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# Effect of long aging on the resistivity properties of aluminium doped $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$ single crystals with a given twin boundary topology

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## Abstract

We investigate the conducting properties in the basal  $ab$ -plane before and after a long time exposure in air atmosphere of aluminium doped  $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$  single crystals. Prolonged exposure leads to an increase of the effective scattering centers of the normal carriers. The excess conductivity in a wide temperature range has exponential temperature dependence and near the critical temperature ( $T_c$ ) can be adequately described within the Aslamazov-Larkin theoretical model. Nevertheless, the description of the excess conductivity through the relation  $\Delta\sigma \sim (1-T/T^*)\exp(\Delta^*_{ab}/T)$ , can be interpreted in terms of the mean-field theory, where  $T^*$  is the mean field temperature of the superconducting transition and the temperature dependence of the pseudo-gap is satisfactorily described by the BEC-BCS crossover theory. It is determined that the prolonged exposure increases significantly the temperature range of the implementation of the pseudogap state, thus narrowing the linear section of the resistivity dependence  $\rho_{ab}(T)$ .

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## Introduction

The change in the composition of high- $T_c$  superconductors (HTSC) such as  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , is an important method to devise empirical ways to improve their critical parameters and to extent their technological applications [1]. A complete or partial substitution of yttrium with rare-earth elements, with the exception of praseodymium (praseodymium anomaly) that suppresses the superconducting parameters of the compound [2], slightly affects their physical characteristics in the normal and superconducting state [3,4]. The substitution of copper by calcium leads to a deterioration of their superconducting properties (see for example [5] and references therein). The partial substitution of copper by gold and by silver is an exception [6,7]. At low concentrations, it improves conductivity and prevents the degradation of the superconducting properties in the aging process [7]. It is of particular interest to investigate the partial substitution of copper by aluminium [8-11]. Around the trivalent aluminium atoms is the typical octahedral environment from oxygen atoms, facilitating a severalfold reduction of the period of twin superstructure [10] and at high concentrations, the formation of structures intersecting the "tweed" type twin domains [11]. The twin boundaries (TB), being extended planar defects, promote strengthening of pinning processes [12], which extend their range in obtaining high magnetic fields. At the same time, the presence of TB complicates the investigation of the resistance characteristics due to the difficulty of defining their contribution to the electrical conductivity in the HTSC compounds [4,10]. Previous studies on the impact of the aluminium on the electro-transport properties of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  are contradictory. For example, van Dover *et al.* [8] reported a slight increase in resistivity in the basal plane  $\rho_{ab}$  in  $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$  crystals at  $z \leq 10\%$ , whereas Oh *et al.* [9], is observed a twofold increase of the  $\rho_{ab}$  at the same concentration of aluminium. The reason for this discrepancy is probably, a non-homogeneous distribution of aluminium over the crystal's volume, since the crystal growth in alundum crucibles the introduction of aluminium occurs in an uncontrolled way. In particular, the non-homogeneous distribution of aluminium shows the broad transitions to the superconducting state  $\Delta T_c \geq 2\text{K}$  and their stepwise form [8,9]. There is also substantial variation in the parameters of the superconducting state. The influence of aluminium doping on the aging process also remains an open question. The present study investigates the effect of prolonged exposure in air atmosphere in different conductivity regimes of pure and aluminium doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals with a high critical temperature  $T_c$  and with unidirectional TB system with the orientation of transport current  $\mathbf{I} \parallel \text{TB}$ , when the influence of TB on the carriers' scattering is minimal.

## Experimental methodology

The  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (Re=Y, Al) single crystals were grown in a gold crucible with the solution-melting method, under small temperature gradient along the crucible [3,4,10]. As source components we used the  $\text{Y}_2\text{O}_3$ ,  $\text{BaCO}_3$ ,  $\text{CuO}$  powder compounds. To obtain aluminium doped single crystals  $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$ , we added 0.2wt.%  $\text{Al}_2\text{O}_3$ . For the resistivity measurements the single crystal with dimensions  $2.3 \times 0.75 \times 0.03 \text{ mm}^3$  was selected. In the sample the c-axis was oriented along the smallest dimension. The standard four-contact scheme was used to form the electric contacts. In this silver paste was applied onto the crystal surface and the connection of gold conductors (0.05mm in diameter) followed by 3 days annealing at  $420 \text{ }^\circ\text{C}$  in an oxygen atmosphere, in order to obtain the optimal oxygen concentration and high  $T_c$ . The technique provided a contact transition resistance of less than  $1\Omega$  and made it possible to measure the resistivity at transport currents up to 10mA in the ab-plane. After saturation with oxygen and fabrication of electric contacts, a bridge was cut of a crystal of thickness 0.02 mm as shown in the inset (a) of Fig. 1. The bridge width was 0.2 mm, and the separation between potential contacts was 0.3 mm. Twin boundaries in the bridge were oriented in the same direction. The bridge was cut so that the transport current vector  $I$  was parallel to twinning planes. The temperature was measured with a copper-constantan thermocouple. The first measurements of the electrical resistivity in the basal ab-plane were performed immediately after removing the crystal from the melt and their oxygen saturation to the optimum value ( $\delta \leq 0.1$ ). After these measurements, the crystal was stored in a glass container until the re-measurements, which were carried out after 6 years.

## Results and Discussion

The temperature dependence of the resistivity in the ab-plane  $\rho_{ab}(T)$ , measured before and after long time exposure in air atmosphere, is shown in Fig. 1. The resistive transitions to the superconducting state are shown in the inset (b) of Fig. 1, in the  $\rho_{ab}$  versus  $T$  and  $d\rho_{ab}/dT$  versus  $T$  coordinates. It is observed that in both cases (prior and after aging), the dependence is quasi-metallic, but the ratio  $\rho_{ab}(300\text{K})/\rho_{ab}(0\text{K})$ , before and after the long aging in air is significantly ( $\approx 34\%$ ) decreased from 12 to 8. In this respect, the value  $\rho_{ab}(0\text{K})$  was determined by interpolation of the linear section of the temperature dependence of the  $\rho_{ab}(T)$  dependence, as is shown in the Figure 1.

The electrical resistivity in the ab-plane at room temperature, increased from 421 to 453  $\mu\text{Ohm}\cdot\text{cm}$ , and the critical temperature dropped from 92.05 to 90.85 K. The width of the

resistive transition to the superconducting state  $\Delta T_c$ , increased two times, from 0.5 to  $\approx 1$  K and the transition is of a stepped form. The parameters of the samples are given in the Table 1.

Using previously published data on the dependence of  $T_c$  on the oxygen concentration [13], we can conclude that its oxygen content with aging is slightly decreases (by 1-2%) and remains within the limits  $\delta \leq 0.15$ . The increase in the width of the resistive transition, suggests a significantly reduced degree of homogeneity of the samples [3,4]. Additionally, it is consistent with the step-like form, which is observed after aging, showing signs indicating the appearance of phase separation in their volume [4,14]. This is also supported by the presence of a clearly pronounced additional peak in the dependence  $d\rho_{ab}/dT$  versus  $T$ . According to previous work [3,4,14], these peaks correspond to the  $T_c$  of different phases, differing in oxygen content within the volume in the crystal and therefore having different  $T_c$ .

Since **I** prior and after long aging is in the same orientation with the TB structure, the increase in electrical resistance cannot be due to the influence of the twin boundaries. In that respect the observed increase in  $\rho_{ab}$  is likely caused by a decrease of the density of the carriers or the emergence of effective scattering centers, as it is also evident from the change in the ratio  $\rho_{ab}(300K)/\rho_{ab}(0K)$ . This in turn is consistent with increasing the number of vacancies forming during the long time exposure in air and the increase of the degree of the oxygen's nonstoichiometry.

From Fig. 1 it is determined that when the temperature falls below a certain characteristic value  $T^*$ , a deviation of the  $\rho_{ab}(T)$  from the linear dependence occurs indicating the appearance of excess conductivity, which can be due to the transition to the pseudogap regime (PG) [15-17]. Currently there are two main scenarios of the pseudogap anomalies in HTSC systems. According to the first scenario, the occurrence of PG is associated with short-range order fluctuations of "dielectric" type, taking place in underdoped compounds (see for example the survey in [15]). The second scenario assumes the formation of Cooper pairs at temperatures significantly above the critical,  $T^* \gg T_c$ , followed by the establishment of its phase coherence at  $T < T_c$  [16,17].

Among the theoretical studies for the second scenario is the crossover theory from the BCS mechanism to the mechanism of the Bose-Einstein condensation (BEC) [17] in which the temperature dependence of the pseudogap in the cases of weak and strong coupling can be obtained. Generally, these relationships are described by the equation:

$$\Delta(T) = \Delta(0) - \Delta(0) \sqrt{\frac{\pi}{2}} \sqrt{\frac{T}{\Delta(0)}} \exp\left[-\frac{\Delta(0)}{T}\right] \left[1 + \operatorname{erf}\left(\sqrt{\frac{\sqrt{x_0^2 + 1} - 1}{T/\Delta(0)}}\right)\right], \quad (1)$$

where  $x_0 = \mu/\Delta(0)$  ( $\mu$  is the chemical potential of the carrier system and  $\Delta(0)$  is the value of energy gap at the zero temperature and  $erf(x)$  is the error function. In the case that of  $x_0 \rightarrow \infty$  (weak coupling), the Eq. (1) becomes:

$$\Delta(T) = \Delta(0) - \Delta(0)\sqrt{2\pi\Delta(0)T} \exp\left[-\frac{\Delta(0)}{T}\right], \quad (2)$$

that is well known in the BCS theory. In the limit of the strong interactions in the three-dimensional case ( $x_0 < -1$ ), the Eq. (1) becomes:

$$\Delta(T) = \Delta(0) - \frac{8}{\sqrt{\pi}} \sqrt{-x_0} \left(\frac{\Delta(0)}{T}\right)^{3/2} \exp\left[-\frac{\sqrt{\mu^2 + \Delta^2(0)}}{T}\right]. \quad (3)$$

As was shown previously [16], with a sufficiently high accuracy the values of the pseudogap can be determined in a wide temperature range from the dependency  $\rho_{ab}(T)$  (resistivity in the basal plane) at temperatures below a characteristic value  $T^*$  (pseudogap opening temperature).

From Table 1 and Figure 1, the prolonged annealing leads to a significant narrowing of the field of linear dependence  $\rho_{ab}(T)$  as compared to the original samples. Additionally, the temperature  $T^*$  is shifted to higher temperatures by about 30 K, indicating a corresponding increase of temperature interval of the excess conductivity existence.

The temperature dependence of the excess conductivity can be determined by the equation:

$$\Delta\sigma = \sigma - \sigma_0, \quad (4)$$

where  $\sigma_0 = \rho_0^{-1} = (A + BT)^{-1}$ , is the conductivity, determined by extrapolating the linear part to zero temperature and  $\sigma = \rho^{-1}$ , is the experimentally determined value of the conductivity in the normal state. The experimental dependencies of  $\Delta\sigma(T)$  are shown in Fig. 2 in  $\ln\Delta\sigma$  versus  $1/T$  coordinates. For a wide temperature range, these curves are straight lines corresponding to the description of the exponential dependence by the formula:

$$\Delta\sigma \sim \exp(\Delta^*_{ab}/T), \quad (5)$$

where  $\Delta^*_{ab}$ , is the value which defines the thermal activation process through the ‘‘pseudogap’’.

The exponential dependence of  $\Delta\sigma(T)$  was also observed earlier, in YBCO samples [16]. In previous work [16] it was demonstrated that the description of experimental data can be significantly expanded by introducing the factor  $(1 - T/T^*)$ . The excess conductivity becomes proportional to the superconducting carrier density  $n_s \sim (1 - T/T^*)$  and inversely proportional to the pairs  $\sim \exp(\Delta^*/kT)$ , that are destroyed by thermal motion:

$$\Delta\sigma \sim (1 - T/T^*) \exp(\Delta^*_{ab}/T), \quad (6)$$

where the temperature interval  $T_c < T < T^*$ , in which the PG regime exists is defined by the phase of the order parameter that depends on either the oxygen hypostoichiometry or the dopant concentration. Another specific mechanisms of the quasiparticle interaction, such as those caused

by structural or kinematic anisotropy of the system, can also be relevant [18-22]. Therefore, by using the method of previous work [16] we can construct the experimental curve  $\ln\Delta\sigma$  for the temperature dependence  $\Delta^*_{ab}(T)$  until the  $T^*$ .

Fig. 3 shows the temperature dependence of the PG in  $\Delta^*(T)/\Delta_{max}$  versus  $T/T^*$  coordinates, where  $\Delta_{max}$  is the  $\Delta^*$  value in the plateau, away from  $T^*$ , measured under different pressures. The  $\Delta^*(T)/\Delta(0)$  versus  $T/T^*$  dependence is calculated using Eqs (2) and (3) in the mean-field approximation within the context of the BCS–BEC crossover theory [17]. The values of the crossover parameters  $\mu/\Delta(0) = 10$  (limit of the BCS) and  $\mu/\Delta(0) = -2, -5, -10$  (limit of the BEC) are shown in Fig. 3 as dotted lines. From this figure it is observed that after long aging there is a shifting of the experimental curves from Eq. (2) (weak coupling) to Eq. (3) (strong coupling).

The value of  $\Delta^*$ , obtained from Eq. 5 is shown in the table. It is evident that prolonged annealing leads to a significant reduction in the absolute value of the pseudogap  $\Delta^*_1/\Delta^*_2 \approx 1.28$ . From Fig. 2 it can be deduced that by approaching the  $T_c$ , a sharp increase in the value of  $\Delta\sigma$  takes place. From theory [23] it is known that near the  $T_c$ , the excess conductivity is caused due to fluctuation pairing of the carriers' processes, whose contribution to the conductivity at  $T > T_c$  for the 2D and 3D cases is determined by the degree of the dependence by the following relations:

$$\Delta\sigma_{2D} = \frac{e^2}{16\hbar d} \varepsilon^{-1}, \quad (7)$$

$$\Delta\sigma_{3D} = \frac{e^2}{32\hbar\xi_c(0)} \varepsilon^{-1/2}, \quad (8)$$

where  $\varepsilon = (T-T_c)/T_c$ ,  $e$  is the electron charge,  $\xi_c(0)$ , is the coherence length along the  $c$ -axis at  $T \rightarrow 0$  and  $d$ , is the characteristic size of a two-dimensional layer. Here  $T_c$  is defined by the maximum on  $d\rho_{ab}(T)/dT$  dependencies in the superconducting transition region (inset (b) to Fig. 1).

The inset of Fig. 2, represents the temperature dependence of  $\Delta\sigma(T)$  in  $\ln\Delta\sigma$  versus  $\ln\varepsilon$  coordinates. Near  $T_c$ , these relationships are satisfactorily approximated by straight lines with a slope  $\text{tg}\alpha_1 \approx -0.5$ , corresponding to the exponent  $-1/2$  in Eq. 8. This clearly indicates the three-dimensional nature of the fluctuation superconductivity in this temperature interval. At higher temperatures the rate of decrease of  $\Delta\sigma$ , is significantly increases ( $\text{tg}\alpha_2 \approx -1$ ), which, in turn is an indication of the change in the dimension of the fluctuation conductivity. As follows from Eq. 7 and Eq. 8, at the 2D-3D crossover:

$$\xi_c(0)\varepsilon_0^{-1/2} = d/2. \quad (9)$$

In this case, determining the value  $\varepsilon_0$  and using the published data regarding the dependence of the inter-plane distance from  $\delta$  [10] ( $d \approx 11.7 \text{ \AA}$ ), we can calculate the value of  $\xi_c(0)$ . Calculations have shown that in the process of aging, a change in the value of the coherence length from  $\xi_c(0) = 2.32 \text{ \AA}$  up to  $\xi_c(0) = 2.54 \text{ \AA}$  occurs. Finally, the 2D-3D crossover point is significantly shifted by the temperature (Table 1 and inset in Fig. 2).

## Conclusions

The long time exposure in air of the aluminium doped  $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$  single crystals, leads to the partial degradation of the conducting properties and the emergence of effective scattering centers of the carriers. The excess conductivity,  $\Delta\sigma(T)$  of the  $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$  single crystals, in a wide temperature range  $T_f < T < T^*$  has an exponential temperature dependence and in the case of approximation to  $T_c$ , is well described within the Aslamazov-Larkin theoretical model. In this case, the temperature dependence of the pseudogap, is satisfactorily described in the framework of the BCS-BEC crossover theory. Prolonged annealing of  $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$  single crystals facilitates to a great extent of the temperature range of the implementation of the pseudogap state in the ab-plane, thus narrowing the linear portion of the dependence  $\rho_{ab}(T)$ . In the aging process there are signs of phase separation in the volume of the experimental samples, which are shown in the presence of additional peaks in the curves  $d\rho_{ab}(T) / dT$  in the superconducting transition region.

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**Table 1** The experimental parameters.

Sample	$T_c$ , K	$\rho_{ab}(300)$ , $\mu\Omega.cm$	$T^*$ , K	$\Delta^*_{ab}$ , meV	$\epsilon_0$	$\alpha_{3D}$	$\alpha_{2D}$	$\xi_c(0)$ , Å
Before aging	92.05	421	199	58.1	0.157	-0.506	-0.990	2.32
After aging	90.85	453	228	45.5	0.188	-0.495	-1.017	2.54

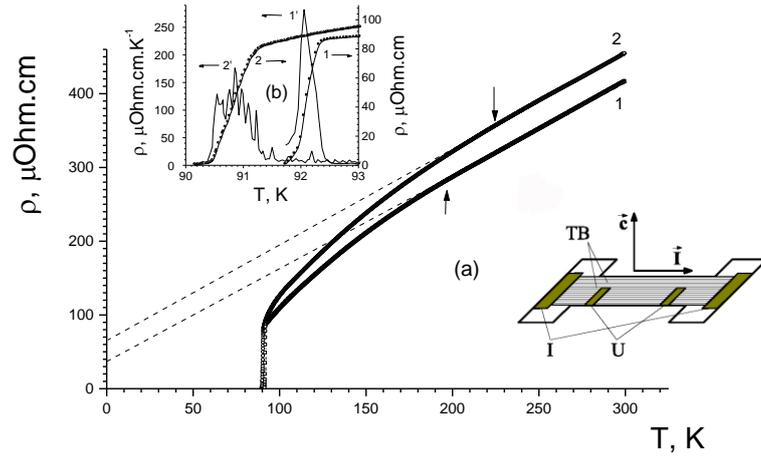


Fig.1. The  $\rho_{ab}(T)$  dependence of the  $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$  single crystals, before and after long aging in air, curves 1 and 2, respectively. The arrows show the mean field transition temperature to the pseudogap regime,  $T^*$ . The inset (a) shows the transitions to the superconducting state in  $\rho_{ab} - T$  and  $d\rho_{ab}/dT - T$  coordinates for the same samples. The numbering of the curves in the inset corresponds to the numbering in the figure. The inset (a) shows the experiment geometry.

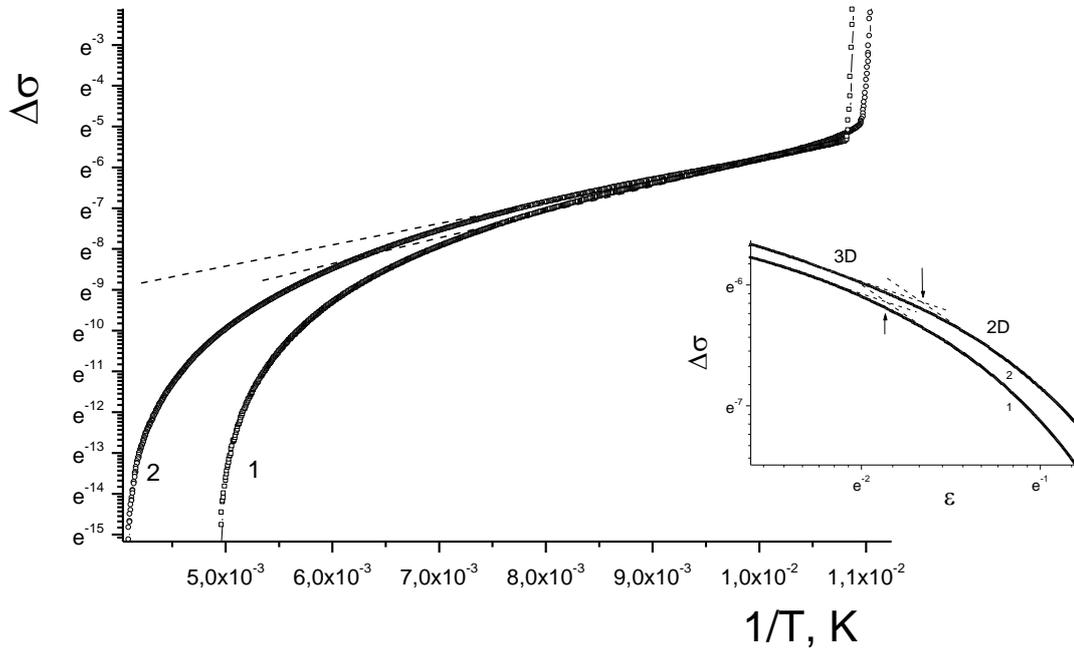


Fig. 2. The temperature dependence of the excess conductivity in the ab-plane for the  $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$  single crystal in  $\ln\Delta\sigma - 1/T$  versus  $\ln\Delta\sigma - \ln\varepsilon$  coordinates (inset). The names (notation) of the curves is relevant to the figure 1. The dashed lines in figure 2 shows the approximation of equation (5), and in the inset, the straight lines – the approximation with a slope  $\text{tg}\alpha_1 \approx -0.5$  (3D regime) and  $\text{tg}\alpha_2 \approx -1.0$  (2D regime). The arrows show the point of the 2D-3D crossover.

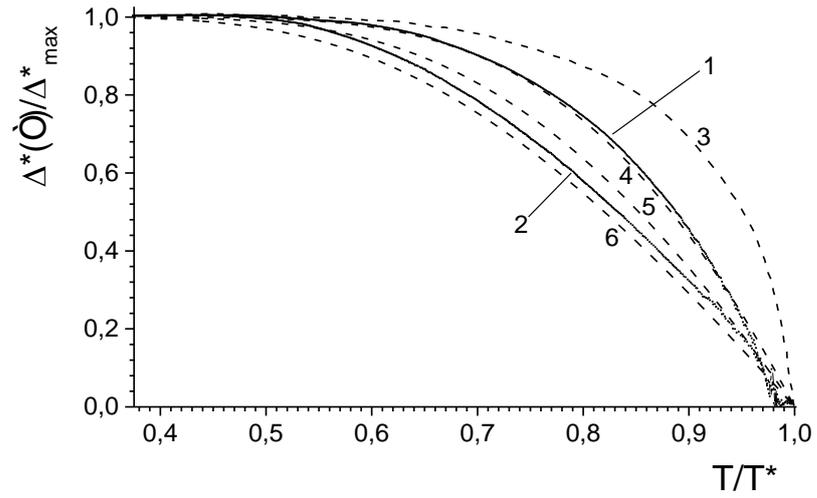


Fig. 3. The temperature dependence of the PG for the  $\text{YBa}_2\text{Cu}_{3-y}\text{Al}_y\text{O}_{7-\delta}$  single crystal, in  $\Delta^*(T)/\Delta_{\max}$  versus  $T/T^*$  coordinates, where  $\Delta_{\max}$  is the  $\Delta^*$  value in the plateau, away from  $T^*$ . The numbering of the curves corresponds to the numbers in the Fig. 1. Dotted lines for the  $\Delta^*(T)/\Delta(0)$  versus  $T/T^*$  dependence were calculated according Eq. (2) and (3) for the values of the crossover parameters  $\mu/\Delta(0)=10$  (limit of the BCS, curve 3) and  $\mu/\Delta(0) = -2, -5, -10$  (limit of the BEC, curves 4-6).