

# The influence of hydrostatic pressure up to 10 kbar on the electrical resistance and critical temperature of single crystals



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The effect of high hydrostatic pressure up to 10 kbar on the electrical resistance in the basic ab- plane of praseodymium-doped ( $x \approx 0.34$ ) single crystals  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  was investigated in the work. It was found that, in contrast to samples with a praseodymium content of  $x \approx 0.05$ , the application of high pressure leads to a multiple increase in the value of the baric derivative  $dT_c/dP$ . It was established that within the limits of experimentally achieved pressures, there was no change in the sign of baric derivatives  $dT_c/dP$  with increasing pressure, which was observed on polycrystalline samples with close values of praseodymium concentration. Possible mechanisms of influence of high pressure on  $T_c$  are discussed, taking into account peculiarities in the electronic spectrum of carriers.

**Keywords:** YBaCuO single crystals, praseodymium doping, hydrostatic pressure, phase separation, baric derivatives.

**Вплив гідростатичного тиску до 10 кбар на електричний опір та критичну температуру монокристалів  $Y_{0.66}Pr_{0.34}Ba_2Cu_3O_{7-\delta}$ .** Гресь В.Ю., Хаджай Г.Я., Комісаров А.О., Врагов О.Ю., Сайнчин В.В., Ковригін В.О., Вовк Р.В.

У роботі досліджено вплив високого гідростатичного тиску до 10 кбар на електричний опір в базовій площині легованих празеодимом ( $x \approx 0,34$ ) монокристалів  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ . Встановлено, що на відміну від зразків із вмістом празеодиму  $x \approx 0,05$ , застосування високого тиску призводить до багаторазового збільшення значення баричної похідної  $dT_c/dP$ . Встановлено, що в межах експериментально досягнутих тисків не відбувається зміни знака баричних похідних,  $dT_c/dP$ , зі збільшенням тиску, що спостерігалось на полікристалічних зразках з близькими значеннями концентрації празеодиму. Обговорюються можливі механізми впливу високого тиску на  $T_c$  з урахуванням особливостей електронного спектра носіїв.

## 1. Introduction

As is known [1-10], changing the degree of deviation from oxygen stoichiometry and doping with praseodymium impurities are effective methods of modifying the electrotransport characteristics and changing the critical parameters of high-temperature superconducting

compounds of the system  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ . At the same time, however, an increase in the degree of oxygen deficiency to  $\delta \geq 0.15$  often leads to the emergence of a non-equilibrium state in the system [1,2], which can be quite easily induced by the application of high pressure [3], a sudden change in temperature [4] or arise as a result of long-term storage [5] of experimen-

tal samples. Such a non-equilibrium state, as a rule, is accompanied by fairly intensive structural relaxation processes [1-5], which, in turn, have a significant impact on a number of physical characteristics of this compound, including magnetoresistive ones [6]. The introduction of praseodymium impurities, in the case of optimally oxygen-doped  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  samples, as follows from known literature sources (see, e.g. [7,8]), does not affect the realization of the non-equilibrium state in this compound, which allows, by means of a comparative analysis, to highlight the role and mechanisms of the influence of the redistribution of labile oxygen on the electrotransport characteristics of the studied samples. At the same time, in the case of applying high pressure, doping with praseodymium leads to the emergence of some peculiarities in the behavior of the baric dependences of the resistive characteristics of this HTSC system [9,10].

As is known [11,12], the application of high pressure to samples of high-temperature superconducting compounds of the  $ReBa_2Cu_3O_{7-\delta}$  system ( $Re=Y$ , or lanthanides), in the vast majority of cases, leads to a significant increase in the value of the critical temperature of the transition to the superconducting state with rates of  $dT_c/dP \approx (0.1\div 3)$  K/kbar. An exception to this are compounds of the  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  system, in which, at certain values of the praseodymium content, both positive and negative values of the baric derivative  $dT_c/dP$  can be observed, and in some cases, a change in its sign as the applied pressure increases [11]. The authors, as a rule, explain such features as the manifestation of the so-called “praseodymium anomaly” - inhibition of the conducting and superconducting characteristics of the  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  compound when praseodymium impurities are introduced. At the same time, the critical temperature decreases from  $T_c \approx 90$  K at  $x=0$  to the complete disappearance of superconductivity at  $x \approx 0.7$  [11-13]. At the same time, it should be noted that the dielectric–superconductor transition in a compound with complete replacement of yttrium by praseodymium,  $PrBa_2Cu_3O_{6.6}$ , was registered in [14].

Currently, there are several theoretical models dedicated to explaining the occurrence of the “praseodymium anomaly”, the most famous of which are the “hole filling model” [15], “pair breaking phenomena” [16], as well as models that predict the localization of hole carriers and, due to interaction with praseodym-

ium ions, various mechanisms of band state rearrangement (review [11]). The discussion on this issue continues to this day. Given the limited scope of our report, we do not conduct a detailed analysis of these phenomena here, leaving it for a separate work. It should also be noted that a significant part of the experimental data obtained during the study of the effect of high pressure on the transport characteristics of  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  compounds was obtained on ceramic, film, and textured samples with very different technological backgrounds [17-19]. At the same time, the most informative, in this sense, are studies on single-crystal samples, as the most perfect objects, in which there are practically no such structural defects as intergranular boundaries and mismatch dislocations at the film-substrate separation boundary, etc.

In the cycle of our previous works [20-25], the results of measurements of the temperature dependence of electrical resistance  $\rho(T)$  on samples  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  with praseodymium concentrations  $0 \leq x \leq 0.23$  were presented when high hydrostatic pressure up to  $P \approx 18$  kbar was applied. In this paper, measurements of  $\rho(T)$  in the basal ab-plane were carried out on single-crystal samples of  $Y_{0.66}Pr_{0.34}Ba_2Cu_3O_{7-\delta}$  in the case of applying high pressure up to  $P \approx 10$  kbar.

## 2. Experimental

HTSC single crystals of compounds  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  were grown by solution-melt technology in a gold crucible, according to the methodology [13,20,21]. To carry out resistive studies, rectangular crystals with a size of  $3 \times 0.5 \times 0.03$  mm<sup>3</sup> were selected. The smallest size of the crystal corresponded to the c - axis direction. Electrical contacts were created according to a standard 4-contact scheme by applying silver paste to the surface of the crystal, followed by the attachment of silver conductors with a diameter of 0.05 mm and three-hour annealing at a temperature of 200°C in an oxygen atmosphere. This procedure made it possible to obtain a transient contact resistance of less than one Ohm and carry out resistive measurements at transport currents up to 10 mA in the ab-plane. Hydrostatic pressure was created in a piston-cylinder type multiplier [20,21]. The pressure value was determined using a managanin manometer, and the temperature value was determined by a copper-constantan thermocouple mounted on the outer surface of the chamber at the level of the sample position.

### 3. Result and discussion

Figure 1 shows the dependences of  $\rho_{ab}(T)$  in the temperature range from room to critical, obtained in the case of high pressure application up to 9.7 kbar. As can be seen from the figure, with increasing pressure, the critical temperature increases, and the electrical resistance  $\rho_{ab}$  decreases, which is qualitatively consistent with literature data [11] and our data obtained for  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  samples with lower concentrations of praseodymium content  $x \leq 0.23$  [20, 23]. At the same time, it should be noted that, in contrast to the previous samples [20,23], which were characterized by a clearly expressed quasi-metallic behavior of the temperature dependences  $\rho_{ab}(T)$ , in our sample with a praseodymium concentration of  $x \approx 0.34$ , the nature of these dependences has changed significantly. The  $\rho_{ab}(T)$  curves for the  $Y_{0.66}Pr_{0.34}Ba_2Cu_3O_{7-\delta}$  single crystal acquired the so-called S-shaped shape with a characteristic “thermoactivation bend”, which may indicate the strengthening of the role of localization effects [8].

In the insert (a) of Fig. 1, the dependences  $\rho_{ab}(T)$  are shown in semi-logarithmic coordinates, in which it can be seen that in the “thermoactivation region” the experimental dependences are significantly “straightened out”. As noted in [8], in this case, at this temperature range, the dependences of  $\rho_{ab}(T)$  can be described by the relation:

$$\rho(T) \sim T \exp\left(\frac{\Delta}{T}\right), \quad (1)$$

where  $\Delta$  - is certain activation energy. With the subsequent decrease in temperature, a faster decrease in  $\rho_{ab}(T)$  is observed, which, according to the classical criteria of Mott [8], can serve as a reliable sign of the realization in the system of the metal-dielectric (MD) transition of the “Anderson” type.

The insert (a) of Fig. 1 shows the resistive transitions to the superconducting state in the coordinates  $\rho_{ab}(T)$  and  $d\rho_{ab}(T)/dT$ . As can be seen from the insert, the step structure observed for the two previous concentrations of praseodymium [20-24] is practically absent at the superconducting transitions, which could be considered a reliable sign of the presence of phase separation in the system [22,23,25-27]. At the same time, the absence of such steps at the resistive transitions of our sample may be a sign of the existence of percolation paths for the flow of transport current in the volume of

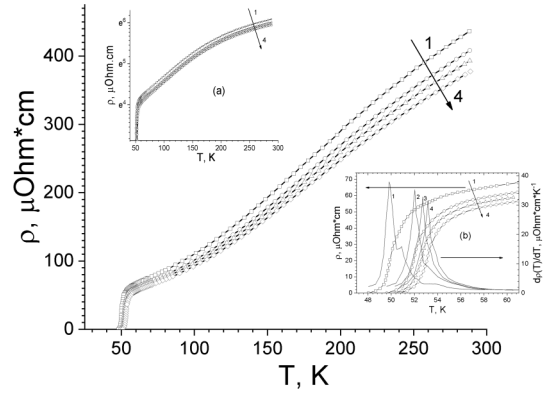


Fig. 1. Temperature dependences of electrical resistance in the base plane  $\rho_{ab}(T)$  of a  $Y_{0.66}Pr_{0.34}Ba_2Cu_3O_{7-\delta}$  single crystal, measured at pressures of 0; 3.72; 6.53; 9.7 kbar – curves 1-4, respectively. Insert (a): the same dependences in semi-logarithmic coordinates. Insert (b): resistive transitions to the superconducting state in the coordinates  $\rho_{ab}(T)$  and  $d\rho_{ab}(T)/dT$ , the numbering of the curves corresponds to the numbering in the figure.

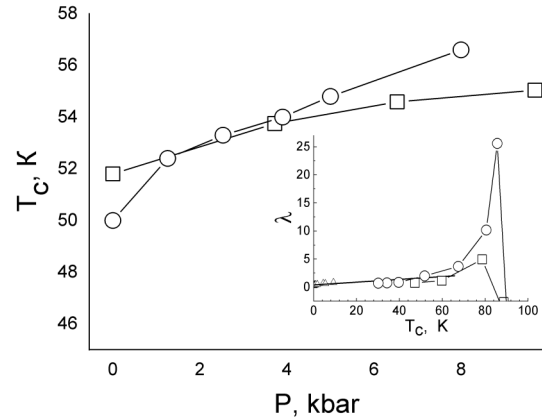


Fig. 2. Baric dependences of  $T_c$  of  $YBa_2Cu_3O_{7-\delta}$  single crystals for  $x=0$  and  $\delta \approx 0.45$  (circles) [26] and  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  for  $x = 0.34$  (squares). On the insert: correlation between  $T_c$  and  $\lambda$ , calculated according to McMillan's formula:  $\square$  –  $YBa_2Cu_3O_{7-\delta}$ ;  $\circ$  –  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ;  $\Delta$  – 12 transition metals [33].

the sample [28], when one of the phases shunts the other.

Dependencies of  $T_c(P)$  for the studied sample are shown in Fig. 2. Similar dependences obtained by us earlier [26] for the pure  $YBa_2Cu_3O_{7-\delta}$  sample with an oxygen deficit of  $\delta \approx 0.45$  and close value of critical temperature  $T_c \approx 50$  K are shown here. It can be seen that the rate of growth of the critical temperature with pressure for our sample  $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$  reaches  $dT_c/dP \approx 0.33$  K/kbar, in the range of experimentally achieved pressures. At the same time, the baric derivative for the oxygen-deficient sample  $YBa_2Cu_3O_{7-\delta}$  reaches values that are actually twice as large

as  $dT_c/dP \approx 0.64$  K/kbar. It can also be seen that for both compounds the maximum increase in the critical temperature is observed in the range of pressures from 0 to approximately 3 kbar, after which the value of  $dT_c/dP$  decreases several times, which will be discussed in more detail below.

As we noted in previous works, the value of the baric derivative up to  $dT_c/dP \leq 0.2$  K/kbar can be successfully explained using the well-known McMillan formula for qualitative analysis of  $T_c(P)$  dependencies [29]:

$$T_c = \frac{\theta_D}{1.45} \exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right], \quad (2)$$

where  $\theta_D$  - is the Debye temperature,  $\mu^*$  - is the shielded Coulomb pseudopotential characterizing electron repulsion,  $\lambda$  - constant of the electron-phonon interaction ( $\lambda, \mu > 0$ ) [30], which, in turn, depends on the parameters of the electronic and phonon spectrum superconductor:

$$\lambda = \frac{N(\varepsilon_F) \langle I^2(\vec{k} - \vec{k}') \rangle}{M\theta_D^2}, \quad (3)$$

where  $N(\varepsilon_F)$  is the density of states at the Fermi level,  $I$  - is the matrix element of the electron-phonon interaction averaged over the Fermi surface, and  $M$  is the mass of the ion.

As it was shown in [10], McMillan's formula is completely correct only for  $\lambda \leq 1.5$ . Values of  $\lambda \leq 1.5$  can be obtained if the value of  $\ln(1.45^*T_c/\theta)$  is in the range from  $-10$  to  $-2$ . For  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  this is realized in the region  $x, \delta > 0.3$ . In [30], data on the correlation between  $T_c$  and  $\lambda$  ( $\lambda$  calculated by McMillan's formula for  $\mu^*=0.13$ ) for 12 transition metals are presented.

Figure 2 shows these data together with similar data for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (see [10]) and  $\text{Pr}_{1-y}\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  [22]. It can be seen (insert) that only for  $T_c \leq 65$  K the data for transition metals agree with the data for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  and indicate a strong electron-phonon interaction in these substances. For  $T_c \geq 65$  K ( $x, \delta < 0.3$ ),  $\lambda$  increases sharply (in [30]  $\lambda \geq 10$  is called superstrong coupling), and then it passes into the negative region. Thus, HTSC  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  can be called superconductors with strong coupling only after  $x, \delta > 0.3$ , with smaller values of  $x$  or  $\delta$  the McMillan formula is obviously inapplicable.

## 4. Conclusion

In addition, as mentioned above, in [11] a change in the sign of  $dT_c/dP$  was observed. All of the above does not allow us to unambiguously interpret the obtained data within the framework of the theory of BCS. Probably, such  $dT_c/dP$  values, as well as a multiple decrease in  $dT_c/dP$  with increasing pressure, may be related to the features of the band structure of the compound  $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  [31], which we discuss in more detail in [21]. A 2-fold increase in the baric derivative  $dT_c/dP$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  compared to  $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  is probably due to the redistribution of labile oxygen [1,2]. The presence of structural and kinematic anisotropy in the system may play a role in this [32-37].

It should also be noted that within experimentally achieved pressures, we did not register a change in the sign of baric derivatives  $dT_c/dP$  with increasing pressure, which was observed in [11] on samples with close values of praseodymium concentration. It is obviously that additional studies on samples with a higher content of praseodymium and in a wider range of applied pressure are needed for the final clarification of this issue.

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