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Transverse conductivity of $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals in a wide range of praseodymium concentrations

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The temperature dependences of the transverse resistance of $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ — single crystals are measured at high concentrations of praseodymium, where "semiconducting" behaviour prevails. The approximation of the transverse electrical resistivity of $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ — single crystals is performed using a relation describing the total hopping and fluctuation conductivity with an accuracy of no worse than 3 %. The obtained dependences of the approximation parameters on the praseodymium content give grounds to assume that in the concentration range $0.34 < y < 0.43$, the sample breaks down into separate conducting regions.
Keywords: transverse electrical resistance, YBaCuO, fluctuations, coherence length.

Поперечна провідність монокристалів $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ в широкому інтервалі концентрацій празеодиму. *М.В.Кислиця, Г.Я.Хаджай, Р.В.Вовк, А.В.Мацепулін, М.Є.Пивовар*

Виміряно температурні залежності поперечного опору монокристалів $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ при великих концентраціях змісту празеодіма, де переважає "напівпровідникова" поведінка. Апроксимацію поперечного електроопору монокристалів $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ виконано за допомогою співвідношення, що описує сумарну стрибкову і флуктуаційну провідність з точністю не гірше 3 %. Отримані залежності параметрів апроксимації від змісту празеодіма дають підставу припустити, що в області концентрацій $0.34 < y < 0.43$ зразок розпадається на окремі провідні області.

Измерены температурные зависимости поперечного сопротивления монокристаллов $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ при больших концентрациях празеодима, где преобладает "полупроводниковое" поведение. Аппроксимация поперечного электросопротивления монокристаллов $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ выполнена посредством соотношения, описывающего суммарную прыжковую и флуктуационную проводимость с точностью не хуже 3 %. Полученные зависимости параметров аппроксимации от содержания празеодима дают основание предположить, что в области концентраций $0.34 < y < 0.43$ образец распадается на отдельные проводящие области.

1. Introduction

High-temperature superconductors (HTSC) are a family of materials with a common structural feature — the presence of relatively well-separated copper-oxygen planes [1, 2]. They are also called cuprate-based superconductors. The superconducting transition temperature, which has been achieved in some compositions of this family at atmospheric pressure, is the highest among all known types of superconducting compounds [1–4].

The discovery of this class of high-temperature superconducting materials has significantly expanded the possibilities for the practical use of the phenomenon of superconductivity to create new technology [5]. At present, the main applied problem of HTSC physics is to increase the critical parameters of these compounds (superconducting transition temperature, current, field) [5–7]. For its successful solution, it is necessary to complete the construction of the microscopic theory of HTSC, which is still absent, despite the nearly 35-year intensive efforts of numerous research teams [8, 9]. To a large extent, the key to understanding the microscopic nature of HTSCs can be the study of the physical properties of these materials in the normal state [8]. At the same time, it is important to obtain the dependences of a number of electrophysical characteristics of HTSC materials on external parameters — temperature [4, 10], pressure [2, 7, 11], composition [12–14], uniformity [15, 16], etc.

Among the numerous family of high-temperature superconductors, the most promising for research are HTSC compounds of the so-called 1–2–3 $YBa_2Cu_3VO_{7-\delta}$ system [2, 17]. This is due to several reasons at once: such compounds have a relatively high critical temperature $T_c \approx 90$ K, which exceeds the boiling point of liquid nitrogen [18]; their conducting properties can be relatively merely varied by replacing the constituents [12–14, 16] and changing the oxygen content [4, 10]; it is relatively easy to obtain their cast and single-crystal samples [2, 5]. The latter circumstance is especially important for experimental research.

The layered structure of HTSC compounds leads to significant anisotropy of their physical properties, in particular, electrical conductivity [19, 20]. In this aspect, it is crucial to accurately determine the mechanisms of transport and scattering of charge carriers [4, 7, 21], which most significantly affect the electric transport in HTSC cuprates.

As is known, the replacement of yttrium with other rare-earth elements does not significantly affect the electrical transport parameters of these compounds [2, 22]. The only exception is the replacement of yttrium with praseodymium, which leads to suppression [7, 12, 14, 16, 22, 23] of the conducting characteristics as the percentage of the latter increases. At a praseodymium content of $y > 0.6$, the $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ compound completely loses its superconducting properties [22, 23] and becomes an antiferromagnetic insulator. There are currently a number of theoretical models that explain this behaviour. The most famous of them is the so-called "hole filling model" [24], "pair breaking phenomena" [25], models assuming the localization of hole carriers [26] and various mechanisms of the rearrangement of band states caused by the interaction with praseodymium ions [27–29].

Even though the literature has accumulated a reasonably extensive material devoted to the study of the effect of praseodymium on electric transport in compounds $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ [22, 23, 30], intense discussions on this issue continue to this day. A certain role in this is played by the fact that a significant part of the experimental material was obtained on film [31] and polycrystalline [32] samples of a very different technological background. The study of normal and fluctuation conductivity in $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals of a high degree of perfection is of great interest, since in such, the purest, objects it becomes possible to controllably vary the scattering and electrical conductivity parameters by changing the praseodymium concentration [33].

For the $YBa_2Cu_3O_{7-\delta}$ compounds, there is a fundamental difference in the temperature dependence of the resistivity when measured in the *ab* plane, $\rho_{ab}(T)$, and along the *c* axis, $\rho_c(T)$. For $\rho_{ab}(T)$, a "metallic" temperature dependence is characteristic in a wide range of δ values [2, 4]. For $\rho_c(T)$, even a small deviation from oxygen stoichiometry leads to a transition from quasi-metallic to semiconductor behaviour of resistance [19, 22]. Despite a large number of studies of longitudinal and transverse transport in the 1–2–3 system [1, 2, 19, 22], many aspects of this issue are still not fully clarified.

It was shown in [34] that the electrical resistance of $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals in the plane of the layers, $\rho_{ab}(T)$, in the temperature range T_c — 300 K and praseodymium concentrations $0 < y < 0.34$ is characterized by a "metallic" temperature dependence, which can be approximate, taking into account the scattering of electrons by phonons and defects, as well as the fluctuation conductivity in the form of the 3D Aslamazov-Larkin model. At the same time, it was established [34] that the change in the approximation pa-

rameters depending on the praseodymium content indicates that the residual electrical resistance is mainly determined by the defectiveness of the lattice; the Debye temperature passes through a maximum due to the competition between changes in the interatomic interaction and lattice parameters; changes in the parameters of scattering by phonons indicate an increase in the density of electronic states upon the introduction of praseodymium. The parameters of fluctuation conductivity — the transverse coherence length and the temperature range of superconducting fluctuations — tend to increase with increasing praseodymium content. However, with a further increase in the concentration of praseodymium [34], a structural transition occurs in the region of $0.35 \le y \le 0.43$, leading to the appearance of a "semiconducting" behaviour of $\rho_c(T)$ at low temperatures immediately before the superconducting transition.

In [35], for the electrical resistance of $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals along the c axis, $\rho_c(T)$, at sufficiently large values of y, the dependence $\rho_c(T) \sim T \cdot \exp[(T_0/T)^{1/2}]$, was found, which is characteristic for hopping conductivity with variable jump length.

The hopping conductivity is due to the tunnelling of electrons between localized states. The physical reason for localization is the interaction of electrons with a random potential, leading to increased scattering and a strong dependence of the dynamics of electrons on the degree of disorder in the system.

At sufficiently low temperatures, the hopping conductivity is determined by states with energies in a narrow strip near the Fermi level [36]. If the density of states at the Fermi level is nonzero, then the temperature dependence of the resistivity has a universal form:

$$\rho(T) \propto \exp\left[(T_0/T)^{1/4}\right], \qquad (1)$$

$$T_0 = \frac{\beta}{kg(\mu)a^3}, \qquad (2)$$

where $g(\mu)$ is the density of state at the Fermi level, a is the radius of localized states close to the Fermi level, and β is a numerical coefficient.

However, numerous experiments carried out with nanocomposites of various compositions show that the resistance $\rho(T)$ of such systems is described by the universal 1/2 law:

$$\rho(T) \propto \exp\left[(T_0/T)^{1/2}\right], \qquad (3)$$

where T_0 is the temperature parameter, the value of which strongly depends on y and tends to zero as $y \to y_c$. Therefore, attempts were made to modify the Mott model by postulating some "selection rules" for admissible hops (for example, the relationship between the hopping length and the size of actual granules [37] in nanocomposites with granules of different sizes) in order to obtain the 1/2 law within its framework.

Usually, the 1/2 law is interpreted as a manifestation of the Coulomb gap in the density $g(E)$ of states of electrons on impurities, when, near the Fermi energy, E_F, the density of states turns to zero according to the law $g(E) \sim (E - E_F)^2$. In this regard, there are numerous works in which, on the one hand, the question of the nature of the Coulomb gap in the density of states of electrons on nanocomposite granules is investigated [37] and, on the other hand, the effect of such a gap on the temperature dependence of its conductivity is studied [38]. It is clear that if the "gap" scenario is realized, then the region of its applicability is low temperatures at which the Coulomb gap is not washed out by thermal excitations (actually at $T < 1 - 10$ K). At higher temperatures, the Coulomb gap plays no role, and the problem of thermoactivation conductivity of granular metals remains open.

It follows from this that the 1/2 law is not associated with any artificial selection rules for tunnel junctions between granules, but is a simple consequence of the wide scatter of the granule size inherent in real nanocomposites.

At $T = 0$ K and in the absence of an external electric field, all-metal granules in the dielectric phase of the nanocomposite are neutral, like the electrostatic energy of the configuration of charged granules $E > 0$. However, at a temperature other than zero, due to the tunnelling transitions of electrons between the granules, thermodynamic equilibrium is established in which part of the granules acquires a positive (and the same part — negative) charge. As a result, conditions arise for the appearance of conductivity due to tunnel transitions (hopping) of charges (electrons or "holes") from charged metal granules to neutral granules. The number of such charges increases with increasing temperature. This means that the conductivity of dielectric nanocomposites is associated with thermal activation of current carriers [37].

On the other hand, as was shown in [35], relation (3) has a much more universal char-

acter and can be fulfilled for a broad class of conducting compounds with a sufficiently high degree of structural disordering. In particular, it is known that doping of HTSC cuprates by means of alternative substitution or changing the oxygen content leads to the decomposition of the system into electrically neutral regions of two types — metallic, with a high carrier concentration, and dielectric [39]. In this case, the type of domains can also be "imposed" by ordering dopants. Obviously, with a sufficiently small size of inclusions with metallic conductivity, the system can acquire the features characteristic of granular metals. In our case, the decisive effect on the structural order in the system is apparently played by doping with praseodymium, which leads to an increase in the degree of structural disordering in the conducting layers [16] and an enhancement of the "semiconductor" character of the behaviour of the temperature dependence of the conductivity across the basal plane [33].

This work aim is to analyze the transverse resistance of high-temperature superconductors $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ in a wide temperature range for high concentrations of praseodymium to elucidate the mechanisms of conduction and scattering of charge carriers, based on currently known models.

2. Experimental

Single crystals of $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ were grown by the solution-melt method in a gold crucible at a temperature of 850–970°C, as described in detail in [4, 7, 9, 16]. To obtain crystals with partial replacement of Y by Pr ($Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$), Pr_5O_{11} was added to the initial charge in the appropriate percentage. The regimes of growing and saturating crystals with oxygen were the same as for undoped YBCO single crystals [4]. Compounds Y_2O_3, $BaCO_3$, CuO, and Pr_5O_{11} were used as initial components for growing crystals. The characteristic dimensions of the samples were $2\times0.3\times0.02$ mm^3. The smallest crystal size corresponded to the c-axis. To obtain samples with optimal oxygen content, $\delta \le 0.1$, the selected crystals were annealed in an oxygen flow at a temperature of 400°C for five days. The electrical contacts were made from silver conductors, which were connected to the surface of the crystals using silver paste. The electrical resistance was measured by the Montgomery method [40].

The temperature was measured with a copper-constantan thermocouple, the voltage across the sample and the exemplary resistance — with V2-38 nanovolmeters. Data from voltmeters were automatically transmitted to a computer via the interface. The measurements were carried out in the temperature drift mode, which was about 0.1 K/min for measurements near T_c, and about 5 K/min at $T > T_c$. In this case, all measurements were carried out three days after the completion of annealing, which ensured an equilibrium oxygen distribution over the sample volume at room temperature [4]. A specially written program was used to approximate the results obtained.

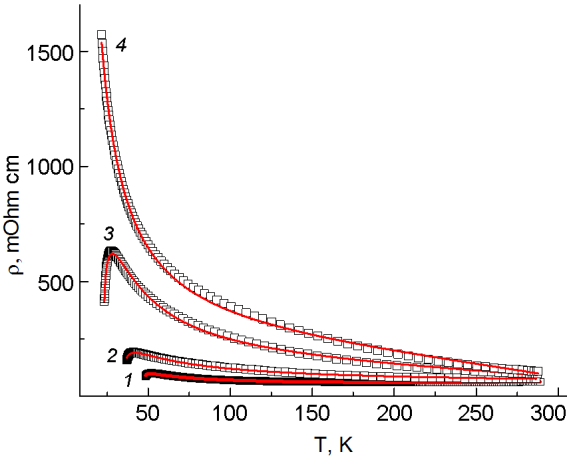

Fig. 1. Curves of electrical resistance $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ along the c axis with different Pr concentration: *1* — 0.34; *2* — 0.43; *3* — 0.48, *4* — 0.5. Points correspond to the experiment; the line corresponds to the approximation.

3. Results and discussion

In Fig. 1 shows the temperature dependences of the electrical resistance of $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ — single crystals in the Pr concentration range of 0.34–0.5.

As seen from Fig. 1, the nature of the temperature dependence of resistance is a semiconductor. The critical temperature, T_c, decreases, and upon reaching $y \approx 0.5$, superconductivity suppresses.

The presented temperature dependences of the transverse resistivity of $YBa_2Cu_3O_{7-\delta}$ were approximated by a relation that takes into account the semiconductor behaviour through the activation exponent and fluctuation conductivity in the form of the 3D Aslamazov-Larkin model [41]:

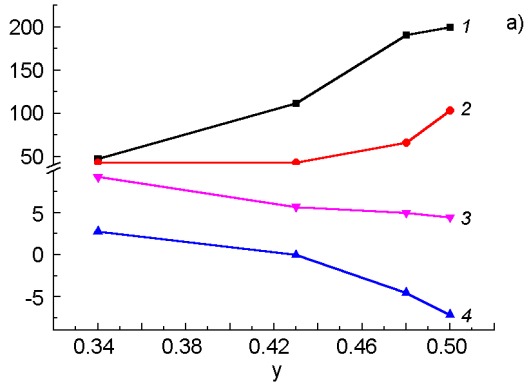

Fig. 2. Dependence of the approximation parameters on the Pr concentration. *1* – T_0, K, *2* – A, mOhm·cm, *3* – $B{\cdot}10^6$, K^{-2}, *4* – $n{\cdot}10$

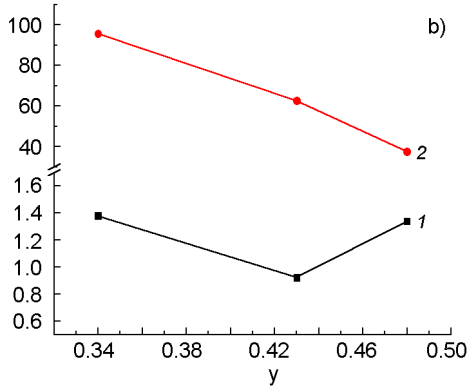

Fig. 3. Dependence of the coherence length $\xi_{ab}(0)$ and temperature T on the Pr concentration. *1* – $\xi_{ab}(0){\cdot}10^6$, cm, *2* – T^*, K

$$\rho_c(T) = \left[\frac{1}{A \cdot \exp\left(\frac{T_0}{T}\right)^n \cdot (1 + B \cdot T^2)} + \sigma_{AL}(T)\right]^{(-1)}, \quad (4)$$

$$\sigma_{AL}(T) = \frac{s \cdot e^2}{32 \cdot h \cdot \xi_{ab}^2 \cdot \sqrt{2 \cdot \varepsilon_0 \cdot \sinh\left(\frac{2 \cdot \varepsilon}{\varepsilon_0}\right)}}. \quad (5)$$

Here ε is the reduced temperature, $\varepsilon = \ln(T/T_c)$; $\varepsilon_0 = \ln(T^*/T_c)$, T^* is the conditional boundary for the existence of superconducting fluctuations [42]; A, B, n — approximation parameters, $T_0 = E/k$ (E — activation energy, k — Boltzmann constant; $\sigma_{AL}(T)$ — fluctuation conductivity [2, 7, 41].

The adjustable parameters in (4) are A, which has the physical meaning of the residual resistance, the exponent n in the modified version of Mott's law, and the parameter B, which is probably related to the electron mobility. The fitting parameters in (5) are $\xi_{ab}(0)$ — the coherence length in the layer, and ε_0.

The fitting was carried out using the program for searching for the minimum of the absolute error, the values $\rho_{exp.} - \rho_{calc.}$, $\rho_{calc.}$ calculated by formulas (4)–(5). Table 1 shows the values of the fitting parameters that were obtained. They provide a minimum approximation error of no more than 3 %. The results of approximation showed that the region of existence of superconducting fluctuations, characterized by the parameter T^*, decreases as the concentration $y = 0.5$ is approached, where superconductivity is not observed. The coherence length in the layer, $\rho_{ab}(0)$, remains in the region of 100 Å. If we take into account that the transverse coherence length $\xi_c(0)$ is of the order of 1 Å [2, 42–44], it turns out that the superconducting pair in the $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ — HTSC exhibits a pronounced anisotropy. In Fig. 2 and 3 show the dependences of the approximation parameters on the praseodymium concentration.

It can be seen that starting from $y = 0.48$, parameter A demonstrates a sharp increase — when it reaches 0.5, more than two times. This confirms the transition of this compound at $y > 0.6$ in the state of an antiferromagnetic dielectric. Parameter B decreases monotonically from $2{,}80{\cdot}10^6$ to $7{,}10{\cdot}10^6$. Note that in (4) the first term is the role of a semiconductor, then the expression $1/(1 + BT^2)$ plays the mobility. At $B > 0$, the mobility decreases with increasing temperature, which is typical for the scattering of electrons by phonons in semiconductors (see, e.g., [45]).

However, with an increase in the concentration of praseodymium, parameter B becomes negative (see Table), that is, the mobility now increases with increasing temperature, which is typical for electron scattering by impurities — in this case, praseodymium. Thus, the change in the sign of the parameter B with increasing y can be associated with the transition from the predominant scattering of electrons by phonons to the predominant scattering by Pr impurities.

The exponent, n, in the concentration range $0.34 < y < 0.43$ drops sharply from $n \approx 1$ to the level $n \approx 0.5$, which indicates a

Table 1. Parameters of approximation of resistance $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$

y	0.34	0.43	0.48	0.5
A, mOhm·cm	43.0	43.3	66.1	103.6
$B{\cdot}10^6$, K^{-2}	2.8	0.01	−4.5	−7.1
n	0.93	0.57	0.50	0.445
T_0, K	47.2	111.6	191	200
$\xi_{ab}(0){\cdot}10^6$, cm	1.4	0.9	1.3	–
T^*, K	95.7	62.6	37.4	–

transition at $n > 0.43$ to hopping conductivity with a variable hopping length — "law 1/2". It was shown in [36] that the "1/2 law" is due to electron hopping between conducting granules of different sizes. A more accurate calculation of the conductive granule size optimal for electrical conductivity leads to $n = 4/9$ [36]. It is this value of the exponent that was obtained at $y = 0.5$ (see Table).

It can be assumed that, in our case, the presence of conducting and non-conducting regions is associated with significant variations in the concentration of praseodymium, y, when approaching the state of an antiferromagnetic dielectric.

Note that, in the model [36], the parameter T_0 vanishes when percolation over the conducting phase occurs. In our case, an increase in the concentration of praseodymium leads to a transition from metallic to semiconducting conductivity and then to the state of an antiferromagnetic insulator. Thus, it can be assumed that in the concentration range $0.34 < y < 0.43$ (where n falls from 1 to 0.5), the common conducting cluster breaks up into separate conducting regions (granules) and the conductivity acquires a hopping character. Therefore, in our case, T_0 grows with increasing y.

4. Conclusion

Thus, it can be concluded that the experimental temperature dependences of the transverse resistance of $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals in the region of the "semiconductor" resistance curve can be approximated by a relationship that includes thermally activated jumps of variable length and fluctuation conductivity in the 3D Aslamazov-Larkin model. An analysis of the concentration dependences of the approximation parameters shows that the concentration range $0.34 < y < 0.43$, the sample disintegrates into separate conducting regions (granules) and the conductivity acquires the character of hopping with variable length. With an increase in the concentration of praseodymium, the effect of scattering of charge carriers by praseodymium impurities increases monotonically. The region of existence of superconducting fluctuations decreases with an increase in the concentration of praseodymium. A pronounced anisotropy of the coherence length, that is, the size of the superconducting pair, is found — the size across the layer is two orders of magnitude smaller than along the layer.

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