ISSN 1063-7761, Journal of Experimental and Theoretical Physics, 2010, Vol. 110, No. 4, pp. 584–593. © Pleiades Publishing, Inc., 2010.
Original Russian Text © D.A. Balaev, A.A. Dubrovskiy, S.I. Popkov, K.A. Shaikhutdinov, O.N. Mart'yanov, M.I. Petrov, 2010, published in Zhurnal Éksperimental'noĭ i Teoreticheskoĭ Fiziki, 2010, Vol. 137, No. 4, pp. 664–674.

ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

Nonmonotonic Behavior of Magnetoresistance, *R*(*H*) Hysteresis, and Low-Temperature Heat Capacity of the $BaPb_{0.75}Bi_{0.25}O_3$ Superconductor in a Magnetic Field: Possible Manifestations of Phase Separation

D. A. Balaev[a,b,*], **A. A. Dubrovskiy**[a], **S. I. Popkov**[a,b], **K. A. Shaikhutdinov**[a,b], **O. N. Mart'yanov**[c], **and M. I. Petrov**[a]

[a] *Kirenskii Institute of Physics, Siberian Branch, Russian Academy of Sciences, Krasnoyarsk, 660036 Russia*
[b] *Siberian Federal University, Krasnoyarsk, 660041 Russia*
[c] *Boreskov Institute of Catalysis, Siberian Branch, Russian Academy of Sciences, Novosibirsk, 630090 Russia*
**e-mail: smp@iph.krasn.ru*

Received May 13, 2009

Abstract—The transport properties (*R*(*T*) and *R*(*H*) dependences at various values of the transport current in magnetic fields up to 65 kOe) and low-temperature heat capacity in magnetic fields up to 90 kOe of the $BaPb_{0.75}Bi_{0.25}O_3$ superconductor ($T_C \approx 11.3$ K) are investigated with the goal of clarifying the mechanisms determining the nonmonotonic behavior and hysteresis of its magnetoresistance *R*(*H*). The type of *R*(*H*) hysteretic dependences for $BaPb_{0.75}Bi_{0.25}O_3$ is analogous to that observed in granular high-T_c superconductors (HTSCs); however, unlike classical HTSC systems, the field width of the magnetoresistance hysteresis loop for polycrystalline $BaPb_{0.75}Bi_{0.25}O_3$ depends on the transport current. This means that although the mechanisms responsible for the magnetoresistance hysteresis (the influence of the magnetic flux trapped in superconducting regions on the effective field in Josephson interlayers) are identical in these objects, the transport current in $BaPb_{0.75}Bi_{0.25}O_3$ may considerably affect the diamagnetic response of the superconductor. A considerable effect of transport current on the field in which the *R*(*H*) dependences have a peak and exhibit hysteretic properties is observed. Such a behavior can be adequately interpreted using the model of the spatially inhomogeneous superconductor–insulator state proposed by Gorbatsevich et al. [JETP Lett. **52**, 95 (1990)]. The nonmonotonic dependence of quantity C/T (*C* is the heat capacity) on the magnetic field discovered in the present study also agrees with the conclusions based on this model.

DOI: 10.1134/S1063776110040059

1. INTRODUCTION

In spite of the long history of investigating the $BaPb_{1-x}Bi_xO_3$ system (in which superconductivity was discovered in 1975 [1]), some questions have remained unanswered. The maximal value of $T_C \approx 12$ K is observed for $x \approx 0.25$. In the range of values of x for which superconductivity exists, $BaPb_{1-x}Bi_xO_3$ polycrystalline samples exhibit a number of interesting magnetoresistive effects such as recurrent superconductivity [2, 3] and current–voltage (*I–V*) characteristics typical of Josephson structures [4, 5]. These features were attributed to the granular structure of the samples and the existence of Josephson bonds at grain boundaries. In addition, anomalous behavior of the magnetoresistance (i.e., the peak on the *R*(*H*) dependence at $T =$ const) was observed [6, 7]. Similar properties were also discovered in the $Ba_{0.6}K_{0.4}BiO_3$ system ($T_C \approx 30$ K) [8]. A possible explanation for the nonmonotonic behavior of magnetoresistance in $Ba_{0.6}K_{0.4}BiO_3$ was also proposed in [8] using the model of a spatially inhomogeneous superconductor–insulator state [9]. In view of the similarity of these systems, this model [9] can probably also explain anomalies in the *R*(*H*) dependence of $BaPb_{1-x}Bi_xO_3$. The goal of this study is to analyze the magnetoresistive properties and low-temperature heat capacity of polycrystalline $BaPb_{0.75}Bi_{0.25}O_3$ in a wide range of magnetic fields.

Interest in the magnetoresistive properties of $BaPb_{1-x}Bi_xO_3$ also stems from the fact that the magnetoresistance of this material exhibits field hysteresis which could not be explained in earlier studies [6, 7]. The Josephson properties of grain boundaries in polycrystalline $BaPb_{0.75}Bi_{0.25}O_3$ [4] render this system similar to grained HTCSs to a certain extent. This suggests that the mechanism responsible for the hysteretic behavior of magnetoresistance is similar to that in granular HTSCs. The *R*(*H*) hysteresis in the latter materials is due to the fact that their grain boundaries are in the effective field, viz., the superposition of the external field and the fields induced by the dipole magnetic moments of superconducting grains [10–12], which also exhibit magnetic hysteresis. In most

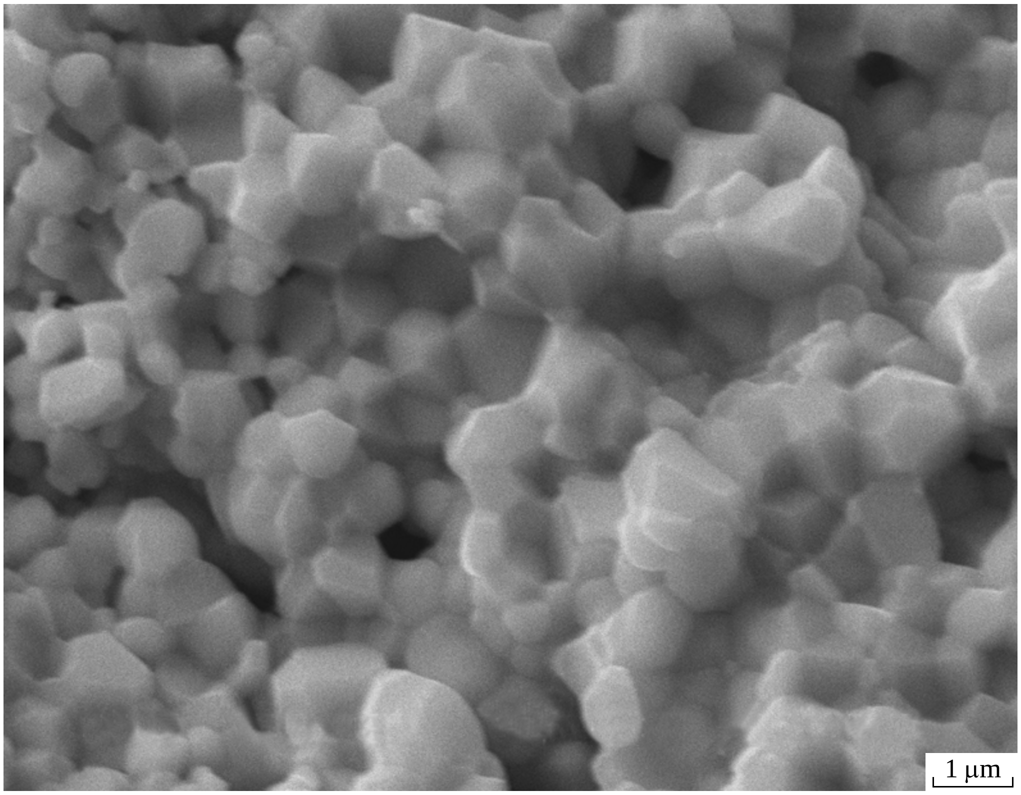

Fig. 1. Microphotograph of the surface of experimental $BaPb_{0.75}Bi_{0.25}O_3$ sample.

cases, the critical density j of transport current in granular HTSC materials is several orders of magnitude lower than the critical current density j_{CG} of superconducting grains. Therefore, the transport current cannot affect the pinning of vortices in grains, and variation in transport current leads only to a change in the value of the magnetoresistance itself (i.e., dissipation in grain boundaries), while the field width of the $R(H)$ hysteresis loop is determined only by the magnetic state of superconducting grains and is independent of transport current. Comparison of the hysteretic behavior of $R(H)$ in $BaPb_{0.75}Bi_{0.25}O_3$ and grained HTSCs is of special interest. In this respect, the effect of transport current on the hysteretic behavior of $R(H)$ of granular $BaPb_{0.75}Bi_{0.25}O_3$ is a logical continuation of [10, 12].

2. EXPERIMENT

Polycrystalline $BaPb_{0.75}Bi_{0.25}O_3$ was obtained by solid-phase synthesis from BaO_2, PbO, and Bi_2O_3. We decided not to use $BaCO_3$ since it decomposes to BaO at $T \geq 1400°C$. At such a high temperature, PbO is noticeably volatile, which makes Ba–Pb–Bi–O synthesis problematic. The use of barium peroxide makes it possible to avoid this complication and to carry out synthesis at low temperatures, at which the pressure of PbO vapor is admissibly low. Primary annealing of thoroughly mixed and pressed oxides was performed at $T = 650°C$ for 18 h. The intermediate product was crushed, pressed, and annealed for 12 h at $T = 700°C$. X-ray structural analysis showed only reflections of the perovskite structure. Figure 1 shows the results of scanning electron microscopy (SEM) of a natural cleave of the $BaPb_{0.75}Bi_{0.25}O_3$ sample. It can clearly be seen that crystallites in this sample have a characteristic size of about 1 μm, the role of crystallite boundaries being played by the cleavage regions formed during the growth of microctystallites.

Transport properties were measured using the standard four-probe technique; the sample was cooled in zero external field. The potential and current contacts were gold-plated and pressured. The specimen parameters were 1 × 1 × 9 mm (sample no. 1 in the figures) for measurements in fields of up to 7 kOe and ~0.5 × 0.2 × 5.0 mm in fields of up to 65 kOe (sample no. 2). Since the cross-sectional area and the distance between the potential contacts for sample no. 2 were determined with a substantial error, the data on magnetoresistance $R(H) = U(H)/I$ are given either in absolute values or normalized to resistance $R(T_C)$ immediately after the transition.

In experiments on magnetoresistance $R(H) = U(H)/I$, the field was applied at right angles to the direction of transport current I. The current–voltage (I–V) characteristics (sample no. 1) were measured in

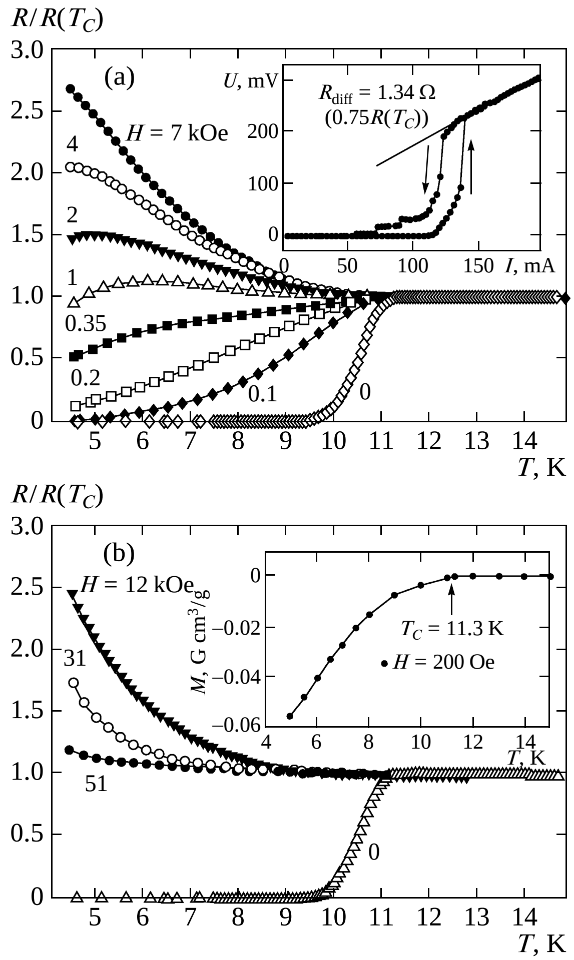

Fig. 2. $R(T)$ curves for polycrystalline $BaPb_{0.75}Bi_{0.25}O_3$ measured in various external magnetic fields for (a) $I = 1$ mA on sample no. 1 and (b) $I = 0.1$ μA on sample no. 2. The insets show (a) the I–V curves for $H = 0$, $T = 4.2$ K; arrows indicate the direction of scanning in current; (b) the $M(T)$ dependence; the value of T_C from magnetic measurements is indicated.

the stable current regime. According to the results of magnetic measurements (on a vibration sample magnetometer [13]), the transition temperature was 11.3 K (see the inset to Fig. 2b). The value of $\rho(T_C = 11.3$ K) (obtained from the data on sample no. 1) was about 0.13 Ω cm. The critical current density at $T = 4.2$ K according to the criterion $U = 1$ μV/cm was approximately 10 A/cm^2.

Heat capacity measurements were taken on a PPMS-6000 setup (Quantum Design). The heat capacity was measured by the relaxation method in the regime of increasing temperature at constant applied magnetic fields of 0, 1, 10, 20, 35, and 90 kOe. The $C(H)$ dependences were measured at $T = 2$, 4, and 6 K in the regime of increasing magnetic field. At temperatures $T = 3$, 5, and 7 K, the $C(H)$ dependences were plotted from the temperature dependences of heat capacity.

3. RESULTS AND DISCUSSION

3.1. Nonmonotonic Behavior of R(H). Effect of Transport Current

Figure 2 shows the $R(T)/R(T_C = 11.3$ K) dependences measured in various external magnetic fields. It can be seen that in addition to conventional broadening of the transition in magnetic fields of up to $H \approx 1$ kOe, a quasi-semiconducting increase in the resistance at low temperature takes place in fields of $H \approx 2$–30 kOe. However, at $H = 31$ kOe, the increase in resistance is less pronounced than in lower fields, and at $H \approx 50$ kOe, the $R(T)$ curve preserves the behavior almost completely independent of temperature, which is observed above T_C.

The I–V curves for the sample at $T = 4.2$ K in zero external field exhibit a hysteresis for relatively low values of transport current (see the inset to Fig. 2a). During I–V measurements, the sample was in liquid helium. In the current range above the voltage jump, the forward and backward $U(I)$ curves coincide, and the shape of I–V curves is preserved for various rates of current variation. These facts lead to the conclusion that the observed hysteresis is not associated with self-heating of the sample. Differential resistance R_{diff} in the range of currents and voltages above the jump in U amounts to approximately 0.75 of the resistance above T_C, which correlates with the magnetoresistance value in a field of about 50 kOe (Fig. 2b). An analogous hysteresis on the I–V curve was observed in $BaPb_{0.75}Bi_{0.25}O_3$ [3–5] as well as in $Ba_{0.6}K_{0.4}BiO_3$ [8]. It is assumed [4] that such a hysteretic behavior reflects the multiple Josephson structure existing in grained superconductors.

The $R(H)$ dependences measured in the range of fields up to several kilooersteds for various values of transport current I are also typical of a network of Josephson junctions with quasi-tunnel conduction. First, magnetoresistance R in a constant field is a nonmonotonic function of transport current. This can be seen from Fig. 3 representing the $R(H) = U(H)/I$ dependences at $T = 4.2$ K, which were measured in fields up to $H = 5$ kOe in increasing field. In weak external fields ($H \lesssim 300$ Oe), an increase in R with current is observed. For $H > 300$ Oe, quantity R has a peak at a certain current. Such a behavior is manifested on the I–V curves measured in external magnetic fields (see the inset to Fig. 3). The nonmonotonic variation of "effective resistance" $R(I) = U(I)/I$ with increasing transport current in a constant external field can be due to percolation of current through Josephson interlayers possessing semiconducting properties [14]. This correlates with the quasi-semiconducting behavior of $R(T)$ in fields of 1–30 kOe (see

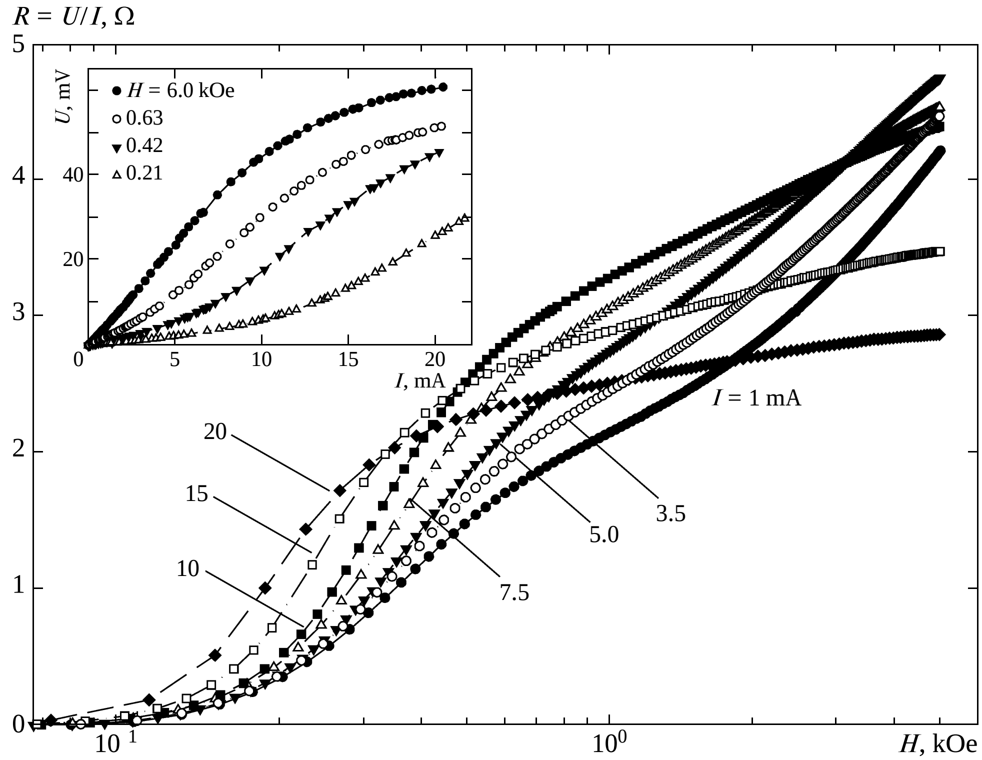

Fig. 3. Dependences $R(H) = U(H)/I$ (sample no. 1) measured for different values of transport current in an increasing external field. The inset shows the I–V curves for this sample in various external fields at $T = 4.2$ K.

Fig. 2).[1] Second, the fastest increase in the magnetoresistance occurs in weak fields ($H \lesssim 1$ kOe). This is typical of granular HTSC systems.

Thus, the form of the $R(T)$, $R(H)$, and $R(I)$ dependences indicates that the magnetoresistance in the range of weak and moderate fields ($H \lesssim 10$ kOe) is due to dissipation in "nonsuperconducting" interlayers.

The data in Fig. 2 show that at $T =$ const, dependence $R(H) = U(H)/I$ is a nonmonotonic function of the external field. Figure 4 shows the $R(H)$ curves measured for different values of transport current in the range of fields up to 65 kOe at $T = 4.2$ K. It can be seen that for small transport currents (1–100 μA), the sample resistance in the range of intermediate fields ($H \approx 10$ kOe) exceeds the sample resistance in the normal state. In addition, the effect of transport current on the field H_{max} at which the resistance peak is observed is substantial (see Fig. 4). Since dissipation in this range of magnetic fields is associated with processes in Josephson junctions, the resistance of these junctions at low temperatures in an external field considerably exceeds their resistance above T_C.

It was observed in [16] that the resistance of $BaPb_{1-x}Bi_xO_3$ bicrystals with $x = 0.27 \pm 0.03$ at $T =$ 4.2 K is an order of magnitude higher than the resistance above T_C. Such a behavior was explained by one-particle tunneling through grain boundaries.

We believe that an alternative approach to interpreting the results is possible on account of our results on $R(T)$ and $R(H)$, as well as on the magnetoresistance hysteresis and low-temperature heat capacity.

The nonmonotonic behavior of the magnetoresistance and the closeness of the values of $R(H)$ and R_{diff} during degradation of superconductivity by a strong magnetic field and transport current, respectively, can be interpreted using the approach based on the model of the superconductor–insulator spatially inhomogeneous state [9], which was used for the first time in [8]. In systems like Ba–Pb–Bi–O, Ba–K–Bi–O, and Nd–Ce–Cu–O, which can be regarded as strongly degenerate semiconductors [8], the Fermi level lies near the band gap; this may lead to phase separation into dielectric and superconducting regions during the emergence of superconductivity [9]. The dielectric region may play the role of Josephson interlayers between superconducting "islands." In these dielectric regions, the band gap is broader and the number of free charge carriers is smaller. We can logically assume that dielectric interlayers are predominantly formed in the region of grain boundaries, where the deviation from oxygen stoichiometry or local variation in the bismuth concentration may take place [16], although phase separation is also observed in $Ba_{0.6}K_{0.4}BiO_3$ sin-

[1] Analogous behavior of the I–V and $R(T)$ curves was observed in Y–Ba–Cu–O + $Cu_{1-x}Li_xO$ HTSC composites [15], in which Josephson interlayers between HTSC grains are formed by the $Cu_{1-x}Li_xO$ semiconducting ingredient.

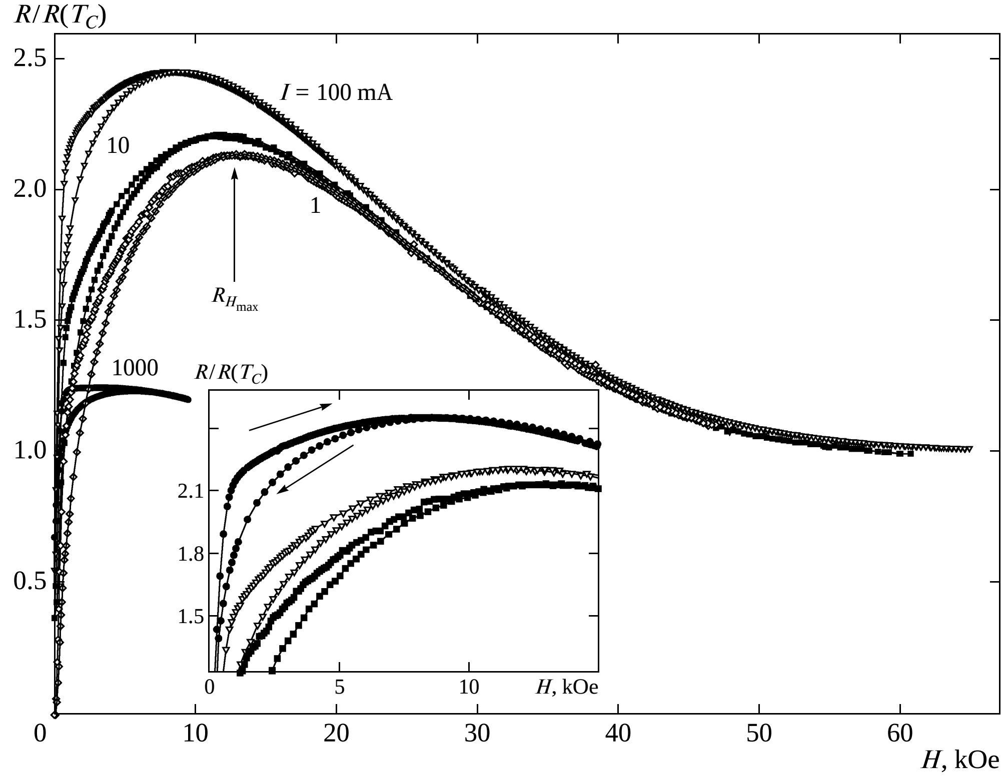

Fig. 4. Dependences $R(H)$ (sample no. 2) measured for different values of transport current I at $T = 4.2$ K. The inset shows the $R(H)$ hysteresis (the direction of field variation is indicated by arrows) on enlarged scale.

gle crystals [17]. In a magnetic field, weak superconductivity in Josephson bonds is the first to pass to the resistive state; as a result, the magnetoresistance becomes higher than in the normal state due to the dielectric nature of Josephson bonds. Degradation of superconductivity by the magnetic field, transport current, or temperature simultaneously leads to disappearance of dielectric regions. This is manifested in the nonmonotonic behavior of magnetoresistance at $T =$ const [9], which was observed for $BaPb_{1-x}Bi_xO_3$ [6, 7], $Ba_{0.6}K_{0.4}BiO_3$ [8, 17], and $Nd_{1-x}Ce_xCuO_4$ systems [18]. The existence of a spatially inhomogeneous state in $Ba_{0.6}K_{0.4}BiO_3$ was confirmed in magnetooptical experiments [19, 20].

It can be seen from Fig. 4 that an increase in transport current also shifts the field $H_{R\max}$ at which the magnetoresistance peak is observed towards lower values. In accordance with the model [9], the $R(H)$ dependence reflects two competing mechanisms: (i) an increase in the magnetoresistance under the action of the field, which is associated with passage of Josephson junctions to the normal state, and a possible additional contribution due to dissipation in superconducting regions; and (ii) a decrease in the "normal" resistance of Josephson junctions in accordance with the model scenario [9]. The first mechanism obviously dominates in the range of fields lower than $H_{R\max}$, while in fields exceeding $H_{R\max}$, the resistance decreases in increasing fields in accordance with the second mechanism. The decrease in the value of $H_{R\max}$ upon an increase in the measuring current indicates that the transport current together with the magnetic field leads to degradation of superconductivity in superconducting regions, and the second mechanism prevails in the range of lower fields.

3.2. Mechanism of R(H) Hysteresis

Apart from the nonmonotonic behavior, $R(H)$ dependences exhibit hysteresis (see insets to Fig. 4). In Fig. 5, the $R(H)$ hysteresis is shown in detail for the range of intermediate fields for various values of transport current (for sample no. 1). For all measurements represented in the figure the maximal applied field was $H = 5$ kOe. The magnetoresistance in increasing field is higher than the value of R recorded in decreasing field H: $R(H_\uparrow) > R(H_\downarrow)$ (subscripts $\uparrow$ and $\downarrow$ correspond to increasing and decreasing fields, respectively). In this respect, the hysteresis of the $R(H)$ curves for $BaPb_{0.75}Bi_{0.25}O_3$ is similar to that observed in grained HTSCs [10–12].

We believe that the approach used for explaining the hysterestic dependences for grained HTSCs [10, 12] can also be applied for analyzing the $R(H)$ dependences of granular $BaPb_{0.75}Bi_{0.25}O_3$. The magnetic-field dependence of the magnetic moment of

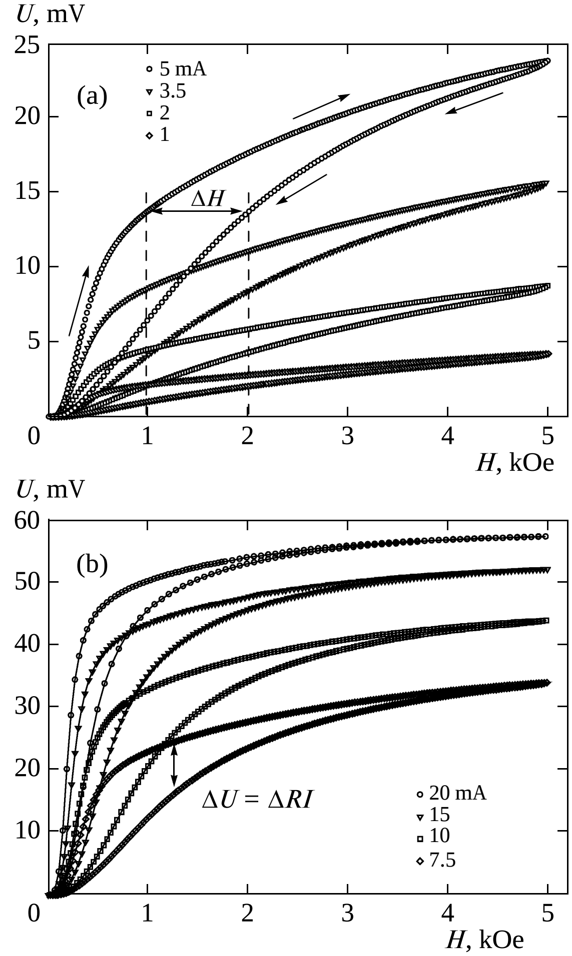

Fig. 5. $U(H)$ dependences (sample no. 1) for different values of transport current I at $T = 4.2$ K. The maximal preset field is 5 kOe in all measurements. Arrows in (a) indicate the direction of variation of the external field; parameter $\Delta H = R(H_\downarrow) - R(H_\uparrow)$ is determined by way of example for $H_\downarrow = 2$ kOe. (b) Example of determining the difference $\Delta R = \Delta U/I = R(H_\uparrow) - R(H_\downarrow)$.

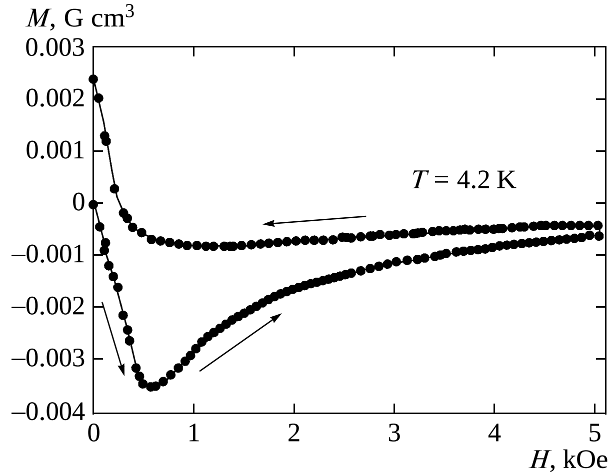

Fig. 6. $M(H)$ dependences at $T = 4.2$ K (sample mass is 18.5 mg). The maximal applied field is 5 kOe. Arrows indicate the direction of variation of the external field.

$BaPb_{0.75}Bi_{0.25}O_3$ exhibits a hysteresis typical of type II superconductors (Fig. 6). The diamagnetic signal is produced by superconducting regions. The magnetic induction lines from dipole moments must be closed by dielectric interlayers (a similar pattern is observed for granular HTSCs [10]). It is logical to assume that dissipation during the passage of transport current through $BaPb_{0.75}Bi_{0.25}O_3$ in a magnetic field mainly occurs in weak links (dielectric interlayers through which Josephson tunneling takes place). We can speak of the effective field $\mathbf{B}_{eff} = \mathbf{H} + \mathbf{B}_{ind}$ in Josephson interlayers; the field B_{ind} induced by the magnetic moments of superconducting regions is proportional to magnetic moment M of the superconductor, and vector $\mathbf{B}_{ind}$ is parallel to $\mathbf{H}$ if $M < 0$ [10]. The $M(H)$ hysteresis associated with pinning of Abrikosov vortices in superconducting regions leads to different values of $\mathbf{B}_{ind}$ (and, hence $\mathbf{B}_{eff}$) in increasing and decreasing external fields in a wide range of the fields (at least, for large values of field H, for which $M(H_\downarrow) = 0$) for $H_\uparrow = H_\downarrow$, $B_{eff}(H_\uparrow) > B_{eff}(H_\downarrow)$. Since dissipation is determined by the value of B_{eff}, $R \sim B_{eff}$, the magnetoresistance exhibits a hysteresis.

The maximal difference $\Delta M = |M(H_\uparrow) - M(H_\downarrow)|$ must generally coincide with the maximal value of $\Delta R = R(H_\uparrow) - R(H_\downarrow)$ for $H_\uparrow = H_\downarrow$. Such a correlation is indeed observed. Figure 7 shows the value of ΔM as a function of H. The $\Delta R(H)$ dependences depicted in Fig. 5 (for $I = 1$ and 10 mA) also demonstrate peaks near $H \sim 0.5$ kOe. However, apart from B_{eff}, the transport current also strongly affects the $R(H)$ dependences (see Fig. 3). For this reason, we cannot expect complete coincidence of the peaks on the $\Delta R(H)$ and $\Delta M(H)$ curves. In the range of weak fields, the $\Delta M(H)$ curve has another peak; however, the $R(H)$ curves have no other singularities. This does not contradict the proposed model of the magnetoresistance hysteresis because R is proportional to the vector sum of $\mathbf{B}_{ind}$ and $\mathbf{H}$.

The presence of a hysteresis on the $R(H)$ curves in the range of fields lower than $H_{R\max}$ implies in the given approach that effective field B_{eff} in grain boundaries has different values for $H_\uparrow$ and $H_\downarrow$ (for $H_\uparrow = H_\downarrow$), and this field affects tunneling of carriers. Since the effect of the magnetic field on the contact resistance in the one-particle tunneling mode is unlikely, we can state that it is Cooper pairs that experience tunneling, while one-particle tunneling does not make a predominant contribution to dissipation.

In the approach explaining the $R(H)$ hysteresis [10], in the external field in which the hysteresis of magnetization disappears, the hysteresis on the $R(H)$ curve also disappears simultaneously. For $BaPb_{0.75}Bi_{0.25}O_3$ at $T = 4.2$ K, the irreversibility field is about 27 ± 3 kOe according to our measurements, and this value coincides with the result obtained in [21] for

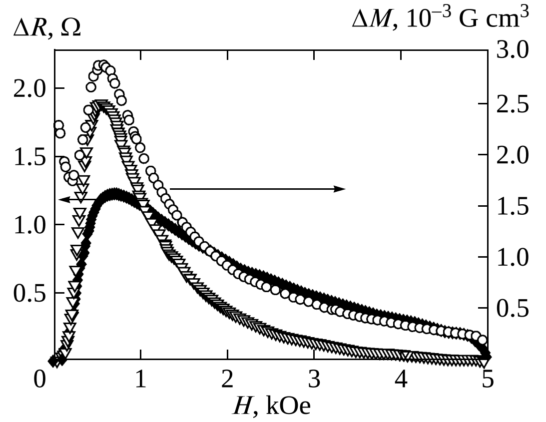

Fig. 7. Dependences $\Delta R(H) = R(H\uparrow) - R(H\downarrow)$ obtained from the results presented in Fig. 5. ($I = 1$ mA (◆), 10 mA (▽)), and $\Delta M(H) = |M(H\uparrow) - M(H\downarrow)|$ obtained from the data in Fig. 6 (○).

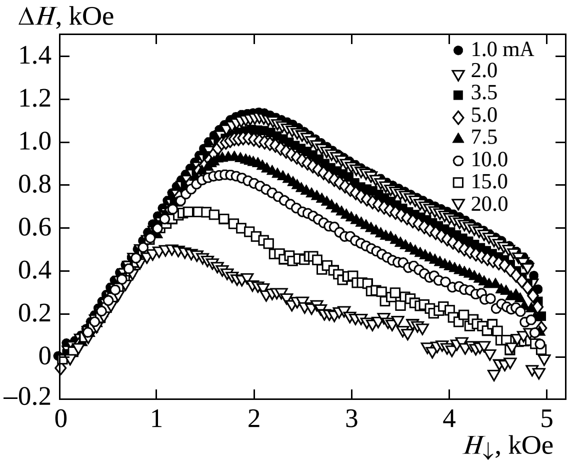

Fig. 8. Dependences of the field width $\Delta H = H_\downarrow - H_\uparrow$ of the magnetoresistance hysteresis loop on $H_\downarrow$ obtained from the data in Fig. 5 for various values of transport current.

the same granular system. However, in accordance with the results presented in Fig. 4 (inset), the "irreversibility" field H^* for magnetoresistive measurements is much smaller (12.0 ± 0.5 kOe for $I = 1$ mA, 10.0 ± 0.5 kOe for $I = 10$ mA, 8.5 ± 0.5 kOe for $I = 0.1$ mA, and 7.0 ± 0.5 kOe for $I = 1$ mA). In addition, an increase in the transport current leads to a decrease in the value of this field. For most of the $R(H)$ dependences shown in Fig. 4 (except that for $I = 1$ mA), the field H^* corresponding to irreversible behavior of magnetoresistance coincides with $H_{R\max}$ to within the error of measurements.

In our earlier works [10, 12], we introduced the field width $\Delta H = H_\downarrow - H_\uparrow$ of magnetoresistance for $R(H_\uparrow) = R(H_\downarrow)$ (here, $H_\uparrow \neq H_\downarrow$) as a parameter for analyzing the hysteretic dependences $R(H)$. The condition $R(H_\uparrow) = R(H_\downarrow)$ for the $R(H)$ dependence measured for a certain current indicates that the effective fields at points $H_\uparrow$ and $H_\downarrow$ are identical ($B_{\rm eff}(H_\uparrow) = B_{\rm eff}(H_\downarrow)$). Consequently, the value of ΔH is proportional to the difference in the values of the magnetic moment at points $H_\uparrow$ and $H_\downarrow$ [10, 12]. In magnetoresistance measurements on granular HTSCs, the critical intragrain current j_{CG} is usually several orders of magnitude larger than the transport current that can be used in experiments. Therefore, the transport current does not affect vortex pinning in HTSC grains and, hence, the diamagnetic response of grains either. Consequently, the field width of the magnetoresistance hysteresis loop $\Delta H = R(H_\downarrow) - R(H_\uparrow)$ does not depend on current (at least, in classical YBCO, BiCaSrCuO, and LaSrCuO compounds) [12].

The $BaPb_{0.75}Bi_{0.25}O_3$ system under investigation exhibits a different behavior. In field H^* for which the forward and backward $R(H)$ curves coincide, the field width of the hysteresis loop is $\Delta H = 0$, and the value of H^* depends on the transport current (see Fig. 4). The $\Delta H(H)$ dependence for various values of I in fields lower than H^* additionally confirms the effect of current on the magnetoresistance hysteresis. Figure 8 shows the $\Delta H(H_\downarrow)$ dependences for the values of $R(H)$ from Fig. 5. An example of determining the field width of the $R(H)$ hysteresis loop for $H_\downarrow = 2$ kOe is given in Fig. 5a. It can be seen that the transport current changes the value of ΔH in practically the entire range of experimental fields. Apart from its effect on the value of $R = U/I$ (see Fig. 3), an increase in current I considerably reduces the width of the magnetoresistance hysteresis loop in the range of fields smaller than $H_{R\max}$.

We interpret such a behavior as the effect of transport current on the magnetic moment of superconducting domains. This can be due to additional depinning of vortices in superconducting regions and percolation of transport current under the action of the Lorentz force. Indeed, the I–V curves recorded in zero field at $T = 4.2$ K (see Fig. 2a) show that critical current I_C (according to criterion of 1 μV/cm) amounts to ≈110 mA, while for currents exceeding ≈140 mA, the I–V curve acquires a linear segment. Consequently, we can assume that this current (140 mA, ≈13 A/cm^2) coincides in order of magnitude with j_{CG} (in this case, $j_{CG} \sim 140$ mA s, where s is the cross-sectional area of "superconducting regions"). In the $R(H)$ measurements (see Fig. 5), the transport current is of the same order of magnitude as j_{CG}.

3.3. Low-Temperature Heat Capacity in Magnetic Fields

Figure 9 shows the dependence of C/T on T^2, which can be used to obtain an approximate value of the electron heat capacity from the linear segment of this dependence. For type II superconductors, we can easily obtain the linear coefficient γ for determining the exact value of the electron component of heat capacity from the relation $C_{\rm el} = \gamma T$. However, since we know that the

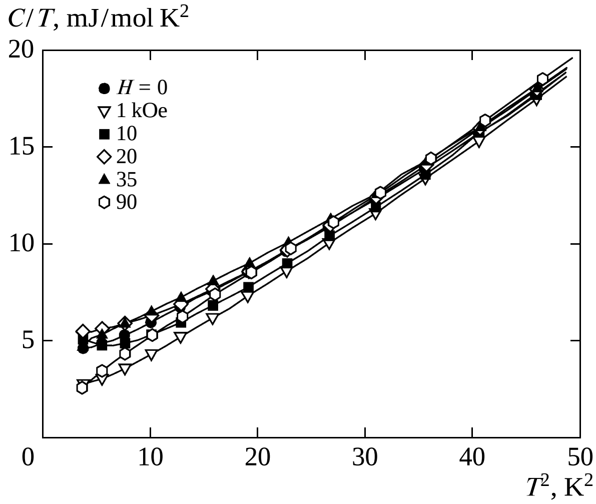

Fig. 9. Dependences of C/T on T^2 in various magnetic fields.

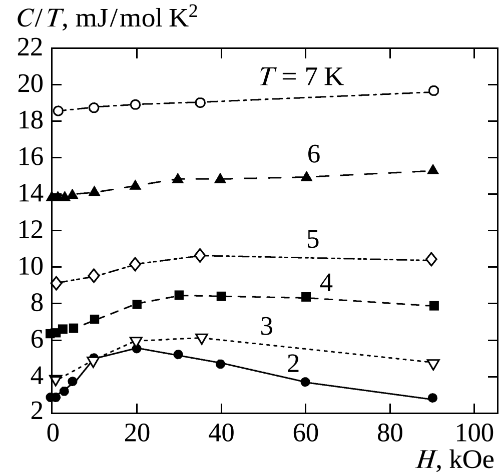

Fig. 10. Dependences of C/T on H at various temperatures. For $T = 2$, 4, and 6 K, the $C(H)$ dependences are measured directly in increasing magnetic field. For $T = 3$, 5, and 7 K, the $C(H)$ dependences are plotted from the results represented in Fig. 9.

electron component of heat capacity of type II superconductors can be described by this relation [22–25], we can estimate qualitatively the temperature dependence of the electron heat capacity. The dependence of C/T on T^2 at low temperatures exhibits a deviation towards increasing heat capacity upon cooling, which is typical of type II superconductors [22–25].

Figure 10 shows the dependence of C/T on H at various temperatures. For $T = 2$, 4, and 6 K, the $C(H)$ dependences were measured in increasing magnetic field. At temperatures $T = 3$, 5, and 7 K, the $C(H)$ dependences were plotted from the temperature dependences of heat capacity. It can be seen from the figure that both the data constructed from dependences $C(T)$ and the directly measured data (dependences $C(H)$) demonstrate identical behavior.

It can be seen that at low temperatures, the value of C/T first increases in a magnetic field, attains a certain maximal value, and then decreases. With increasing temperature, this effect is gradually suppressed and the dependence exhibits a slight increase in the value of C/T in an increasing magnetic field. Since the electron heat capacity depends on the density of states on the Fermi surface (i.e., the charge carrier concentration per unit volume [26]),

$$C_{\rm el} = \frac{1}{3}\pi^2 k_{\rm B}^2 \eta(\varepsilon_{\rm F})T = \frac{k_{\rm B}^2 m}{\hbar^2}\left(\frac{V^2\pi^2 N}{9}\right)^{1/3} T,$$

were N is the total number of charge carriers in the system, $\eta(\varepsilon_{\rm F})$ is the density of states at the Fermi level, m is the mass of the charge, and V is the sample volume, we can state that at low temperatures (2–4 K), the charge carrier concentration as a function of the applied field first increases and then decreases, while at higher temperatures (5–7 K), the concentration of the carried charge slowly increases with the applied magnetic field.

Such a behavior can be interpreted as follows. At low temperatures, the sample in a weak magnetic field is in the state of phase separation [9] (see above); i.e., the sample contains local superconducting domains in which the charge carrier concentration is high, and dielectric regions in which electrons are localized [9]. When the magnetic field is increased to a certain value (of about 20 kOe), the superconducting phase is preserved, while dielectric regions pass to the semimetal state (see Fig. 2). Therefore, the concentration of free electrons (i.e., charge carriers) also increases, which affects the electron component of heat capacity. Upon a further increase in the magnetic field, the superconducting state also degrades and the sample also acquires the properties of a semimetal; consequently, the concentration of charge carriers decreases. At higher temperatures (5–7 K), the nonmonotonic behavior of C/T as a function of H is not observed, although the value of C/T rapidly increases with H in the range of fields up to 20 kOe. Obviously, phase separation in this temperature range is manifested less strongly than at lower temperatures, and a slight increase in the magnetic field transforms the sample to the normal state. Thus, in this temperature range, a gradual transition from the semiconducting to semimetallic state takes place, which leads to a slight increase in the charge carrier concentration and, accordingly, to a slight increase in the electron heat capacity (Fig. 10).

4. CONCLUSIONS

Summarizing our experimental results on the magnetoresistive effect in polycrystalline $BaPb_{0.75}Bi_{0.25}O_3$, we can conclude that at temperatures below the superconducting transition temperature, the magnetoresistance exhibits a nonmonotonic behavior. An increase in the transport current density shifts the peak on the $R(H)$ curve towards lower fields.

Admitting the possible interpretation of our results taking into account tunneling of charge carriers through the Josephson network in a $Ba(Pb,Bi)O_3$ polycrystal, we can show that the observed behavior fits the model of the superconductor–insulator spatially inhomogeneous state [9], according to which a system like $BaPb_{0.75}Bi_{0.25}O_3$ can exhibit phase separation into superconducting and dielectric regions. In comparatively weak fields ($H \lesssim 10$ kOe), dissipation in the magnetic field as a result of passage of the transport current takes place in dielectric regions in which weak Josephson bonds are formed. After the degradation of superconductivity in superconducting regions by the external magnetic field and transport current, phase separation disappears also; i.e., the transparency of dielectric regions for charge carriers increases.

In this range of external fields ($H \lesssim 10$ kOe), the magnetoresistance is of the hysteretic type, the hysteresis mechanism being analogous to the mechanism of giant hysteresis of $R(H)$ in granular HTSCs. It includes the effect of the magnetic moments of superconducting regions on the effective field in Josephson interlayers. The difference from HTSCs is that the transport current in $BaPb_{0.75}Bi_{0.25}O_3$ can additionally affect depinning of Abrikosov vortices due to low values of the critical current density in superconducting regions. This leads to the dependence of the field width of the magnetoresistance hysteresis loop on the transport current and a decrease in the field value above which the $R(H)$ hysteresis disappears upon an increase in the transport current.

The experimental data on heat capacity also confirm the phase separation model [9] as applied to the system under investigation. In the range of low temperatures (2–4 K), a nonmonotonic dependence of C/T on the magnetic field is observed. Since the value of C/T is proportional to the density of states at the Fermi level, such a behavior illustrates an increase in the free electron concentration in dielectric regions in the field range 10–30 kOe, in which phase separation becomes less pronounced in view of partial degradation of superconductivity in the superconducting regions by the field and a decrease in the free electron concentration in strong fields (close to H_{C2}).[2]

[2] According to the results of different magnetic and magnetoresistive measurements [4, 20, 27, 28], the value of H_{C2} for $BaPb_{0.75}Bi_{0.25}O_3$ amounts to about 50 kOe at $T = 4.2$ K, which is in conformity with the results depicted in Fig. 4 (the resistance becomes close to $R(T_C)$ in field $H \approx 60$ kOe).

ACKNOWLEDGMENTS

The authors thank V.V. Val'kov, V.I. Zinenko, and A.D. Balaev for fruitful discussions.

This study was performed under the program no. 5 of the Russian Academy of Sciences (project no. 7).

REFERENCES

1. A. W. Sleight, J. L. Gillson, and P. E. Bierstedt, Solid State Commun. **17**, 27 (1975).
2. T. H. Lin, X. Y. Shao, M. K. Wu, P. H. Hor, X. C. Jin, and C. W. Chu, Phys. Rev. B: Condens. Matter **29**, 1493 (1984).
3. V. M. Svistunov, Yu. F. Revenko, D. P. Moiseev, V. M. Postrikov, and A. P. Gaevskis, Fiz. Nizk. Temp. (Kharkov) **11** (11), 1133 (1985) [Sov. J. Low Temp. Phys. **11** (11), 623 (1985)].
4. A. M. Gabovich and D. P. Moiseev, Usp. Fiz. Nauk **150** (4), 599 (1986) [Sov. Phys.—Usp. **29** (12), 1135 (1986)].
5. M. Bel'ski, O. Vendik, A. P. Gaevskis, A. B. Kozyrev, V. P. Morozik, and T. B. Samoilova, Zh. Tekh. Fiz. **56** (2), 389 (1986) [Sov. Tech. Phys. **31** (2), 235 (1986)].
6. E. A. Protasov, S. V. Zaĭtsev-Zotov, Yu. N. Venevtsev, and V. V. Bogatko, Fiz. Tverd. Tela (Leningrad) **20** (11), 3503 (1978) [Sov. Phys. Solid State **20** (11), 2028 (1978)].
7. S. V. Zaĭtsev-Zotov and E. A. Protasov, Fiz. Tverd. Tela (Leningrad) **26** (5), 1374 (1984) [Sov. Phys. Solid State **26** (5), 834 (1984)].
8. N. V. Anshukova, V. B. Ginodman, A. I. Golovashkin, L. N. Zherikhina, L. I. Ivanova, A. P. Rusakov, and A. M. Tskhovrebov, Zh. Éksp. Teor. Fiz. **97** (5), 1635 (1990) [Sov. Phys. JETP **70** (5), 923 (1990)].
9. A. A. Gorbatsevich, Yu. V. Kopaev, and I. V. Tokatly, Pis'ma Zh. Éksp. Teor. Fiz. **52** (2), 736 (1990) [JETP Lett. **52** (2), 95 (1990)].
10. D. A. Balaev, D. M. Gokhfel'd, A. A. Dubrovskiĭ, S. I. Popkov, K. A. Shaikhutdinov, and M. I. Petrov, Zh. Éksp. Teor. Fiz. **132** (6), 1340 (2007) [JETP **105** (6), 1174 (2007)].
11. T. V. Sukhareva and V. A. Finkel, Zh. Éksp. Teor. Fiz. **134** (5), 922 (2008) [JETP **107** (5), 787 (2008)].
12. D. A. Balaev, A. A. Dubrovskiĭ, K. A. Shaikhutdinov, S. I. Popkov, D. M. Gokhfeld, Yu. S. Gokhfeld, and M. I. Petrov, Zh. Éksp. Teor. Fiz. **135** (2), 271 (2009) [JETP **108** (2), 241 (2009)].
13. A. D. Balaev, Yu. V. Boyarshinov, M. M. Karpenko, and B. P. Khrustalev, Prib. Tekh. Éksp., No. 3, 167 (1985).
14. V. N. Alfeev, *Semiconductors, Superconductors, and Paraelectrics in Cryoelectronics* (Sovetskoe Radio, Moscow, 1979) [in Russian].
15. M. I. Petrov, D. A. Balaev, K. A. Shaikhutdinov, and K. S. Aleksandrov, Supercond. Sci. Technol. **14**, 798 (2001).
16. V. N. Stepankin, E. A. Protasov, A. V. Kuznetsov, and S. V. Zaĭtsev-Zotov, Pis'ma Zh. Éksp. Teor. Fiz. **41** (1), 23 (1985) [JETP Lett. **41** (1), 27 (1985)].
17. A. I. Golovashkin, L. N. Zherikhina, and A. M. Tskhovrebov, in *Proceedings of the International*

Conference on Fundamental Problems of High-Temperature Superconductivity (FPS-06), Zvenigorod, Moscow region, Russia, 2006 (Zvenigorod, 2006), p. 125.

18. A. I. Golovashkin, L. N. Zherikhina, and A. M. Tskhovrebov, in *Proceedings of the International Conference on Fundamental Problems of High-Temperature Superconductivity (FPS-08), Zvenigorod, Moscow region, Russia, 2008* (Zvenigorod, 2008), p. 179.
19. A. I. Golovashkin, L. N. Zherikhina, A. M. Tskhovrebov, and L. S. Uspenskaya, in *Proceedings of the International Conference on Fundamental Problems of High-Temperature Superconductivity (FPS-06), Zvenigorod, Moscow Region, Russia, 2006* (Zvenigorod, 2006), p. 127.
20. A. I. Golovashkin, L. N. Zherikhina, A. M. Tskhovrebov, and L. S. Uspenskaya, in *Proceedings of the International Conference on Fundamental Problems of High-Temperature Superconductivity (FPS-08), Zvenigorod, Moscow region, Russia, 2008* (Zvenigorod, 2008), p. 84.
21. D. N. Zheng, Z. X. Zhao, and A. M. Campbell, Phys. Rev. B: Condens. Matter **61**, 14 804 (2000).
22. S. E. Stupp and D. M. Ginsberg, Physica C (Amsterdam) **158**, 299 (1989).
23. D. M. Ginsberg, in *Physical Properties of High-Temperature Superconductors I*, Ed. by D. M. Ginsberg (World Sci., Singapore, 1989; Mir, Moscow, 1990).
24. J. W. Loram, K. A. Mirza, and P. F. Freeman, Physica C (Amsterdam) **171**, 243 (1990).
25. J. W. Loram, K. A. Mirza, J. R. Cooper, and W. Y. Liang, Phys. Rev. Lett. **71**, 1740 (1993).
26. C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1953; Nauka, Moscow, 1978).
27. T. D. Thanh, A. Koma, and S. Tanaka, Appl. Phys. **22**, 205 (1980).
28. S. V. Zaĭtsev-Zotov, E. A. Protasov, and M. N. Khlopkin, Fiz. Tverd. Tela (Leningrad) **26** (10), 2933 (1984) [Sov. Phys. Solid State **26** (10), 1772 (1984)].

Translated by N. Wadhwa