Re-entrant Critical Current Behavior as a Common Feature of Single Grain Boundary Josephson Junctions in Bicrystals of Copperless Oxide Superconductors

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***Abstract*—Critical current and electrical transport properties of single grain boundaries in bulk bicrystals of the copperless oxide superconductors $BaPb_{1-X}Bi_XO_3$ and $Ba_{1-X}K_XBiO_3$ were measured in order to investigate quasiparticle tunneling and Josephson properties of these materials, which hold promise for the creation of low temperature microwave electronics and cryogenic particle detectors. An unusual and remarkable feature of the temperature dependencies of critical current is the non-monotonous behavior with sharp maximum at temperatures well below the critical temperature. This anomalous low temperature re-entrant behavior appears to be a new universal feature of single grain boundaries in copperless oxide superconductors. Our studies of grain boundary transport properties in $BaPb_{1-X}Bi_XO_3$ and $Ba_{1-X}K_XBiO_3$ bicrystals lead us to the conclusion that re-entrant behavior of critical current originates from changes in oxygen electronic states due to heterovalent substitution in copperless materials.**

I. Introduction

The copperless oxide superconductors $BaPb_{1-X}Bi_XO_3$ and $Ba_{1-X}K_XBiO_3$ comprise a class of superconducting materials, which is important for prospective applications, in spite of relatively low critical temperatures. Applications are expected in connection with the creation of tunneling junctions and the interesting Josephson properties of these materials. Low leakage and high-quality tunneling junctions were found on $BaPb_{1-X}Bi_XO_3$ bicrystals a few years ago [1], good quality tunneling structures were obtained by $Ba_{1-X}K_XBiO_3$ thin film with bicrystal substrate technology [2], and Josephson junctions in bulk bicrystals of the $Ba_{1-X}K_XBiO_3$ were found recently [3]. Tunneling and Josephson contacts in bicrystals could be useful in producing microwave electronic elements and cryogenic particle detectors. From this point of departure we performed intensive studies of transport properties in the bulk bicrystals $BaPb_{1-X}Bi_XO_3$ and $Ba_{1-X}K_XBiO_3$ in order to analyze the effects and properties of the single grain boundary as a macroscopic defect and to reveal the common features of Josephson junctions in bicrystals of the whole class of copperless oxide superconductors.

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II. Experimental Details

$BaPb_{1-X}Bi_XO_3$ bicrystals were grown by crystallization in a melt of Bi_2O_3, PbO and $BaCO_3$. $Ba_{1-X}K_XBiO_3$ bicrystals were synthesized by electrochemical deposition technique. Details of growth technique, chemical analysis and sample preparation were published earlier, in [1] and [3]. A few bicrystals with chemical composition, corresponding to the formula $BaPb_{1-X}Bi_XO_3$, and with the three values of the concentration parameter $x = 0.21 \pm 0.03$, $x = 0.27 \pm 0.03$ and $x = 0.29 \pm 0.03$ were synthesized. $Ba_{1-X}K_XBiO_3$ bicrystals were of the concentration parameter $x = 0.45 \pm 0.03$. The structural properties were similar for all the bicrystals: each block was a single domain crystal with well-defined structural order and typical geometric size of a few cubic millimeters. Three characteristic components of misorientation angles of the samples were determined using the Laue patterns in accordance with the procedure described in [1]. For all the bicrystals investigated in the work the misorientation was a combination of tilt and rotation of the [010] and [100] axes. Transport properties, to be discussed further in this paper, were found to be similar for all bicrystals with the values of all the components of the misorientation angle more than 15°, i.e., for bicrystals with entirely large misorientation. Typical properties of samples with such misorientations are reported here without an analysis of misorientation angles value effects.

III. Experimental Results

A. Critical current behavior

The Josephson properties of the bicrystals under study were established by measurements of RF-induced resonance on their current-voltage characteristics, observation of the oscillating critical current vs. applied low magnetic field dependencies, and studies of the thermally activated phase slippage (TAPS) effect. Details of the characterization of the bicrystals as Josephson junctions and their parameters could be found in [1], [3]. It should be mentioned that all the bicrystals investigated in this work have the values of Josephson penetration depth, comparable or larger than the dimensions of the single grain boundary, i.e., they should be treated as small Josephson junctions.

The temperature dependencies of the Josephson critical current of the bicrystals are presented in Fig. 1. The most remarkable feature of the dependencies is the clear maxima of the critical current, which take place at temperatures well below the critical temperature. (Dependencies are normalized with respect to maximal critical current density J_{cm}.) This feature was found to be universal for all the bicrystals with the structural, composition, and superconducting parameters mentioned above. Different temperatures of the critical current maximum were found, but two significant details were observed. The first is that the temperature of the maximum was found to be sensitive to the chemical composition of $BaPb_{1-X}Bi_XO_{3-Y}$ bicrystals and insensitive to the value of critical current density J_{cm} (taken at maximum). Unfortunately, we could not check this feature for $Ba_{1-X}K_XBiO_{3-Y}$ bicrystals, but for several studied samples with x = 0.45, the temperature of the maximum was found to be nearly the same. The second common feature of the critical current vs. temperature behavior of the bicrystals under study is power law dependence $(1-T/T^*_c)^\alpha$ near the critical temperature. The critical exponent $\alpha = 1.5 \pm 0.2$ was found to be the best fit for the measured data for all the $BaPb_{1-X}Bi_XO_{3-Y}$ and $Ba_{1-X}K_XBiO_{3-Y}$ bicrystals studied in this work. The characteristic critical temperatures used in fittings are presented in Fig. 1. It is necessary to note that these characteristic temperatures were exactly the same as we found in studies of the TAPS behavior of the crystals. Furthermore, a TAPS analysis of the critical behavior was performed to establish the value of the critical exponent by another method. The results of the TAPS effect studies are important enough to be published separately, but we will just say here that the critical exponent $\alpha = 1.5$ fit our data perfectly. Thus, the application of these two independent analysis methods gives us the proof that the above value of the critical exponent is a common feature of all samples we studied.

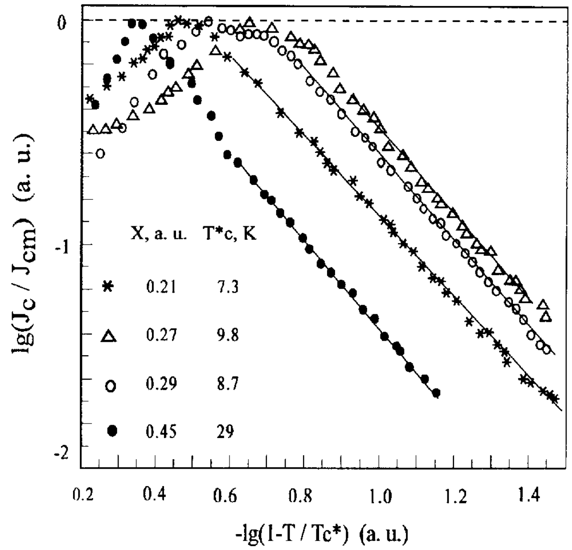

Fig. 1. Critical current vs. reduced temperature dependencies for grain boundary Josephson junctions in $BaPb_{1-X}Bi_XO_{3-Y}$ and $Ba_{1-X}K_XBiO_{3-Y}$ bicrystals.

B. Re-entrant Resistive Transitions

Studying the temperature dependence of the resistance of the single grain boundary under dc bias current, we found re-entrant behavior for all the bicrystals investigated. The typical temperature dependence for one of the investigated $BaPb_{0.73}Bi_{0.23}O_3$ bicrystals is presented in Fig. 2.

The dependencies were measured at different fixed dc bias currents through the grain boundary junction; the values of the currents are shown near the curves. The resistive transition, measured at I = 4 μA ($I/I_{cm} = 10^{-2}$) bias current, looks typical for a Josephson junction. The foot structure at the bottom of this resistive transition is caused by TAPS effect. The re-entrant resistive transitions were observed at 300-600 μA (I/I_{cm} ~ 0.5-0.8) range of currents. The differnce between the temperature, where measured resistivity becomes smaller than the resistance criterion of the superconducting state and the temperature of re-entrant appearance of the resistance decreases when the bias current increases. Under a bias exceeding the maximum Josephson current, a minimum of resistance without transition to a superconducting state was observed. The temperatures of these minima were the same as the temperatures of the maxima of critical current. No significant differences in behavior of the bicrystals of both compounds were observed, except the temperatures of minima. Thus, we conclude that the re-entrant resistive transitions should be treated as the second important common feature of the bicrystals under study.

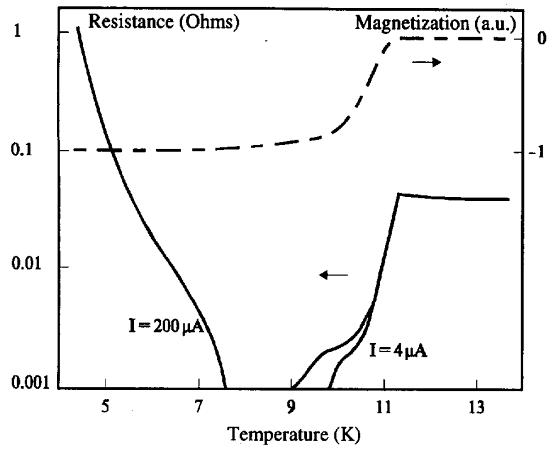

Fig. 2. Resistance and magnetization as functions of temperature.

We presume that the reason for the re-entrant behavior at temperatures lower than the critical temperature of the crystalline blocks is related to the decreasing of the critical current and the switching of the grain boundary junction in resistive mode if the bias dc current becomes larger than the critical current at a given temperature. To verify this supposition, we measured temperature dependencies of magnetic moment of the bicrystal in the absence and presence of a bias current. The resulting curve is shown by a dashed line in the upper part of Fig. 2. A clear transition to diamagnetic response below T_c without any peculiarities was observed under the same bias current.

We conclude that the re-entrant resistive transition in the superconducting bicrystals is not a bulk effect, but is completely defined by the properties of the grain boundary Josephson junction. The transition of this junction to a resistive state under bias current due to the degradation of critical current seems to be responsible for the re-entrant behavior. The Josephson supercurrent degradation may be connected with the growth of resistivity of the barrier of the grain boundary junction.

IV. Discussion and Conclusions

In this work we established that the bulk $BaPb_{1-X}Bi_XO_{3-Y}$ and $Ba_{1-X}K_XBiO_{3-Y}$ bicrystals demonstrate the anomalous behavior of critical current with the maximum at a temperature well below the critical one. Another common feature of the bicrystals with the non-monotonous behavior of critical current is their power law behavior, with the critical exponent of 3/2 at relatively high temperatures close enough to the critical temperature. These two most clear and most significant features were used earlier to propose an explanation for the non-monotonous critical current dependencies in $BaPb_{1-X}Bi_XO_{3-Y}$, based on the theory of Josephson junction with the semiconducting barrier [4]. This explanation, using the similarity in the Josephson properties, could also be applied to the bulk $Ba_{1-X}K_XBiO_{3-Y}$ studied in this work. It is necessary to point out that the aforementioned common features of Josephson properties of different bicrystals take place for Josephson junctions with basic parameters that varied widely. The insensitivity of the critical current anomaly to the value of the critical current density and other parameters, like Josephson penetration depth, indicates to us that this effect is not due to a random cause, such as an inhomogeneous current distribution in the junctions. The supposition, which could be used as a general one to explain the origin of the critical current anomaly, is concerned with the difference in electronic states between the oxygen ions, situated near the grain boundary and inside the single crystalline block. In the $BaPb_{1-X}Bi_XO_{3-Y}$ and $Ba_{1-X}K_XBiO_{3-Y}$ bicrystals studied in this work, the electronic state differences could be initiated and stabilized by local changes in the chemical compound due to the grain boundary segregation of heterovalent dopant. Further studies of the transport and structural properties of the bicrystals are necessary to reach a definitive conclusion about the origin of the anomaly.

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