

# Coherence properties and current transport in a ceramic Josephson medium, $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$

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The effect of heating on the I-V characteristics of the superconducting ceramic  $\text{BaPb}_{0.75}\text{Bi}_{0.25}\text{O}_3$  has been studied. The results show that this characteristic and also the temperature ( $T$ ) dependence of the critical current  $I_c$  may be altered substantially by changes in the heat transfer to liquid helium. The  $I_c(T)$  behavior found under single-pulse conditions is not affected by heat evolution in the interior or at the contacts. This behavior agrees well with the Ambegaokar-Baratoff equation for the Josephson current at temperatures up to  $T \lesssim 0.9T_c$ , where  $T_c$  is the critical temperature. This agreement proves that the switching events on the voltage-current characteristic are of a multiple tunneling nature. The  $I_c(T)$  behavior has been studied in detail for various samples. The nature of reentrant superconductivity, observed previously [T. H. Lin *et al.*, Phys. Rev. **B29**, 1493 (1984)], is explained. The critical current  $I_c$  has been measured as a function of a weak magnetic field  $H$ . For certain samples,  $I_c(H)$  is a monotonically decreasing function, while for others there are oscillations with a period  $H_{\text{osc}} \approx 0.13$  Oe near the maximum of  $I_c(H)$ . This behavior is attributed to a frustration of weak links between grains of the ceramic in the magnetic field.

## 1. INTRODUCTION

Recent years have seen extensive and multifaceted studies of inhomogeneous superconducting systems of two types. First, there are the ordered or partially ordered arrays consisting of Josephson junctions which are coupled through current-carrying contacts and/or an electromagnetic field.<sup>1–3</sup> Second, there are the granular or inhomogeneous composites, which either are prepared from several components by a special technique<sup>4,5</sup> or are prepared by sintering a powder to a ceramic<sup>6,7</sup> (similar structures form during the deposition of films<sup>8</sup>).

Such systems are of considerable research interest since they are pertinent to some new and rapidly developing fields in the theory of superconductivity: 1) the electrodynamics of coupled Josephson junctions<sup>3,9,10</sup> and 2) the theory of mixtures of a superconductor and a normal metal (or an insulator or a semiconductor), incorporating the percolation nature of the current transport in the superconducting regions and the complex topological structure of an infinite cluster (of fractal dimensionality<sup>11–15</sup>).

There are several questions to be resolved here. For example, in a study of the possibility of synchronizing several junctions (the studies have actually been restricted to one-dimensional series connections of junctions) by means of electromagnetic coupling it has been asserted<sup>9</sup> that a random scatter in the parameters should make the coherent operation unstable. Although this assertion does agree with experiments<sup>1</sup> on a regular two-dimensional Josephson array of lead-based tunnel junctions in which contacts between the superconducting grains are ruptured by a laser, the granular ceramic  $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$  (BPB; Refs. 6 and 7) has exhibited switching to a single-particle branch of the I-V characteristic

with a voltage  $U \approx 0.1\text{--}1$  V, which implies the simultaneous operation of many junctions. The resolution of this discrepancy between theory and experiment (which is apparently a consequence of the fact that the theoretical model of Refs. 3 and 9 is simpler than the actual situation) would also be useful from the standpoint of practical applications.<sup>16</sup>

There is also the more academic but exceedingly interesting question of whether superconductivity can coexist, in the same sample, with a negative temperature coefficient of the resistivity  $\rho$  above the critical temperature  $T_c$ . Various measurements which have been carried out for inhomogeneous systems—the ceramic BPB<sup>17,18</sup> and granular Al in a Ge matrix<sup>19</sup>—appear to provide evidence in favor of this possibility.

Finally, structural features found on the curve of  $T_c$  as a function of the magnetic field  $H$  in regular two-dimensional superconducting networks of In (Ref. 20) and Al (Ref. 21) and in a Pb-Cu-Pb array<sup>22</sup> have been explained on the basis of a homogeneous frustration<sup>23</sup> which is characteristic of these systems. If an inhomogeneous superconductor is furthermore irregular, e.g., if it consists of weakly linked granules (or grains) which do not occupy definite sites in a spatial lattice, then the frustration of the weak links will modify the  $T_c(H)$  dependence in this case.<sup>24</sup> Specifically, with increasing degree of disorder the oscillatory features should disappear, and  $T_c(H)$  should become a monotonically decreasing function. This behavior of superconducting granular composites is a consequence of a profound analogy between such systems and spin glasses.<sup>25</sup> As was pointed in Ref. 24, the frustration of the Josephson links between grains should also be manifested in the  $H$  dependence of other properties, e.g., the critical current  $I_c$ . In this regard it would appear to be worthwhile to carry out an experimental study

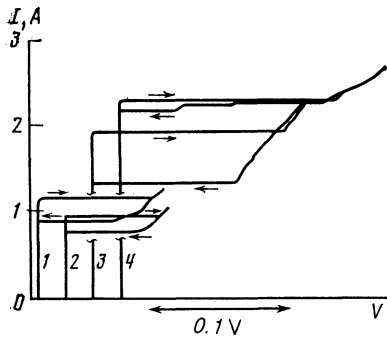


FIG. 1. I-V characteristics of sample 1 at several temperatures. 1— $T = 3.9$  K; 2—2.19 K; 3—2.13 K; 4—1.67 K.

of three-dimensional objects, since two-dimensional superconductors exhibit additional features which stem from the Kosterlitz-Thouless topological transition<sup>26</sup> and the weak localization of current carriers.<sup>27</sup>

The ceramic BPB solid solutions mentioned above occupy a special place among superconductors with a complex macrostructure. They exhibit a nonmonotonic composition ( $x$ ) dependence of  $T_c$  (Refs. 17 and 28), a metal-insulator transition at  $x \gtrsim 0.4$  (Ref. 28), and numerous structural transitions (for each given value of  $x$ ).<sup>29</sup> The metal-insulator transition, whose nature has not been completely resolved, is probably related to a change in the valence of bismuth ions,<sup>30</sup> which is facilitated at grain boundaries.<sup>6-8,17</sup> It is the conversion of the electron spectrum to that corresponding to an insulator in interlayers between grains<sup>17,31-33</sup> which causes the superconducting ceramic BPB to convert into a multiply connected Josephson medium during sintering. BPB is therefore a convenient model system, with easily adjustable parameters, for studying the questions outlined above. Our previous study<sup>6,7,16,34</sup> of its electrical and magnetic properties has shown that at  $T < T_{c1}$  BPB exhibits a superconductivity in each individual grain, while at  $T < T_{c2} < T_{c1}$  the tunneling of Cooper pairs between grains is responsible for the superconducting properties of the system as a whole, in agreement with the well-known model.<sup>35</sup>

In the present paper we are reporting a detailed study of the I-V characteristics of two groups of superconducting samples ( $x = 0.25$ ;  $T_c \approx 10$  K) with different critical currents  $I_c$ . In contrast with Refs. 6 and 7, the present measurements were carried out at temperatures down to  $T \gtrsim 1.5$  K, and a pulsed technique was used. It therefore became possible to determine the role played by ordinary heating and to prove that the switching from the superconducting state to a single-particle branch of the I-V characteristic is of a coherent Josephson nature. This study is the subject of Section 2 of this paper. In Section 3 we examine the temperature dependence of the critical currents and thereby find it possible to identify possible types of weak links in a ceramic and to explain the reentrant superconductivity in BPB. In Section 4 we report measurements of the  $I_c(H)$  behavior for various samples. We show that the disordered arrangement of grains with respect to each other leads to a frustration of weak links between them in a magnetic field.

## 2. EFFECT OF HEATING ON THE I-V CHARACTERISTICS OF THE CERAMIC $\text{BaPb}_{0.75}\text{Bi}_{0.25}\text{O}_3$

In this section we report experimental results on samples with a relatively high critical current, on the order of 1 A. Assuming in this case that the current is distributed uniformly over the cross section of the samples, whose dimensions are on the order of  $5 \times 5 \times 1$  mm, we conclude that the critical current density is  $j_c \approx 10-100$  A/cm<sup>2</sup> and is less than the typical values of good tunnel junctions.<sup>36</sup> Even more emphatically, we can conclude that these values do not contradict the interpretation that some of the junctions in the ceramic are of an S-N-S or S-N-I-N-S type (Section 3).

Figure 1 shows I-V characteristics of sample 1 at various temperatures. The nature of the characteristics changes substantially when the temperature of the liquid helium in which the sample is immersed is lowered. Below  $T = T_\lambda$  ( $T_\lambda = 2.18$  K is the temperature at which the helium goes into a superfluid state) there is an increase in the switching current  $I_c$ . The temperature dependence of this current is shown for various strengths of the magnetic field in Fig. 2 (the inset is the temperature dependence of the critical heat flux density to liquid helium<sup>37</sup>). The differential resistance of a sample with a current  $I > I_c$  does not change at the transition through the  $\lambda$ -point. An extrapolation of the sloping part of the I-V characteristic shows that the abscissa intercept is a nonzero voltage. This nonohmic behavior implies that single-particle tunneling is affecting this branch. At temperatures well below  $T_\lambda$ , the hysteresis on the I-V characteristic becomes less pronounced. When the I-V characteristic is traced out in the reverse direction, there is a return

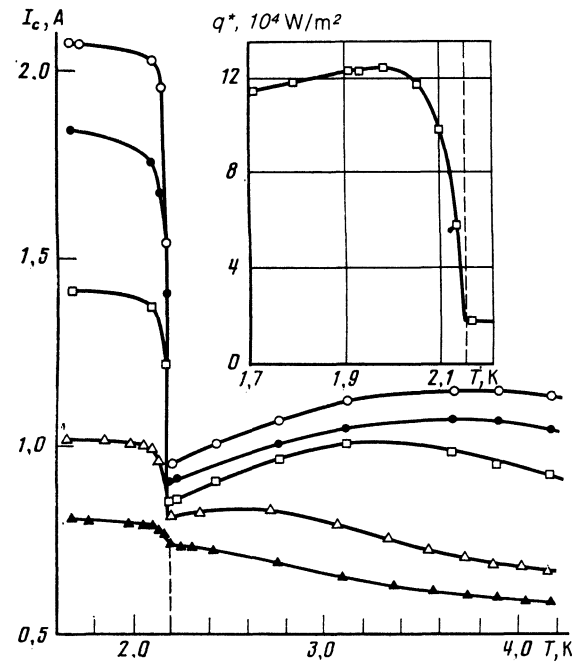


FIG. 2. Temperature dependence of the critical current  $I_c$  in sample 1 for various strengths of the external magnetic field:  $\circ$ — $H = 0$ ;  $\bullet$ —10 Oe;  $\square$ —20 Oe;  $\triangle$ —30 Oe; and  $\blacktriangle$ —50 Oe. The inset shows the temperature dependence of the critical density ( $q^*$ ) of the heat flux into the liquid helium.<sup>37</sup>

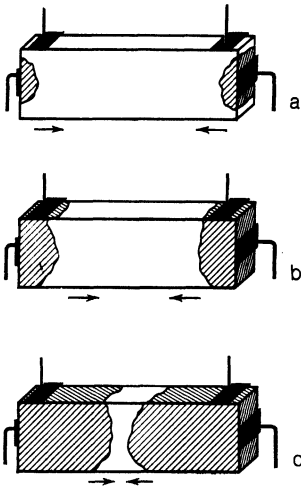


FIG. 3. Propagation of regions of film boiling (hatching) over the surface of sample 1.

to the  $I(U)$  dependence characteristic of temperatures  $T > T_\lambda$ ; this return occurs in the temperature interval in which  $I_c$  changes sharply. The sharp change in  $I_c$  at the  $\lambda$ -point is due to a change in the phase state of the helium; the value of  $I_c$  itself is determined at least in part by energy dissipation.

Studies of granulated In films<sup>38</sup> have also revealed a jump in the voltage and hysteresis on the reverse path of the I-V characteristic. These features have been attributed to the appearance of mobile, resistive thermal domains at inhomogeneities. In view of the granular structure of the ceramic BPB, it may be suggested that a thermal instability is occurring in our experiments also. There are two possibilities here: 1) Current-conducting contacts (junctions) are serving as an extended inhomogeneity; 2) a point inhomogeneity is localized somewhere in the volume of the ceramic. We will examine the first of these possibilities below. The second necessarily implies the assumption that the vertical part of the I-V characteristic corresponds not to a superconducting state but a resistive state with a very low resistance. In this case the maximum observable current in the vertical region should be determined by a transition from a "film-free" boiling to "film" boiling,<sup>7</sup> with the result that the sample can undergo an avalanche heating. This chain of arguments, however, leads to a contradiction: If the flux density ( $q$ ) of the heat which is evolved is to reach the critical value  $q^*$ , at which there is a change in the type of heat removal, we would have to also assume that the heat is evolved at a single "hot spot," so that we would have  $q = q^* = IU/S \approx 10^4$  W/m<sup>2</sup>, where  $I \approx 1$  A, and  $U \approx 0.1$   $\mu$ V. The area of the heat-transfer surface can then be estimated to be  $S \approx 10^{-11}$  m<sup>2</sup>. In view of the pronounced porosity of the sample, it may be assumed that the heat-transfer surface area agrees in order of magnitude with the cross-sectional area of the current path under consideration. In this case the transport current density near the hot spot would be  $j_c \approx 10^{11}$  A/m<sup>2</sup>, which would be higher than the maximum critical current densities which have been achieved for BPB single crystals.<sup>39</sup> If, on the other

hand, the current is distributed over an area much greater than the grain contact area, we would conclude that the current at which the differential resistance changes sharply is totally unrelated to a change in the nature of the heat transfer from the sample to the HeII, since in this case the critical heat flux density  $q^*$  would not be reached.

We are left with the other possibility, that heating affects the superconducting critical current, if we assume that the heat evolved at the normal contacts to the sample leads to a change in the nature of the heat transfer at a sufficiently high current.

If we were to set the current through the sample at a value below that which is the critical value at the given temperature, then there would be a disruption and a return to the normal state after a certain time. Visual observation through the window of the optical cryostat would reveal that a film is propagating along the surface of the sample over this time interval. The sequence of events would be as follows: A region of film boiling would appear near the current contacts (Fig. 3a). This region would then sweep over the entire cross section of the sample. Finally, it would collapse (Fig. 3, b and c; a potentiometer would reveal a switching to the normal state). One would obviously be observing the propagation of a normal zone through the volume of the inhomogeneous superconductor, accompanied by a change in the nature of the heat transfer. The jump in the current at the  $\lambda$ -point would be determined in this case by a change in several properties of the helium and by a sharp increase in the heat-transfer surface area at  $T < T_\lambda$ , while the normal helium would not penetrate into the pores of the sample. This conclusion follows from estimates of the time which would be required to fill the pores under actual experimental conditions. Assuming that the porosity of the sample is<sup>40</sup>  $\theta = V_{\text{hole}}/V \approx 0.3$ , that the height of the layer of liquid helium above the sample is  $\Delta h \approx 20$  cm, that the pore radius is  $R \approx 3 \cdot 10^{-4}$  cm, and that the length of the channel is equal to half the minimum linear dimension of the sample,  $l \approx 0.05$  cm, and using the tabulated data on the density ( $\rho_d = 0.1455$  g/cm<sup>3</sup>) and the dynamic shear viscosity [ $\eta = 3.15 \cdot 10^{-3}$  g/(s·cm)] for normal helium, we find the time for the penetration of the helium for viscous flow from the Poiseuille formula:

$$t = 8V_{\text{hole}}\eta l / \pi \Delta h g R^4 \rho_d \approx 17.5 h, \quad (1)$$

where  $g$  is the acceleration due to gravity, and the volume of the sample is  $V = 0.1 \times 0.2 \times 0.6$  cm<sup>3</sup> =  $1.2 \cdot 10^{-2}$  cm<sup>3</sup>.

The temperature dependence of the critical current was also measured in various magnetic fields,  $0 < H < 50$  Oe (Fig. 2). We see from this figure that the supercurrent is suppressed over the entire temperature range studied, and there is essentially no jump in it below  $T_\lambda$  at  $H \approx 50$  Oe. The reason for the lowering of the critical current is that the sample goes into a mixed state (as was shown in Ref. 41, the first critical magnetic field for BPB single crystals with  $x = 0.25$  in this temperature range is  $H_{c1} \approx 12$  Oe). The current anomaly at the  $\lambda$ -point disappears as a result of a decrease in the heat evolution at the contacts. Here one can observe a purely Josephson critical current  $I_c$  during a partial screening of the external magnetic field.

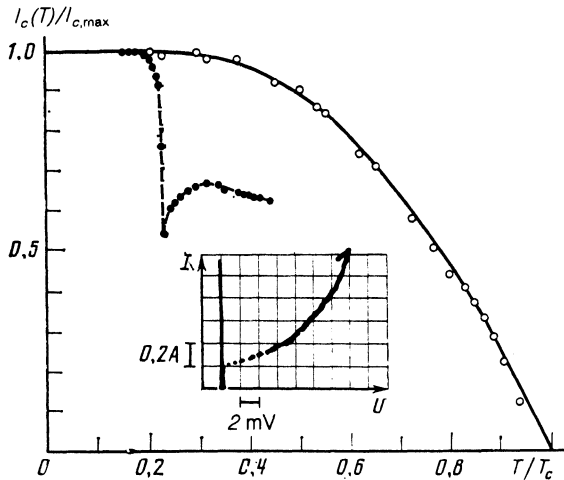


FIG. 4.  $I_c(T)/I_{c,max}$  versus  $T/T_c$  in sample 2.  $\circ$ —Pulsed experiments;  $\bullet$ —dc experiments. The inset shows the voltage-current characteristic of sample 2 in a pulse experiment at  $T = 1.97$  K.

If the arguments above are correct, then in order to achieve Josephson switching undistorted by thermal effects it will be necessary to prevent the penetration of heat propagating away from the normal contact into the interior of the ceramic as the I-V characteristic is swept out.

For this purpose, we supplemented the *dc* measurements in the case of sample 2 with pulsed experiments, using a highly linear generator of single triangular pulses of length  $\tau = (0.1-10) \cdot 10^{-3}$  s (this generator was developed especially for the purpose). The inset in Fig. 4 shows the I-V characteristic at  $T = 1.97$  K ( $\tau = 4 \cdot 10^{-4}$  s). We see that there is multiple Josephson tunneling with  $I_c = 1.15$  A and with a switching voltage  $\Delta U \approx 12.5$  mV  $\approx 4 \cdot 2(T)/e$ , where  $\Delta(T)$  is the superconducting energy gap, and  $e$  is the charge of an electron. Figure 4 also shows the temperature dependence of the critical current, in reduced coordinates,  $I_c(T)/I_{c,max}$  (the open circles). Up to the point  $T = 0.9 T_c$ , this behavior agrees well with the Ambegaokar-Baratoff equation

$$\frac{I_c(T)}{I_c(0)} = \frac{\Delta(T)}{\Delta(0)} \text{th} \frac{\Delta(T)}{2T} \quad (2)$$

for tunnel junctions, where  $I_c(T)$  is the critical current at the temperature  $T$ , and the Boltzmann constant is  $k_B = 1$ . Also shown here is the corresponding behavior found for sample 2 in the dc measurements. The maximum critical current is substantially lower here than in the pulsed case ( $I_{c,max} = 176$  mA). At  $T = T_\lambda$ , we see a jump in  $T_c$ , as for example 1.

Figure 5 shows oscilloscope traces of the pulses of the current, which is swept, and the voltage which arises across the sample ( $T \approx 6$  K). The jump occurs in a time much shorter than  $5 \cdot 10^{-7}$  s. It follows that the thermal contribution to the switching of a large number of junctions has in fact been eliminated, since the resistance, estimated from the I-V characteristic, essentially reaches the resistance corresponding to the normal state at the time of the switching, and we find that the velocity of the thermal wave, for a sample

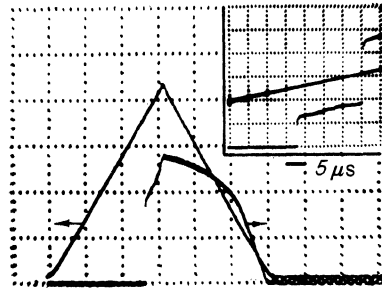


FIG. 5. Oscilloscope traces of the current, which is swept, and of the voltage (the vertical scales are 0.2 A and 5 mV per division, while the horizontal scale is 50  $\mu$ s per division). The inset shows the same behavior during double switching, on a longer time scale.

1.5 cm long, would have to be  $v > 3 \cdot 10^6$  cm/s, which would be unlikely.

What would be responsible for mutual synchronization of a significant number of tunnel junctions in a bulk sample? Synchronization through an electromagnetic field seems the most likely explanation. In particular, this explanation is suggested by the results of Ref. 7, where something on the order of 500 contacts were synchronized by an external microwave field, and superradiance was detected in a system consisting of two thin-film samples separated by an insulating film.<sup>42</sup> While an injection current was flowing through the lower film (the generator) in Ref. 42, a voltage with a maximum amplitude of about 2 mV was measured on the voltage-current characteristic of the upper film (the detector); this voltage blurred the initial voltage-current characteristic.

Indirect evidence for the possibility of a mutual synchronization of junctions through a high-frequency electromagnetic field comes from the observation in sample 3, with a critical current  $I_c \approx 5.5$  mA at  $T = 2$  K, of a scatter in the voltages determining the horizontal part of the I-V characteristic (Fig. 6). This scatter may be due to a self-detection of the radiation, when a voltage appears across one group of contacts in a sample as a result of radiation produced during the switching of another group to the one-particle branch.

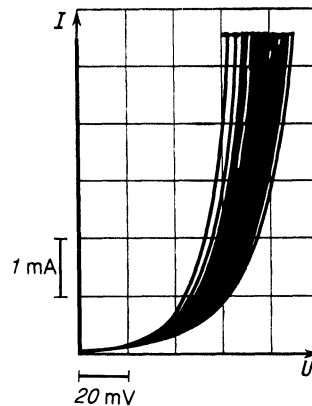


FIG. 6. Voltage-current characteristics of sample 3 at  $T = 2$  K. The black region is the region of possible reverse branches of the voltage-current characteristic for identical initial conditions.

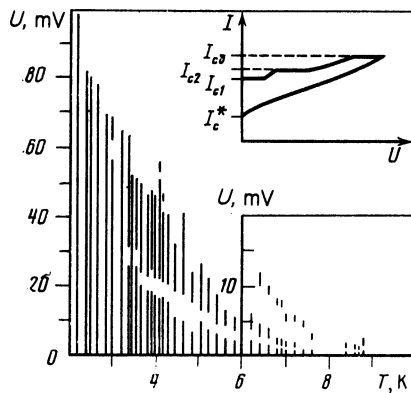


FIG. 7. Temperature dependence of the switching voltage  $U$  for sample 3. The inset shows a typical I-V characteristic and the corresponding critical currents: the reentrant supercurrent  $I_c^*$  and the critical currents corresponding to different groups of Josephson junctions ( $I_{c1}$ ,  $I_{c2}$ , and  $I_{c3}$ ).

However, the electromagnetic mechanism for switching in BPB cannot be solidly established until further experiments are carried out.

In summarizing the experiments described in this section of the paper, we should stress the main conclusion: It has been proved that the switching<sup>6,7</sup> in the ceramic BPB is of a multiple Josephson nature. It has also been shown that the I-V characteristic and the temperature dependence of the critical current may be modified substantially when the heat-removal conditions change.

### 3. STRUCTURE OF THE CURRENT PATHS; TEMPERATURE DEPENDENCE OF THE CRITICAL CURRENTS

The samples in the second group which we studied have a low critical Josephson current ( $\leq 10$  mA). The temperature dependence of the critical current of the samples does not have an anomaly at  $T = T_\lambda$ . Sharp switching events and a hysteresis on the reverse path are found on the I-V characteristics of these samples, recorded on a recording potentiometer. In an effort to eliminate heating effects to the maximum extent possible, we also studied the I-V characteristics of sample 3 with the help of a storage oscilloscope. This approach made it possible to find the temperature dependence of the first three voltage steps (Fig. 7); their critical currents  $I_{c1}$ ,  $I_{c2}$ ,  $I_{c3}$ ; and also the reentrant supercurrent  $I_c^*$  (Fig. 8:  $\circ - I_c^*$ ;  $\blacktriangle - I_{c1}$ ;  $\blacksquare - I_{c2}$ ;  $\bullet - I_{c3}$ ). From the behavior here we can draw some conclusions about the structure of the current paths in the ceramic.

The curves of the temperature dependence of the critical currents are the characteristics of three weak links; in the first two we can clearly see proximity effects (the steep slope over the entire temperature range).<sup>43</sup> The curve  $I_{c3}(T)$  is typical of an asymmetric tunnel junction.<sup>36</sup>

In the conducting network of the centered grains of the ceramic,  $S-N-S$  contacts shunt the tunnel junctions. In the case of a parallel connection with the tunnel junctions, the voltage steps become anomalously small at high temperatures (Fig. 7). The switching, which is proportional to the width of the energy gap, can thus be observed only at the lowest temperatures, where the  $S-N-S$  shunts disappear

(Fig. 8). The increase in the voltages which determine the steps with decreasing temperature is therefore due, on the one hand, to a synchronization of the junctions with approximately equal critical currents and, on the other, to a decrease in the effect of the  $S-N-S$  shunts.

By means of typical values<sup>6,7</sup> of the switching voltage at low temperatures,  $\Delta U \approx 0.1$  V, we can calculate the number of synchronized Josephson junctions from the relation  $\Delta U = n \cdot 2 \Delta / e$ ; we find  $n \approx 40$ . Taking the average size of the granules to be on the order of  $5-10 \mu\text{m}$ , for a sample  $1.5$  cm long, we conclude that, thought of as a current-conducting system, the sample is a three-dimensional set of clusters consisting of many granules. The boundaries between clusters are weak links, which exhibit either tunneling or proximity characteristics. This conclusion agrees with the arguments above.

The reentrant supercurrent  $I_c^*$  goes through a rounded maximum and falls to zero at low temperatures. Since the reverse path is determined primarily by the  $S-N-S$  junctions, the current  $I_c^*$  is a sort of critical current of the  $S-N-S$  junction, at which, as the temperature is lowered, there is a change in the resistance of the interlayer of normal metal. If the sample contains junctions of exclusively this type, one should observe a "through" ("reentrant") superconductivity, with the resistance dropping to zero at  $T = T_{R1}$ , and with a resistive state reappearing at  $T = T_{R2} < T_{R1}$ . Precisely this effect was apparently discovered in Ref. 44. The I-V characteristic found in that study revealed no switching of a tunneling nature, although Lin *et al.*<sup>44</sup> interpreted their results on the basis of a different model. That model was one of a superconductor-semiconductor-superconductor transition,<sup>45</sup> for which the following expression would hold at an intermediate degeneracy of the current carriers in the semiconducting interlayer:

$$I_c \sim \frac{T^{3/2} \Delta^2(T)}{\Delta^2(T) + \pi^2 T^2} \exp(-AzT^{1/2}), \quad (3)$$

where  $z$  is the thickness of the interlayer, and  $A$  is independent of the temperature.

In our case the presence of tunnel junctions masks the "reentrant" superconductivity. At measuring currents near the maximum of  $I_c^*$ , the  $R(T)$  behavior observed is similar to that in Ref. 44, with the difference that below  $T_{R2}$  the resistance tends toward a nonzero value determined by the

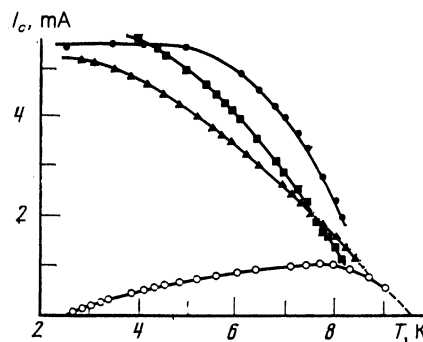


FIG. 8. Temperature dependence of the critical currents of sample 3.  $\circ - I_c^*$ ;  $\blacktriangle - I_{c1}$ ;  $\square - I_{c2}$ ; and  $\bullet - I_{c3}$ .

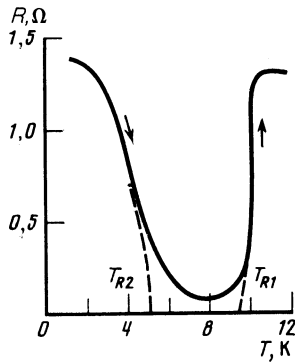


FIG. 9.  $R(T)$  of a sample with a low critical current. Solid line— $I > I_c^*$ ; dashed line— $I < I_c^*$ .

single-particle branch of the I-V characteristic (Fig. 9; the solid line corresponds to  $I > I_c^*$ , the dashed line corresponds to  $I < I_c^*$ , and the sample goes into a resistive state at  $T < T_{R2}$ ).

#### 4. DEPENDENCE OF THE CRITICAL CURRENT ON A WEAK MAGNETIC FIELD

The presence in the interior of a ceramic of a set of randomly oriented junctions, either tunnel junctions or something quite similar, coupled above the percolation threshold<sup>14,15</sup> in a common electric circuit, should make the functional dependence  $I_c(H)$  different from that of a single junction and also from that of a SQUID.<sup>36</sup> (The possibility that the current flows by a percolation mechanism in BPB at  $x \gtrsim 0.25$  should be borne in mind because the surface layer of the grains goes into an insulating state.<sup>6-8,17,31,32</sup>) The theoretical work on the properties of granular systems of this sort has been based for the most part on a Hamiltonian which is written as follows in the absence of a magnetic field<sup>2,24,43</sup>:

$$\hat{\mathcal{H}}_0 = - \sum_{ij} J_{ij} \cos(\phi_i - \phi_j), \quad (4)$$

where  $J_{ij}$  is a  $T$ -dependent quantity whose explicit form should be chosen to correspond to the  $S$ - $I$ - $S$  or  $S$ - $N$ - $S$  links used in the given experiments. In (4), the quantity  $\phi_i$  is the phase of the complex order parameter  $\psi_i \equiv \Delta_i \exp(i\phi_i)$  in the  $i$ th granule, and the summation runs over all possible pairs of granules. As was shown in Ref. 46, this model is isomorphic with respect to the classical  $XY$  model for a set of spins. In a magnetic field, the phase of each of the terms in Hamiltonian (4) changes:

$$\hat{\mathcal{H}} = - \sum_{ij} J_{ij} \cos(\phi_i - \phi_j - A_{ij}), \quad (5)$$

where

$$A_{ij} = \frac{2\pi}{\Phi_0} \int \mathbf{A} \cdot d\mathbf{l}. \quad (6)$$

The contour integral goes along the paths connecting the centers of grains  $i$  and  $j$ ;  $\mathbf{A}$  is the vector potential of the field  $\mathbf{H}$ ; and  $\Phi_0 = hc/2e$  is the quantum of flux. This expression ignores<sup>24</sup> a possible  $H$  dependence of  $J_{ij}$  and the difference (because of the Meissner effect) between the local field in a

pore and the applied field. If the structure is regular, the Hamiltonian  $\hat{\mathcal{H}}$  is periodic in  $H$  with a period  $\Phi_0/d^2$ , where  $d$  is the spatial period of this structure. Consequently, the field dependence of the critical temperature  $T_c$  or of the critical current  $I_c$  should have an oscillatory component.<sup>24</sup> Such oscillations of  $T_c(H)$  would be a generalization of the classical "single-loop" Little-Parks effect.<sup>36</sup>

As we mentioned in the Introduction, weakly coupled superconductors with a regular structure should experience a uniform frustration of intergranular bonds in a magnetic field under certain conditions,<sup>23,47</sup> and this effect has been observed.<sup>20,21</sup> The most interesting point, however, is the behavior of a Josephson system when the arrangement of granules is not perfectly periodic.<sup>24,48</sup> In this case the frustration of the bonds should lead to a damping of the oscillations in  $T_c(H)$  with increasing degree of disorder and also to a complete transformation of this behavior in the case of a pronounced disorder.<sup>24</sup> The disruptive effect of a disorder in the arrangement of granules on the oscillations in  $I_c(H)$  was discussed previously by Rosenblatt.<sup>46</sup>

In this section of the paper we are reporting measurements of the critical current of BPB Josephson composites with  $x = 0.25$  in a weak magnetic field. The results of these experiments confirm the arguments above and the calculations of Ref. 24.

For a typical sample, 4, with  $I_c \approx 15$  mA and  $T_c \approx 10$  K, the dependence  $I_c(H)$  measured by the method of Ref. 34 at  $T = 4.2$  K in fields much weaker than  $H_{c1}$  ( $4.2$  K)  $\approx 12$  Oe for BPB,<sup>41</sup> in which case the Abrikosov vortices do not yet penetrate into the interior of a grain, is a monotonically decreasing function (Fig. 10) reminiscent of the functional dependence  $T_c(H)$  found in Ref. 24. We see that a pronounced disorder of the system prevents a coherence of the order parameters in the different granules (or grains) because of the frustration of the weak intergranular bonds in an external magnetic field. A similar behavior was observed in Ref. 49 for a granular BPB film of the same composition. This film may be regarded as three-dimensional in terms of the superconducting properties of an individual grain of the ceramic, since its thickness is  $d_f \approx 0.3 \mu\text{m} \gg \xi \approx 60\text{--}70 \text{ \AA}$ , where  $\xi$  is

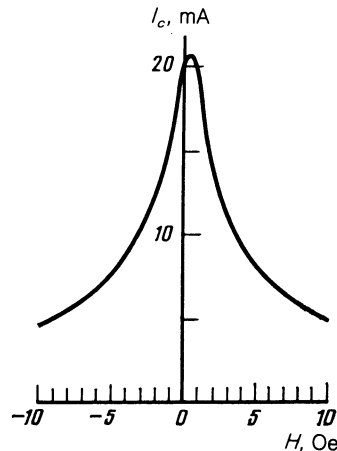


FIG. 10. Critical current of sample 4 versus the magnetic field.

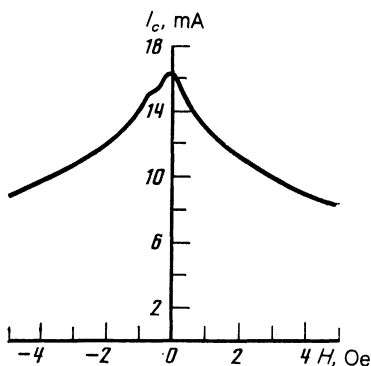


FIG. 11.  $I_c(H)$  for sample 5.

the coherence length at  $T=0$  (Ref. 41). However, as an inhomogeneous Josephson medium, the film obtained in Ref. 49 was two-dimensional, since the typical grain size in it was  $\approx 0.2 \mu\text{m}$ .

Although it is an exceedingly complicated matter to change the degree of disorder of a ceramic in a controlled way during the fabrication, it was found possible to select samples in which the coherence of the bonds was preserved in part even at  $H=0$ . Figure 11 shows  $I_c(H)$  for sample 5. Rotating the superconductor in the volume between Helmholtz coils made it possible to change the orientation of the superconductor with respect to the external field  $H$ . The  $I_c(H)$  curve in Fig. 11 was measured with  $H$  parallel to the sample. Figure 12 shows four curves, which show the behavior of the maximum of  $I_c(H)$  at the positions specified in the insets. The effect of the random nature of the orientation of the planes of the tunnel junctions between grains and of the macroscopic inhomogeneity of the entire composite is seen as a shift of the  $I_c(H)$  maxima with respect to each other in the various experimental configurations. The picture depends strongly on the angle between the current and the magnetic field. In Fig. 12a we can clearly see damped oscillations with an average period  $H_{\text{osc}} \approx 0.13$  Oe. We then find the typical linear dimension of a pore to be  $d = (\Phi_0/H_{\text{osc}})^{1/2} \approx 12 \mu\text{m}$ , in order-of-magnitude agreement with (slightly greater than) the actual dimensions of the pores in the ceramic. With increasing  $H$ , the oscillations fade away (Fig. 11), and the  $I_c(H)$  curve becomes essentially the same as that for samples of type 4 (Fig. 10).

In conclusion we should emphasize that the experiments which we are reporting here provide a comprehensive demonstration of the fact that the ceramic BPB is a multiple solid-state Josephson medium. It has been found possible to distinguish thermal effects from coherence effects during a switching between the superconducting and single-particle branches of the I-V characteristic. The observed disruption of the coherence of the individual intergranular bonds in a magnetic field is a result of a disordering of the medium and a frustration of these bonds. A circumstance which makes the ceramic BPB a model object, with unique properties is the conversion of the surfaces of the grains to an insulating state, so that the  $S$ - $I$ - $S$  and  $S$ - $N$ - $S$  bonds are inherent in the material itself.

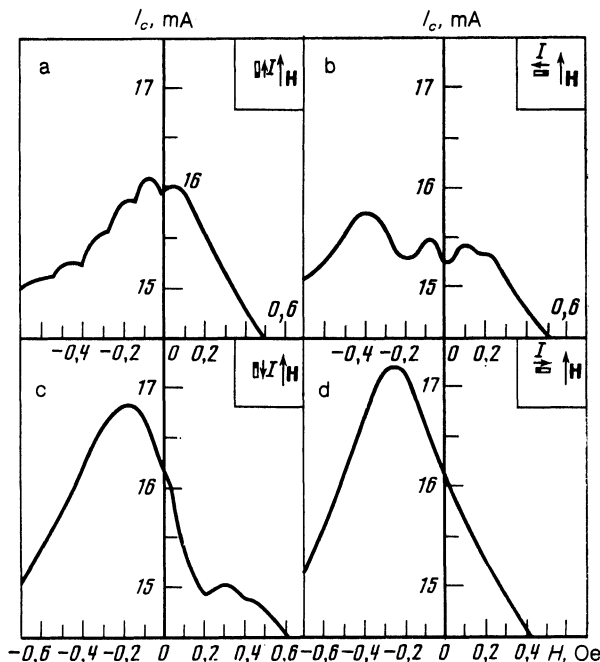


FIG. 12. Oscillations of the critical current of sample 5 at the maximum of the current as a function of the magnetic field. The insets show the orientation of the sample with respect to the magnetic field.

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