Effect of Partial Doping with La Atoms on Some Superconducting and Electrotransport Characteristics of Y_{1-x} La_xBa₂Cu₃O_y Polycrystals

S.K. Nikoghosyan^{1,2}, A.G. Sargsyan¹, Y.G. Zargaryan¹, A. Aivazyan¹, A. Hakopyan¹

¹International Scientific-Educational Center of NAS RA, Armenia, 0019, Yerevan, M. Baghramyan 24d ²A.I. Alikhanyan National Science Laboratory (Yerevan Physics Institute) Foundation, Armenia, 0036, Yerevan, Alikhanyan Yeghbayrneri St., 2 Building

Email: yerjanik.zargaryan@isec.am

(Received: November 21, 2023; Revised: December 15, 2023; Accepted: December 23, 2023)

Abstract. The effect of the location with different concentrations of La atoms (x = 0; 0.02; 0.028; 0.11) on some superconducting and electrotransport characteristics of $Y_{1-x}La_xBa_2Cu_3O_y$ polycrystalline compounds was studied by recording the temperature dependence $\rho(T)$ curves of resistivity in the case of direct current in the range of temperatures (77K - 300K). The temperatures at the onset (T_c^{on}) and offset (T_c^0) of the transition, as well as the width of the transition - $\Delta T_c = T_c^{on}$ - T_c^0 , were considered as the superconducting characteristics. The absolute values of $\rho(300K)$ and $\rho(100K)$ and their ratio $a = \rho(300K)/\rho(100K)$ were considered as electric transport characteristics, as well as the temperature coefficient of resistivity d ρ/dT , which was determined by $\rho(T)$ in the range of linear dependence with the slope of the curve. From the comparison of these characteristics, it was concluded that there is a certain correlation between the superconducting and electrotransport properties of the studied compounds depending on the concentration *x* of the partial location of Y atoms with La.

Keywords: high temperature superconductor, superconducting transition, superconducting transition width, onset and offset temperature of superconducting transition (ΔT_c , T_c^{on} , and T_c^{0}), phase separation phenomenon

DOI:10.54503/18291171-2023.16.4-114 **1. Introduction**

The mechanism of superconductivity actually operating in high-temperature superconductors (HTSCs) at the microscopic level has not been elucidated so far. It is an important fundamental problem both from the point of view of solid-state physics and its application. The application of HTSCs is limited due to their still low values of critical temperature (T_c) and current (J_c) [1, 2]. Therefore, it is a topical issue to study the changes in the critical characteristics of $YBa_2Cu_3O_y$ (Y-123) systems under the influence of various factors, which will contribute to the discovery of the mechanism of superconductivity, as well as the obtaining of compounds with higher characteristics [2-5]. In order to change the critical characteristics of Y-123 superconductors, hydrostatic pressure or displacement of Y atoms with other rare earth elements (R) is applied ("internal" or "chemical pressure") [3-5]. It is known that the nature of the change in critical and structural characteristics of the $Y_{1-x}R_xBa_2Cu_3O_y$ compound largely depends on the ionic radius of R, its concentration (x) and valence [1, 3, 4, 6-10]. Thus, it is important to study the effects of Pr and La in Y-123 among the R elements that replace Y, because the first of them can appear in both trivalent and tetravalent states, and the second only in trivalent. [1, 3, 5, 10-12]. In addition, the ionic radius of La is larger than the ionic radii corresponding to those two valence states of Pr, as well as the ionic radius of Y [3]. It should also be noted that the ion of Pr has its own large magnetic moment, while La does not, which in the case of Pr, unlike La, can significantly worsen the superconducting characteristics of Cooper pairs [1, 7, 8, 11, 13]. In our previous work, the effect of Pr and La on the superconducting and electrotransport properties of $Y_{0.89}R_{0.11}Ba_2Cu_3O_{7-\delta}$ compounds was studied [14]. It was found that the temperature of the superconducting transition for Pr is much smaller compared to La, and

the transition is significantly narrower. Moreover, if in the first case, the resistivity in the fluctuation range shows incomparably large values, then in the normal state, contrary to the result obtained in [1], at the same concentration of La (x = 0.11), it is significantly smaller, which is the reason has not yet been fully dissected [14].

In the present work the effect of partial substitution of Y atoms with smaller concentrations of La atoms on some superconducting and electrotransport characteristics of $Y_{1-x}La_xBa_2Cu_3O_y$ (x = 0; x= 0.02; 0.028; 0.11) polycrystals is also studied.

2. Experimental methods

The studied solid solutions of the type $Y_{1-x}La_xBa_2Cu_3O_{7-\delta}$ (x=0; 0.02; 0.028; 0.1) were synthesized by standard solid phase technology and for this purpose Y₂O₃, La₂O₃, CuO and BaCO₃ were used as starting materials [1]. Appropriate amounts of the indicated materials, after thorough mixing in an agate mortar, were pressed into disc-shaped pellets and synthesized at 930°C. The time required for the synthesis process, including seven intermediate grindings and pressings, was 160 hours. It should also be noted that after the synthesis was completed, the samples were further heated at 300°C for 3 hours and then slowly cooled in the furnace to room temperature to achieve oxygen saturation. The long procedures of the synthesis process contributed to the deposition of La in the Y junctions [1]. The curves of the temperature dependence of the resistivity $\rho(T)$ were measured by the standard four-contact method at a constant current of 10 mA in the range of temperatures (77-300)K [14-17]. The samples prepared for this purpose have the following concentrations of La: x = 0: 0:02: 0.028 and 0.11, which will be numbered as 0. respectively; 1; 2; and 3. They have a rectangular cut and characteristic dimensions of 1.8 mm×1.7 mm×4 mm, and the distance between the potential contacts was 2 mm. The onset (T_c^{on}) and offset (T_c^{0}) of transition temperatures to the superconducting state were determined by the drop in the normal state resistivity (ρ_n) of the sample by 10 and 90%, respectively, and the width of the transition: $\Delta T_c = T_c^{\text{on}} - T_c^0$ [14-17]. At the same time, T_c^{on} is the temperature of the beginning of the transition to the superconducting state of the grains, and T_c^{0} is the temperature of the establishment of complete superconducting connections between them, when the resistivity of the sample finally disappears. In order to gain an additional understanding of the electrical transport processes taking place in the samples, the absolute values of $\rho(300K)$ and $\rho(100K)$ and their ratio [a = $\rho(300K)/\rho(100K)$], as well as dp/dT (slopes) determined from the range of linear dependence of the ρ (T) curves have been compared with superconducting characteristics [14, 15]. From that combination, one can get an idea about the arrangement of the elementary cell of the studied sample [14, 15, 18-23].

3. Results and discussions

The dependence curves of some superconducting and electrophysical characteristics of the studied samples $Y_{1-x}La_xBa_2Cu_3O_y$ (x = 0; x = 0.02; 0.028; 0.11) on the concentration *x* of La alloy atoms are shown in Fig.1 and Table 1. As can be seen, as *x* varies from 0 to 0.02, 0.028, and 011, T_c^{on} continuously decreases from the initial 91.7K to 90.2K, reaching a minimum of 88.6K when x = 0.028 and increases again to 90.5K. Meanwhile, for those same concentrations, T_c^{0} first increases from 87.5K to 88.5K, then decreases to a minimum of 84.8K and increases again to 86.5K. As *x* increases in the range $0 \le x \le 0.028$, T_c^{on} decreases, which is accompanied by an increase in $\rho(300K)$ and $\rho(100K)$, as well as a monotonous decrease in their ratio *a*. Also, at x = 0.028, $\rho(300K)$, $\rho(100K)$ and $d\rho/dT$ reach their maximum values (4060 $\mu\Omega \cdot cm$, 3150 $\mu\Omega \cdot cm$ and 3.91 $\mu\Omega \cdot cm/K$) respectively, then T_c^{on} and T_c^{0} , on the contrary, to their minimum values (88.6K and 84.8K). Note that as x increases, ΔTc decreases sharply from 4.2K (x = 0) and shows a minimum

value of 1.7K at x = 0.02, and continues to increase up to 3.8K and 4K for x values of 0.028 and 0.11, respectively.

Sample number	Х	dρ/dT, μΩ·cm/K	ρ(300K), μΩ·cm	ρ(100K), μΩ·cm	<i>a</i> , A.U.	T _c ^{on} ,K	T _c ⁰ ,K	$\Delta T_c, K$
0	0	2.13	461	243	1.89	91.7	87.5	4.2
1	0.02	1.51	980	650	1.51	90.2	88.5	1.7
2	0.028	3.91	4150	3060	1.36	88.6	84.8	3.8
3	0.11	2.36	2700	2150	1.26	90.5	86.5	4
Pr*	0.11	2.67	1720	1180	1.46	83.6	80.5	3.1

Table 1. Some superconducting and electrotransport characteristics of $Y_{1-x}La_xBa_2Cu_3O_y$ compounds.

(* for comparison, the last line also shows the characteristics of Pr when x = 0.11)



Fig. 1. Dependence of some superconducting $(T_c^{on}, T_c^{0}, \Delta T_c)$ (a) and electrophysical [$\rho(300K)$, $\rho(100K)$, a, d ρ/dT] (b) characteristics on concentration x of La atoms in $Y_{1-x}La_xBa_2Cu_3O_y$ compounds.

When La concentration $x = 0.11 \Delta T_c = 4 \text{ K}$, that it approaches its starting value (4.2K), which is accompanied by a further increase in the critical temperatures T_c^{on} and T_c^0 (90.5K and 86.5K), as well as with the simultaneous decrease of $\rho(300K)$ and $\rho(100K)$, slope (dp/dT) and ratio a (see Fig.1 and Table 1). Note, however, that when x varies from 0 to 0.11 in the $Y_{1-x}La_xBa_2Cu_3O_y$ samples, all characteristics exhibit non-monotonic behavior, while a monotonically decreases. The decrease in *a* means that the concentration-dependent $\rho(100K)$ increases faster than $\rho(300K)$ (see Table 1). On the other hand, the observed increase in ρ and T_c^{0} characteristics, as well as the accompanying decrease in ΔT_c , T_c^{on} and $d\rho/dT$ when x = 0.02 can be explained as follows. The increase of La leads to an increase in the elastic deformation of the spatial lattice of the sample, because it substitutes the atoms of Y with a much smaller radius [3]. This leads to a change in the distances between the atoms of the spatial lattice, which causes new defects in the oxygen sublattice and a rearrangement of the existing ones, which determines the increase in the absolute values of $\rho(300K)$ and $\rho(100K)$. Moreover, the defects formed in the grains lead to a decrease in T_c^{on} by 1.5 degrees on the one hand due to the increase of Cu₂-O₃ interatomic distances in the Cu-O planes, and on the other hand to an increase in T_c^0 by 1 degree due to the reduction of the distances between these planes [3]. The latter means the strengthening of intergranular weak bonds, which ultimately leads to a decrease in ΔT_c from 4.2K to 1.7K. All this is accompanied by a significant decrease of a and $d\rho/dT$ (see Table 1). The decrease of $d\rho/dT$ indicates that the role of the electronphonon interaction in the scattering of current carriers decreases, which is probably related to the decrease in the frequency of the phonons responsible for the Raman scattering [3]. A further increase in La concentration leads to a sharp increase in p of the sample and the appearance of a peak at x = 0.028, which is also accompanied by a sharp increase in dp/dT and ΔT_c , which coincidentally means the role of electron-phonon interaction and inhomogeneity increase. It is a consequence of the fact that the addition of La promotes the formation of different superconducting (with T_c close to each other) and non-superconducting phases in the sample and their subsequent coexistence. It is known as the "phase transition phenomenon" and is also manifested in sample 3 (x = 0.11) [3, 14]. Y_{1-x}La_xBa₂Cu₃Oy and La123 compounds act as superconducting phases, and BaCuO₂ is not superconducting [3,14]. However, in this case (x = 0.11), this phenomenon is manifested by a simultaneous sharp decrease in ρ and $d\rho/dT$, which is also accompanied by an increase in T_c^{on} , T_c^0 and ΔT_c , which are close enough to their origin values in the pure sample. It turns out that T_c is much less affected by La than Pr [1-14, 19-21]. If for T_c in the range $0 \le x \le 0.11$ this change is only a few degrees, then for ρ it is more than one order. Note that ΔT_c drops sharply from the initial value of 4.2 K (x = 0) to a minimum (1.7 K) when x = 0.02, and with the further increase of x it reaches 4 K (x = 0.11) (see Table 1). Interestingly, the resistivity exhibits a maximum as La increases, accompanied by a minimum in ΔT_c . However, the maximum of ρ is observed at the concentration value x = 0.028, and the minimum of ΔT_c at the concentration x =0.02. If the presence of Pr leads to a strong reduction of T_c at the concentration x = 0.11, then the resistivity around T_c is 2.5 orders of magnitude higher than for sample containing La with the same concentration [14]. Meanwhile in the normal state it is significantly reduced compared to the sample containing La. As can be seen from the table, this difference in the case of a lower content of La (x = 0.028) is much greater than in the case of a high concentration of Pr (x = 0.11) [14]. Such behavior in the fluctuation region, which was previously observed by other authors as well, is expressed by the large influence of T_c and p on Pr, is known as the "anomaly of Pr ", and its nature has not yet been definitively determined [10, 14]. And in the normal state, the smaller value of p shown by our sample containing Pr compared to La, can also be classified as an additional "anomalous" phenomenon. One of the possible reasons for this may be that Pr, in contrast to La, has an intrinsic magnetic moment, the effect of which on carrier scattering at high temperatures is probably greatly reduced [14].

Conclusions

Thus, the influence of La introduction on some superconducting and electrotransport characteristics of $Y_{1-x}La_xBa_2Cu_3O_y$ (x = 0; x = 0.02; 0.028; 0.11) compounds was studied using the temperature dependence $\rho(T)$ curves of resistivity. The obtained main results can be formulated in the form of the following statements:

- 1. The temperature of the beginning (T_c^{on}) and the end (T_c^{0}) of the superconducting transition depending on the concentration of La shows a minimum for the value x = 0.028, which is accompanied by a large maximum at $\rho(300K)$ and $\rho(100K)$. It is attributed to the "phase separation phenomenon", which is due to the coexistence of superconducting and non-superconducting phases close to each other at T_c , accompanied by a maximum of $d\rho/dT$.
- 2. At the concentration x = 0.02, the value of the weak maximum displayed by T_c^{0} (88.6K) corresponds to the minimum of ΔTc and $d\rho/dT$, that is, a decrease in the role of electron-phonon interaction on the dispersion of current carriers.
- 3. Unlike other characteristics, the ratio $a = \rho(300\text{K})/\rho(100\text{K})$ has a monotonically decreasing nature depending on x, which is due to the fact that $\rho(100\text{K})$ increases faster with x, than $\rho(300\text{K})$.

References

- M.I. Petrov, Yu.S. Gokhfeld, D.A. Balaev, S.I. Popkov, A.A. Dubrovskiy, D.M. Gokhfeld, K.A. Shaykhutdinov, Supercond. Sci. Technol. 21 (2008) 085015.
- [2] P.N. Barnes, J.W. Kell, B.C. Harrison, T.J. Haugan, C.V. Varanasi, M. Rane, F. Ramos, Appl. Phys. Lett. 89 (2006) 012503.
- [3] A. Gantis, M. Calamiotou, D. Palles, D. Lampakis, E. Liarokapis, Phys. Rev. B 58 (2003) 15238.
- [4] H. Huhtinen, V.P.S. Awana, A. Gupta, H. Kishan, R. Laiho, A.V. Narlikar, Supercond. Sci. Technol. 20 (2007) S159.
- [5] S.K. Gaur, R.K. Singhal, K.B. Garg, T. Shripathi, U.P. Deshpande, E.M. Bittar, P.G. Pagliuso, E.M. Baggio Saitovitch, J. Phys.: Condens. Matter. **22** (2010) 509802.
- [6] L.M. Ferreira, P. Pureur, H.A. Borges, P. Lejay, Phys. Rev. B 69 (2004) 212505.
- [7] H.A. Blackstead, J.D. Dow, D.B. Chrisey, J.S. Horwitz, M.A. Black, P.J. McGinn, A.E. Klunzinger, D.B. Pulling, Phys. Rev. B 54 (1996) 6122.
- [8] A.V. Narlikar, A. Gupta, S.B. Samanta, C. Chen, Y. Hu, F. Wondre, B.M. Wanklyn, J.W. Hodby, Philosophical Magazine B 79 (1999) 717.
- [9] R.K. Singhal, Mat. Lett. 65 (2011) 825.
- [10] G.Ya. Khadzhai, A.L. Solovjov, N.G. Panchenko, M.R. Vovk, R.V. Vovk, Low Temp. Phys. 48 (2022) 576.
- [11] V.N. Narozhnyi, E.P. Khlybov, V.N. Narozhnyi, E.P. Khlybov, Supeconductivity: Phys., Chem., Tech. 5 (1992) 923.
- [12] G. Cao, J. Bolivar, J.W. O'Reilly, J.E. Crow, R.J. Kennedy, P. Pernambuco-Wise, Phys. B 185-188 (1993) 1004.
- [13] A.L. Solovjov, L.V. Omelchenko, R.V. Vovk, S.N. Kamchatnaya, Low Temp. Phys. 43 (2017) 1050.
- [14] S.K. Nikoghosyan, A.G. Sargsyan, Y.G. Zargaryan, A. Aivazyan, A. Hakopyan, Arm. J. Phys. 15 (2022) 182.
- [15] S.K. Nikoghosyan, A.G. Sargsyan, E.G. Zargaryan, Arm. J. Phys. 11 (2018) 11.
- [16] S.K. Nikoghosyan, V.V. Harutunyan, V.S. Baghdasaryan, E.A. Mughnetsyan, E.G. Zargaryan, A.G. Sarkisyan, Solid State Phenomena 200 (2013) 267.
- [17] S.K. Nikoghosyan, V.V. Harutyunyan, V.S. Baghdasaryan, E.A. Mughnetsyan, E.G. Zargaryan, A.G. Sarkisyan, IOP Conf. Series: Mat. Sci. Eng. **49** (2013) 012042.
- [18] A.P. Voitovich, A.G. Bazilev, N.V. Bandalet, S. Ovseichuk, V. Dobryankii, V. Malyshevskii, I. Ovseichuk, Supercoductivity: Phys., Chem., Tech. **3** (1990) 263.
- [19] R.V. Vovk, N.R. Vovk, A.V. Samoilov, I.L. Goulatis, A. Chroneos, Solid State Commun. 170 (2013) 6.
- [20] A.L. Solovjov, V.M. Dmitriev, Low Temp. Phys. 35 (2009) 227.
- [21] J.C. Zhang, Z.P. Qin, G. Jin, M.Q. Chen, X. Yao, C.B. Cai, S.X. Cao, J. Phys.: Conf. Series 153 (2009) 012039.
- [22] M.I. Petrov, D.A. Balaev, Yu.S. Gokhfeld, A.A. Dubrovskiy, K.A. Shaykhutdinov, Fizika Tverdogo Tela 49 (2007) 1953.
- [23] T.P. Orlando, K.A. Delin, S. Foner, E.J. McNiff, Jr., J.M. Tarascon, L.H. Greene, W.R. McKinnon, G.W. Hull, Phys. Rev. B 36 (1987) 2394.