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Research Article

Electrical conductivity of $YBa_2Cu_3O_{7-\delta}$ single crystals under conditions of anionic ordering in $Cu(1)O_{1-\delta}$ layers

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Abstract

The influence of thermocycling annealing processes on the oxygen ordering degree (order parameter) in $YBa_2Cu_3O_{7-\delta}$ single crystals has been studied. It has been shown that an increase in the critical onset temperature of the transition to the superconducting state during thermocycling annealing procedures is consistent with the decrease of the σ_c/σ_{ab} parameter. This fact indicates a redistribution of the electronic density between the structurally inhomogeneous $Cu(2)O_2$ and $Cu(1)O_{1-\delta}$ planes, due to the formation of oxygen long-range order in the O(4)–Cu(1)–O(4) linear groups along the ***(b)*** crystal structure axis of the unit cell, and elimination of oxygen defects in the square nets of the $Cu(2)O_2$ planes. The existence of the critical value of the conductivity anisotropy σ_c/σ_{ab}, below which its behavior does not correlate with the change of T_c, has been confirmed. In this case an increase in T_c and orthorhombic distortion of the crystal structure during isothermal annealing are caused by the amplification of the "interlayer" interaction between the $Cu(2)O_2$ and $Cu(1)O_{1-\delta}$ planes. As a result, the contribution of the $Cu(1)O_{1-\delta}$ chain layers to the electron state density at the Fermi level increases. These layers can acquire superconducting properties due to tunneling of Cooper pairs from the $Cu(2)O_2$ planes resulting in the formation of the induced superconductivity in these planes.

Keywords

high-temperature superconductivity, $YBa_2Cu_3O_{7-\delta}$ single crystals, oxygen non-stoichiometry, electrical conductivity, order parameter.

1. Introduction

An urgent task in the research of high-temperature superconductivity is to improve the technology of high-quality specimens including the $YBa_2Cu_3O_{7-\delta}$ compound having reproducible superconducting properties and to study their physico-chemical properties. One condition of the existence of the superconducting state in cuprate compounds is that planes perpendicular to the crystallographic C_4 axis and those parallel to that axis should contain virtually square nets with minor rhombic distortion. The sites of the squares should be occupied by O^{-2} oxygen anions and their centers should accommodate variable valence copper cations, i.e., $Cu^{+1,+2,+3}$, the average valence evaluated from the length of the Cu–O bond being ~2.33 [1, 2]. Analysis of the dependence of the superconducting properties of $YBa_2Cu_3O_{7-\delta}$ on oxygen non-stoichiometry has shown that the critical temperature of the onset of the transition to the superconducting state (T_c) is controlled by the density of electronic states $N(E)$ at the Fermi level E_F which are in turn associated with the concentration of oxygen vacancies (δ) and their distribution in the $YBa_2Cu_3O_{7-\delta}$ structure [3–7].

T_c is known to depend on the concentration of mobile oxygen distributed in the chain $Cu(1)O_{1-\delta}$ planes and may

reach the highest level (~92 K) at $\delta = 0 \div 0.2$ [8]. This correlation is however not definitive since T_c may vary at a constant δ due to the effect of not only the concentration of oxygen vacancies but also their ordering in the anionic sublattice of $YBa_2Cu_3O_{7-\delta}$ crystals [9–14]. The order parameter of oxygen vacancies (ηv_0) is in turn controlled by temperature and annealing time and therefore affects T_c [15–18]. Thus ordering of oxygen vacancies in $YBa_2Cu_3O_{7-\delta}$ can be considered as one method to change the carrier concentration in the square nets of the $Cu(2)O_2$ structural planes which determine the superconducting properties of the compound [19–22].

Despite the large number of works on the topic, the ordering conditions of oxygen vacancies between the (0 1/2 0) and (1/2 0 0) structural planes in the $YBa_2Cu_3O_{7-\delta}$ anionic sublattice, especially at $\delta \to 0$, have been studied insufficiently yet. It is therefore an important task to evaluate the threshold temperature (T_{thr}, K) at which the energy of the thermal atomic oscillations becomes higher than the oxygen bond energy in the –Cu(1)–O(4)–Cu(1)–O(4)– chains and starts to violate the order of oxygen vacancies in the anionic sublattice.

2. Experimental

The $YBa_2Cu_3O_{7-\delta}$ single crystals were zone melt grown with directional mass transport in the system of $\{Ba_3Cu_5O_8 + xBaCuO_2\}/Y_2BaCuO_5$ diffusion pairs due to the component concentration gradient between the contacting layers [23, 24]. The Y_2BaCuO_5 and $BaCuO_2$ compounds were synthesized using high purity grade Y_2O_3, BaO and CuO oxides. The compounds were synthesized in thermal plants at 1220 K, 1270 K and $pO_2 = 0.21 \times 10^5$ Pa for $BaCuO_2$ and Y_2BaCuO_5, respectively. The temperature in the thermal plants was maintained using a RIF-101 high-precision temperature controller and was monitored with a Pt-Pt/Rh(10%) thermocouple accurate to ± 0.5 K. The as-grown single crystals had sizes of $1 \times 1 \times 0.5 \div 5 \times 4 \times 2$ mm^2, $\delta = 0.6$–0.7 and the superconducting parameters $T_c = 31$–36 K and $\Delta T_c = 11$–18 K, where $\Delta T_c = 90 \div 10$ % is the width of the temperature transition to the superconducting state. The structure of the single crystals was studied on a DRON-3 diffractometer in CuK_α radiation. The lattice parameters were determined using the asymmetrical method accurate to $\pm 5 \times 10^{-5}$ nm for $YBa_2Cu_3O_{7-\delta}$ powders.

Since the saturation rate and subsequent oxygen ordering in $YBa_2Cu_3O_{7-\delta}$ triple cuprate are far lower for single crystals and dense ceramics ($\rho = 6.0$–6.2 g/cm^3) than for moderate density ceramic specimens ($\rho = 4.4$–4.7 g/cm^3), the parameter value $\delta \le 0.1$ was achieved using three stage thermocycling annealing [25–27]. At the initial stage the $YBa_2Cu_3O_{7-\delta}$ crystals were annealed at 820 K for 25 h, and the second stage, at $T = 1020$ K for 2 h and at the third stage they were stepwise cooled at a 40–50 K/h rate within the 1020–870 K range and at a 1–5 K/h rate in the 870–720 K range. The electrical conductivity of the $YBa_2Cu_3O_{7-\delta}$ crystals was measured in the 77–800 K range using the four-probe method with platinum contacts.

3. Results and discussion

The highest diamagnetic response was observed in the crystal after the fourth annealing stage. In that crystal the diamagnetic response was 3.7 times higher than after single-stage annealing. Therefore the superconducting transition temperature increases and the transition width decreases as indicated by the single crystal magnetization temperature functions (Fig. 1). Further increase of the number of annealing stages did not improve the superconducting parameters of the crystals.

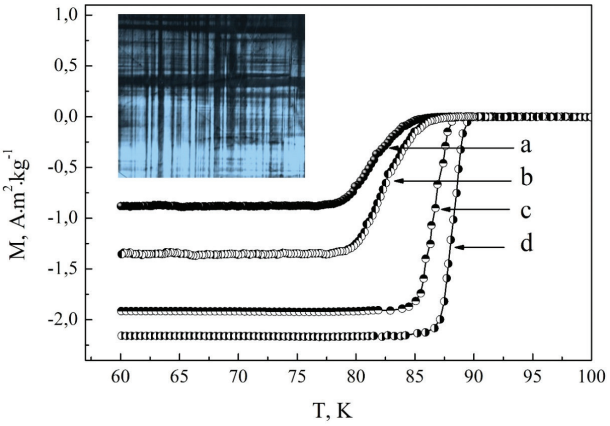

Figure 1. $YBa_2Cu_3O_{7-\delta}$ single crystal magnetization as a function of temperature after thermocycling annealing: a, b, c and d after the first, second, third and fourth annealing stages, respectively. Inset shows crystal surface image in polarized light.

These results combined with data on field functions of magnetization allowed evaluating the critical current of the crystal using the Bean model:

$$J_c = 20|-M^+ + M^-|/h, \quad (1)$$

where M^+ and M^- are the magnetizations of the crystal for opposite magnetic induction vectors of the outer magnetic field. Analysis of the field functions of magnetization showed that the plateaus on the hysteresis loops are almost symmetrical (Fig. 2). It is therefore sufficient to substitute $|-M^+ + M^-|$ in Eq. (1) for the double residual moment of magnetization M_{res} which equals to the crystal magnetization in a zero field after application of a strong magnetic field (14 T). Then the equation of the critical current density in the crystal takes on as follows:

$$J_c \approx 40\, M_{res}/h, \quad (2)$$

The magnetization curves $M(B)$ at $T = 7$ K in magnetic field B parallel to the (c) axis show that with an increase in the number of annealing stages, the areas of the hysteresis loops and hence M_{res} increase significantly (Fig. 2).

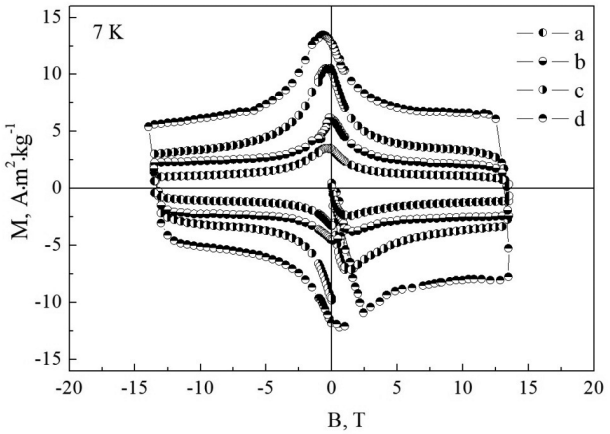

Figure 2. Field dependences of the magnetization of the $YBa_2Cu_3O_{7-\delta}$ single crystal; a, b, c and d after the first, second, third and fourth annealing stages, respectively.

In accordance with Eq. (2) the annealing process described above increases the critical current density $J_c \approx 0.68$; 1.21; 2.05; 2.59×10^4 A/cm^2 for the first, second, third and fourth annealing stages, respectively.

The effect of gas thermal annealing of the $YBa_2Cu_3O_{7-\delta}$ crystal on the concentration and ordering of oxygen vacancies in the (***ab***) plane between different crystallographic positions (0 1/2 0) and (1/2 0 0) was determined by measuring the electrical conductivity in different crystal directions: along the ***c***(σ_c) axis and in a direction parallel to the (***ab***) (σ_{ab}) plane. This study showed that thermocycling annealing of the crystals caused correlated changes in T_c and σ_c/σ_{ab} (Fig. 3). The decrease in the σ_c/σ_{ab} ratio due to a faster increase in σ_{ab}, than in σ_c is caused by different mechanisms of the effect of thermocycling annealing on the electrical conductivity of the crystal in different directions.

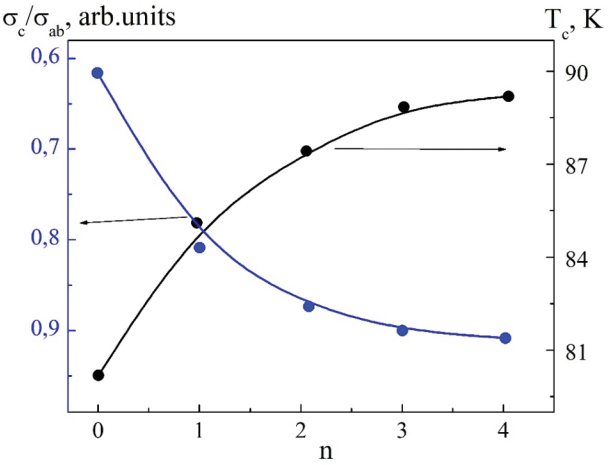

Figure 3. Influence of the number of thermocycling annealing processes on the anisotropy of conductivity and the onset temperature of the $YBa_2Cu_3O_{7-\delta}$ crystal transition to the superconducting state.

One can assume that the growth of σ_c is caused by an increase in the degree of covalence of the bond along the *c* axis of the $YBa_2Cu_3O_{7-\delta}$ lattice leading to an increase in the overlapping of the wave functions of electrons located on the Cu3d_{z2} orbitals of copper and the O2p_z orbitals of oxygen. This assumption is confirmed by a decrease in the lattice parameter along the (*c*) axis (Table 1).

Table 1. Dependence of the superconducting characteristics and crystal lattice parameters of the $YBa_2Cu_3O_{7-\delta}$ single crystal on the number of thermocycling annealing stages (*n*).

n	T_c, K	ΔT, K	$\Delta_{(b-a)}$, nm	(*c*), nm	ηv_0	δ
1	84.2	4	0.0052	1.17085	0.3333	0.15
2	87.1	2	0.00572	1.17034	0.3666	0.13
3	88.5	1.5	0.00597	1.17010	0.3826	0.11
4	89	1	0.00606	1.17001	0.3884	0.10

The increase in the electrical conductivity σ_{ab} after thermocycling annealing is caused by a redistribution of the electronic density from the square nets of the $Cu(2)O_2$ layers to the $Cu(1)O_{1-\delta}$ chain layers which leads to an increase of $N(E)_F$ in $Cu(2)O_2$. The redistribution of the electronic density is affected by the concentration and ordering of oxygen vacancies along **(*a*)** or an increase in the occupation density of (0 1/2 0) crystallographic positions by oxygen anions leading to an increase in the orthorhombic distortion $\Delta_{(b-a)}$.

After constant T_c and σ_c/σ_{ab} were achieved we started isothermal annealing in the 720–560 K range at $pO_2 = 5 \times 10^5$ Pa for 15 h. T_c increased at ΔT_c = const at temperatures below the threshold one T_t = 600 K (Fig. 4). Furthermore σ_c/σ_{ab} remained constant during isothermal annealing in the 660–560 K range whereas T_c increased. The increase in T_c can be arbitrarily split in two regions I and II with T_c increasing faster in the region I than in the region II (Fig. 4).

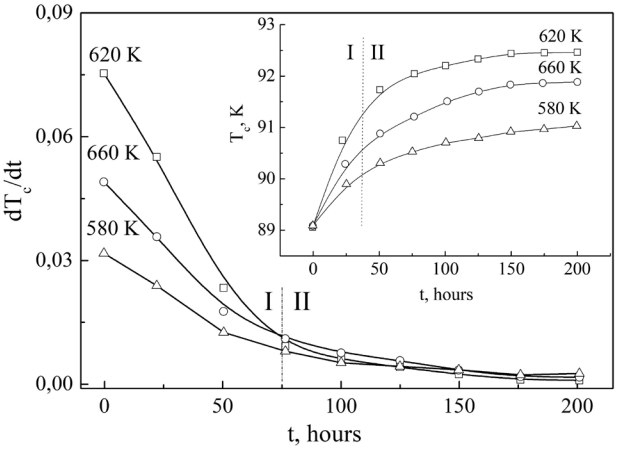

Figure 4. Kinetic dependence of the superconducting transition onset temperature (T_c, K) for the $YBa_2Cu_3O_{7-\delta}$ crystals annealed at $pO_2 = 5 \times 10^5$ Pa and at various temperatures under isothermal conditions.

For determining T_c as a function of oxygen vacancy concentration and ordering, the order parameter was introduced which depends linearly on the orthorhombic distortion $\Delta_{(b-a)}$ and is expressed analytically as $\Delta_{(b-a)} = \alpha\eta v_0$, where α is the proportion coefficient. This latter proportion

coefficient is calculated for the maximum value $\max(\Delta_{(b-a)})$ = 0.00780 nm for the stoichiometric composition of $YBa_2Cu_3O_{7-\delta}$ corresponding to η_{max} = 0.5 [28]. The T_c parameters of the thermocycled crystals are more sensitive to the concentration of oxygen vacancies (δ) than to their ordering (ηv_0) (Tables 1, 2). During isothermal annealing in the 660–560 K range the ordering of oxygen vacancies makes the largest contribution to the changes in T_c. Then only ηv_0 change whereas δ = const. An increase in ηv_0 is caused by the ordering of oxygen anions accompanied by an increase in the length of the –Cu(1)–O(4)–Cu(1)–O(4)– chain fragments. This is auspicious for an increase in the covalence degree of the bonds along the structural direction *c*, a decrease in the length of the –Cu(1)–O(1)–Cu(2)– bond with a redistribution of the electronic density from the square nets of the $Cu(2)O_2$ layers to the $Cu(1)O_{1-\delta}$ chain layers and an increase in the free carrier concentration at the antibonding $Cu3d_{(x2-y2)}$-$O2p_{xy}$ hybridized orbitals. The difference in the T_c growth rates between the regions I and II stems from the fact that oxygen ordering in the –Cu(1)–O(4)–Cu(1)–O(4)– chains along the **(*b*)** axis (region I) requires atomic movements through an order of one interatomic distance whereas for the region II long chain ordering along the **(*b*)** axis requires anion movements through quite large distances.

Table 2. Dependence of the superconducting characteristics and crystal lattice parameters of the $YBa_2Cu_3O_{7-\delta}$ single crystal on the temperature of isothermal annealing.

Isothermal Annealing (*T*, K)	T_c, K	Δ*T*, K	$\Delta_{(b-a)}$, nm	(*b*), nm	ηv_0	δ
660	91.7	1	0.0691	1.16900	0.4429	0.06
620	92.4	1	0.0750	1.17036	0.4807	0.06
580	90.7	1	0.0683	1.16932	0.4378	0.07

Analysis of the change in the anisotropy of the electrical conductivity after thermocycling and isothermal annealing showed that the increase in T_c does not necessarily correlate with the changes in σ_c/σ_{ab}. On the one hand the increase in T_c for the heat treated specimens can be accounted for by an increase in the free carrier concentration in the $Cu(2)O_2$ planes and an increase in the strength of the interlayer interaction (σ_c/σ_{ab}) between the $Cu(2)O_2$ and $Cu(1)O_{1-\delta}$ planes. On the other hand annealing at 660–560 K and $pO_2 = 5 \times 10^5$ Pa increases T_c of the crystals without changing their σ_c/σ_{ab}. This suggests that the anisotropy of the electrical conductivity of the $YBa_2Cu_3O_{7-\delta}$ single crystals depends on the oxygen nonstoichiometry parameter δ and is not determined by ηv_0. It is safe to assume that annealing at 660–560 K and $pO_2 = 5 \times 10^5$ Pa changes the mechanism that controls the superconducting properties of the crystals. Then the increases in σ_{ab} and T_c originate from the ordering of oxygen ions and are caused by the contribution of the $Cu(1)O_{1-\delta}$ chain layers to the electronic density of states at the Fermi level. The $Cu(1)O_{1-\delta}$ chain layers can be superconducting due to the proximity effects, and this fact makes possible the existence of induced superconductivity in these layers due to tunneling of Cooper pairs from the $Cu(2)O_2$ planes.

4. Conclusion

Study of the regularities of oxygen interaction with yttrium/barium cuprate single crystals for the first time justified the necessity of using multistage gas thermal treatment in order to increase the superconducing parameters of $YBa_2Cu_3O_{7-\delta}$ due to intentional impact on oxygen sorption and ordering processes in its anionic sublattice.

An increase in the critical onset temperature of the transition to the superconducting state during this annealing is consistent with the decrease of the σ_c/σ_{ab} parameter. This fact indicates a redistribution of the electronic density between the structurally inhomogeneous $Cu(2)O_2$ and $Cu(1)O_{1-\delta}$ planes, due to the formation of oxygen long-range order in the O(4)-Cu(1)-O(4) linear groups along the **(*b*)** crystal structure axis of the unit cell, and elimination of oxygen defects in the square nets of the $Cu(2)O_2$ planes.

The existence of the critical value of the conductivity anisotropy σ_c/σ_{ab}, below which its behavior does not correlate with the change of T_c, was confirmed. In this case an increase in T_c and orthorhombic distortion of the crystal structure during isothermal annealing are caused by the amplification of the "interlayer" interaction between the $Cu(2)O_2$ and $Cu(1)O_{1-\delta}$ planes. As a result, the contribution of the $Cu(1)O_{1-\delta}$ chain layers to the density of electronic state at the Fermi level increases. These layers can acquire superconducting properties due to tunneling of Cooper pairs from the $Cu(2)O_2$ planes resulting in the formation of the induced superconductivity in these planes.

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