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METALS AND SUPERCONDUCTORS

Andreev Reflection and Experimental Temperature Dependences of the Critical Current in Heterogeneous High-Temperature Superconductors (Polycrystals and Related Composites)

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Abstract—The temperature dependences of the critical current of $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ polycrystalline high-temperature superconductors subjected to annealing for different times and those of $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ + $BaPbO_3$ composites were measured. The results obtained were analyzed in terms of the Gunsenheimer–Schüssler–Kümmler theory considering the Andreev reflection of carriers in a "superconductor–normal metal–superconductor" (*S*–*N*–*S*) junction. Good agreement between the experimental and theoretical temperature dependences of the critical current over a wide temperature range for polycrystals with both natural and artificially created boundaries (composites) made it possible to estimate the effective length of grain boundaries in the high-temperature superconductors. It was revealed that the critical current density measured at 4.2 K is an exponential function of the length of grain boundaries in samples of both types.

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1. INTRODUCTION

The transport properties of polycrystalline high-temperature superconductors are determined for the most part by the quality of the boundaries between crystallites, which are considered the main factor limiting the critical current in high-temperature superconductors (HTSCs). These boundaries are Josephson-type weak links for which the larger the geometric length, the smaller the critical current. The transport properties of granular high-temperature superconductors are governed by the percolation structure of the sample, the grain misorientation [1], and the internal physical mechanisms affecting the Josephson effects in the "superconducting grain–grain boundary–superconducting grain" structure [2]. In turn, the Josephson effects depend on the character of the grain boundaries (metal, insulator, etc.), which ultimately determines the critical current density and its specific temperature dependence [2–6]. In this work, we analyzed the experimental temperature dependences of the critical current $I_c(T)$ of high-temperature superconductor polycrystals annealed for different times and those of "high-temperature superconductor + $BaPbO_3$ normal metal" composites. It was shown that the Gunsenheimer–Schüssler–Kümmler theory [7] developed for "superconductor–normal metal–superconductor" (*S*–*N*–*S*) Josephson junctions adequately describes the experimental dependences $I_c(T)$ and offers reasonable estimates of the effective length of *N* boundaries between superconducting grains.

Earlier analyses [8, 9] of the current–voltage characteristics of yttrium-based polycrystalline high-temperature superconductors showed that the grain boundaries in these systems have a metallic character. In this work, we studied samples prepared according to the same technology as was used in [8, 9]. This gives grounds to analyze the experimental dependences $I_c(T)$ in the framework of the theories developed for *S*–*N*–*S*-type Josephson junctions. In composites based on high-temperature superconductors, the nonsuperconducting component plays the part of interlayers separating the high-temperature superconductor grains [4, 5, 10–13], thus permitting one to purposefully choose the type of weak link. The metal oxide $BaPbO_3$ is known to exhibit metallic properties [14, 15]. Therefore, the experimental data obtained for the HTSC + $BaPbO_3$ composites can be treated with the theories developed for *S*–*N*–*S* junctions.

The Josephson current in *S*–*N*–*S* junctions was considered in [7, 16–18]. It was shown that a transfer of Cooper pairs through *S*–*N*–*S* junctions occurs as a result of Andreev reflection. The theories developed in recent years to treat the Andreev reflection in calculating the critical current and current–voltage characteristics of *S*–*N*–*S* junctions adequately describe the experimentally observed features in the transport properties of these structures [7, 16–28]. For example, the theory formulated in [20] for describing the current–voltage characteristics of *S*–*N*–*S* structures accounts for the subharmonic gap structure, excess current, and portions of the current–voltage characteristics with negative differential resistance. Therefore, when analyzing high-temperature superconductor polycrystals with grain boundaries of a metallic nature, it is reasonable to consider the mechanisms of Andreev reflection in the "HTSC grain–grain boundary–HTSC grain" structure.

A polycrystal is characterized by a distribution of grain boundaries over their length. Hence, a polycrystalline high-temperature superconductor can be used as a physical model of a random array of Josephson junctions, which is characterized by a distribution over coupling energies and critical currents [1, 29]. We assume that, as a first approximation, the behavior of the critical current of an array of Josephson junctions can be identified with the behavior of the critical current of a single junction with an effective geometric length. As will be shown below, the calculations conducted within the Gunsenheimer–Schüssler–Kümmler theory [7] account for the experimentally observed reversal of the curvature sign in the dependence $I_c(T)$ of the samples studied. The inclusion of the junction distribution over geometric sizes with the use of the results obtained from microscopic theories of the Josephson effect leads to a more realistic pattern of current flow through a high-temperature superconductor polycrystal. This is, however, a problem in itself.

Earlier [11], the experimental dependences $I_c(T)$ of the composites consisting of a high-temperature superconductor and metal oxide $BaPbO_3$ were analyzed using the theoretical dependences $I_c(T)$ derived in [7] for the *S*–*N*–*S* junction based on the classical low-temperature superconductor. Starting from the agreement between the theoretical curves calculated in [7] for the lengths of *N* interlayers $2a = 1.57\xi_0$ and $5.23\xi_0$ (where ξ_0 is the coherence length) and the experimental dependences $I_c(T)$, as well as from the assumption that $2a \sim V^{1/3}$ (where V is the volume concentration of the metal in the composite), we obtained a rough estimate of the effective length of intergranular interlayers in the composite. In order to make a more accurate comparison of the experimental and theoretical dependences $I_c(T)$ and to estimate the effective thickness of the grain boundaries, both calculation of the curves $I_c(T)$ with the use of microscopic parameters of high-temperature superconductors and a thorough fitting of the theoretical dependences to the experimental data are required. This paper reports on the results of such a comparison and on the determination of the effective thickness of the grain boundaries according to the best fit of the Gunsenheimer–Schüssler–Kümmler theory [7] to experiment.

2. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUE

Polycrystalline high-temperature superconductors of the composition $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ were synthesized by the standard solid-state reaction technique. The Debye powder patterns revealed reflections only from the 1–2–3 structure. The last stage of sample preparation included final baking at 930°C for different times. The baking time was varied from the shortest procedure (5 min) to 150 h. We denote the samples in accordance with their baking times, namely, YBCO-5 min, YBCO-5 h, YBCO-30 h, and YBCO-150 h. Electron microscope images of the samples baked for 5 and 150 h showed that long baking brings about an increase in the grain size from ~2 to 10 μm [30].

The nonsuperconducting component of the composites, namely, $BaPbO_3$, was prepared by the solid state reaction technique from BaO_2 and PbO at temperatures of 650–700°C with three intermediate grindings for 20 h. The Debye powder patterns showed reflections from the perovskite structure only.

The $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7 + BaPbO_3$ composites were prepared by the fast sintering technique described earlier in [11]. The temperature regime was as follows: 5 min at 930°C, followed by 6 h at 400°C. The Debye powder patterns of the composites contained reflections only from the two phases of the initial ingredients. Electron microscope images showed that the average size of high-temperature superconductor grains in the composite is ~1.5 μm. In what follows, the composite samples will be denoted by YBCO + VBaPbO$_3$. Here, V is the volume content (in %) of the $BaPbO_3$ metal oxide; the volume content of the high-temperature superconductor (YBCO) is, accordingly, $100\% - V$.

Electrical measurements were performed by the standard four-point probe method. The sample was cut out in the form of a parallelepiped 1 × 1 × 10 mm in size. Next, the sample was attached to a sapphire substrate. The central part of the sample, to which gilded potential contacts were subsequently pressed, was ground to a thickness of ~200 μm. Bulk silver current contacts were mechanically pressed to sample pads coated with an In–Ga eutectic. In the high-temperature superconductor with the highest critical current density, finely dispersed silver paste was fired into the contact pads of the sample. The measurements were performed with the sample placed in a helium heat-exchange atmosphere. All the above precautions permitted us to decrease the heat release at the current contacts. In the case where the transport critical current at liquid-helium temperature reached ~4.5 A (the YBCO-5 h sample), the sample was observed to heat (by no more than ~2 K). We measured the initial parts of the current–voltage characteristics at different temperatures. The critical current I_c was derived from the initial part of the current–voltage characteristics in accordance with the standard criterion of 1 μV/cm [31]. The value of I_c at 4.2 K was rechecked by measuring the current–voltage characteristic of the sample immersed directly in liquid helium. In this way, the temperature dependences of the critical current of the composites and high-temperature superconductors without additions were measured.

The superconducting transition temperature of the composites was derived from magnetic measurements and amounted to 93.5 K, which corresponds to the critical temperature T_c of the $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ initial compound. The dependences $R(T)$ for the YBCO + VBaPbO$_3$ composites are presented in [11]. They exhibit a two-step structure characteristic of granular superconductors [32], namely, a sharp jump at 93.5 K corresponding to the superconducting transition in high-temperature superconductor grains, followed by a smooth transition to the state with $R = 0$, which can be ascribed to grain boundaries.

3. GUNSENHEIMER–SCHÜSSLER–KÜMMLER THEORY FOR THE CRITICAL CURRENT OF *S–N–S* JUNCTIONS: ESTIMATION OF THE RANGE OF ITS APPLICABILITY TO HIGH-TEMPERATURE SUPERCONDUCTOR POLYCRYSTALS

Out of all the above papers dealing with the calculation of the Josephson current, only the theories explicated in [7, 16] permit one to calculate the temperature dependence of the critical current of *S–N–S* junctions with metal layers of different thicknesses. Both models predict the change in the curvature sign of the temperature dependence of the critical current with increasing thickness of the metal layer. The theory formulated in [16] also predicts the existence of a plateau in the dependence $I_c(T)$ in the low-temperature range (near 0 K). Because the experiments we are aware did not reveal such a feature for *S–N–S* junctions, we used the theory proposed in [7] for describing the experimental data obtained.

In the Gunsenheimer–Schüssler–Kümmler theory [7], the transfer of Cooper pairs through an *S–N–S* junction is considered to occur as a result of the Andreev reflection and the critical current of clean *S–N–S* junctions (for which the mean free path of carriers l in the N interlayer is larger than the length of the N interlayer, $l > 2a$ [33]) is calculated throughout the temperature range below T_c and for arbitrary values of $2a$. The dependence of the current density in *S–N–S* junctions on the phase difference Φ of superconducting banks is represented by the following relationship [7]:

$$
\begin{aligned}
j(\Phi) = e_z - \frac{e}{L_x L_y}\Bigg\{ \sum_{k}^{E \le \Delta} \frac{\hbar k_h}{2ma^*}\bigg[& \tanh(E_k^+/2k_B T)\frac{a^*}{a^* + \lambda_k^+} \\
& - \tanh(E_k^-/2k_B T)\frac{a^*}{a^* + \lambda_k^-}\bigg] \qquad (1) \\
+ \sum_{k}^{E > \Delta} \frac{\hbar k_h}{mD} & \tanh(E/2k_B T)[|\eta^+|^2 - |\eta^-|^2]\Bigg\},
\end{aligned}
$$

where $L_x L_y$ is the area of the normal region, $2a^*$ is the effective length of the normal region, $2D$ is the length of the *S–N–S* junction, Δ is the superconductor energy gap, λ_k is the penetration depth, e is the elementary charge, m is the effective mass of the electron, k_B is the Boltzmann constant, $\hbar$ is the Planck constant, η^+ and η^- are the components of the quasiparticle wave functions, E_k is the quasiparticle energy derived from the relationship $E = \hbar^2 k_{zF}^2/(2a^*m)[n\pi + \arccos(E/\Delta) \pm \Phi]$, k_{zF} is the projection of the quasiparticle wave vector on the current flow direction z, and $n = 0, 1, 2, \ldots$. The critical current density j_c is determined as the maximum in the dependence $j(\Phi)$. In order to obtain the temperature dependence of the critical current, one has to calculate expression (1) for different temperatures with due regard for the dependence $\Delta(T)$. We took the dependence $\Delta(T)$ from the Bardeen–Cooper–Schrieffer theory for $\Delta(0) = 17.5$ meV and $m = 5m_e$, $E_F = 0.323$ eV [34, 35]. Note that, for $2a/\xi_0 \longrightarrow 0$, the Gunsenheimer–Schüssler–Kümmler theory [7] leads to both the well-known result of Kulik and Omel'yanchuk [36] for short microbridges in the clean limit and the result of Kupriyanov [37] for clean *S–N–S* sandwiches.

Different estimates of the length of grain boundaries indicate that this length ranges from several angströms to several tens of angströms for different high-temperature superconductor polycrystals [3, 9, 38, 39]. According to Delin and Kleinsasser [38], the mean free path l for an yttrium high-temperature superconductor is equal to 6.5 nm [38]. This parameter for grain boundaries should be apparently somewhat smaller than that in crystallites. Nevertheless, in most cases, the clean limit condition (equivalent to the inequality $l > 2a$) for natural boundaries of yttrium high-temperature superconductor polycrystals should be satisfied. In composites with BaPbO$_3$, the effective length of N boundaries is larger than that in pure polycrystals; however, the mean free path of carriers in the material of N interlayers is also larger. According to the estimates obtained by Kitazawa et al. [14], the mean free path l for BaPbO$_3$ is approximately equal to 220 Å in the low-temperature range. Thus, we come to the conclusion that, in this case, the clean limit condition for the HTSC + BaPbO$_3$

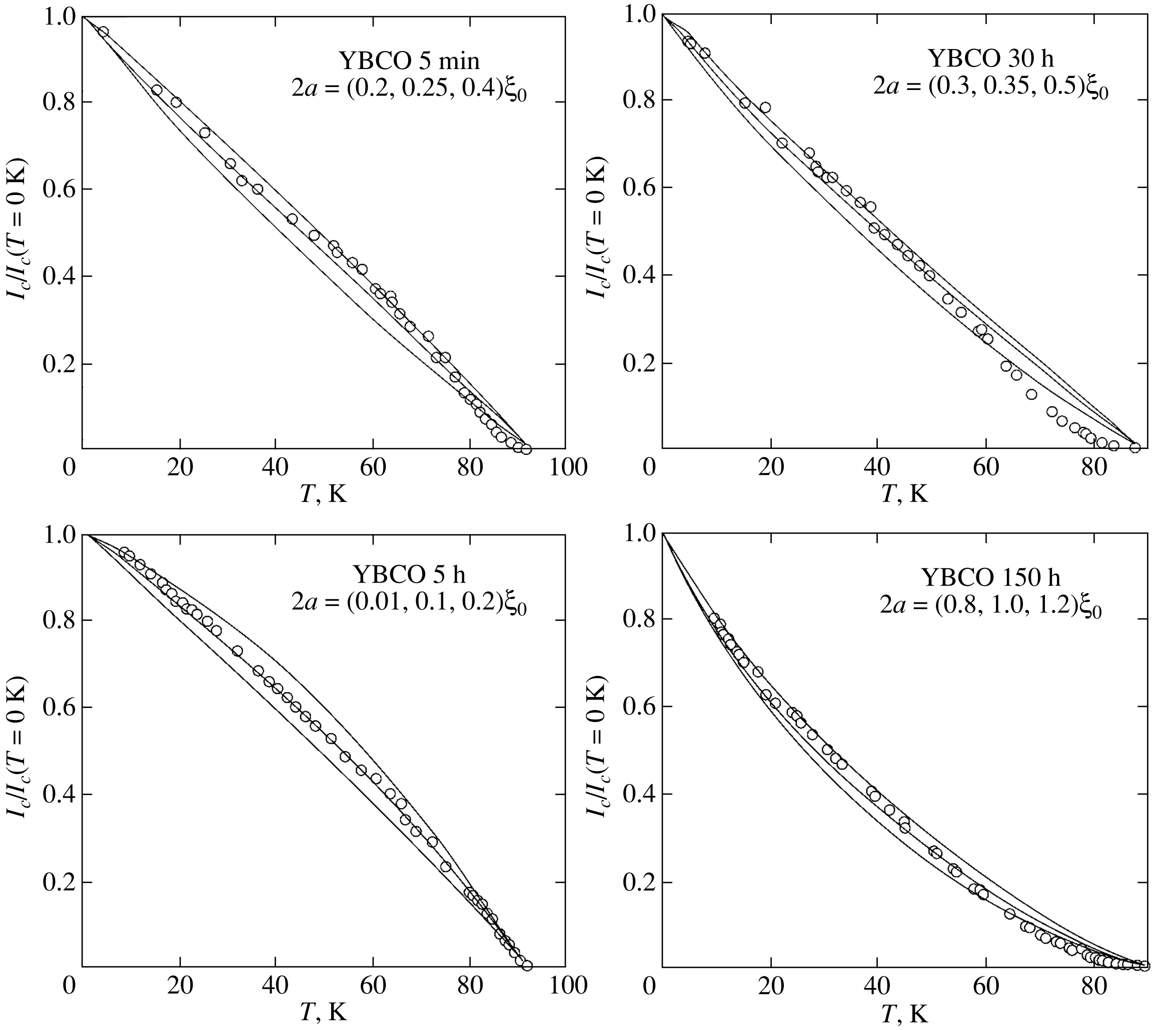

Fig. 1. Temperature dependences of the critical current density in polycrystalline high-temperature superconductors synthesized with different baking times (circles) and theoretical dependences $I_c(T)$ normalized to the value of $I_c(T = 0)$ for the S–N–S junction [7] (solid curves). The values of $2a$ for the theoretical dependences are specified in the figure. The smaller value of $2a$ corresponds to the upper theoretical curve.

composite is also satisfied and the Gunsenheimer–Schüssler–Kümmler theory [7] is valid.

4. RESULTS AND DISCUSSION

4.1. $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ Polycrystalline High-Temperature Superconductors Synthesized at Different Baking Times

Figure 1 shows the normalized experimental dependence $I_c(T)$ for high-temperature superconductors baked for different times. The transformation of the dependence $I_c(T)$ is noteworthy. As the baking time increases to 150 h, the curvature of the dependence $I_c(T)$ reverses sign. We calculated the theoretical dependences $I_c(T)$ from the Gunsenheimer–Schüssler–Kümmler theory [7]. Using the thickness $2a$ of the N interlayers (in units of the coherence length ξ_0) as a fitting parameter, we obtained the best fit of the theoretical dependence to the experimental curve $I_c(T)$ over a wide temperature range. In this case, the low-temperature range was taken as a basis. The solid curves in Fig. 1 are the theoretical dependences. The middle of the three curves is the result of the best fit. The flanking curves indicate values of I_c obtained under a slight variation in $2a$ and illustrate the limits of reasonable fitting. We readily observe good agreement between the experimental and fitting dependences $I_c(T)$. Figure 2a plots the dependence of the fitting parameter $2a$ on the baking time of the high-temperature superconductors. Different authors quote different values of ξ_0 ranging from 2 to 3 nm for the YBCO system [34, 35]. Assuming $\xi_0 = 24$ Å [34], we obtain 5.0 ± 1.0 and 2.5 ± 1.0 Å for the effective length of grain boundaries in samples baked for 5 min and 5 h, respectively. It is difficult to conceive of an grain boundary of such a small length. One should take into account, however, that the Gunsenheimer–Schüssler–Kümmler theory [7] makes use of the effective thickness of the N layer, which is always smaller than the real value because of the proximity effect [33]. The high critical current densities in high-temperature superconductor polycrystals in strong magnetic fields are likewise accounted for by the presence of superconducting shorts between the grains [5], i.e., between grain boundaries of extremely short length. Prolonged baking brings about an increase in the effective length of grain boundaries up to 8.5 ± 1.0 Å (30 h) and 24 ± 2 Å (150 h). Such a transformation of the boundaries appears to be within reasonable limits for the polycrystals under study, because, as was shown by electron microscopy [30], prolonged baking leads to a substantial increase in the size of the grains. Figure 2b presents the dependence of the critical current density j_c at 4.2 K on the baking time of the high-temperature superconductors. Samples with smaller effective lengths of grain boundaries are seen to exhibit the highest critical current densities. Thus, the data derived from our experimental dependences $I_c(T)$ suggest that the effective length of boundaries in polycrystalline high-temperature superconductors can reach a few angstroms in the best samples. Similar estimates of the length of boundaries between grains were obtained by other researchers for films [3], bicrystals [40, 41], and also by us in an analysis of the experimental current–voltage characteristics of high-temperature superconductor polycrystals [9, 39]. The maximum critical current can obviously be obtained with baking of an optimum duration. For our series of high-temperature superconductor samples fabricated at different baking times, a 5-h final baking was found to be optimum.

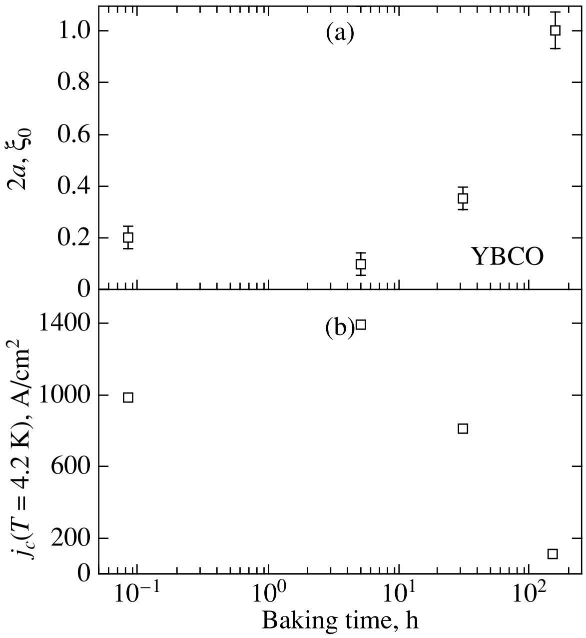

Fig. 2. (a) Effective length of grain boundaries $2a$ (in terms of ξ_0) in the high-temperature superconductors synthesized with different baking times and (b) critical current density j_c at 4.2 K as a function of the baking time of polycrystals (log scale).

4.2. $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7 + BaPbO_3$ Composites

Figure 3 shows the experimental dependences $I_c(T)$ of YBCO + VBaPbO$_3$ composites. To begin with, all the graphs have positive curvature, as is the case with a pure high-temperature superconductor subjected to long baking (Fig. 1). The temperature of the disappearance of the critical current decreases with increasing BaPbO$_3$ concentration in the composite. The solid curves in Fig. 3 are the best fits (the middle curves) of the experiment to the Gunsenheimer–Schüssler–Kümmler theory [7]. The flanking theoretical curves depicted in Fig. 3 have the same meaning as those in Fig. 1; i.e., they bound the reasonable fitting stripe. Theory is seen to agree well with experiment over a wide temperature range. Figure 4a plots the dependence of the best-fit value of the parameter $2a$ on the volume concentration of the nonsuperconducting component of the composites. The result obtained was both expected and logical; namely, the effective length of the N boundaries between grains increases with increasing volume concentration of BaPbO$_3$. Using $\xi_0 = 24$ Å [34], we arrive at the conclusion that, as the volume concentration of BaPbO$_3$ in the composite increases from 3.75 to 45.00%, the effective length of BaPbO$_3$ grain boundaries increases monotonically from 35 ± 5 to 105 ± 7 Å. Figure 4b shows the dependence of the critical current density j_c at 4.2 K (log scale) on the volume concentration of BaPbO$_3$. As in the case of pure high-temperature superconductors (Fig. 2), the critical current density j_c (T = 4.2 K) decreases because of the increase in the effective length of grain boundaries.

4.3. Dependence of the Critical Current Density j_c (T = 4.2 K) on the Effective Length of Grain Boundaries

Figure 5 shows the dependence of the critical current density j_c at 4.2 K (on a log scale) on the best-fit value of the parameter $2a$ derived from the experimental curves $I_c(T)$. The points fall well on a straight line, even though a considerable scatter is observed near $2a = (1–2)\xi_0$ (the data for YBCO-150 h, YBCO + 3.75BaPbO$_3$, YBCO + 7.5BaPbO$_3$). Nevertheless, we can conclude that the dependence of the critical current density j_c on the thickness of N boundaries between high-temperature superconductor grains has the form $j_c \sim \exp(–2a)$, which is characteristic of many Josephson structures [31, 36] based on both low-temperature and high-temperature superconductors.

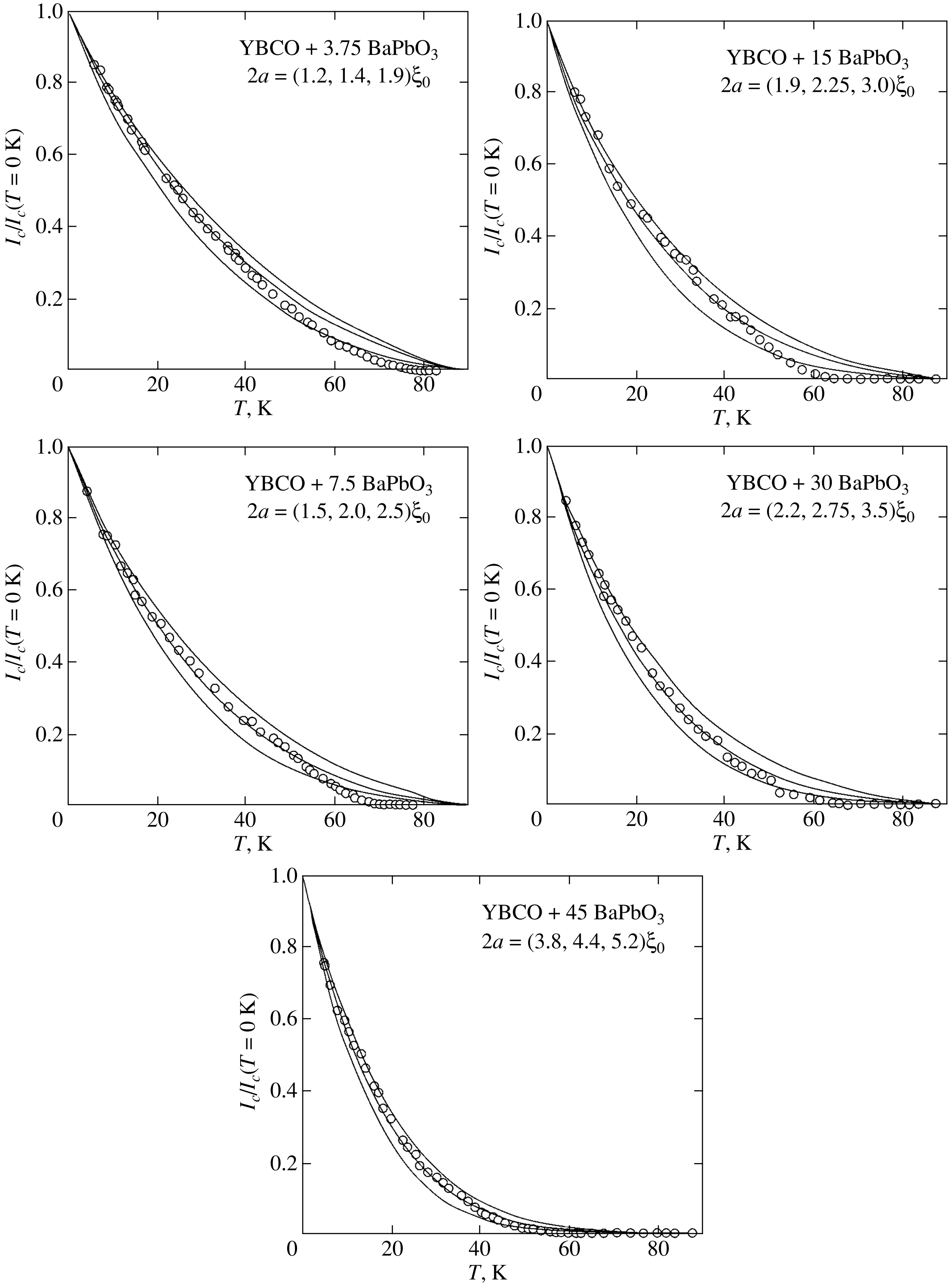

Fig. 3. Temperature dependences of the critical current density of YBCO + VBaPbO$_3$ composites (circles) and theoretical dependences $I_c(T)$ normalized to the value of $I_c(T = 0)$ for the S–N–S junction [7] (solid curves). The values of 2a accepted for the theoretical dependences are specified in the figure. The smaller value of 2a corresponds to the upper theoretical curve.

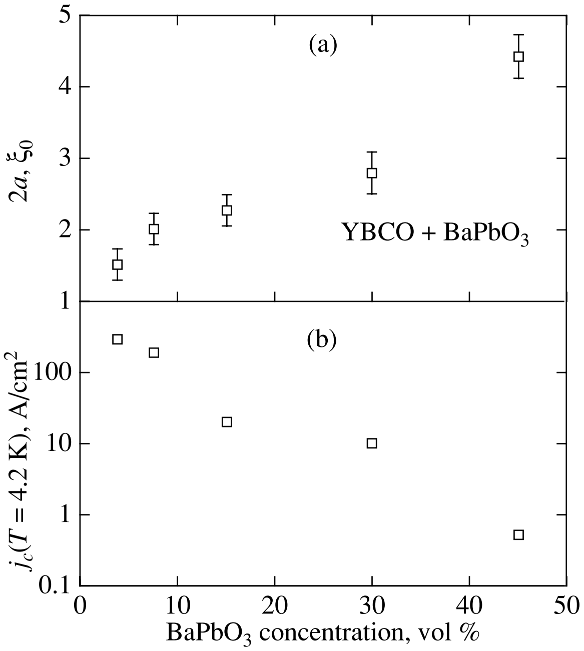

Fig. 4. (a) Effective length $2a$ (in terms of ξ_0) of $BaPbO_3$ interlayers in YBCO + $V$$BaPbO_3$ composites and (b) critical current density j_c at 4.2 K as a function of the volume concentration of $BaPbO_3$ in the composite.

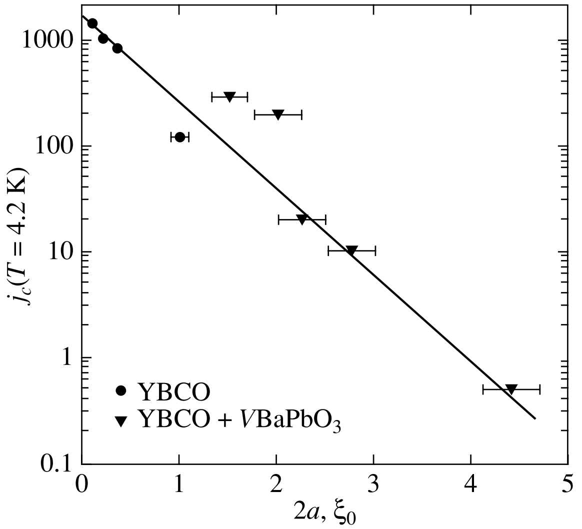

Fig. 5. Dependence of the critical current density j_c at 4.2 K (log scale) on the effective length of N boundaries (in terms of ξ_0) between high-temperature superconductor grains in the samples under study. The straight line is the best fit with an exponential function.

5. CONCLUSIONS

Thus, we performed an analysis of the temperature dependences of the critical current of heterogeneous yttrium-based high-temperature superconductors of two types, namely, with natural grain boundaries and artificially created N ($BaPbO_3$) interlayers. It was demonstrated that the experiment is in good agreement with the Gunsenheimer–Schüssler–Kümmler theory [7] accounting for the Andreev reflection in S–N–S junctions. A comparison of the theoretical and experimental dependences $I_c(T)$ made it possible to estimate the effective length $2a$ of metallic boundaries between the high-temperature superconductor grains. These values are in reasonable agreement with the data available in the literature; more precisely, prolonged baking brings about an increase in the size of the high-temperature superconductor grains and an increase in the length of the grain boundaries. For HTSC + $BaPbO_3$ composites, the value of $2a$ increases monotonically with increasing content of the nonsuperconducting component. It was found that the critical current density of bulk high-temperature superconductors decreases exponentially with increasing effective length of the grain boundaries. The results obtained suggest that, in order to adequately analyze the transport properties of polycrystalline superconductors, the percolation pattern of current flow should be complemented by inclusion of the internal physical processes occurring in weak links. The Gunsenheimer–Schüssler–Kümmler theory [7] taking into account the Andreev reflection has permitted us not only to obtain quantitative estimates but also to find an explanation for such a nontrivial effect as the curvature sign reversal of the temperature dependence of the critical current with increasing effective length of grain boundaries in polycrystalline high-temperature superconductors.

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