ISSN 1027-5495. Functional Materials, 24, No.4 (2017), p. 527-529.
doi:https://doi.org/10.15407/fm24.04.527 © 2017 — STC "Institute for Single Crystals"

Single-file diffusion in oxygen underdoped $ReBa_2Cu_3O_{7-x}$ (Re=Y, Ho) single crystals

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Received March 1, 2017

Analysis of the kinetics of relaxation processes observed at room temperature in the $ReBa_2Cu_3O_{7-x}$ (Re=Y, Ho) compounds, having a high-temperature superconductivity is made. It was concluded that the initial (faster) stage of these processes is controlled by a single-file diffusion of oxygen along one-dimensional chains of vacancies, forming in the oxygen sub-lattice of compound with a deficit of oxygen, and final (slow) stage controls by the usual classical diffusion. Presented and discussed the results of two-stage relaxation of the electrical resistance of above mentioned compounds which were a long time (more than three days) at room temperature in air.

Keywords: $ReBa_2Cu_3O_{7-x}$ single crystals, oxygen vacancies, annealing processes, diffusion mechanism.

Проведен анализ кинетики релаксационных процессов, наблюдаемых при комнатной температуре в высокотемпературных сверхпроводящих соединениях $ReBa_2Cu_3O_{7-x}$ (Re=Y, Ho). Сделан вывод о том, что начальная (более быстрая) стадия этих процессов контролируется однофайловой диффузией кислорода вдоль одномерных цепочек вакансий, образующихся в кислородной подрешетке соединения с дефицитом кислорода, а конечная (более медленная) — с помощью обычной классической диффузии. Представлены и обсуждены результаты двухступенчатой релаксации электрического сопротивления вышеупомянутых соединений, которые находились длительное время (более трех дней) при комнатной температуре на воздухе.

Однофайлова дифузія у недодопованих киснем монокристалах $ReBa_2Cu_3O_{7-x}$ (Re=Y, Ho). *Ю.І.Бойко, В.В.Богданов, Р.В.Вовк, О.Г.Орт, Ю.В.Литвинов*

Проведено аналіз кінетики релаксаційних процесів, які спостерігаються при кімнатній температурі в високотемпературних надпровідних сполуках $ReBa_2Cu_3O_{7-x}$ (Re=Y, Ho). Зроблено висновок про те, що початкова (більш швидка) стадія цих процесів контролюється однофайловою дифузією кисню вздовж одновимірних ланцюжків вакансій, що утворюються у кисневій підгратці сполуки з дефіцитом кисню, а кінцева (більш повільна) — за допомогою звичайної класичної дифузії. Представлено та обговорено результати двохступінчастої релаксації електричного опору вищезазначених сполук, котрі перебували тривалий час (більше трьох днів) при кімнатній температурі на повітрі.

The $YBa_2Cu_3O_{7-x}$ crystal is a well-known and one of the most studied ionic compounds regarding the so called "high-temperature" superconductivity [1]. An important feature of the structural state of this crystal is the presence in its lattice one-dimensional ordered clusters formed by oxygen vacancies [2]. The formation of such structural defects is due to the layered nature of the crystal lattice, as well as due to the deficiency of the oxygen ions, characterized by the parameter (x).

It should be noted that the superconducting properties of the $YBa_2Cu_3O_{7-x}$ system (the transition temperature to the superconducting state T_c, the electrical resis-

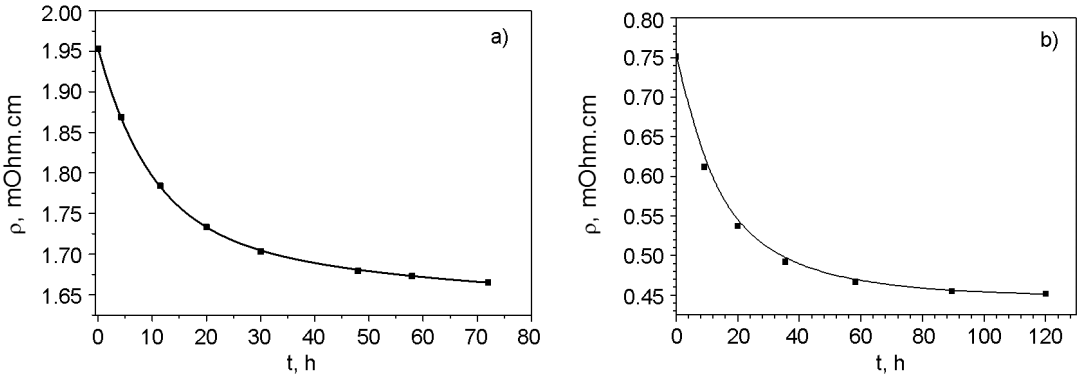


Fig. 1. The dependence of electrical resistivity of $YBa_2Cu_3O_{7-x}$ (a) and $HoBa_2Cu_3O_{7-x}$ (b) single crystals from the time of exposure in air at room temperature.

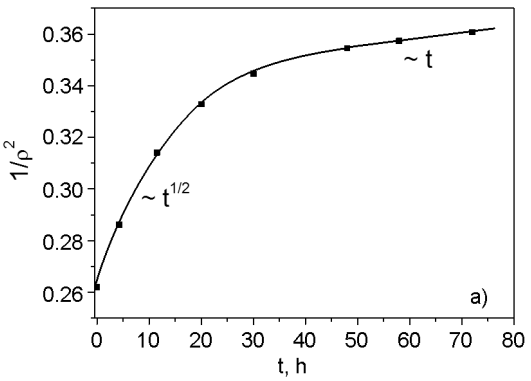
tivity ρ, the critical current density J_c et al.) are substantially dependent from the degree of the vacancies' concentrations filling by oxygen ions, i.e. depends by the value of the parameter (x). Thus, for example, if when we change the value of the parameter (x) from ≈0.1 to ≈0.4, a decrease for the T_c is observed from T_c≈92 K to T_c≈50 K, and for values of $x \geq 0.5$ the superconductivity in this crystal disappears [3]. Similar dependences from the value of the parameter (x) are also observed in the study of other characteristics of the above described superconductor. This effect is associated with the formation of one-dimensional chains of oxygen ions (clusters) that is formed as the clusters of structural vacancies is filling. Oxygen clusters with Cu ions forming the so-called U-centers that are capable in generating of coupled electric charge carriers [4–6].

The formation of clusters occurs by the diffusion of the oxygen ions during the high temperature annealing process of the crystal in an oxygen atmosphere. As evidenced by the results of [7], the diffusion filling of the $YBa_2Cu_3O_{7-x}$ crystal by oxygen ions takes place by two different mechanisms. At the initial stage when we have a large deficit of oxygen ions ($(x) \geq 0.4$) the process of filling the crystal lattice by oxygen ions occurs in the mode of the "single file" diffusion [8]. This accelerated transport of oxygen ions is replaced by the usual classical diffusion mechanism at the final stage of the process, when the parameter $(x) \rightarrow 0.1$. Based on this fact, it is natural to assume that many of the relaxation processes observed in the studied crystal at room temperature, can also be controlled by these oxygen ions diffusion mechanisms. Oxygen is sufficiently contained in the conventional air atmosphere in which usually — most often — the crystals are stored. In this study, the relaxation of the electrical resistance of $ReBa_2Cu_3O_{7-x}$ (Re=Y, Ho) single crystals that is keeping for a long time (more than three days) at room temperature in air atmosphere is investigating (to prove the correctness of the above assumption).

The $ReBa_2Cu_3O_{7-x}$ (Re=Y, Ho) single crystals grown by gold crucible solution — melt technology as described in [9]. Measurement of the electrical resistance of the samples was carried out by a standard four-point scheme using two pairs of silver contacts. After cooling the samples until room temperature for 2–3 minutes, the electrical resistivity measurements were occurred. We subsequently repeated the measurements at intervals of about 5–7 h at a total exposure of the sample for three days.

The results of these measurements are presented in Fig. 1. As in [7], we can clearly observe the two-stage process of reducing the resistivity with an increase in the exposure time. At the initial stage (approximately 20 h), accelerated kinetics is recorded, which is replaced by a slower one, emerging to the saturation at exposure up to three days.

To explain this result, as in [7], we used the fact that at room temperature and above, the $ReBa_2Cu_3O_{7-x}$ (Re=Y, Ho) crystals are common semiconductors. Accordingly, an increase in oxygen ions concentration causes an increase in the electrical conductivity and the corresponding decrease in electrical resistivity ρ. At the same time, the change in the oxygen concentration in the crystal from the time t is described by the same law as the average displacement of the diffusing oxygen ions into the crystal: $L> = (2Dt)^{1/2}$ (where D is the diffusion coefficient) [10]. Consequently, we can judged from the $1/\rho^2 = f(t)$ the time dependence of



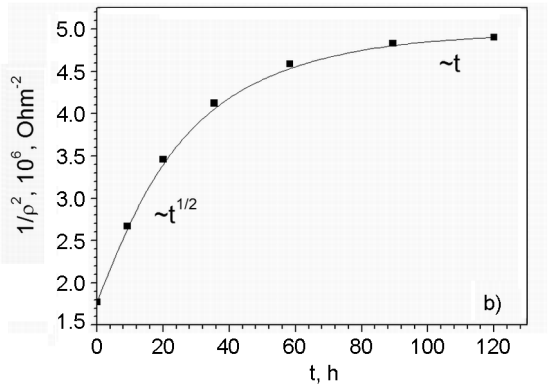


Fig. 2. The plots of $1/\rho^2 = f(t)$ dependence.

the mean square displacement of oxygen ions in the process of their diffusional introduction $<L^2> = f(t)$. In turn, the $<L^2> = f(t)$ dependence defines the ion diffusion mechanism: The $<L^2> \sim t^{1/2}$ dependence corresponds to a single file diffusion, and the dependence $<L^2> \sim t$ consistent with the common classical diffusion [8]. Using the experimental data presented in Fig. 1, we processed them by building the $(1/\rho^2) = f(t)$ dependence (see Fig. 2). From this figure, it follows that the initial stage of the relaxation process $\rho = f(t)$ is controlled by a single-file diffusion mechanism, and at the final stage this process occurs in the classical diffusion regime. Thus, in this case, a certain influence could have the specific mechanisms of quasiparticle scattering [11–15], due to the presence in the system kinetic and structural anisotropy.

In all high-temperature oxide superconductors, characterized by an oxygen ions deficiency, and in particular, in the $ReBa_2Cu_3O_{7-x}$ (Re=Y, Ho) compounds, relaxation of the electrical resistivity at room temperature is observed, controlled by a single file diffusion and by the classical oxygen ions diffusion. It should be noted, that some other relaxation processes that observed in the oxide high-temperature superconductors, can also be described by the same kinetic laws. As an example, we can mention the time dependence of temperature T^* at which the pseudo-gap opens, during exposure of the sample at room temperature (Fig. 3).

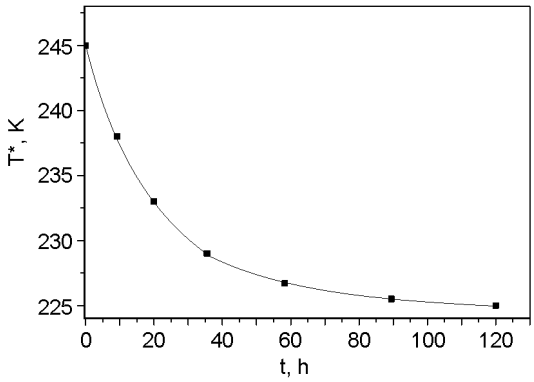


Fig. 3. Dependence of the temperature T^* at which the pseudo-gap opens, during exposure of the sample at room temperature in air atmosphere [6].

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