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Abstract

The electrical resistivity in the *ab*-plane of the high degree of perfection $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals in the interval of T_c–300 K was investigated. The increasing of praseodymium content leads to the reduction of the critical temperature (T_c) of 92 to 30 K. The experimental results can be approximated by the expression, taking into account the scattering of electrons by phonons, defects, the fluctuation conductivity in the 3D Aslamazov – Larkin model, as well as the transition to a "semiconductor" type behavior of the resistivity at the high praseodymium concentrations. The concentration dependences of all fitting parameters indicate a structural transition in the region $0.35 \leq y \leq 0.43$. In particular, the Debye temperature changes in this range of from 350 to 550 K, and the transverse coherence length passes through a maximum $\xi_C(0) \approx 5$ Å. The concentration dependence of the critical temperature testifies the *d*-pairing of the BCS model.

KEYWORDS: A. $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals; D. electrical resistivity; E. 3D Aslamazov – Larkin model.

Introduction

According to recent concepts one the issues to construct a microscopic theory of high-temperature superconductivity (HTSC), is the explanation of unusual phenomena observed in the normal (or non-superconducting) state of HTSC compounds [1-3]. Among the latter, in the field of electrical transport properties are the presence of a wide temperature range of fluctuation para-conductivity (FP) [4,5], the so-called pseudogap anomaly (PG) [6,7], the incoherent electrotransport [3,8,9], the metal-insulator transitions [10,11] and other effects. To understand the nature of these phenomena in HTSC it is important to study the carriers' scattering processes [12-14]. The most promising system to investigate are the HTSC compounds of the 1-2-3 system – $ReBa_2Cu_3O_{7-\delta}$ (Re = Y or another rare earth element). This is because (a) they have a relatively high critical temperature $T_c \approx$ 90 K, exceeding the liquid nitrogen temperature [15,16], (b) their conductive properties can be relatively simply varied by replacing the Re [17,18] and the oxygen stoichiometry [19, 20] and (c) it is relatively easy to grow single-crystal samples [17,20-22]. The latter is very important for the experimental studies.

The substitution of yttrium by rare earths ions normally has no significant impact on the electro-transport parameters of these compounds [7,9,17,18]. The only exception is the substitution of yttrium by praseodymium, which leads to the suppression [23-32] of the superconductive characteristics with the increasing content of Pr (known as the praseodymium anomaly). When the praseodymium content is $y \geq 0.6$ the $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ compound loses the superconducting properties [3,17] and becomes an antiferromagnetic insulator. Currently, there are a number of theoretical models to explain this behavior. The most well-known are the "hole filling model" [23], and the "pair breaking phenomena" models [24], which assume the localization of hole carriers [25] and, caused by the interaction of praseodymium ions, various mechanisms of adjustment band states [26-28]. Despite the extensive literature regarding the influence of praseodymium to electric transport in $Y_{1-z}Pr_zBa_2Cu_3O_{7-\delta}$ compounds [29], intensive discussions on this issue are ongoing.

Herewith, a role is played by the fact that a significant portion of the experimental data was obtained on the film [30] and polycrystalline [31-32] samples with very different processes.

Electrical conductivity can be represented as $\sigma \approx \frac{Se^2L}{12\pi^3\hbar}$, where S is the area of the Fermi surface and L is the mean free path of electrons [33]. It is characteristic for the metallic conductivity that $d\rho/dT > 0$ and $0 < \rho = \sigma^{-1} \leq \rho_{max}$, where $\rho_{max} = \frac{3\hbar\alpha}{e^2}$ is the Ioffe - Regel limit (a is a lattice constant), at a = 3 Å $\rho_{max} \approx 400$ μOhm*cm [33]. In this case, the resistance changes due to changes in the mean free path of electrons ($0 < L < \alpha$) due to scattering on the periodicity violations, that primarily is phonons (the temperature dependence of L) and structural defects (the concentration dependence of L). Therefore, if the experimental values of the resistance are close to the mentioned limits and $d\rho/dT > 0$, then, the investigation for scattering by phonons and by defects in such high-T_c materials is natural.

Scattering by phonons leads at temperatures $T \geq \theta$ (θ is the Debye temperature) to almost linear temperature dependence, which is typical for HTSC with a high degree of perfection at high temperatures. With decreasing temperature, the phonon resistance is deflected downward from the high temperature extrapolation ($\rho \propto T$) already at $T \leq \theta/3$ [14], where the fluctuation conductivity can be shown. Therefore it is quite difficult to set experimentally the region that the fluctuation conductivity exists.

In the present study we investigated the temperature dependence of the resistivity of $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single-crystals in a wide range of praseodymium concentrations ($0.0 \leq y \leq 0.5$) and temperature (T_C–300 K) within a single scheme that takes into account the scattering of electrons by phonons and defects, the transition to "semiconductor" behavior of resistance together with the influence of the fluctuation conductivity.

Experimental techniques

The $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals were grown by the solution-melt method in a gold crucible at 850-970 °C as described in detail in previous studies [3, 14]. The typical sample's size is $2 \times 0.3 \times 0.02$ mm^3. The smallest size of the crystal corresponded to *c*- axis. For samples with an optimal oxygen content (defined here as $\delta \leq 0.1$) selected crystals were annealed in oxygen flow at 400 °C for five days. The electrical contacts were made of silver wire that was connected to the crystal surface using a silver paste. Resistivity measurements were carried out by the standard four-probe method at a constant current of 1 mA at two opposite current directions in zero magnetic fields. The temperature was measured by a copper-constantan thermocouple, the voltage by the use of B2-38 nanovoltmeters. Measurements were carried out in the mode of temperature drift, which was about 0.1 K/min for measurements near T_c and about 5 K/min at $T > T_c$.

Results and discussion

The obtained temperature dependencies of the resistivity for various concentrations of praseodymium are shown in Fig. 1a. It can be seen that at low praseodymium concentrations (samples 1-5, $20 < \rho < 130$ μOhm*cm) $\rho(T)$ shows the "metallic" behavior, but at bigger concentrations of praseodymium (samples 6-8, $\rho > 200$ μOhm*cm) with decreasing temperature ($T < 60$ K), $\rho(T)$ dependencies deviating upward from the "metallic" behavior, showing the transition to the "semiconductor" type dependence. We believe that at such praseodymium concentrations, mean free path reaches its minimum value, $L \leq a$ and ρ(T) dependence is now associated with a change in the band structure, which is characterized, inter alia, the area of the Fermi surface, *S*.

Fig. 1b shows the concentration dependences of the derivatives, $d\rho_{ab}/dT$, at fixed temperatures (at $T \geq 150$ K). It is determined that the values of $d\rho_{ab}/dT$ pass through a maximum

at $y \approx 0.43$, i.e. in the region of "metallic" behavior $d\rho_{ab}/dT$ increases with y, but at the transition to the "semiconductor" depending $d\rho_{ab}/dT$ decreases with increasing y.

We approximate the temperature dependence of the resistivity on our samples in the interval T_c –300 K with expressions taking into account the interband scattering of electrons by phonons [34] and by defects, the fluctuation conductivity in the 3D Aslamazov-Larkin model [35], as well as the transition to the dependencies of the “semiconductor” type. The general expression for the conductivity is:

$$\sigma = \rho_1^{-1} + \Delta\sigma_{AL}; \qquad \Delta\sigma_{AL} = \frac{e^2}{16\hbar\xi_c(0)\sqrt{2\varepsilon_0 \sinh(2\,\varepsilon/\varepsilon_0)}} \tag{1}$$

Expression for the fluctuation conductivity is chosen to limit the scope of its influence [36], $\varepsilon = ln(T/T_C)$ is the reduced temperature, T_c is the critical temperature in the mean-field approximation, $T > T_c$, $\xi_c(0)$ is the transverse coherence length at $T=0$, ε_0 determines the temperature interval of the superconducting fluctuations – $\varepsilon_0 = ln(T^*/T_C)$, T^* is a characteristic temperature, which determines - together with $\xi_C(0)$ - the collapse of the superconducting fluctuations.

$$\rho_1 = \frac{(\rho_0 + \rho_3)(1 - bT^2)}{1 - n[1 - \exp(-\Delta E / 2kT)]} \qquad \rho_3 = C_3\left(\frac{T}{\theta}\right)^3 \int_0^{\theta/T} \frac{x^n e^x}{(e^x - 1)^2} dx \tag{2}$$

In Eqn. (2) ρ_0 is the residual resistance characterizing the scattering by defects; ρ_3 is the contribution to the resistance due to interband scattering on phonons [34]; θ is the Debye temperature; b_0 depends on the shape of the curve of the electronic density of states, effective mass of the carriers and the Fermi energy [37-38]; n is the share of the semiconductor phase: $n=0$ – the “metallic” behavior; $n>0$ – the “semiconductor” type behavior; ΔE is a characteristic energy [39].

Optimal set of the fitting parameters, which provides a minimum average error of about 1% (over the interval T_C –300 K), is shown in Table 1. The derivatives $d\rho/dT$, calculated from

samples (1) - (3), adequately approximate the behavior of $d\rho/dT$, calculated from the experimental data. These curves are shown in Fig. 1a with solid lines.

Notably, for the fitting of samples 1-5 we needed six adjustable parameters and for the samples 6-8, we needed eight adjustable parameters. Each parameter has a physical meaning and their dependence from the praseodymium concentration is discussed below.

Fig. 2 shows the experimentally obtained dependence of T_c on the concentration of praseodymium, y, and its fitting to the universal Abrikosov-Gor'kov equation describing the T_c suppression by nonmagnetic defects in the case of d-pairing [24, 40-41]:

$$\ln\left(\frac{T_c}{T_{c0}}\right) = \Psi\left(\frac{1}{2}\right) - \Psi\left(\frac{1}{2} + \frac{e^{-C} y T_{c0}}{4 y_{cr} T_c}\right) \ .$$

Here, T_{c0} is the superconducting transition temperature of a defect-free sample, $\Psi(z)$ is the digamma function, y_{cr} is the critical concentration of superconductivity disappearance, and C is Euler's constant.

The curve in Fig. 2 is carried out for x_{cr} = 0.62 and T_{c0} = 92 K. It is observed that the theory [40] describes very well the experiment that suggests the d-pairing in the BCS model. Dependence of T_C(y) which is given is fully consistent with the data [24].

The same figure shows the dependence of the residual resistance from the concentration of praseodymium. In the interval $0 \le y \le 0.35$, this dependence is close to linear, but further $\rho_0(y)$ is sharply increased (212 $\rightarrow$ 472 μOhm*cm, samples 6-8) and in the entire range from $0 \le y \le 0.50$ this dependence is close to exponential: $\rho_0 \sim \exp(y/y^*)$, $y^* \approx 0.42$ (curve 2). This behavior of ρ_0(y) indicates, in our opinion, that when $y > 0.35$ the electron mean free path, L, can reach its minimum value ($L \sim a$) and ρ_0 increases due to changes in the electronic characteristics.

In Figure 3, shows the dependencies of the fitting parameters of the praseodymium concentration on the scattering parameters (refer to Fig. 3a) and the fluctuation conductivity parameters (refer to Fig. 3a).

The values of the Debye temperature, θ, correspond to the literature data (see, for example [42-43]); θ(y) of the samples (6-8) was 1.5 times higher than for the (1-5), i. e. θ exhibits the jump in the range $0.35 \le y \le 0.40$ and as C_3, passes through a maximum at $y_m \approx 0.43$. Since $\Delta\theta(y)/\theta \approx -\alpha\Delta V(y)/V + \beta\Delta f/f$ (ΔV is the change in volume of the unit cell, Δf is the change of the force constants) and all the lattice parameters of $Y_{1-y}Pr_yBa_2Cu_3O_{7-x}$ increase with increasing y (at least for x~0.1) [24], the sharp increase of θ in samples (6-8) indicates a significant increase in the force constants. At a still higher y content (samples 7-8), θ is reduced probably due to the expansion of the lattice. The θ increase can be associated with a structural transition from orthorhombic to tetragonal phase [29], which is accompanied by a transition to the "semiconductor" type conduction behavior.

Dependence of $C_3(y)$ passes through a maximum at $y_m \approx 0.43$. Parameter b_0 is qualitatively behaves oppositely to C_3, passing through minimum when $y_m \approx 0.43$. The parameters C_3 and b_0 are determined by the electronic structure of the sample. $C_3 \propto N_d(E_F)$ ($N_d(E_F)$ is the electron density of states at the Fermi level; E_F is the Fermi energy) [37], b_0 depends mainly on the values $\left[\frac{1}{N}\frac{dN}{dE}\right]_{E_F}$ and $\left[\frac{1}{N}\frac{d^2N}{dE^2}\right]_{E_F}$ [37-38]. The change of these parameters by increasing y is connected with the change in the density of electronic states when introducing praseodymium (refer to Fig. 3a, curves 1 and 2). This result is consistent with the results of [45], assuming that the main reason for the suppression of superconductivity in $Y_{1-y}Pr_yBa_2Cu_3O_{7-x}$ is caused by the significant change in the band structure due to Pr.

Thus, in the interval $0.35 \le y \le 0.43$ the parameters characterizing the normal scattering of electrons by phonons and impurities are related with the restructuring changes of the electronic characteristics.

Fig. 3b shows the dependence of the fluctuation conductivity parameters, $\xi_c(0)$ and ΔT, on the concentration of praseodymium y. These dependences, $\xi_c(0)$ and ΔT, are qualitatively

similar. They both have maxima in the region $y \approx 0.4 \div 0.5$, where the maximums of scattering parameters on phonons and impurities are also occurred. This points to the correlation between phonons, electrons and fluctuation parameters. Notably, the qualitative changes in the properties at $y \approx 0.4 \div 0.5$ are consistent with previous data (for example refer to [46-47]).

Fig. 4 shows that the dependence $\xi_c(0)$ from $1/T_C$ is linear, which corresponds to the BCS theory. In sample 8 (y = 0.5) there occurs the greatest suppression of superconductivity, and consequently it is the only sample failing to fit in the model. Finally, this is possibly due to the impact of specific mechanisms of quasiparticle scattering [48-51], which are related to the presence of structural and kinematic anisotropy in the system.

Transition to a “semiconductor” type temperature dependence of resistance is represented by 3 samples. So it is difficult to say that the relevant parameters are depending on the praseodymium concentration. Table 1 shows that with the increase of y the characteristic energy ΔE decreases, and n, the share of the semiconductor phase, demonstrates a tendency to increase.

Conclusions

It is determined that the temperature dependence of the resistivity of $Y_{1-z}Pr_zBa_2Cu_3O_{7-\delta}$ single crystals (studied along the plane of the layers in the range of T_C –300 K) can be approximated by a general expression that takes into account the scattering of electrons by phonons and defects, fluctuation conductivity in the 3D Aslamazov-Larkin model, as well as the transition to "semiconductor" type temperature dependence of resistivity. The concentration dependence of the critical temperature and the correlation $\xi_c(0) \propto 1/T_C$ evidence in favor of d-pairing in the BCS model. In the interval from $0.35 \leq y \leq 0.43$ all fitting parameters – phonons, electrons and fluctuation - are showing signs of structural adjustment. In particular, the residual resistivity, ρ_0, increases with increasing praseodymium content much faster than linearly, the Debye temperature, θ, has a jump; the derivatives, $d\rho_{ab}/dT$, electronic parameters b_0 and C_3 as well as coherence length, $\xi_c(0)$, passes through the extremes. This restructuring is accompanied

by a transition to "semiconductor" type behavior, i. e. the regime in which the resistance changes occur due to changes in the mean free path turns into a regime in which the resistance changes occur due to changes in the electronic structure.

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Table 1. Fitting parameters of electrical resistance.

Samples	1	2	3	4	5	6	7	8
T_c, K	91.738	85.663	80.540	67.460	51.757	39.654	34.212	30.139
c_{Pr}	0	0.05	0.19	0.23	0.34	0.43	0.48	0.5
ρ_0, Ohm·cm	$2.12\cdot10^{-5}$	$4.50\cdot10^{-5}$	$8.13\cdot10^{-5}$	$1.42\cdot10^{-4}$	$1.26\cdot10^{-4}$	$2.12\cdot10^{-4}$	$4.36\cdot10^{-4}$	$4.72\cdot10^{-4}$
C_3, Ohm·cm	$3.3\cdot10^{-4}$	$3.32\cdot10^{-4}$	$3.97\cdot10^{-4}$	$4.61\cdot10^{-4}$	$8.68\cdot10^{-4}$	$2.01\cdot10^{-3}$	$1.20\cdot10^{-3}$	$1.12\cdot10^{-3}$
θ, K	352	366	366	367	358	561	557	545
b_0, K^{-2}	$2\cdot10^{-8}$	$1.28\cdot10^{-6}$	$1.3\cdot10^{-6}$	$6.95\cdot10^{-7}$	$-5.65\cdot10^{-7}$	$-1.2\cdot10^{-6}$	$-1.85\cdot10^{-7}$	$-4.50\cdot10^{-7}$
ξ_{C0}, Å	0.7	1.7	0.7	0.5	2.6	3.2	5.2	2.1
$\Delta T=T^*-T_C$, K	8.3	22	18	24	18	49	2.1	2.2
n_0	0	0	0	0	0	0.063	$7.5\cdot10^{-3}$	0.60
T^*						50	45	29

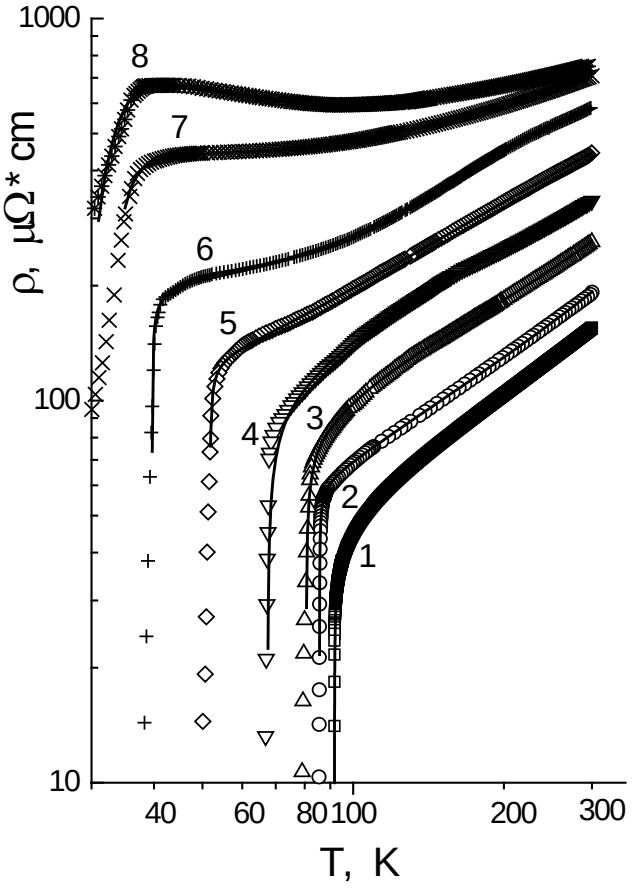

Fig. 1a. Plot of temperature dependencies ρ_{ab} for different $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals with praseodymium concentration y: 1 – 0.0, 2 – 0.05, 3 – 0.19, 4 – 0.23, 5 – 0.34, 6 – 0.43, 7 – 0.48, 8 – 0.5. The numbers of the curves correspond to the number of the samples in the table. Points – the experiment; lines – fitting in accordance with Eqs. (1) and (2).

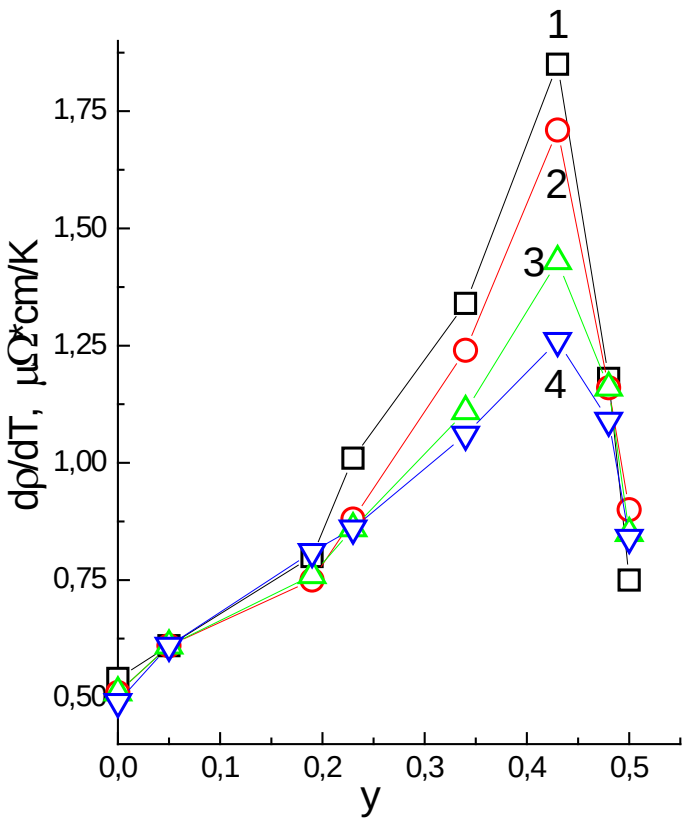

Fig. 1b. The concentration dependencies of the derivatives, $d\rho_{ab}/dT$, for $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ single crystals at different fixed temperatures: 1 – 150 K; 2 – 200 K; 3 – 250 K and 4 – 300 K.

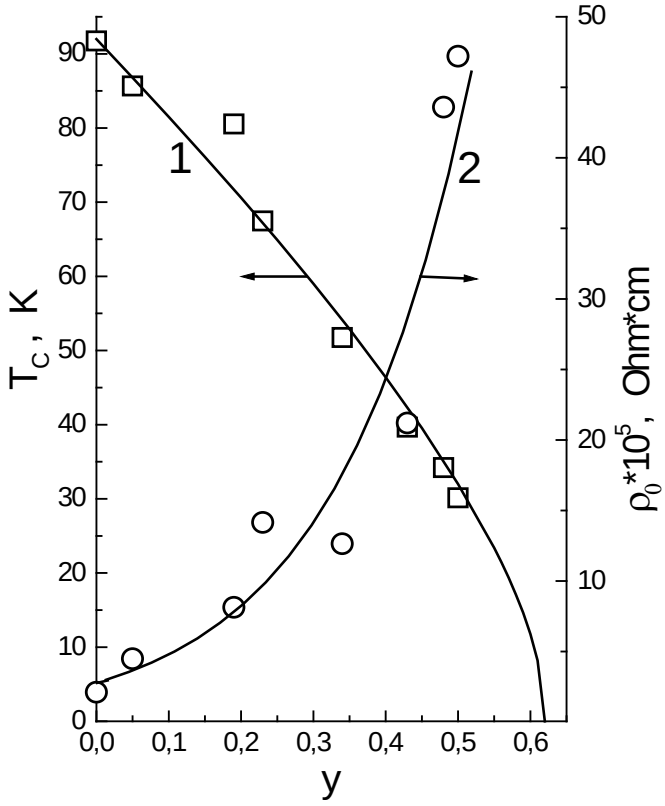

Fig. 2. Plot of T_C (1) and ρ_0 (2) dependences with respect to the praseodymium concentration *y*.

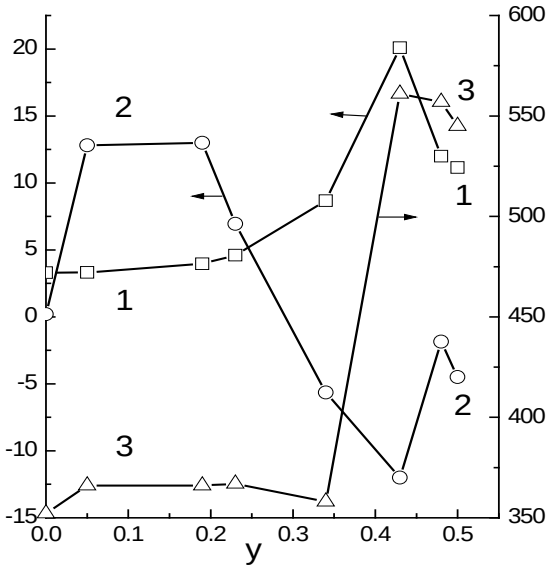

Fig. 3a. Phonon scattering parameters versus *y*: 1 – C_3*10^4 Ohm*cm; 2 – b_0*10^7 K^{-2}; 3 – θ, K.

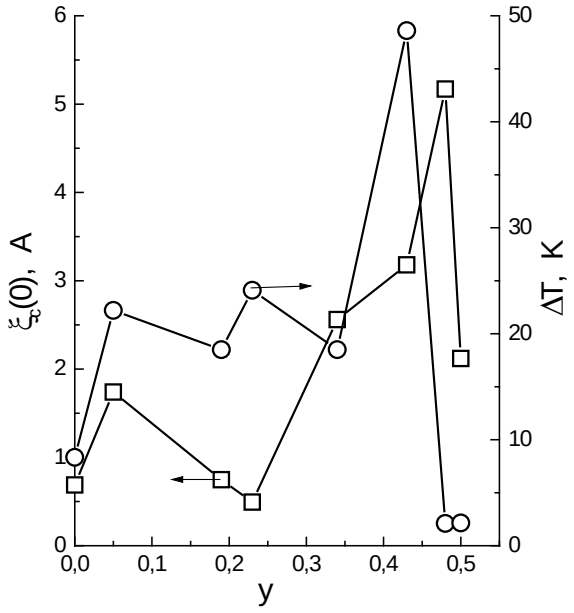

Fig. 3b. Fluctuation conductivity parameters dependencies with respect to the praseodymium concentration, y: 1 – $\xi_c(0)$, Å, and 2 – ΔT, K.

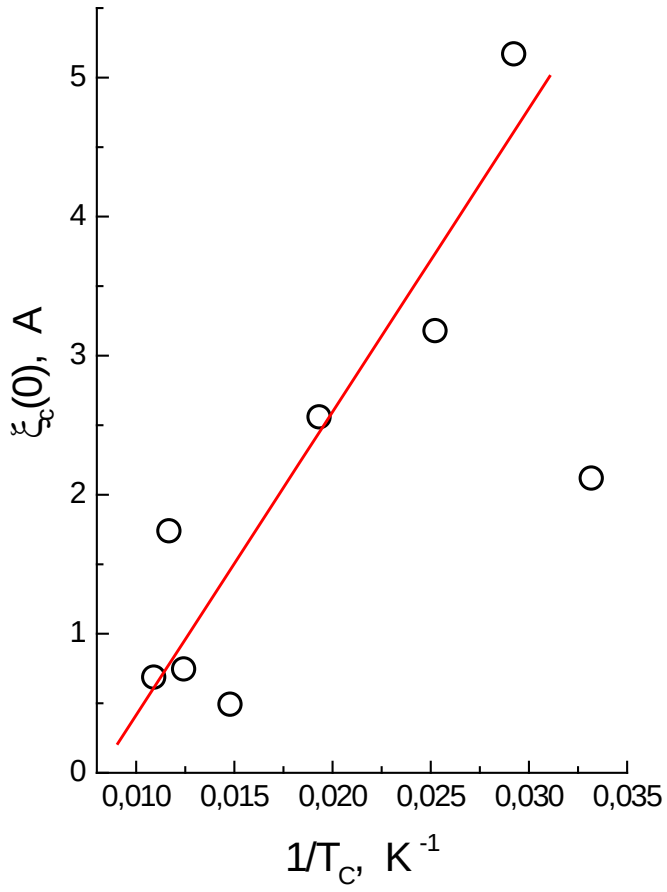

Fig. 4. Plot of $\xi_c(0)$ with respect to $1/T_C$.