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Influence of Short-Term Heat Treatment on Superconducting and Normal-State Properties of Polycrystalline $YBa_2Cu_3O_x$

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Abstract. In this study, the change of superconducting and normal characteristics was studied in (77-290)K temperature range by means of determining the resistivity of the polycrystalline $YBa_2Cu_3O_x$ sample stored in room conditions for 30 years after a short-term heat treatment at 400°C and slowly freezing. It has been shown that in this case, the heat treatment does not affect the width of the superconducting transition of the sample, but results in a significant increase in its resistivity in the fluctuating region and a slight reduction in the critical T_c temperature of the superconducting transition. It has been found out that both before and after the heat treatment a temperature $T^* \gg T_c$ is observed in the sample below which the pseudogap mode is established characterized by a faster reduction than the linear resistivity. Moreover, if the T_c is reduced by only one degree after the heat treatment, then the T* decreases by 26.5K, i.e. the narrowing of pseudogap mode temperature range and, hence, the extension of metal behavior temperature range is observed. In addition, the resistivity and its temperature slope are substantially reduced in comparison with the initial one from 110K to room temperature. These results are qualitatively interpreted within the frames of the redistribution model of defects previously available in the Cu-O chains and planes as well as new structural ones of the spatial lattice of the sample caused by the heat treatment.

Keywords: high-T_c polycrystalline $YBa_2Cu_3O_x$, critical transition temperature (T_c), fluctuation conductivity, pseudogap state

1. Introduction

The study of the stability of heat treatment of high temperature superconducting (HTSC) critical and electro-transport characteristics is an important fundamental and practical problem for modern solid-state physics. On the one hand, this is due to the fact that in recent years the production of technological equipments has been intensively developing on the basis of HTSC, and the electrophysical characteristics stability issue acquires importance. On the other hand, though over 30 years have passed since the detection of HTSC, the superconducting mechanism in them has not been revealed so far at the microscopic level. According to some viewpoints, the unusual features observed in the normal state near the critical temperatures and at elevated temperatures can be the key factor to detect the HTSC superconducting mechanism. The fluctuation conductivity [1-4] arising in wide temperature range of HTSC compounds, the existence of the so-called pseudogap anomalies [5-11], metal-to-dielectric transitions [12,13], incoherent electro-transport [6,11] and other phenomena can be classified among them. The listed properties can be influenced by the heat treatment, but the extent of that effect is considerably dependent both on its performance conditions (temperature, duration, heating and freezing speeds), and the prehistory of the sample preparation [1-25]. From this point of view, the low temperature processing is of great interest, which stimulates the appearance of metastable conditions in the sample [10,11,15-25] not changing its oxygen content, while simultaneously causing the change of the weak bound oxygen atoms arrangement degree in the elementary cell, due to their high mobility [23]. It should be noted, that according to [14], the change in the characteristics of superconducting compound of yttrium type, stored in room conditions for a long time (10-year period), corresponds to the changes occurred after being heated for 100 hours at 200^0C. The phenomenon of aging has been studied in various superconducting compounds after having been stored up to 17 years in room conditions [11, 15-25]. The study of the effects of the heat treatment on the prolonged aged samples is important both in revealing the superconductivity mechanism and in restoring their initial characteristics [20, 21, 22]. The number of similar research works is small and some of the obtained results, such as the reaction to the nature of the origin of pseudogap mode and various factors, sometimes have a controversial character [8, 11, 23, 24]. Recently, the short-term low-temperature heat treatment effect on electro-transport and superconducting properties of yttrium samples stored in room conditions for approximately 30 years after the synthesis has been studied by us in the $(77-92)K$ temperature range [26].

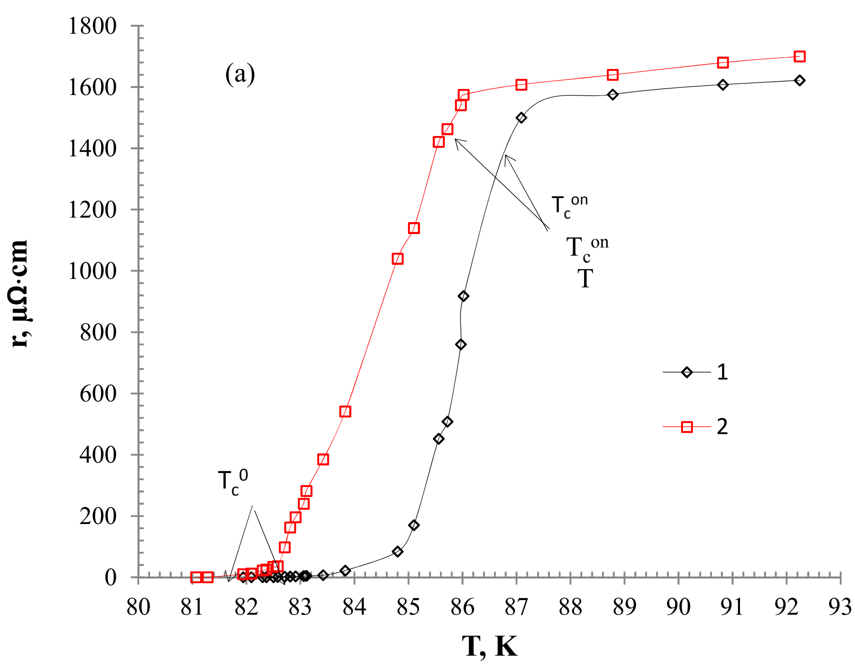
The goal of the current research work is to continue and expand the temperature range of those studies up to $290K$, paying special attention to the manifestation of the pseudogap state.

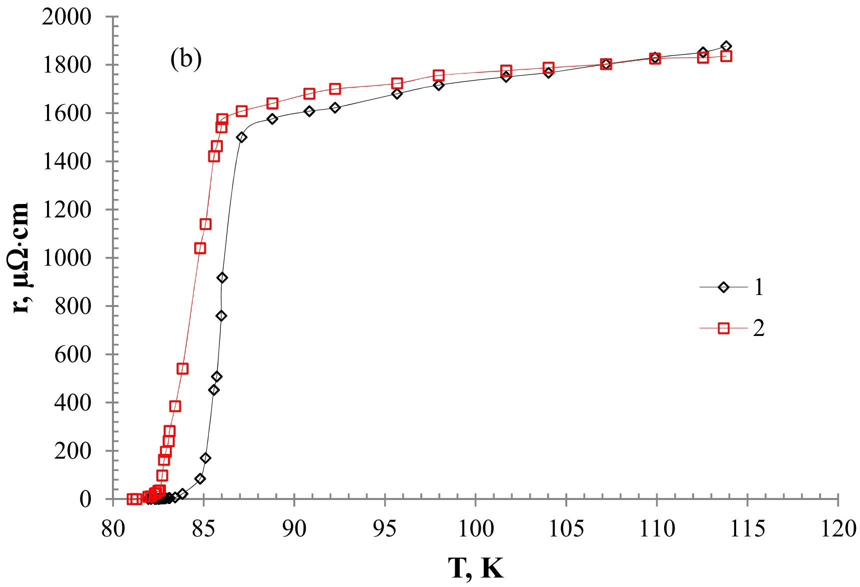
2. Materials and Technique

The investigated HTSC polycrystalline $YBa_2Cu_3O_x$ compound has been synthesized in the furnace by ordinary method for 10 hours at 960^0C by means of heat treatment in the air [22, 25-28] and stored in room conditions for approximately 30 years. The X-ray fluorescence measurements have shown that the weight concentration of barium atoms in the sample is approximately 3 percent less than that of the samples prepared by us at 950^0C [22, 25,28]. The sample was frozen slowly (3-4 degrees / minute) together with the furnace after an additional heat treatment in the air for 30 minutes at 400^0C and during a month in the range of $(77-290)K$ regularly determined the resistivity temperature dependence of r(T) curves using the four-contact method for measuring the current-voltage characteristics [22,25-28]. These measurements determine the superconducting and normal characteristics of the sample before and after the heat treatment. In these states the samples are numbered as 1 and 2. It should also be noted that the characteristics of sample 2 a month later change very slowly depending on time, i.e. stabilization takes place in the sample. Thus, in this research work after the heat treatment only the characteristics of the stabilized state of sample 2 are given, and the results of their time dependence will be published in future works. The width of the superconducting (SC) transition state was determined as follows: $\Delta T_c = T_c^{0.9} - T_c^0$, where the $T_c^{0.9}$ and $T_c^{0.1}$ are the temperatures corresponding to the normal state resistivity level $r_n\,(0.9\div0.1)$. $T_c^{0.9}$ and $T_c^{0.1}$ are respectively the onset and offset (zero resistance) critical temperatures T_c^{on} and T_c^0 of the superconducting transition state.

3. Results and Discussion

Figs.1 (a, b, c) show the temperature-dependence curves of the resistivity in three different temperature ranges before and after the heat treatment (1 and 2 curves, respectively). As it is seen from this picture, in both cases the resistivity (r) of the sample in normal (non-superconducting) state manifests a linearly diminishing character typical of metals, as the temperature drops to a specific temperature T^*. However, at lower temperatures a faster decrease is observed than the linear dependence of r. It should be noted that the heat treatment in the $T_c^0 < T < T_c^{on}$ range leads to a much greater increase in the resistivity of the sample (up to a dozen times) than at high temperatures (Fig.1, 1 and 2 curves.). This increase is accompanied by decrease in T_c^{on} and T_c^0 by only one degree, as well as the decrease in the ratio of the resistivity at $290K$ and $100K$ from 1.56 to 1.25 (see table, fig. 1a, 1 and 2 curves). The conductivity in the given region is conditioned by the temperature fluctuations of the density of Cooper pairs [1-3] (fig. 1, 1 and 2 curves). Let's note that if the dr/dT slope in the temperature range of $T > T^*$ before the heat treatment is $5.9\mu\Omega \cdot cm / K$, then after the heat treatment it becomes $3.00\mu\Omega \cdot cm / K$, which is accompanied by the monotonic decrease of its resistivity, too. This decrease is observed at temperatures above $110K$, which reaches up to 18.5% at $290K$ (see table, Fig. 1 c, curve 2).





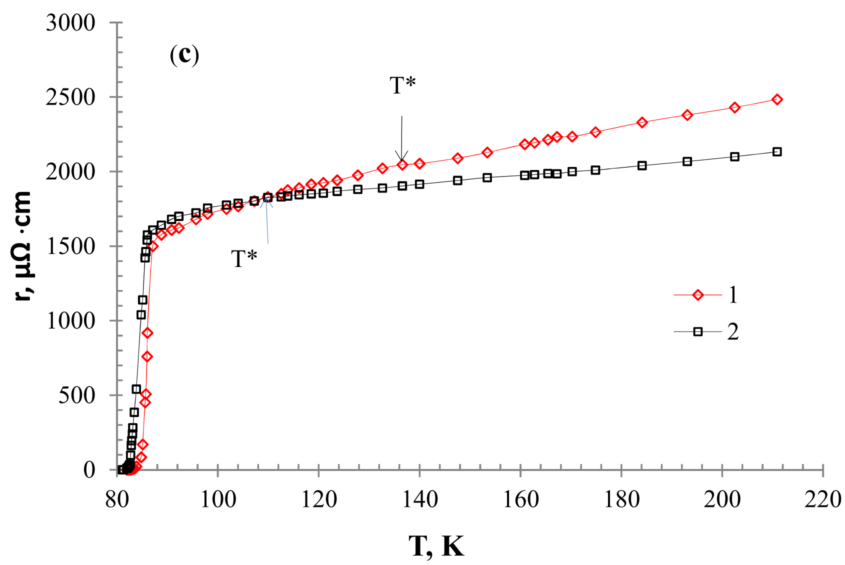


Fig 1. The temperature dependence of the resistivity (r) for samples 1 and 2 in different temperature ranges (a, b, c). The arrows point to the pseudogap mode temperatures (T*), as well as critical transition temperaturs (T_c^{on}, T_c^0).

Table: Some characteristics of the studied SC samples and their normal state

sample	T_c^{on}, K	T_c^0 , K	r(290K), μΩ· cm	r(100K), μΩ· cm	dr/dT, μΩ· cm/K	T*, K	ΔTc, K
1	86.7	82.5	2700	1733	5.90	136.5	4.2
2	85.6	81.4	2200	1765	3.00	110	4.2

Similar behavior of the resistivity r and its temperature slope dr/dT was also observed in $YBa_2Cu_3O_{6.55}$ and $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals both after the application of high pressures [29] and impurity substitution [30], which is due to the reduction of the role of phonons during the scattering process of normal current carriers [30]. However, the resistivity (fluctuating conductivity) in the low temperature range is conditioned by the weak bound O_4 and O_1 atoms arrangement in the Cu-O chains, and at high temperatures – by strong bound oxygen atoms arrangement in Cu-O planes [14-18,31,32]. Since the chain oxygen atoms are weaker bounded compared to plane atoms, the heat treatment in the chains results in greater number of defects that contributing to the weakening and destruction of Cooper pairs, lead to the above-mentioned great increase of the resistivity in low temperature range. However, if such a great increase of resistivity in the sample is accompanied by the critical transition temperature T_c reduction by only 1 degree (see table), then the T^* temperature in that case decreases from $136.5K$ to $110K$. The following facts should be considered for the explanation of the obtained results. It is interesting to note that after the low temperature heat treatment, in long-term stored samples in room conditions [14] with the presence of water vapors in the air, defects occur in the oxygen, Y and Ba sublattices of HTSC elementary cell. The characteristic T^* temperature is associated with the establishment of pseudogap mode, with two approaches regarding its origin. According to the first one, it is conditioned by the so-called "dielectric" type of short-range fluctuations that are observed in the underdoped compounds [5, 33]. The second approach refers to the formation of Cooper pairs in the temperature range of $T^* \gg T_c$, which is much higher than the critical one, where the density of local SC pairs is still small enough and does not provide the necessary coherence so that the sample can transit into SC state with the entire volume. This coherence is established only in the case of $T < T_c$ [1-4]. The reduction of T^* is caused by defects occurred both in the chains and planes of spatial lattice. The increase of defects density hinders the formation of superconducting pairs [1]. However, it is interesting to note that in [8, 11, 23, 24] the opposite result has been observed: the increase of T^* by 29 degrees has been observed in pure and partially aluminum doped yttrium type crystalline samples prolonged stored at room temperature. In other words, in HTSC samples the influence of defects on the change of T^* and T_c is not the same [8, 9, 23], and as recent evidence suggests, the nature of the origin of the pseudogap has not been completely determined yet [3, 4, 10, 33]. In addition, under different influences T^* changes to a much greater extent than T_c [8, 9, 11, 23]. On the other hand, the low-temperature heat treatment in the yttrium superconductors stimulates an unstable state stipulated by disorder and their subsequent relaxation of high-mobility chain oxygen atoms [15-19]. The major defects occurring after the long-term heat treatment at low temperature are antisite interchangable atoms of Ba and Y that lead to the re-arrangement of oxygen atoms, which, in its turn, implies changes in the sample characteristics. These defects have dimensions of nanometers and are distributed in the elementary cell strongly inhomogeneous, which can enhance the appearance of pseudogap anomalies even with optimal oxygen content in HTSC $YBa_2Cu_3O_{6.93}$ single crystals, though, according to the previously accepted view, these phenomena are observed in samples with oxygen deficiency [10,11,33]. And a much weaker change in T_c, as a result of heat treatment, compared to T^* can also be explained by the occurrence of nanodefects in the sample. It is known that T_c's change is largely conditioned by the change in the populations of oxygen $O4$ sites. However, according to [10], the occurrence of the above-mentioned antisite defects contributes to the fact that the population of oxygen $O4$ sites changes slightly, whereas in the chain $O5$ positions (which are vacant in an ideal state) it increases, resulting in the maintenance of high T_c values or their small change. Further researches are needed to provide a final and unambiguous interpretation for the obtained results.

4. Conclusions

By determining the temperature dependence curves of the resistivity, the electro-transport properties of the HTSC $YBa_2Cu_3O_x$ compound, synthesized and 30-year stored in room environment, were investigated before (sample 1) and after (sample 2) short-term heat treatment. The main obtained results can be summarized as follows:
After the 30-minute heat treatment at 400^0C the resistivity of the sample 2, slowly frozen up to room temperature, increases in the low temperature range for dozens of times as compared to sample 1, which is simultaneously accompanied by the decrease of the critical temperature by only one degree, whereas the transition width remains unchanged $-4.2K$. Meanwhile, the reduction of T^* temperature of the formed local pairs is observed from $136.5K$ to $110K$, which is explained by the defects occurred in Cu-O weak bound chains. The huge increase of the resistivity is also due to these defects in the fluctuating mode. It should also be noted that after the heat treatment the resistivity of the sample is considerably reduced in the temperature range $T^* - 290K$, which is caused by the reduction of the role of the phonons on the scattering of current carriers. The obtained results will allow the use of a sufficiently simple method to correlate with the superconducting and normal characteristics of the compound, which will enable to obtain compounds with high performance characteristics which are of great importance in the use of power and measuring techniques.

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