

## Research Article

# The Stratified Superconductivity in $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ Single Crystal: Direct Measurement of Energy Gap between Homo-, and Inhomogeneous States

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The existence of space inhomogeneous superconductor insulator state (SISIS) found out earlier in polycrystalline samples of high- $T_C$  system  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  ( $T_C \approx 30$  K) is confirmed on  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  single crystal. At  $T^*$  ( $T^* < T_C$ ,  $T^* \approx 17$  K) the transition from the homogeneous superconducting state into the SISIS occurs. SISIS is characterized by the appearance of two gaps on the Fermi surface, semi- and superconducting, which are modulated in space in antiphase, the electric transport between superconducting regions being carried out due to Josephson tunneling. Thus the whole sample becomes a multiple Josephson system. Nonlinear  $I$ - $V$  curves are observed on  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  single crystal at temperatures below  $T^*$ . Dependence of  $I$ - $V$  curves on temperature and magnetic field, typical to a Josephson system, was found out. Besides, a step-like peculiarity at the values of voltage of the order of one and two superconducting gaps shows up. These peculiarities are suppressed by magnetic field much earlier than critical current. The new data firstly correlate with the model of SISIS and secondly permit for the first time to determining directly the energy gap between homogeneous and stratified superconductor states.

## 1. Introduction

High-temperature superconductor (HTSC)  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  has a cubic lattice and has no copper atoms or any other magnetic ions (has been found out in 1988 [1, 2]). These features distinguish it from other HTSC compounds and do not allow one to justify its HTSC properties on the bases of layered structure or internal magnetic ions. Relative towards high- $T_C$   $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$  is the system  $\text{BaPb}_Y\text{Bi}_{1-Y}\text{O}_3$  [3] famous even before the discovery of high-temperature superconductivity [4] and in behaviour of which different anomalies [5], answering to the spatially inhomogeneous superconductivity, were observed already at the end of the seventies. In subject plan the spectrum of implemented to date researches of high- $T_C$  system  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  appears to

be rather wide. Not being exhaustive, the list displayed below illustrates this thematic latitude:

- (i) problems of synthesis, composition, and structure of high- $T_C$   $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  [6–11];
- (ii) transport, phonon, and electron-phonon effects [12–14];
- (iii) heat capacity, thermal expansion, and so forth [15–20];
- (iv) investigation of magnetic response, accompanying superconducting transition, anisotropy of magnetic properties, and irreversibility effects in remagnetization [21–24];

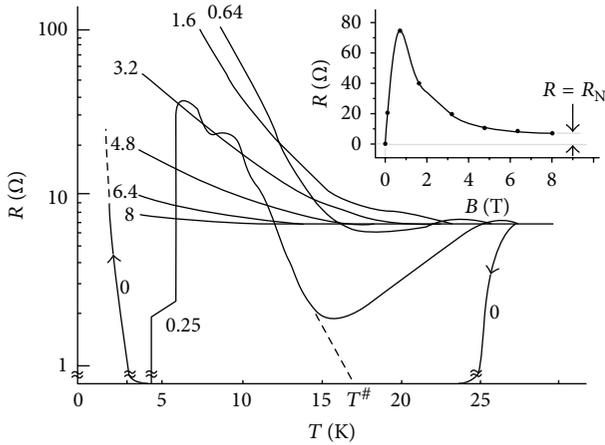


FIGURE 1: Temperature dependence of electric resistance in polycrystals of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  when  $T < T_C$  for different magnetic fields at “big” operating current (1 mA). Fields in  $T$  are put at corresponding curves. On inset: magnetoconductance of the sample at fixed temperature ( $T = 10 \text{ K}$ ,  $I = 1 \text{ mA}$ ). The unusual dependence of resistance from magnetic field (the inset to Figure 1) is that  $R$  varies with field nonmonotonically, demonstrating negative magnetoresistive effect: at first resistance increases and then decreases sharply down to  $R_N$ —the value of resistance in the normal state just above the superconducting transition. The maximum value of resistance depends on temperature and at low temperatures can exceed  $R_N$  for several orders of magnitude.

- (v) electronic structure and mechanisms of superconductivity [25–36];
- (vi) superconductivity stratification and phase transitions metal-dielectric [37–40];
- (vii) Josephson and microwave properties and nonlinear effects under the microwaves action [41–46].

We chose the  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  system for investigation, since, being a HTSC, this compound has relatively low critical temperature  $T_C$  and low critical field  $H_{C2}$ . As a result,  $H_{C2}(T)$  curves can be measured and hence phase transition can be observed over a wide temperature range. In experiment we managed to investigate different parameters, characterizing superconducting transition, in the temperature interval  $0,08T_C < T < T_C$ . Measurements of critical field in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  performed by us [47] revealed negative curvature of temperature dependence  $H = H_{C2}(T)$ , which is rather typical feature for majority of high- $T_C$  systems, but only in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  this negative curvature happened to be followed down to  $0,08T_C$ . At the same time we have found a number of unusual anomalies in behaviour of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  investigated initially in polycrystalline form.

Figures 1–4 demonstrate the observed anomalies which are

- (i) the recovery of the resistive state from the superconducting state (reentrant behavior) as the temperature decreases at  $T < T_C$  (Figure 1, curve  $B = 0 \text{ T}$ ), which is caused by the nonmonotonic temperature dependence of the critical current [47];

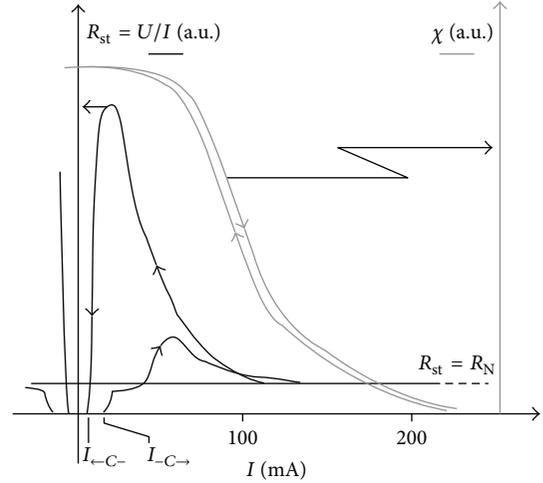


FIGURE 2: Dependence of static resistance from current, introduced in a sample,  $R_{st} = R_{st}(I)$  shows that when  $I > I_{C-}$ , the system transfers into resistive ( $R_{st} > 0$ ) state, while its magnetic susceptibility  $\chi$  remains virtually permanent relative to the homogenous superconductivity state. On return course when current decreases the nonzero resistance is retained until current value  $I_{C-}$ , which answers hysteresis character of the volt-ampere characteristic, on the basis of which the dependence  $R_{st} = R_{st}(I)$  was obtained.

- (ii) presence of superconducting phase in the volume of the sample, being in the high electric resistance state, that was caused by magnetic field or electric current  $I > I_C$  action; it is apparent from low-frequency contactless induction measurements that presence of superconducting phase keeps safe until full destruction of this high-resistance state by a strong current  $I \gg I_C$  (Figure 2, [48]);
- (iii) hysteretic  $I$ - $V$  curves (Figure 3) with nonmonotonic temperature dependence of the critical current having maximum at  $T^\# \approx 17 \text{ K}$  (Figure 3(b), upper inset [47]);
- (iv) the suppression of a critical current by microwave radiation (Figure 3(b), bottom inset [49]);
- (v) nonstationary Josephson effect in polycrystalline samples at  $T < T^\# < T_C$  (Figure 4 [50]).

Temperature dependence of resistance obtained at different magnetic fields  $R = R(T, B)$ , as it can be seen from Figure 1, agrees with field dependence of voltage-current characteristics (VCC), presented in Figure 3(a). In this case, the nonmonotone dependence of the critical current in zero field  $I_C = I_C(T, B = 0)$  (inset to Figure 3(b)) has a maximum at  $T^\# \approx 17 \text{ K}$ .

In our further investigations, performed on single crystal samples  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ , a number of anomalies have been found in the vicinity of  $T^\# \approx 17 \text{ K}$  (it should be mentioned that critical currents of single crystals turned out to be so large that observation of VCC, at least in first experiences, appeared impossible). The anomalies, observed on single crystals, are presented in Figures 5 and 6:

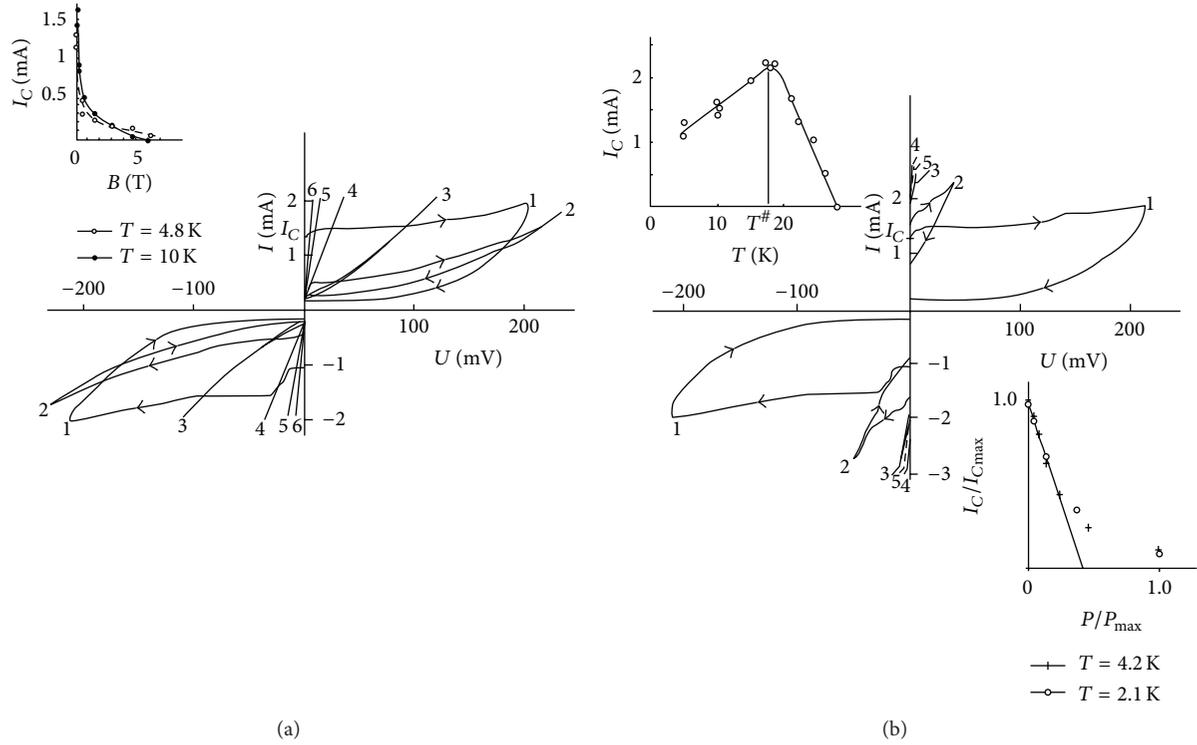


FIGURE 3: Current-voltage characteristics of ceramic sample  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  at different temperatures and magnetic fields: (a)  $T = 4.8$  K; 1;  $H = 0$  T, 2; 0.64 T, 3; 1.6 T, 4; 3.2 T, 5; 4.8 T, and 6; 6.4 T; (b)  $H = 0$  T; 1;  $T = 4.8$  K, 2; 10 K, 3; 15 K, 4; 18 K, and 5; 22 K. On inset to (a) the dependence of the critical current versus magnetic field, built from hysteretic curves on (a), is given. Top inset to (b): the nonmonotone dependence of the critical current versus temperature, with maximum at  $T^\# \approx 17$  K. Bottom inset to (b): the effect of suppression of critical current under the action of microwave radiation, which demonstrates possible presence of Josephson properties of system below  $T^\#$ .

- (i) kink of temperature dependence of residual magnetizations of single crystal near  $T^\# \approx 17$  K (Figure 5) [38];
- (ii) reentrance of resistive state of single crystal sample on “low-current” when  $T < 17$  K (Figure 6) [38];
- (iii) magneto-optic visualization of stratification effect—appearance of spatially inhomogeneous state superconductor-insulator in single crystals  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  at temperatures below 17–18 K [40].

It is necessary to note one more anomaly observed in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  single [44, 45]: the temperature dependence of the second and the third harmonics microwave emission exhibited two peaks, one just below  $T_C$  and the second around  $T = 17$  K. Since a peak of the harmonic signals generally occurs just below the transition temperature, the lower peak might be a sign of the transition from homogeneous to stratified state.

## 2. The Model of Stratified State in High- $T_C$ Superconductor $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$

The data given above can be explained in terms of the model proposed in [51–53], which implies the self-consistent coexistence of superconducting and dielectric phases in

the system under study. The dielectric phase accounts for the field-induced increase of the resistance at temperatures below  $T_C$ , and the complete suppression of superconductivity in a magnetic field is accompanied by the breaking of the coexistent dielectric phase, which results in the decrease of the resistance  $R$  (Figure 1) to its value in the normal state  $R_N$ . According to the model of [51–53], HTSC systems are considered as strongly doped semiconductors with a Fermi level that satisfies the nesting condition, which causes instability with respect to the transition into the ordered space inhomogeneous state. Such a long-range order causes a gap in an excitation spectrum at the Fermi level (correspondingly, a pseudogap in the case of a short-range order). The appearance of additional carriers above the gap upon doping decreases this gap and, hence, the chemical potential  $\mu$ . This decrease in  $\mu$  cannot be compensated by the increase in the kinetic energy that, in turn, is hindered because of the high density of states at the gap edge. As a result, the derivative of the chemical potential with respect to the number of particles turns out to be negative:  $\partial\mu/\partial n < 0$ . This nonmonotonic behavior of  $\mu(n)$  results in the instability of the state with a constant concentration ( $n = \text{const.}$ ): this state decomposes to form many coexistent regions with high (metal or superconductor) and low (dielectric) values of  $n$ . At  $T < T^\# \approx 17$  K, superconducting regions are coupled through Josephson tunneling, which is indicated by the hysteretic  $I$ - $V$  curves

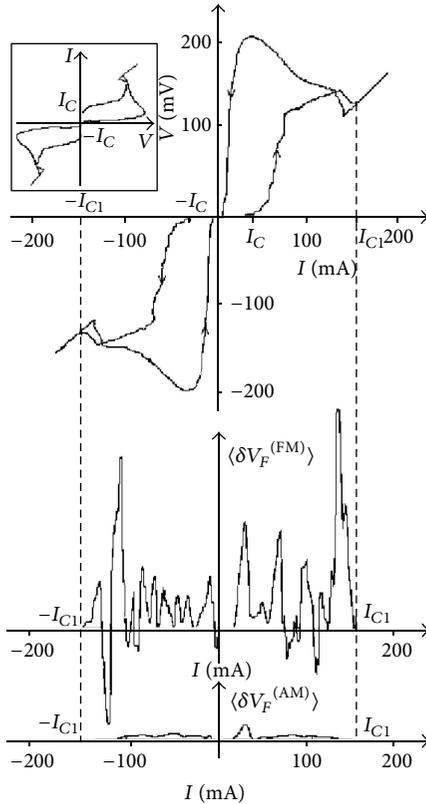


FIGURE 4: The top graph: volt-ampere characteristic—the dependence of sample voltage on operating current (on inset conventional VCC of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  sample with regular arrangement of reference axes  $I = I(V)$ ) is shown. The middle graph: averaged component of sample response  $\langle \delta V_F^{(FM)} \rangle$  to periodical variation of microwave irradiation frequency of backward-wave tube, registered on modulation frequency  $F$ . The bottom graph: averaged component of sample response  $\langle \delta V_F^{(AM)} \rangle$  on amplitude modulation with a depth reduced to deviation factor of frequency modulation (6%). The appreciable exceeding of response on FM relative to response on AM is the evidence for synchronicity of low frequency oscillations of Shapiro voltage steps, corresponding to different Josephson junctions. These junctions are formed below  $T^\# \approx 17$  K due to appearance of spatial inhomogeneity of superconducting phase distribution in the sample.

and the nonstationary Josephson effect; dielectric layers here should be rather thin. In this temperature range  $T < T^\# \approx 17$  K the critical current (the Josephson current  $I_C$ ) decreases with decreasing  $T$  due to an increase of the dielectric-layer thickness or insulator gap width. The strong exponential dependence of the Josephson critical current on the layer thickness or barrier height explains the noticeable decrease in  $I_C$  with decreasing temperature. The breaking of the weak Josephson coupling in a magnetic field causes electrical resistance in the stratified phase, and this resistance turns out to be higher than that in the normal phase, since it is specified by the tunneling resistance of dielectric regions in the stratified phase. Thus, the anomalous temperature dependence  $R = R(T)$  and  $I_C = I_C(T)$ , the hysteresis of the  $I$ - $V$  curves (Figures 1 and 3 inset), and presence of superconducting phase in resistive state (Figure 2) can be explained in terms

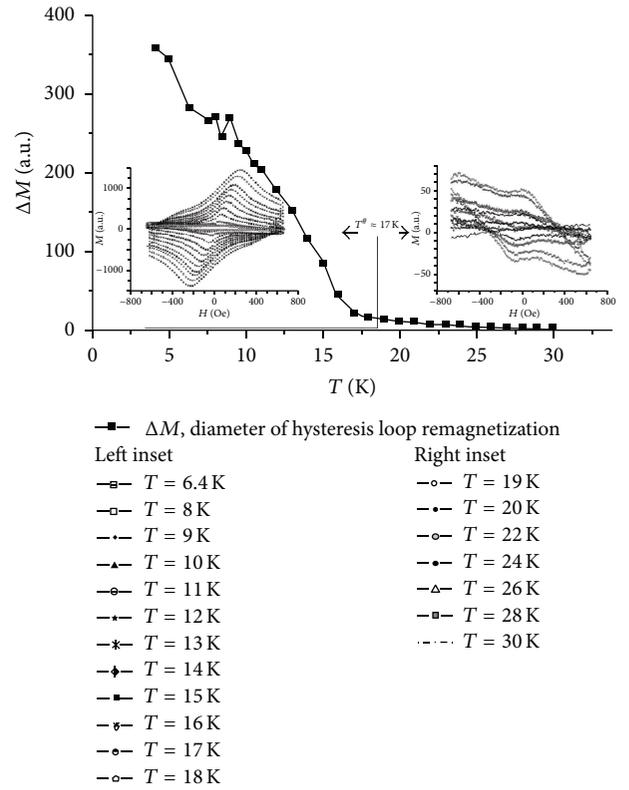
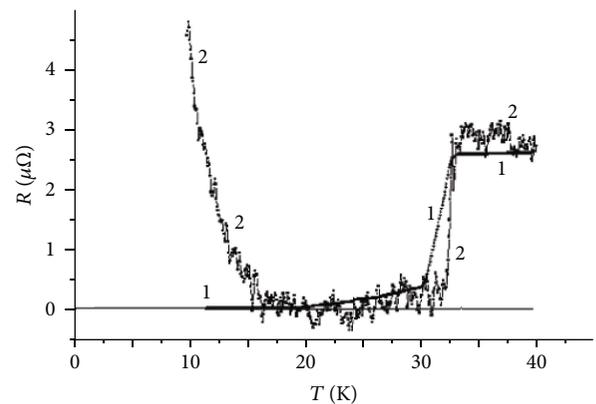


FIGURE 5: Temperature dependence of residual magnetization of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  crystal. Values of residual magnetization were obtained from hysteresis loops of sample magnetization at different temperatures, presented in the insets: low temperatures ( $T < 18$  K): left inset; high temperatures ( $T > 19$  K): right inset. Sample magnetization was measured by the Hall-effect sensor. Rapid growth of irreversibility below  $T^\# \approx 17$  K confirms the fact of appearance of spatial inhomogeneity of superconducting phase distribution in a sample, which can cause additional pinning.



Curve 1 ( $I = 100 \mu\text{A}$ )  
Curve 2 ( $I = 10 \mu\text{A}$ )

FIGURE 6: Temperature dependence of sample resistances in magnetic field 270 Oe. Curve 1 corresponds to “large” current (100 mA) through the single crystal sample  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  and curve 2 to “small” current (10 mA). Reentrance of resistive state on “small” current testifies S-like form of crystal’s VCC. On polycrystalline samples S-like VCC was also observed (the inset of Figure 4).

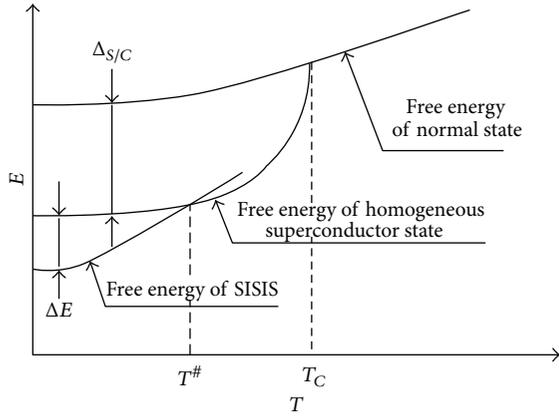


FIGURE 7: Temperature dependencies of free energies of different states in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ ;  $\Delta E$ : energy gap between homogeneous superconductor and stratified (i.e., SISIS) states;  $\Delta_{S/C}$ : superconducting gap.

of the model of a spatially inhomogeneous state insulator-superconductor (SISIS) with Josephson tunneling between superconducting regions.

The set of collected experimental data allows in the context of (SISIS) model speaking about three possible states existing in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ : normal, homogenous superconducting, and stratified (Figure 7 [54, 55]). Below  $T^\#$  superconducting and dielectric regions coexist in the sample; therefore, the stratified state has the lowest energy; the homogeneous superconducting phase is located above this state on the energy scale, and the normal state is located higher (energy diagram in Figure 7).

### 3. Samples

The single crystals of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ , grown up by the method of chemical transport reactions, had dimensions of the order of  $2 \times 2 \times 2 \text{ mm}^3$  [56].

### 4. Direct Measurement of Energy Gap between Homogeneous and Inhomogeneous States

In the course of experiments, set recently with  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  single crystals, we happened to estimate energy parameters of diagram of equilibrium phases. In one of series of these experiments a new phenomenon was observed: a distinct temperature and magnetic field dependent step-like peculiarity on voltage-current characteristics. VCC of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  single crystal obtained at different temperatures in zero magnetic field are presented in Figure 8. On low temperature curves one can see distinct step-like peculiarities. The second step on each curve happened at the voltage twice more than the voltage  $V_S$ , corresponding to the position of the first step on the curve (Figure 8). The peculiarities on the 2 and 4.2 K curves had a complex S-like form.

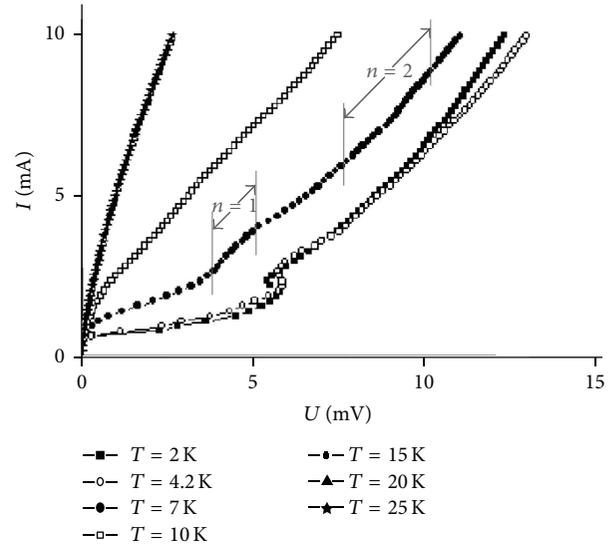


FIGURE 8: Voltage-current characteristics of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  single crystal at different temperatures. The regions analogous to the 1st and 2nd Shapiro steps are outlined on the curve for  $T = 7 \text{ K}$ .

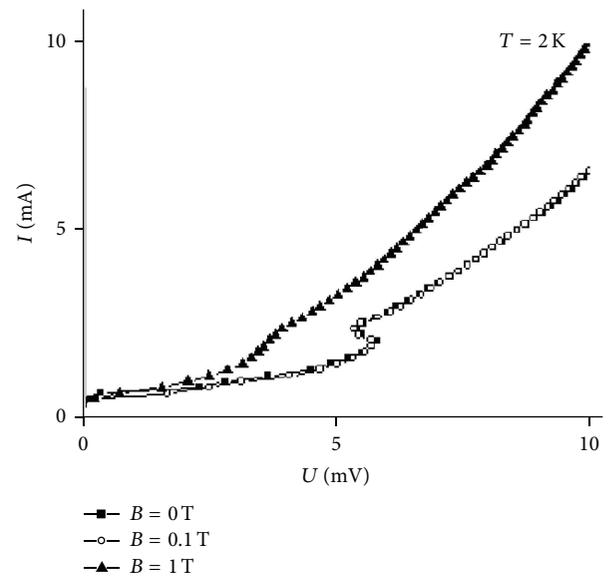


FIGURE 9: Voltage-current characteristics of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  single crystal at  $T = 2 \text{ K}$  in various magnetic fields.

The dependence of VCC on magnetic field is demonstrated in Figure 9.

The step was displaced and spread both with temperature rise and magnetic field enhancing, but influence of magnetic field on  $I = I(V)$  dependence in the magnetic field range 0–0.1 T is explained apparently by the quadratic dependence of effect, and as a result influence of field  $B = 0.1 \text{ T}$  will make just 1% from the effect of  $B = 1 \text{ T}$ .

Figure 10 demonstrates temperature dependence of the first step position  $V_S$ , determined from  $I-V$  curves, registered

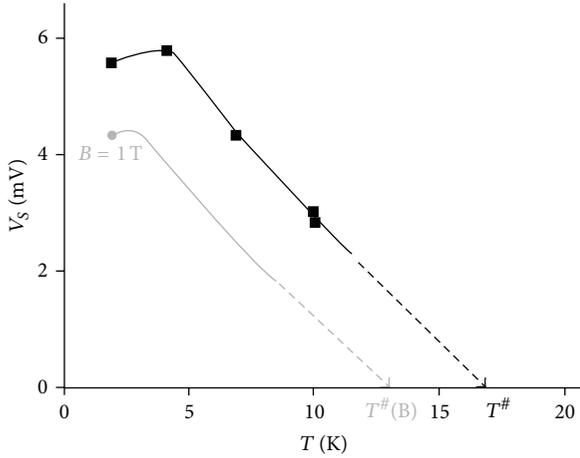


FIGURE 10: Temperature dependence of step position  $V_S$  on VCC. The black curve is built from Figure 8 data and responds to zero magnetic field. The gray curve is based on Figure 9 and responds to the field  $B = 1$  T.

at zero magnetic field (black points) and at  $B = 1$  T (grey points). Black curve is a guide line through experimental data. We drew the gray curve through gray point congruently to the black one supposing that effect of temperature in both cases in zero and nonzero magnetic fields is similar.

$V_S$  decreases with temperature rise. The extrapolation of the  $V_S(T)$  dependence to zero voltage value crosses  $T$  axes at temperature near  $T^\# \approx 17$  K (Figure 10 black curve). Comparison of Figure 7 and the black curve in Figure 10 convinced us that the step is related to the existence of stratified state in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  single crystals, since temperature dependence of step position  $V_S(T)$  and of the difference between free energies of homogeneous and stratified superconducting states  $\Delta E(T)$  is quite similar. It permits us to propose the following explanation of the step appearance. Suppose that occasionally it turns out that between potential contacts is a single Josephson junction. It may be an occasional mechanical microdefect. The voltage drop on potential contacts of the sample (displayed as  $V$  on VCC) due to comparatively high resistance of separately occurred Josephson junction in practice appears to be applied just entirely to this transition. When voltage  $V$  drops on Josephson junction, as is known [57, 58], the last one generates microwave radiation with frequency  $f = eV/\pi\hbar$  (“active” nonstationary Josephson effect). If radiation quantum coincides with energy gap between homogeneous and stratified states of superconductor  $\Delta E$  (Figure 7) the system falls within resonance conditions that should affect its VCC due to nonstationary Josephson effect under self-radiation of the junction (“passive” nonstationary Josephson effect [56, 59]). The manifestation of such [60] “active/passive” nonstationary Josephson effect explains temperature dependence of  $V_S$  (Figure 10) and disappearance of the step at temperatures near  $T^\#$ . Multiple peculiarities on VCC observed in these experiments are quiet analogous to Shapiro steps with different  $n$  (Figure 8,  $T = 7$  K) and it provides evidence in favor of registration the phenomena, related to self-action of Josephson generation (the

“active/passive” effect [59]). Noticeable S-like form of step (Figures 8 and 9,  $T = 2$  K, 4.2 K) seems to be caused by the low  $Q$  of resonator, from which Josephson radiation, affecting thereafter back on the transition, is reflected. Base frequency of this low- $Q$  resonator  $f_0 = \Delta E/(2\pi\hbar)$  corresponds to energetic gap between homogenous and inhomogeneous superconductor states in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  (Figure 7).

The amplitude and phase of reflected emission depend on tuning out of generated frequency  $f = eV/\pi\hbar$  from resonant  $f_0 = \Delta E/(2\pi\hbar)$  one. According to elementary theory of “passive” nonstationary Josephson effect the value of superconducting components of tunnel current, determining the Shapiro step height, in its turn depends on the amplitude and phase of radiation, affecting transition. As voltage drop on Josephson junction overloads “resonant value”  $V_R = \Delta E/2e$  module of superconducting components of tunnel current at first grows out of zero point due to well-known dependence on phase  $\sim \sin \theta$ . Then owing to dependence on amplitude  $\sim J_n(U_0/\Phi_0 f)$  tunnel current starts to fall gradually in concert with  $U_0$  (here  $\theta$  is phase difference between reflected and incident radiation, determined by conventional phase/frequency dependence of resonance system,  $-\pi/2 < \theta < \pi/2$ ,  $J_n$  Bessel function,  $\Phi_0 = \pi\hbar/e$  magnetic flux quantum, and  $U_0$  amplitude of variable voltage component on contact). About so with  $V$  growth from point  $V_R = \Delta E/2e$  the “top” bell-shaped peculiarity, forming the local peak, is to be plotted on the background of undisturbed VCC. Analogously, on the back course starting from value  $V_R = \Delta E/2e$  should be plotted the “bottom” bell-shaped peculiarity, forming the local minimum. It will be noticed that “bottom” peculiarity corresponds to the interval of phase values  $-\pi/2 < \theta < 0$  on phase-frequency resonance characteristic and “top” to  $0 < \theta < \pi/2$ , correspondingly. To explain the observed S-like step it is necessary to show why the “top” bell-shaped peculiarity begins actually before the “bottom” terminates. Josephson oscillator binds together with irradiated resonator either increasing its efficient inertia or raising efficient rigidity, depending on phase sine. In the first case efficient resonance frequency  $f_0 = \Delta E/(2\pi\hbar)$  is reduced and in the second increase. As a result the top peculiarity on VCC crosses with the bottom one. Eventually, the observed S-shaped step is formed (Figures 8 and 9). With temperature increasing the gap  $\Delta E$  is reduced (Figure 7), which leads to additional lowering of effective  $q$ -factor  $Q_{\text{eff}} \cong \Delta E/kT$  and “smoothing” of S-like step. If one regards Shapiro step appearance as the synchronization effect of internal oscillations of current and voltage in Josephson junction (arising when  $I > I_C$  and  $V > 0$ ) by outer oscillations source [61], then “transformation” of step into the S-shaped region in VCC is possible to interpret, to an extent, as the effect of oscillator frequency pulling by the external cavity.

Values of energy gap  $\Delta E$  between homogenous and inhomogeneous states in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ , taken from Figure 10 as  $\Delta E = eV_S$ , it is possible to compare quantitatively with the result of extrapolation of temperature dependence of sample resistance in weak field (Figure 1,  $B = 0.25$  T). Let us present the dependence  $R = R(T, B = \text{const.})$  at low fixed magnetic field in the form  $R = R_{\Delta E}(T) + r(T)$ . The presence of linear region on dependence  $R = R(T, B = 0, 25$  T) in logarithmic

scale (Figure 1) allows, in case  $R_{\Delta E} \gg r$ , representing the contribution  $R_{\Delta T}$  in the form of

$$R_{\Delta E} = R_0 e^{\Delta E(T)/kT}, \quad (1)$$

where energy gap  $\Delta E$  plays the role of barrier height, overcome by the charge carriers, involved in transport current in stratified sample. Since the gap  $\Delta E$  is simultaneously the parameter of stratified phase, its temperature dependence according to Ginzburg-Landau theory can be written as linear in  $(T^\# - T)$  expansion  $\Delta E = A(T)(T^\# - T)$ . Then linear region of the curve  $R(T, B = 0, 25 \text{ T})$ , presented in half log scale (Figure 1) may be approximately described as

$$R_{\Delta E} \approx R_0 e^{\langle A \rangle (T^\# - T)/k\langle T \rangle}. \quad (2)$$

Brackets  $\langle \rangle$  mean the averaging over temperature interval, corresponding to linear region of the curve, which for curve under consideration corresponds to  $\langle T \rangle = (1/2)(11 \text{ K} + 14 \text{ K}) = 12.5 \text{ K}$ . The slope of linear region, according to Figure 1, is

$$\frac{\partial \ln R_{\Delta E}}{\partial T} = \frac{\langle A \rangle}{k \langle T \rangle} \approx 0.42 \text{ K}^{-1}. \quad (3)$$

Using the data obtained, one can estimate the value of energy gap at  $T = 4.2 \text{ K}$  from the formula

$$\Delta E(T = 4.2 \text{ K}) = \frac{k}{e} \langle T \rangle (T^\# - 4.2 \text{ K}) \frac{\partial \ln R_{\Delta E}}{\partial T} \approx 5.8 \text{ mV}. \quad (4)$$

One should note that presented estimation  $\Delta E(T = 4.2 \text{ K}) \approx 5.8 \text{ mV}$  coincides with value, taken from Figure 10, with uncertainty less than one percent.

Returning to dependence of step-wise peculiarity on VCC (Figure 9) from magnetic field, applied to single crystal sample  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ , one can note the likeness of curves corresponding to cases of  $B = 1 \text{ T}$ ,  $T = 2 \text{ K}$  in Figure 9 and  $B = 0 \text{ T}$ ,  $T = 7 \text{ K}$  in Figure 8. This similarity gives an indication of certain equivalence of influence on VCC of magnetic field and temperature: either ways with the increase of  $B$  and with the increase of  $T$  the peculiarity is smoothed, and  $V_S$  stoops down to zero point when  $T = T^\#(B)$ . The dependence of  $T^\#$  from applied magnetic field may be approximately derived from analogy with  $T_C$ :  $T_C \sim \Delta_{S/C}(0)$ . Hence for stratified state  $T^\# \sim \Delta E(0)$ . Then using  $T^\#(B)/T^\# \approx \Delta E(0, B)/\Delta E(0)$  it is possible to supplement the dependence  $V_S = V_S(T)$  on Figure 10 by analogous approximate dependence for nonzero field ( $B = 1 \text{ T}$ , the grey curve on Figure 10). Since there is strong relation between  $V_S$  and gap width  $\Delta E = e \cdot V_S$  it is possible to transfer approximate dependence  $V_S \approx V_S(T)$  at  $B = 1 \text{ T}$  onto the energetic state diagram and in consequence obtain free energy of SISIS at  $B = 1 \text{ T}$  (Figure 11 “grey” curve) on the base of Figure 7. “Dislodged” (grey) curves, built on Figure 11 with maintenance of mutual proportions, show magnetic field induced shifts of phase transitions parameter in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  system (shift of critical temperature, corresponding to  $B = 1 \text{ T}$ ,

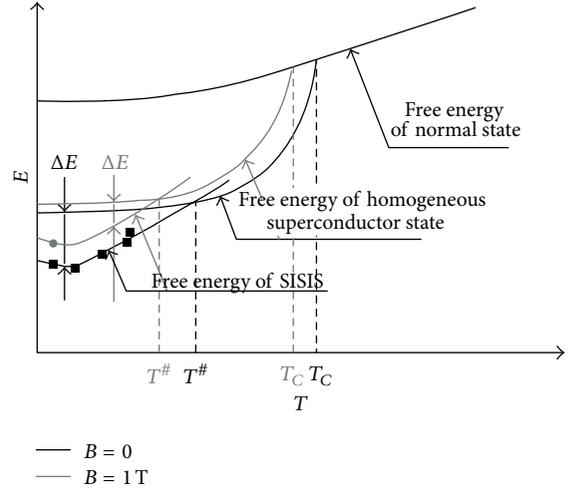


FIGURE 11: Change of temperature dependence of free energies of different phase states in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  under the action of external magnetic field.

was measured directly in the course of experiments with  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  single crystals).

Thus the new data firstly correlate with the model of SISIS and secondly permit for the first time to determine directly the energy gap between homogeneous and stratified superconductor states.

## Conflict of Interests

The authors declare that there is no conflict of interests concerning the publication of this paper.

## Acknowledgment

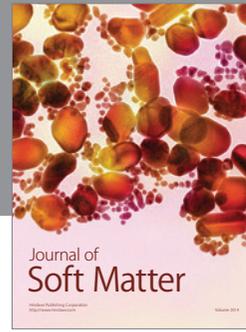
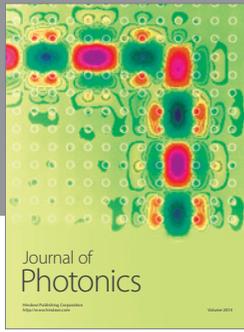
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## References

- [1] L. F. Mattheiss, E. M. Gyorgy, and D. W. Johnson Jr., “Superconductivity above 20 K in the Ba-K-Bi-O system,” *Physical Review B*, vol. 37, no. 8, pp. 3745–3746, 1988.
- [2] R. J. Cava, B. Batlogg, J. J. Krajewski et al., “Superconductivity near 30 K without copper: the  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  perovskite,” *Nature*, vol. 332, no. 6167, pp. 814–816, 1988.
- [3] A. W. Sleight, J. L. Gillson, and P. E. Bierstedt, “High-temperature superconductivity in the  $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$  systems,” *Solid State Communications*, vol. 17, no. 1, pp. 27–28, 1975.
- [4] J. G. Bednorz and K. A. Muller, “Possible high  $T_c$  superconductivity in the Ba–La–Cu–O system,” *Zeitschrift für Physik B Condensed Matter*, vol. 64, pp. 189–193, 1986.
- [5] A. M. Gabovich and D. P. Moiseev, “Metal oxide superconductor  $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ : unusual properties and new applications,” *Soviet Physics Uspekhi*, vol. 29, pp. 1135–1150, 1986.

- [6] M. L. Norton and H. Tang, "Superconductivity at 32 K in electrocrystallized barium potassium bismuth oxide," *Chemistry of Materials*, vol. 3, pp. 431–434, 1991.
- [7] L. Z. Zhao and M. L. Norton, "XPS study of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ ," *Chinese Journal of Chemistry*, vol. 11, no. 5, pp. 425–428, 1993.
- [8] J. L. Rehspringer and D. Niznansky, "Preparation of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  pure superconducting powder," *Journal of Alloys and Compounds C*, vol. 195, pp. 73–76, 1993.
- [9] C. J. Hou, H. Steinfink, L. Rabenberg, C. Hilbert, and H. Kroger, "Superconducting  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ : thin film preparation by RF magnetron sputtering," *Journal of Materials Research*, vol. 8, no. 8, pp. 1798–1804, 1993.
- [10] H.-T. Kim, K.-Y. Kang, W.-J. Kim, Y.-N. Choi, B.-J. Kim, and Y. C. Kim, "Synthesis of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  superconductors and structural analysis using neutron powder diffraction experiment," *Journal of the Korean Physical Society*, vol. 39, no. 6, pp. 1013–1018, 2001.
- [11] V. Raghuvver and A. Manthiram, "Effect of  $\text{BaBiO}_3$  and  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  additives on the rechargeability of manganese oxide cathodes in alkaline cells," *Electrochemistry Communications*, vol. 7, no. 12, pp. 1329–1332, 2005.
- [12] A. I. Golovashkin, A. V. Gudenko, L. N. Zherikhina, A. M. Tskhrovrebov, and M. L. Norton, "The electron-phonon mechanism for resistivity in the high-temperature superconductor system  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ ," *Journal of Experimental and Theoretical Physics*, vol. 79, no. 1, pp. 163–169, 1994.
- [13] M. Braden, W. Reichardt, A. S. Ivanov, and A. Y. Rumiantssev, "Anomalous dispersion of LO phonon branches in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ ," *Europhysics Letters*, vol. 34, no. 7, pp. 531–536, 1996.
- [14] D. Varshney, S. Shah, and R. K. Singh, "Superconductivity and normal state resistivity of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ : an optical phonon approach," *Superlattices and Microstructures*, vol. 24, no. 6, pp. 409–422, 1998.
- [15] J. E. Graebner, L. F. Schneemeyer, and J. K. Thomas, "Heat capacity of superconducting  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  near  $T_c$ ," *Physical Review B*, vol. 39, no. 13, pp. 9682–9684, 1989.
- [16] M. F. Hundley, J. D. Thompson, and G. H. Kwei, "Specific heat of the cubic high- $T_c$  superconductor  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ ," *Solid State Communications*, vol. 70, no. 12, pp. 1155–1158, 1989.
- [17] S. J. Collocott, N. Savvides, and E. R. Vance, "Specific heat of polycrystalline  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  from 0.5 to 20 K," *Physical Review B*, vol. 42, no. 7, pp. 4794–4796, 1990.
- [18] G. K. Panova, A. A. Shikov, B. I. Savel'ev et al., "Specific heat of the noncuprate oxide superconductor in magnetic fields," *Journal of Experimental and Theoretical Physics*, vol. 76, no. 2, p. 302, 1993.
- [19] E. B. Nyeanchi, D. F. Brewer, T. E. Hargreaves et al., "Specific heat of single crystal  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  at low temperatures," *Solid State Communications*, vol. 97, no. 3, pp. 175–178, 1996.
- [20] A. I. Golovashkin and A. P. Rusakov, "Experimental studies of the thermal and electronic properties of  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$  and other perovskite-like oxide HTSC systems," *Physics-Uspokhi*, vol. 43, pp. 184–187, 2000.
- [21] M. E. McHenry, M. P. Maley, G. H. Kwei, and J. D. Thompson, "Flux creep in a polycrystalline  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  superconductor," *Physical Review B*, vol. 39, no. 10, pp. 7339–7342, 1989.
- [22] M. Baran, H. Szymczak, R. Szymczak, S. N. Barilo, V. I. Gatal'skaya, and S. V. Shirayev, "Growth-induced anisotropy in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  superconducting single crystal," *Applied Physics A*, vol. 67, no. 4, pp. 413–416, 1998.
- [23] D. Hall, R. G. Goodrich, C. G. Grenier, P. Kumar, M. Chaparala, and M. L. Norton, "Magnetization measurements on single crystals of superconducting  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ ," *Philosophical Magazine B*, vol. 80, no. 1, pp. 61–79, 2000.
- [24] D. A. Balaev, D. M. Gokhfeld, L. A. Klinkova et al., "Increase of magnetization loop width in superconductor  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ . Possible manifesting of phase separation," *Journal of Experimental and Theoretical Physics*, vol. 118, no. 1, 2014.
- [25] M. S. Hegde, P. Barboux, C. C. Chang et al., "Electronic structure of high- $T_c$   $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  by x-ray photoelectron spectroscopy," *Physical Review B*, vol. 39, no. 7, pp. 4752–4755, 1989.
- [26] Z. Schlesinger, R. T. Collins, J. A. Calise et al., "Superconducting energy gap and a normal-state excitation in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ ," *Physical Review B*, vol. 40, no. 10, pp. 6862–6866, 1989.
- [27] W. Jin, M. H. Degani, R. K. Kalia, and P. Vashishta, "Superconductivity in  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ ," *Physical Review B*, vol. 45, no. 10, pp. 5535–5546, 1992.
- [28] R. G. Goodrich, C. Grenier, D. Hall et al., "De Haas-van Alphen measurements in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ ," *Journal of Physics and Chemistry of Solids*, vol. 54, no. 10, pp. 1251–1258, 1993.
- [29] C. Y. Lee and P. A. Deymier, "Two-electron states in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$  via discretized path integral molecular dynamics," *Solid State Communications*, vol. 102, no. 9, pp. 653–657, 1997.
- [30] V. Merregalli and S. Y. Savrasov, "Electron-phonon coupling and properties of doped  $\text{BaBiO}_3$ ," *Physical Review B*, vol. 57, no. 22, pp. 14453–14469, 1998.
- [31] D. Jung and E. K. Choi, "Differences of structural and electronic properties in  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ ," *Bulletin of the Korean Chemical Society*, vol. 20, no. 9, pp. 1045–1048, 1999.
- [32] A. Yu. Yakubovskii, S. V. Gudenko, N. V. Anshukova, A. I. Golovashkin, L. I. Ivanova, and A. P. Rusakov, "EPR observations of anomalous triplet states in  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$  and  $\text{BaPb}_y\text{Bi}_{1-y}\text{O}_3$ ," *Journal of Experimental and Theoretical Physics*, vol. 88, no. 4, pp. 732–737, 1999.
- [33] D. Varshney and M. P. Tosi, "Pairing mechanism and superconducting state parameters of cubic perovskite  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ ," *Journal of Physics and Chemistry of Solids*, vol. 61, no. 5, pp. 683–688, 2000.
- [34] D. Varshney, R. P. Kumhar, S. Shah, and R. K. Singh, "Effect of plasmons on superconducting transition temperature of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ : a three square well approach," *Indian Journal of Pure and Applied Physics*, vol. 40, no. 12, pp. 879–886, 2002.
- [35] A. P. Menushenkov, I. A. Troyan, and M. I. Eremets, "Resonant Raman scattering in superconducting  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ ," *JETP Letters*, vol. 77, no. 9, pp. 521–525, 2003.
- [36] G. É. Tsydynzhapov, A. F. Shevchun, M. R. Trunin et al., "Observation of a transition from BCS to HTSC-like superconductivity in  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$  single crystals," *JETP Letters*, vol. 83, no. 9, pp. 405–409, 2006.
- [37] N. V. Anshukova, A. I. Golovashkin, L. I. Ivanova, O. T. Malyuchkov, and A. P. Rusakov, "Insulator-metal phase transition and superconductivity in  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ ," *Journal of Experimental and Theoretical Physics*, vol. 81, no. 6, pp. 1163–1170, 1995.
- [38] A. I. Golovashkin, L. N. Zherikhina, G. V. Kuleshova, A. M. Tskhovrebov, and M. L. Norton, "Self-consistent spatially inhomogeneous state in HTSC  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$  single crystals," *Journal of Experimental and Theoretical Physics*, vol. 102, no. 4, pp. 603–615, 2006.
- [39] H. Fujishita, T. Yamada, S. Nakada et al., "Spontaneous strain in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ ," *Physica C*, vol. 470, no. 1, pp. S770–S771, 2010.

- [40] A. I. Golovashkin, A. M. Tskhovrebov, L. N. Zherikhina, L. S. Uspenskaya, and M. Norton, "Visualization of space inhomogeneous superconductor-insulator state in high- $T_c$  system  $Ba_{0.6}K_{0.4}BiO_3$  by magneto-optic method," *Journal of Physics: Conference Series*, vol. 150, no. 4, Article ID 042043, 2009.
- [41] Q. Huang, J. F. Zasadzinski, K. E. Gray, D. R. Richards, and D. G. Hinks, "Ideal tunneling characteristics in  $Ba_{1-x}K_xBiO_3$  point-contact junctions with Au and Nb tips," *Applied Physics Letters*, vol. 57, no. 22, p. 2356, 1990.
- [42] Y. Liu, J. F. Whitaker, and C. E. Platt, "Terahertz spectroscopy of superconducting thin film  $Ba_{0.6}K_{0.4}BiO_3$ ," *OSA Proceeding on Ultrafast Electronics and Optoelectronics*, vol. 14, pp. 238–241, 1993.
- [43] M. R. Trunin, "Surface impedance of HTSC single crystals in the microwave band," *Physica-Uspokhi*, vol. 41, no. 9, pp. 843–863, 1998.
- [44] A. A. Gallitto, M. Guccione, M. Guccione, and M. Li Vigni, "Microwave third harmonic emission by  $Ba_{0.6}K_{0.4}BiO_3$  crystals," *International Journal of Modern Physics B*, vol. 13, no. 9–10, pp. 1163–1168, 1999.
- [45] A. Agliolo Gallitto, M. Guccione, and M. Li Vigni, "Nonlinear microwave emission in  $Ba_{0.6}K_{0.4}BiO_3$  crystals near  $T_c$ ," *Physica C*, vol. 330, no. 3, pp. 141–149, 2000.
- [46] S. Fricano, M. Bonura, A. A. Gallitto, M. Li Vigni, L. A. Klinkova, and N. V. Barkovskii, "Microwave properties of  $Ba_{0.6}K_{0.4}BiO_3$  crystals," *European Physical Journal B*, vol. 41, no. 3, pp. 313–318, 2004.
- [47] N. V. Anshukova, V. B. Ginodman, A. I. Golovashkin et al., "Anomalies in the temperature dependences, resistance, critical current, and critical magnetic field in  $Ba_{1-x}K_xBiO_3$ ," *Journal of Experimental and Theoretical Physics*, vol. 70, no. 5, pp. 923–928, 1990.
- [48] L. N. Zherildfina, A. I. Golovashkin, A. V. Gudenko, and A. M. Tskhovrebov, "The de-current destruction of superconducting state and hysteresis effects in Ba-K-Bi-O," *Czechoslovak Journal of Physics*, vol. 46, supplement 2, pp. 851–852, 1996.
- [49] N. V. Anshukova, V. B. Ginodman, A. I. Golovashkin et al., "I-V characteristics hysteresis in superconducting  $Ba_{1-x}K_xBiO_3$  under microwave radiation," in *Proceedings of the International Conference on High Temperature Superconductivity and Localization Phenomena*, p. J14, Moscow, Russia, 1991.
- [50] A. I. Golovashkin, A. N. Zherikhin, L. N. Zherikhina, G. V. Kuleshova, and A. M. Tskhovrebov, "Observation of latent coherent effects in randomized systems," *Journal of Experimental and Theoretical Physics*, vol. 99, no. 2, pp. 370–378, 2004.
- [51] A. A. Gorbatsevich, V. Y. Kopaev, and I. V. Tokatly, "Stratification and superconducting droplets in high- $T_c$  superconductors," *JETP Letters*, vol. 52, p. 95, 1990.
- [52] A. A. Gorbatsevich, V. Y. Kopaev, and I. V. Tokatly, "Band theory of phase stratification," *Journal of Experimental and Theoretical Physics*, vol. 74, p. 521, 1992.
- [53] A. A. Gorbatsevich, V. Y. Kopaev, and I. V. Tokatly, "Phase separation and dielectric correlations in HTSC," *Physica C*, vol. 223, no. 1–2, pp. 95–105, 1994.
- [54] L. N. Zherikhina, A. I. Golovashkin, A. V. Gudenko, G. V. Kuleshova, A. M. Tskhovrebov, and M. L. Norton, "H-T phase diagram of high- $T_c$  Ba-K-Bi-O," *Physica C*, vol. 388–389, pp. 451–452, 2003.
- [55] A. I. Golovashkin, L. N. Zherikhina, G. V. Kuleshova, and A. M. Tskhovrebov, "Magnetic properties of high- $T_c$  system BaKBiO in superconducting state," in *Proceedings of 1st International Conference on Fundamental Problems of High Temperature Superconductivity*, p. 139, Zvenigorod, Russia, 2004.
- [56] L. A. Klinkova, M. Uchida, Y. Matsui, V. I. Nikolaichik, and N. V. Barkovskii, "Noncubic layered structure of  $Ba_{1-x}K_xBiO_3$  superconductor," *Physical Review B*, vol. 67, no. 14, Article ID 140501, 2003.
- [57] B. D. Josephson, "Possible new effects in superconductive tunnelling," *Physics Letters*, vol. 1, no. 7, pp. 251–253, 1962.
- [58] I. K. Yanson, V. M. Svistunov, and I. M. Dmitrenko, "Experimental observation of the tunnel effect for cooper pairs with the emission of photons," *Journal of Experimental and Theoretical Physics*, vol. 21, pp. 650–652, 1965.
- [59] S. Shapiro, A. R. Janus, and S. Holly, "Effect of microwaves on josephson currents in superconducting tunneling," *Reviews of Modern Physics*, vol. 36, no. 1, pp. 223–225, 1964.
- [60] K. K. Likharev and B. T. Yl'rik, *Sistemy s dzhozefsonovskimi kontaktami (Systems with Josephson Contacts)*, Izd-vo MGU, Moscow, Russia, 1978.
- [61] H. Kanter and F. L. Vernon Jr., "High-frequency response of Josephson point contacts," *Journal of Applied Physics*, vol. 43, no. 7, pp. 3174–3183, 1972.



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