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## Effect of high hydrostatic pressure on various diffusion mechanisms in oxygen deficient $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$ (Re=Y, Ho) single crystals

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In present paper the effect of high pressure on the relaxation of the electrical resistivity at room temperature of oxygen nonstoichiometric  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$  (Re=Y, Ho) single crystals are investigated. It is established that the hydrostatic pressure significantly intensifies the process of diffusion coalescence in the oxygen subsystem. At the same time, the intensity of the redistribution of labile oxygen significantly changes when yttrium is replaced by holmium.

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В работе исследовано влияние высокого давления на релаксацию электросопротивления при комнатной температуре монокристаллов  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$  (Re=Y, Ho) нестехиометрического по кислороду состава. Установлено, что гидростатическое давление существенно интенсифицирует процесс диффузионной коалесценции в кислородной подсистеме. При этом интенсивность перераспределения лабильного кислорода существенным образом изменяется при замене иттрия на гольмий.

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### Introduction

As is well known, the presence of labile oxygen [1,2] in high-temperature compounds (HTSC)  $\text{ReB}_2\text{Cu}_3\text{O}_{7-x}$  (Re = Y or other rare earth element) leads to the appearance of a non-equilibrium state that can be manifested during the application of high pressure [3,4], a step by step change in temperature [5,6], long-term storage [7-9], contribute to phase separation processes [10,11], ascending diffusion [12,13] and the appearance of various superstructures [14-16]. All these processes have a significant effect on the physical properties of HTSC in both the normal and superconducting states and are most clearly manifested in the case of oxygen deficient samples [17,18].

The modification of the structural and electrophysical characteristics of superconducting materials can be achieved by applying various external influences [19-21] and is an important experimental tool of modern solid state physics. In particular, the study of the effect of the external hydrostatic pressure [22,23] on the structural state and transport processes

in  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$  single crystals, is a source of important information necessary for elucidating the microscopic mechanism of “high-temperature” superconductivity, which until now remains unclear [24]. Taking this into account, experimental methods have proved to be of a particular importance, allowing us to reveal the parameters of superconductors, which most significantly affect their physical characteristics in the normal and superconducting states. One of the most important methods is the use of high pressure [25-27], since this not only makes it possible to clarify the role and the influence of structural features of the system on the superconducting states formation, but also enables to model the conductive characteristics and critical parameters of the superconductor.

A characteristic feature of the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  compound is the relative simplicity of the complete or partial substitution of yttrium by its isovalent analogues, which enables the variation of the conductive characteristics, and thereby can verify the adequacy of theoretical models. In this respect,

the substitution of yttrium by the holmium, which has a sufficiently large magnetic moment ( $10.61\mu_B$  and  $9.7\mu_B$  in  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-x}$ ), allows us to predict a qualitatively different behavior of the system due to the paramagnetism of  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-x}$  in the normal state [2]. It is of interest to study oxygen deficient samples, in which the rare-earth ion can serve as a sensor sensitive to the local symmetry of its environment and to the distribution of charge density, since their change affects the crystal field that forms the electronic structure of such an ion [6].

In this work, we study the effect of an external hydrostatic pressure up to 5 kbar on the temperature dependence of the electrical resistance  $\rho(T)$  in the *ab* plane of  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$  ( $\text{Re} = \text{Y, Ho}$ ;  $x \approx 0.3-0.47$ ) single crystals in the temperature interval from 300 K to the temperature of the superconducting transition  $T_c$ .

### Experimental methodology

The crystals were grown by the “solution-melt” method in a gold crucible as was previously described [18]. The samples had the shape of a parallelepiped with dimensions  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  -  $2 \times 1.8 \times 0.5 \text{ mm}^3$  and  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-x}$  -  $1.9 \times 1.9 \times 0.5 \text{ mm}^3$ . The largest area of the sample corresponded to the crystallographic *ab* plane. Initially, the samples were heat-treated in oxygen flow at  $T = 400^\circ \text{C}$  for five days to saturate them with oxygen completely, i.e., to reduce the parameter  $x$  to a value  $\approx 0 - 0.1$ .

The temperature dependence  $\rho(T)$  was first measured during the heating of the multiplier under atmospheric pressure. The pressure was then gradually increased at room temperature. When the desired pressure was reached, the multiplier was cooled to a temperature  $T < T_c$ , and after that the  $\rho(T)$  measurements were carried out. Following the measurement at the maximum pressure, the pressure was lowered to atmospheric pressure and the  $\rho(T)$  measurements were repeated.

In addition to the  $\rho(T)$  dependence, we carried out isothermal measurements of the change of the electrical resistance  $\rho$  at temperature 300 K as a function of time under pressure  $\approx 5$  kbar, as well as after its removal. In this experiment, the maximum exposure time reached  $\approx 80$  hours.

### Results and discussion

In previous work [29], the results of the electrical resistance time relaxation dependence at room temperature: curve 1 - after the application of pressure 5 kbar and curve 2 - after the pressure released were presented. Thus, it was determined that after the pressure was removed, the equilibrium resistance value was reached much faster than when the pressure is applied. To explain the latter result, we proposed that the observed phenomenon is due to the difference in the diffusion paths of the pressure-induced process of elongation-shortening of oxygen ion chains in *Cu-O* planes.

Thereafter [6], we provided evidence that the diffusion of oxygen ions in the  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$  ( $\text{Re}=\text{Y, Ho}$ ) compounds can take place via two mechanisms. At the initial stage of the process, the single-channel diffusion mechanism is dominant, whereas in the final stage the diffusion of oxygen ions is limited by the conventional mechanism of classical diffusion. In the first case, oxygen ion diffusion is characterized by the  $\langle L^2 \rangle \sim t^{1/2}$  dependence and in the second case by the  $\langle L^2 \rangle \sim t$  dependence.

The study of the time dependence  $\rho=f(T)$  carried out previously [29] (relaxation curves) also indicates that in this case there are two stages: a fast initial stage and a slower final stage. This result can also explain the existence of two mechanisms for the redistribution of oxygen ions induced by the application of pressure. To confirm the correctness of this assumption, we processed these relaxation curves by constructing the  $(\rho_0/D\rho)^2=f(T)$  dependences, which, in fact, determine the operation of the one or the other oxygen ions' diffusion mechanism (refer to Figure 1).

Figure 1, in our opinion, confirms the correctness of the previously stated assumption that the application of an external pressure of  $\sim 5$  kbar causes the diffusion redistribution of oxygen ions in the formation-decay of the oxygen clusters in the  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$  ( $\text{Re} = \text{Y, Ho}$ ) compounds. Herewith, the diffusion of oxygen ions process is realized by two mechanisms: By the single-channel diffusion mechanism and by the conventional classical diffusion mechanism (at the final stage of the process).

The entire set of the experimental results obtained, agrees well, if presume that the external hydrostatic pressure intensifies the appearance of several (at least two) phases in the studied samples, characterized by different values of the transition temperature  $T_c$ . In our opinion this is due to the process of the redistribution of the oxygen ions in the volume of  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$  ( $x=0,4$ ) crystals. The pressure accelerates the diffusion coalescence process [30] of microscopic clusters of various sizes oxygen vacancies formed in the crystal due to the oxygen deficiency. As a result of the coalescence, oxygen ions are redistributed in the crystal, which leads to an increase of a number of sufficiently large in size ordered one-dimensional clusters of oxygen ions. At the same time, the parameter  $x$  decreases. Accordingly, the number of specific structural entities - negative U-centers, capable of generating coupled charge carriers [31] increases. In this case, a higher concentration of U-centers corresponds to higher values of  $T_c$  of the superconducting phase.

As it was mentioned above, the establishment of the equilibrium value of the resistance  $R(t \rightarrow \infty)$  after the pressure removal occurs almost twice faster than under the action of pressure. This experimental fact is also explained within the framework of the proposed coalescence of oxygen clusters mechanism. Indeed, under the pressure application conditions, the process of filling clusters with oxygen ions by the accelerated “one-file” diffusion mechanism is

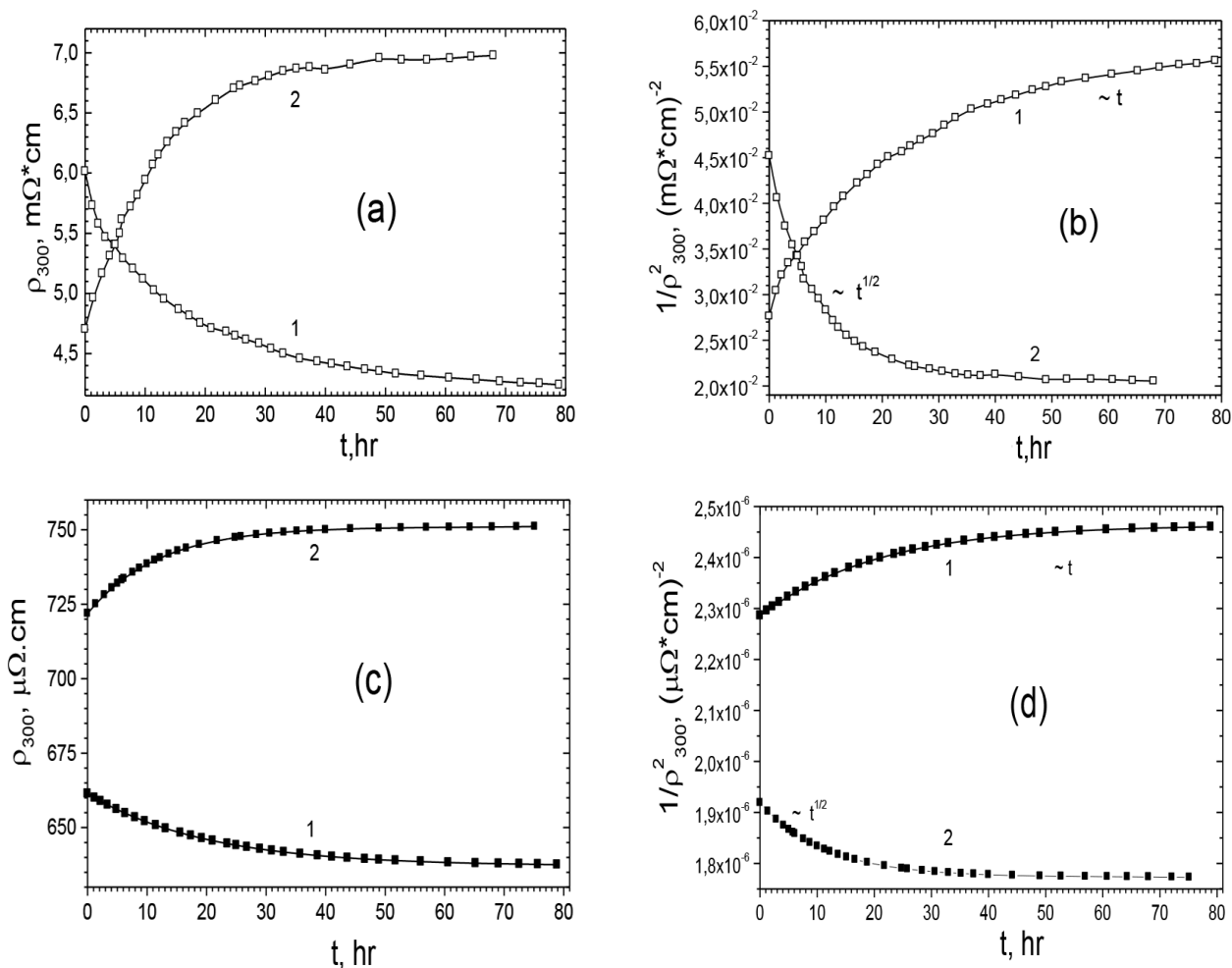


Fig.1. Time dependences of the electrical resistivity at room temperature in coordinates:  $\rho_{300} - t$  and  $1/\rho_{300}^2 - t$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  ((a) and (b)) and  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-x}$  ((c) and (d)) single crystals, measured immediately after application (curves 1) and removal (curves 2) of high hydrostatic pressure.

realized only at the initial stage, and further, this process and the decrease in electrical resistance, is limited by the slower mechanism of classical diffusion. After the pressure removal the process of more intensive increase in the size of oxygen clusters is suspended. Herewith, as the time passes, the number of not fully completed clusters increases and there is a relaxation of the resistance. Under these conditions, the movement of oxygen ions is at much shorter distances and can be realized in the “one-file” diffusion regime, practically, until the equilibrium value  $\rho(t \rightarrow \infty)$ . A certain role in this may be played by the presence of structural and kinematic anisotropy in the system [32-37].

Notably, despite the higher  $T_c$  and the smaller oxygen deficiency, in the case of the  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-x}$  samples, all the characteristic changes in the form of the electrical resistivity temperature dependences and the absolute values of the resistive parameters that were observed during the pressure application-removal process at room temperatures were not less pronounced than for the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  samples. Apparently, in the case of  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-x}$ , the substitution of yttrium by holmium which has a larger ionic radius, can play

a role on the structural order in the system, which in turn leads to a change in the interaction of the oxygen ions in the CuO planes. Indeed, as is known from the literature [1,2,6,29], when yttrium is replaced by rare-earth elements with a larger ionic radius, significant qualitative changes occur in the  $T_c(x)$  dependences. Therefore, the characteristic for the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$   $T_c(x)$  dependence with two plateaus, at 60 and 90 K, degenerates into a much sharper monotonic dependence [1,2]. Thus, it can be assumed that in the case of deviation from oxygen stoichiometry, the  $\text{HoBa}_2\text{Cu}_3\text{O}_{7-x}$  compound should be characterized by a much more disordered oxygen superstructure in comparison with the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ .

Thus, we can conclude, that the external hydrostatic pressure  $P \approx 5$  kbar substantially intensifies the process of the diffusion coalescence of oxygen clusters, i.e., causes the growth of their average size. In turn, the increase in the size of oxygen clusters leads to an increase in the number of negative U-centers, the presence of which leads to the appearance of a phase capable of generating paired carriers of electric charge and, accordingly, characterized by a higher transition temperature  $T_c$ .

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