, alongside with the T_c loweribg from 92 K to 50 K, in a transition from quasi-metallic behavior of the $R_{ab}(T)$ curves (typical of the optimally oxygen doped samples [2]) to curves with the characteristic thermo-activated sagging. Fig. 2 shows these dependences in $\ln[R_{ab}(T)] - 1/T$ coordinates. It is seen that a significant range (115 K $\leq T \leq$ 255 K) segment of the linear behavior of $\rho_{ab}(T)$ is preserved. This corresponds to the description by means of analytical expression:

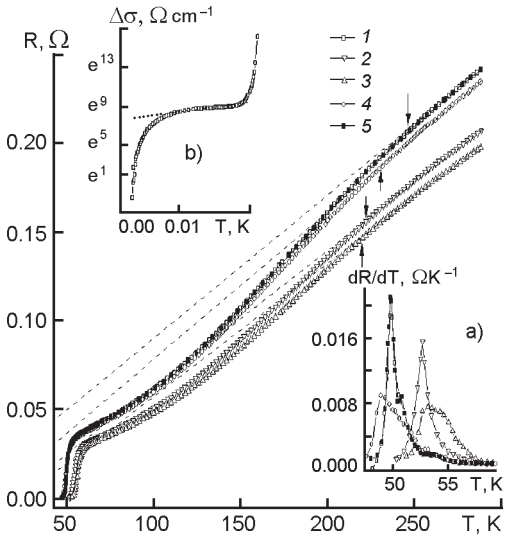

Fig.1. Temperature dependences $R_{ab}(T)$, obtained at different pressures: *1*, prior to pressure application; *2*, immediately after the application of 7.8 kbar pressure; *3*, after the sample keeping at room temperature under 7.8 kbar pressure for a week; *4*, immediately after the pressure removal; *5*, after the sample holding under zero pressure for 3 days. The points indicated by arrows response to T^* (the temperature of the transition onset to the pseudo-gap mode). In the inset (a), presented is the plot $dR_{ab}(T)/dT$ in the vicinity of the superconducting transition. The curves in the inset are denoted as in Fig.1. Inset (b): temperature dependence of the excess conductivity $\Delta\sigma(T)$ of the sample under atmospheric pressure.

$$R = AT\exp(\Delta/T)\,, \tag{1}$$

where A is a constant and Δ is the activation energy. When temperature is lowered, we observed a faster $R_{ab}(T)$ decline which, according to the Mott criteria [6], may serve as a reliable feature of the metal-dielectric (MD) "Anderson" type transition in the system. In fact, as our experimental data analysis shows (Fig. 3), in the temperature interval where we observed a systematic deviation from the linear dependence in $\ln[\rho_{ab}(T)] - 1/T$ coordinates, our curves are described as

$$1/R \propto T^{1/3}\,. \tag{2}$$

Such a behavior in the $\rho(T)$ dependences was observed in [7] for amorphous Gd-Sn alloys. According to [7], the dependence of Eq. (2) type results from the scaling description in the vicinity of the MD transition, in the "critical" mode, where the conductivity is mainly of the quantum character [8]. Here, we do not review these issues in detail.

It has been established [6] that the Anderson transition may occur in non-amorphous materials that possess a certain degree of disordering. In the 1-2-3 compounds, this is the labile component disordering [3, 4]. This is consistent with the presence of a residual displacement of the temperature interval where the MD-transition is realized. From Fig. 2, comparing curves *1* and *2* taken under zero pressure before the application and just after the pressure removal, we can see that in

the curve *2*, the point that corresponds to the MD-transition temperature, is displaced by about 5 K in comparison to the corresponding point in curve *1*. It is important to note that, after the sample holding under room temperature for 3 days, the $R(T)$ dependence matched with that measured prior to the pressure application. This testifies the process reversibility. As it was shown before [4], such processes are due to the labile oxygen redistribution caused by the high pressure.

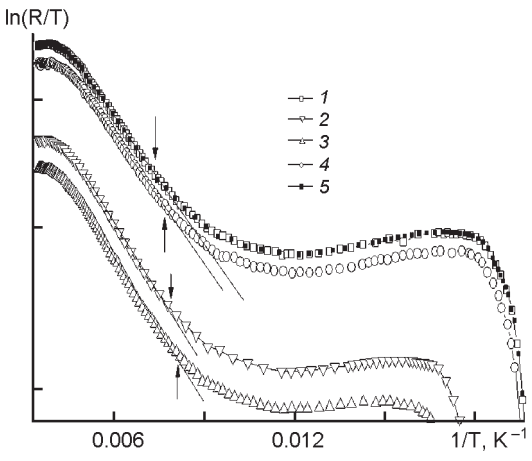

Fig.2. Plot of $\ln(R_{ab}/T) - 1/T$ dependences under different pressures. The curves are numbered as in Fig.1. The arrows show the characteristic value T^* (onset of the experimental data systematical deviation from the linear dependence).

In our case, the questions are how the above mentioned peculiarities may be correlated with the emerging PG anomaly that we mentioned in the introduction. As Mott has mentioned [6], the appearance of the PG (or density of states minimum) could be expected when the conduction and valence bands overlap slightly. This situation can be realized by changing the mean interatomic distance, the composition or the coordination number of the metal-insulator transition [6]. In recent years, one of the dominant models regarding the PG appearance is the cluster model [9-11]. Among the most recent theoretical mechanisms referring to the PG formation are the NUC model (negative U-centers) [9], the impurity mechanism of high T_c superconductivity [10] and the Abrikosov PG percolation theory [11]. According to the impurity model of high T_c superconductivity [10], the appearance of the PG anomaly in HTSC compounds is realized by the existence of numerous oxygen vacancies that may attract an electron from the O^{2-} ion and in conjunction with the nearby O- ion form a 2-level system with a pair of electrons localized within. The dimension of the 2-level system is the minimum coherence length and the T_c is determined from the equation [11]: $T_c \propto \{E_1 - E_2 - 0.5(W_1 - W_2)\}/k_B$, where W_1 and W_2 are the band widths formed by the interference of the wave functions of the set of electrons being at the E_1 and E_2 energy levels of the 2-level system. The Cooper pairs are the electron pairs with opposite spins in the lower levels of these 2-level systems that are in coherently motion when $T < T_c$. For $T > T_c$, the interband gap does not disappear and this explains the PG emergence [10].

As it was shown previously [9, 10], the temperature T^* (the pseudo-gap opening temperature) is uniquely correlated with the size of the superconductive clusters formed by the oxygen ions. The high pressure induced redistribution of the labile oxygen may influence the size of the clusters which could be separate superconducting phases as well as the attainment of the percolation threshold. This, in turn, should be reflected in the $R_{ab}(T)$ dependence. As it is shown in the inset of Fig. 1, the sample holding at room temperatures in the high pressure application-removal procedure affects significantly the width and the form of the superconducting transition. A characteristic peculiarity of the effect of such a holding during ~5 days is the substantial (up to $\Delta T_c \approx 2$ K) upwards and downwards temperature displacement of the peak in the $dR_{ab}(T)/dT$ dependences (according to [12], the corresponding maximum is T_c). This indicates the change in the transport current passing paths. This, in turn, is possible in the case of phase foliation in the oxygen non-stoichiometric samples at changes in the spatial distribution and the cluster sizes in the low and the high temperature phases [3]. This requires that the oxygen diffusion displacement by distances of the order of the cluster size.

In the inset (a) to Fig. 1, curve *1* represents the dependence measured prior to the pressure application, whereas curve *2* was measured immediately after the application of 7.8 kbar pressure, and curve *3*, after holding the sample under room temperature under pressure 7.8 kbar for a week; curve *4* was obtained immediately after the pressure removal and curve *5* obtained after keeping the sample under atmospheric pressure for 3 days. The sample keeping at room tempera-

ture during the pressure application-removal procedure is seen to result, in addition to a change in the T_c value, in significant qualitative changes of the superconducting transition width and shape. Comparing curves *2* and *3*, it can be concluded that after holding the sample under pressure for a week, the absolute value of dR_{ab}/dT is reduced by a factor of 2 and the transfer itself is broadened substantially. In the dependence measured directly after pressure removal (curve *4*), there is change only in the absolute T_c value, while the transfer width and the dR_{ab}/dT value remain unchanged. The comparison between curves *4* and *5* indicates that after the sample was held under atmospheric pressure for three days at room temperature, the the transfer width and shape regain their initial values. Consequently, we suggest that when the pressure increases, oxygen migrates partially from the phase with lower T_c to that with a higher critical temperature. Conversely, when the pressure decreases, the opposite redistribution occurs. This behavior may occur in the case of the realization of the ascending diffusion process [13]. In an disordered field of mechanical stresses (arising in particular due to the external pressure application), resulting from the difference between the volume of the impurity atoms and the atoms of their normal crystallographic positions the impurity atoms exert a force that is proportional to their difference in volume. As a result, the atoms with the larger ionic-radius diffuse to the expansion region whereas the smaller atoms migrate to the compression area. According to contemporary conceptions, defects can also exist in the process of the ascending diffusion [13], in our case the oxygen vacancies. An important peculiarity of this effect is the reversibility. For example, after the removal of the external pressure, we observe a reverse equilibration of the point defect concentration of the within the sample volume.

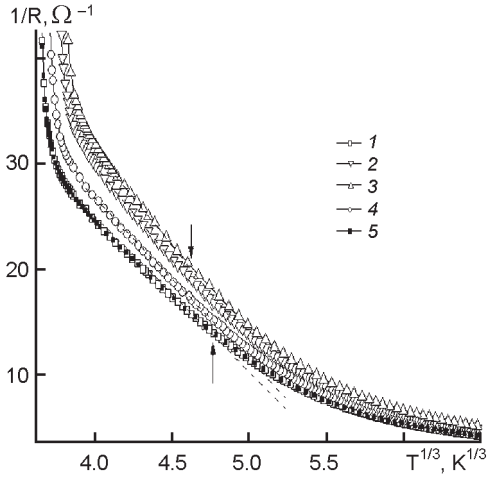

Fig.3. Plot of $1/R_{ab} - T^{1/3}$ dependences under different pressures. The curves are numbered as in Fig.1. The arrows show the characteristic value T^* (onset of the experimental data systematical deviation from the linear dependence).

According to the previous studies [14,15], the critical temperature value in $YBa_2Cu_3O_{7-\delta}$ is uniquely associated with the number of carriers in the CuO_2 layers through the dependence

$$T_c = T_c^{\max}\left[1 - 82.6(n - n_{opt})^2\right] \qquad (3)$$

where $T_c^{\max}$ is the maximum critical temperature and n_{opt}=0.25 is the optimum number of holes in the layer for $YBa_2Cu_3O_{7-\delta}$. This equation predicts that at increasing pressure (at room temperature) the number of holes for the low-temperature phase is reduced by about 2-3%. As to the high-temperature phase, the reverse process occurs and the number of holes increases. This is consistent to the supposition that the high hydrostatic pressure application results in a diffusion redistribution of the labile components from a phase of lower critical temperature to the high-temperature superconducting phase. This is accompanied with various size cluster formation and decomposition processes that may be an additional confirmation of the structural and electro-transport anomalies interrelation in the system.

As noted above, the application of pressure causes also a significant (up to 15 K) expansion of the linear interval in the $\rho_{ab}(T)$ dependence in the high temperature region. The last phenomenon appears to reduce the temperature value T* at which a systematic deviation of experimental points downwards from the linear dependence begins. The decrease of the $R_{ab}(T)$ value, which is observed at temperatures $T<T^*$, indicates the appearance of so-called excess conductivity $\Delta\sigma$ in the crystal. The temperature dependence of excess conductivity is usually determined from the equation:

$$\Delta\sigma = \sigma - \sigma_0 \tag{4}$$

where $\sigma_0 = \rho_0^{-1} = (A+BT)^{-1}$ is the conductivity obtained from the extrapolation of the linear part to zero temperature, and $\sigma = \rho^{-1}$ is the experimental conductivity value in the normal state. The $\Delta\sigma(T)$ dependence is represented in the inset (b) to Fig.1 in the $\ln\Delta\sigma - 1/T$ coordinates. It is seen that in a wide temperature interval the experimental dependence is linear, in agreement with the analytical expression:

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

where Δ^*ab the value that defines a thermally activated process across the energy gap - the pseudo-gap.

The exponential dependence of $\Delta\sigma\ (T)$ was observed before in YBCO film samples [16]. Prokofyev *et al.* [16] have demonstrated that the experimental data approximation can be expanded significantly by introducing the factor $(1-T/T^*)$. The excess conductivity in this case is proportional to the superconducting carrier density $n_s \sim (1-T/T^*)$ and inversely proportional to the number of pairs $\sim\exp(\Delta^*/kT)$, that are destroyed by thermal motion:

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

Here, T^* is considered as the mean field temperature of superconducting transition, and the temperature interval $T_c < T < T^*$ where the PG regime exists is defined by phase rigidity of the order parameter that depends on either the oxygen nonstoichiometry or the dopant concentration. Thus, using the method proposed by Prokofyev et al. [16] and proceeding from the experimental curve $\ln\Delta\sigma$, a temperature dependence $\Delta^*_{ab}(T)$ until the T^* can be constructed. Fig. 4 shows the PG temperature dependence in $\Delta^*(T)/\Delta_{max}$ vs T/T^* coordinates, where Δ_{max} is the Δ^* value in the plateau away from T^*, measured at different pressures.

In [17], the PG temperature dependences are described in the frame of the BCS-BEC crossover theory. In general form,those dependences are described as

$$\Delta(T) = \Delta(0) - \Delta(0)\sqrt{\frac{\pi}{2}}\sqrt{\frac{T}{\Delta(0)}}\exp\left[-\frac{\Delta(0)}{T}\right]\left[1 + erf\sqrt{\frac{\sqrt{x_0^2+1}-1}{T/\Delta(0)}}\right], \tag{7}$$

where $x_0 = \mu/\Delta(0)$ (μ is the chemical potential of the carrier system, $\Delta(0)$ is the energy gap value at $T = 0$, and $erf(x)$ is the error function.

In the case $x_0 \to \infty$ (weak coupling), Eq. (7) takes the form:

$$\Delta(T) = \Delta(0) - \Delta(0)\sqrt{2\pi\Delta(0)T}\exp\left(-\frac{\Delta(0)}{T}\right), \tag{8}$$

that is well known in the BCS theory. In the limit of the strong interactions in the three-dimentional case ($x_0 < -1$), Eq. (7) takes the form

$$\Delta(T) = \Delta(0) - \frac{8}{\sqrt{\pi}}\sqrt{-x_0}\left(\frac{\Delta(0)}{T}\right)^{3/2}\exp\left[-\frac{\sqrt{\mu^2+\Delta^2(T)}}{T}\right] \tag{9}$$

The $\Delta^*(T)/\Delta(0)$ vs T/T^* dependences have been calculated using Eqs. (8), (9) in the mean-field approximation within the context of the BCS–BEC crossover theory [17]. The results for 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. 4 as dotted lines. It is seen that at increasing of the applied pressure the experimental curves are shifted from the shape corresponding to Eq. (9) (strong coupling) to that answering to Eq. (8) (weak coupling). This behavior is qualitatively similar to the transformation effect of pseudogap temperature dependences for YBCO samples observed at the reduction of oxygen nonstoichiometry [2]. It is obvious that these correlations in the behavior of the curves $\Delta^*(T)$ are not random. Indeed, as is well known from literature (see e.g. [12]), the high pressure application to the

HTSC samples of 1-2-3 system, as well as increasing oxygen content, results in improved conductive properties which is manifested as the critical temperature increase and significant reduction of the electrical resistivity. Thus, taking into account some arbitrary definition of the T^* value from the pseudo-gap onset (defined by the deviation $\Delta(T)$ from the linear behavior), the experiment can be considered as consistent with theory.

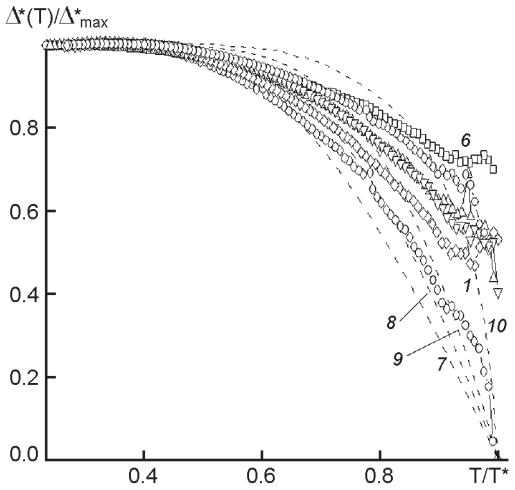

Fig. 4. Temperature dependence of the PG for the $YBa_2Cu_3O_{7-\delta}$ single crystal in $\Delta^*(T)/\Delta_{max}$ vs T/T^* coordinates, where Δ_{max} is Δ^* value in the plateau away from T^*. The curves are numbered as in Fig. 1. The dotted lines for $\Delta^*(T)/\Delta(0)$ vs T/T^* dependence were calculated according to [17] for the crossover parameter values $\mu/\Delta(0) = 10$ (limit of the BCS, curve *7*) and $\mu/\Delta(0) = -2, -5, -10$ (limit of the BEC, curves *8-10*).

4. Conclusion

Thus, the redistribution of the labile oxygen induced by the high pressure in $YBa_2Cu_3O_{7-\delta}$ crystals results in the intensidied phase foliation and the stimulation of the ascending diffusion processes between superconducting clusters with different degree of oxygen deviation from stoichiometry. This, in turn, produces a significant displacement of the temperature range that corresponds to MD type transitions and the PG realization mode. The high pressure application to $YBa_2Cu_3O_{7-\delta}$ ($\delta < 0.5$) single crystals causes a significant expansion of the linear interval in the $R_{ab}(T)$ relationship and a narrowing of the pseudo-gap mode temperature range. In this case, the excess conductivity follows an exponential temperature dependence in a wide temperature range. The temperature dependence of the pseudogap is well described in the framework of the BCS-BEC crossover theory.

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Перехід метал-діелектрик і температурна залежність псевдощілини у монокристалах $YBa_2Cu_3O_{7-\delta}$: вплив тиску і кисневої нестехіометрії

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Досліджено вплив високого гідростатичного тиску на електроопір в *ab*-площині монокристалів $YBa_2Cu_3O_{7-\delta}$ з нестачею кисню. Встановлено, що індукований високим тиском перерозподіл лабільного кисню приводить до посилення фазового розшарування, яке супроводжується процесами структурної релаксації в об'ємі експериментального зразка. При цьому відбувається істотне зміщення температурних ділянок, що відповідають переходам виду метал-діелектрик і режиму реалізації псевдощілинної аномалії. Описання надлишкової провідності $\Delta\sigma$ за допомогою співвідношення $\Delta\sigma \sim (1 - T/T^*)\exp(\Delta^*_{ab}/T)$ може бути інтерпретовано у термінах теорії середнього поля, де T^* представлена як середньопольова температура надпровідного переходу, а температурна залежність псевдощілини задовільно описується у межах теорії кросовера БКШ-БЕК. Збільшення прикладеного тиску приводить до ефекту звуження температурного інтервалу реалізації ПЩ-режима, тим самим розширюючи область лінійної залежності $R(T)$ в *ab*-площині.