

High-pressure-induced relaxation of normal electrical resistance in single-crystal $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ underdoped with oxygen

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The effect of the external hydrostatic pressure $P \approx 4$ kbar on the relaxation of the normal electrical resistance of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ single crystals with oxygen deficiency was investigated. It has been established that a high pressure significantly intensifies the process of diffusion coalescence of oxygen clusters, i.e. causes the growth of their average size. In turn, an increase in the size of oxygen clusters leads to the appearance of a phase, which is characterized by a higher critical temperature of the superconducting transition T_c .

Keywords: $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ single crystals, hydrostatic pressure, diffusion coalescence, relaxation, oxygen deficiency.

Исследовано влияние внешнего гидростатического давления $P \approx 4$ kbar на релаксацию нормального электросопротивления монокристаллов $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ с дефицитом кислорода. Установлено, что высокое давление существенно интенсифицирует процесс диффузионной коалесценции кислородных кластеров, т.е. обуславливает рост их среднего размера. В свою очередь, увеличение размеров кислородных кластеров приводит к появлению фазы, которая характеризуется большей критической температурой сверхпроводящего перехода T_c .

Індукована високим тиском релаксація нормального електроопору у недопованих киснем монокристалах $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$. Ю.І.Бойко, Г.Я.Хаджай, С.Р.Вовк, Р.В.Вовк, М.А.Дригаїло, В.Ю.Гресь, Ясек Гралевські.

Досліджено вплив зовнішнього гідростатичного тиску $P \approx 4$ kbar на релаксацію нормального електроопору монокристалів $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ з дефіцитом кисню. Встановлено, що високий тиск істотно інтенсифікує процес дифузійної коалесценції кисневих кластерів, тобто зумовлює зростання їх середнього розміру. У свою чергу, збільшення розмірів кисневих кластерів приводить до появи фази, яка характеризується більшою критичною температурою надпровідного переходу T_c .

1. Introduction

As is known, the presence of labile oxygen [1, 2] in high-temperature cuprates (HTSC compounds) $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ leads to the

appearance of a non-equilibrium state, which can manifest itself during the application of high pressure [3, 4], a sudden change in temperature [5, 6] and long-term

storage [7–9] and, in turn, contribute to the processes of phase separation [10, 11], upward diffusion [12, 13] and the emergence of various kinds of superstructures [14–16]. All these processes have a significant impact on the physical properties of HTSCs in the normal and superconducting states and are most pronounced in the case of samples of non-stoichiometric oxygen composition [17, 18].

The modification of the structural and electrophysical characteristics of superconducting materials by applying various external influences [19–21] is an important experimental tool of modern solid state physics. In particular, the study of the influence of external hydrostatic pressure [22, 23] on the structural state and transfer processes in single crystals of the $\text{YB}_2\text{Cu}_3\text{O}_{7-x}$ — x system can serve as a source of important information necessary to achieve an understanding of the microscopic mechanism of "high-temperature" superconductivity, which so far remains unclear [24], despite a more than 30-year history of intensive research. Taking this fact into account, the experimental methods have been of particular importance, which make it possible to identify the parameters of superconductors that most significantly affect their physical characteristics in the normal and superconducting states. One of the important methods in this aspect is the use of high pressure [25–27], since this not only clarifies the role and influence of the structural features of the system on the formation of the superconducting state, but also enables us to study the mechanisms of defect redistribution and the relationship of these processes with electric transport characteristics of a superconductor.

In this paper, we present and discuss the results of an experimental study of the influence of external hydrostatic pressure up to 7 kbar on the temperature dependences of the electrical resistance, $R(T)$, in **ab**-the plane of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ single crystals ($x \approx 0.4$) in the temperature range from 300 K to temperature of the transition in superconducting state, T_c .

2. Experimental

The crystals were grown by the "solution — melt" technology in a gold crucible according to the procedure described in [18]. The samples studied had the shape of a parallelepiped with dimensions $\approx 2 \times 1.5 \times 0.2$ mm³. The largest area of the sample corresponded to the crystallographic plane **ab**. Initially, the samples were subjected to heat treatment in

a stream of oxygen at a temperature $T = 400^\circ\text{C}$ for five days in order to completely saturate them with oxygen, i.e. reduced the parameter x to a value of $\approx 0-0.1$.

The temperature dependences of the electrical resistance, $R(T)$, were measured by the standard four-probe method at a constant current. The transition temperature to the superconducting state of samples that underwent initial heat treatment in an oxygen flow reached ≈ 92 K. In order to reduce the oxygen content in the samples under study, they were further annealed for two days in the air atmosphere at temperature of 500°C . The temperature T_c for samples with oxygen deficiency decreased to ≈ 45 K. According to the literature data, this transition temperature corresponds to the value of the parameter $x \approx 0.4$ [2, 28]. Samples of precisely this stoichiometric composition were the subject of our research under the conditions of an external hydrostatic pressure on the electrical conductivity of the crystal under study [29].

The pressure was created using a multiplier according to the method described in [18]. The magnitude of the applied pressure, P , was measured using a manganin gauge and varied in the range of 0 – 7 kbar. The temperature T_c was determined from the middle of the resistive transition to the superconducting state, i.e. at the level $R = R_n/2$, where R_n is the value of the electrical resistance of the sample in the normal (non-superconducting) state.

The temperature dependences, $R(T)$ were first measured during the heating of the multiplier at atmospheric pressure. Then the pressure was gradually increased at room temperature. When the desired pressure was reached, the multiplier was cooled to a temperature $T < T_c$ and already after that, the measurements of $R(T)$ were performed. After the measurement at the maximum pressure, it was lowered to the value of the atmospheric pressure and the measurement of the dependence $R(T)$ was repeated. In addition to the dependences $R(T)$, the isothermal measurements of changes in the electrical resistance R at 300°K as a function of time, t , under a pressure of 4.3 kbar, as well as after its removal were made. In this experiment, the maximum exposure time reached ≈ 80 h.

In [29], the results of measurements of the temporal dependences of the relaxation of the electrical resistivity at room temperature are presented: curve 1 — after applying a pressure of 4.3 kbar and curve 2 —

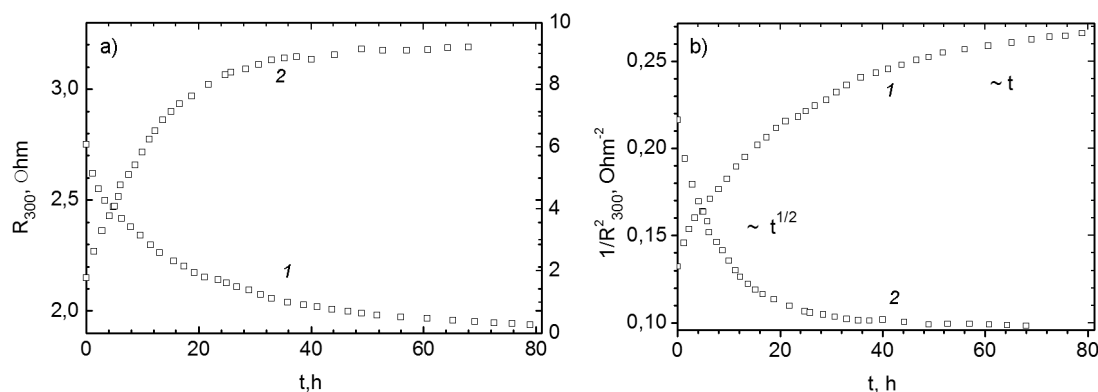


Fig. The dependences of the electrical resistance on time at room temperature for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ single crystals under pressure (1) and after pressure relief (2) in the coordinates $R - t$ (a) and $1/R^2 - t$ (b).

after removing the pressure. It turned out that after removing the pressure, the equilibrium value of resistance is achieved much faster than with the application of pressure. To explain the latter result, the authors suggested that the observed phenomenon is due to the difference in diffusion paths of the pressure-induced process of lengthening and shortening the chains of oxygen ions in the Cu-O planes.

In [6], evidence was given that the diffusion of oxygen ions in the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ compound can occur through two mechanisms. At the initial stage of the process, the "single-file" diffusion mechanism is decisive, and at the final stage, the movement of oxygen ions is limited by the usual classical diffusion mechanism. In the first case, the kinetics of diffusion of oxygen ions is characterized by the dependence of the mean square displacement $\langle L^2 \rangle \sim t^{1/2}$, and in the second case $\langle L^2 \rangle \sim t$.

The study of the time dependences $R = f(T)$, carried out in [29] (relaxation curves), also indicates that in this case two stages of this process are observed: the fast initial stage and the slower final stage. This result also explains the existence of two mechanisms for the redistribution of oxygen ions induced by the application of pressure. To confirm the correctness of the assumption made, we processed the indicated relaxation curves by plotting the dependences $(1/R)^2 = f(T)$ (see Fig.), which, in fact, determine the effect of a diffusion mechanism of oxygen ions.

In Fig. shows the corresponding curves. The obtained data, in our opinion, confirm the correctness of the previously suggested assumption that the application of an external pressure of ~ 4 kbar causes a diffusion redistribution of oxygen ions during the formation-decay of oxygen clusters in the

$\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ compound. In this case, the process of diffusion of oxygen ions is realized by two mechanisms: the mechanism of "single-file" diffusion and the mechanism of ordinary classical diffusion (at the final stage of the process).

3. Results and discussion

The entire set of obtained experimental results is in good agreement if it is assumed that the external hydrostatic pressure intensifies the appearance in the samples under study several (at least two) phases characterized by different values of the transition temperature T_c . In our opinion, this is due to the redistribution of oxygen ions in the volume of the studied $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ crystal ($x = 0.4$). The pressure accelerates the process of diffusion coalescence [30, 31] of microscopic clusters of oxygen vacancies of various sizes, which are formed in the crystal under study due to oxygen deficiency. As a result of coalescence, the redistribution of oxygen ions in the crystal occurs, which leads to an increase in the number of sufficiently large sized ordered one-dimensional clusters of oxygen ions. At the same time, the parameter x decreases.

As noted above, the establishment of the equilibrium value of the resistance $R(t \rightarrow \infty)$ after the pressure is removed occurs almost two times faster than under the conditions of pressure. This experimental fact is also explained within the framework of the proposed mechanism for the coalescence of oxygen clusters. Indeed, under pressure, the process of filling clusters with oxygen ions by an accelerated "single-file" diffusion mechanism is realized only at the initial stage, but later this process and, accordingly, a decrease in electrical resistance, are

limited by the slower mechanism of classical diffusion. After removing the pressure, the process of more intensive increase in the size of oxygen clusters is suspended. At the same time, the number of incomplete clusters increases over time and, accordingly, the resistance R relaxes. Under these conditions, oxygen ions move far less distances and can be carried out in the "single-file" diffusion mode until the equilibrium R value is fully established ($t \rightarrow \infty$). In this case, the presence of structural and kinematic anisotropy in the system may play a certain role [32–37].

4. Conclusions

Based on the processing of the obtained experimental results and their discussion, we can draw the following conclusions. The external hydrostatic pressure significantly intensifies the process of diffusion coalescence of oxygen clusters, i.e. causes the growth of their average size. In turn, an increase in the size of oxygen clusters leads to the appearance of a phase with a higher critical transition temperature T_c .

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