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PHYSICAL REVIEW B VOLUME 36, NUMBER 13 1 NOVEMBER 1987

Current transport in high-T_c polycrystalline films of Y-Ba-Cu-O

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(Received 17 July 1987)

Electrical measurements were performed on polycrystalline superconducting films of Y-Ba-Cu-O to explore tunneling effects in the film. The films were deposited on $SrTiO_3$ (110) substrates via pulsed excimer laser evaporation of bulk superconducting material. Data are reported for two films having T_c (zero-resistance temperature) of 79 and 63 K and transition widths (90% to 10%) of 5 and 15 K, respectively. Temperature-dependent I-V measurements performed over a broad range of T/T_c values (0.05 to 0.99) reveal that in both films the critical current is proportional to $(1-T/T_c)^{3/2}$ near T_c, while at temperatures lower than about $0.8T_c$ it is approximately proportional to $(1-T/T_c)^2$.

The discovery of superconductivity at 40 K in the La-Ba-Cu-O system by Bednorz and Müller[1] and at 93 K in the Y-Ba-Cu-O system by Wu *et al.*[2] has spurred an enormous growth of research activity in this field during the past year. Very recently, further important advances have been made in this area with the observation of electronic anisotropy in single-crystal $Y_1Ba_2Cu_3O_{7-x}$,[3] fabrication of thin-film superconducting quantum interference devices (SQUID),[4] and demonstration of critical current densities higher than 10^6 A/cm^2 in these materials.[5,6] The latter development has eliminated the major barrier for technological utilization of these materials and has added fuel to the already brisk activity in this field. A considerable emphasis of the current activity is on exploring ways of depositing high-quality thin films of the new oxide-based superconducting materials, since a number of novel applications of these materials are envisaged in the field of electronics and optoelectronics, wherein miniaturization is already an established key concept. Attempts have been made to deposit films by such methods as *e*-beam evaporation,[5,7] sputtering,[8] laser evaporation,[9] and molecular-beam epitaxy;[10] it has been demonstrated that polycrystalline films of oxide-based superconductors with good superconducting properties can be obtained with a proper choice of substrate and annealing conditions. These studies have revealed that epitaxial films of the superconductor can be grown only on specific substrates (and their crystallographic orientations) such as $SrTiO_3(100)$ and grain size of 0.5 μm or more can be easily achieved in such cases. On most substrates, however, the films are polycrystalline in nature and hence in the broader context of utilization of the films of the new superconducting materials in applications, it is of interest to understand the physical properties of such polycrystalline films.

The electrical properties of polycrystalline superconducting films depend upon the quantity of superconducting versus nonsuperconducting grains, the nature of the associated percolation network, and the tunneling features of the junctions formed between the grains.[11] Microscopically, the grain boundary tunneling characteristics depend upon the nature of the superconducting state of the participating grains, the nature and density of interface states, the electronic properties of the material in the intergrain space, and the quasiparticle excitation spectrum of the system. Clearly, these aspects can lead to interesting transport behavior in such materials as a function of temperature[11,12-14] and external perturbations;[15] and it can be used to realize novel device features.[16,17] The study of the transport properties of polycrystalline films of the new oxide-based superconducting materials is also of interest from the standpoint of examining whether these materials follow any of the trends predicted by tunneling calculations based on the assumption of a Bardeen-Cooper-Schrieffer (BCS)-like superconducting state for the grains and specific model potentials for the grain boundary region.[18-20] There appears to be only one report in the literature, by Moriwaki, Enomoto, and Murakami,[21] which directly addresses the issue of grain boundary tunneling in the new class of superconductors via electrical measurements. These authors employed thin films of $La_{1.8}Sr_{0.2}CuO_4$, which led to a T_c of 17 K and a transition width of 13 K (almost comparable to T_c itself) and reported measurements over a limited range of T/T_c values. On the basis of their observations these authors concluded that the nature of the Josephson junction itself is different in these materials compared to the well-studied oxide-based superconducting system,[11] $BaPb_{0.75}Bi_{0.25}O_3$ which is electronically similar to the $La_{1.8}Sr_{0.2}CuO_4$ system in the normal state. Since this conclusion can have important consequences it needs to be examined carefully.

In this paper we present data on electrical measurements carried out on polycrystalline films of Y-Ba-Cu-O deposited on $SrTiO_3(110)$ substrates. This substrate orientation was used in the present work because it does not lead to any significant degree of orientation in the deposited films[22] and as such the issues related to anisotropies may be ignored in the analysis. The films used in the present studies were deposited using the pulsed laser evaporation method directly from the parent superconducting bulk material.[9] A pellet of the superconducting material with nominal composition of $YBa_2Cu_3O_{7-x}$ showing onset of superconductivity at 93 K and a transition width of

RAPID COMMUNICATIONS

1 K was used as a target. It was mounted in a vacuum system with a base pressure of 5×10^{-7} Torr and irradiated through a quartz window with pulses from a KrF eximer laser (Lambda Physik EMG200E, 30-ns FWHM, 1 J/shot) at an angle of incidence of 45°. Using a quartz lens the energy density at the target was enhanced to 2 J/cm^2. The pellet was slowly rotated during irradiation to avoid texturing of its surface. The substrates were mounted at a distance of 3 cm from the target surface and were heated to 450 °C. The pressure during deposition was 1×10^{-6} Torr. Subsequent to the deposition the films were annealed at 900 °C for half an hour. The resistivity of the annealed films was typically in the 10^{-3} Ω cm range. The resistance versus temperature measurements were carried out using the conventional four-probe method inside a variable temperature cryostat. The I-V characteristics of the films were recorded at different temperatures by passing a current from 1 μA to 100 mA through the film and measuring the generated voltage. The value of the critical current was chosen to be the one at which the generated voltage rose above 1 μV. There was no evidence of any measurement-induced heating effect in the film.

The resistance-temperature relationships for the films used in this work are shown in Fig. 1. One of the films (hereafter called film A) has a zero-resistance temperature T_c of 79 K and a transition width (ΔT_c, 90%–10%) of less than 5 K; while the other film (hereafter called film B) has T_c and ΔT_c of 63 and 15 K, respectively. Clearly the transition shown by film A is much superior as compared to the one exhibited by film B. We intentionally selected these films with differing quality of transition with a view to examine whether the quality of transition is reflected by and is related to the nature of the tunneling process itself. The critical current density in the films, as measured by the conventional four-probe method, was found to be more than 10^4 A/cm^2 at $0.9T_c$. Considering the intrinsic limitations of the method this indeed is a lower limit.

In Fig. 2 typical I-V curves are shown for sample A ($T_c = 79$ K) at five different temperatures. It can be seen that at temperatures below T_c a finite voltage appears only when the current exceeds the value of critical current for the given temperature. At the temperature of 79.5 K, which happens to be just above T_c and in the superconducting transition region, the voltage begins to rise as the current is raised above zero and exhibits a significant degree of nonlinearity. This interesting feature was also reported by Moriwaki, Enomoto, and Murakami,[21] and was attributed to the intrinsic nature of superconducting transition rather than extraneous effects. The nonlinearity gradually decreases as the temperature is raised and at a temperature of 89 K, which is close to the onset temperature of superconductivity, the I-V behavior shows a pure Ohmic character (see Fig. 2).

In Fig. 3 the dependence of critical current is shown as a function of $1-T/T_c$ for sample A (open circles) and sample B (solid circles). First, it is interesting to note that the nature of this dependence is very similar for both the samples even though the quality of superconducting transition exhibited by the two films is different (see Fig. 1). This indicates that the basic features of the dependence of critical current on temperature are intrinsic to the tunneling process rather than the nature of the percolation network and inhomogeneities which control the quality of transition. It can be clearly seen that the nature of this dependence is different over different regions of $1-T/T_c$. For the values of $1-T/T_c$ over the range between 0.02 and 0.1 (i.e., for temperatures near T_c) the dependence is

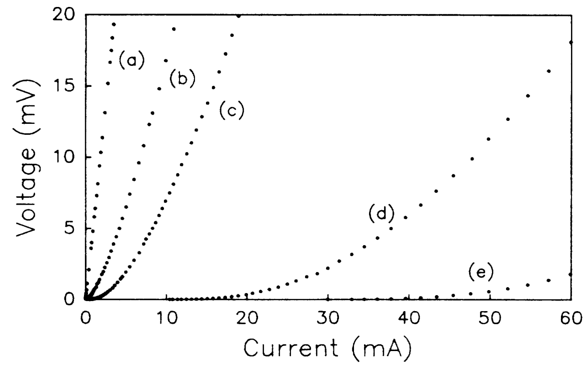

FIG. 2. I-V curves at (a) 89 K, (b) 79.5 K, (c) 76 K, (d) 51 K, and (e) 10 K.

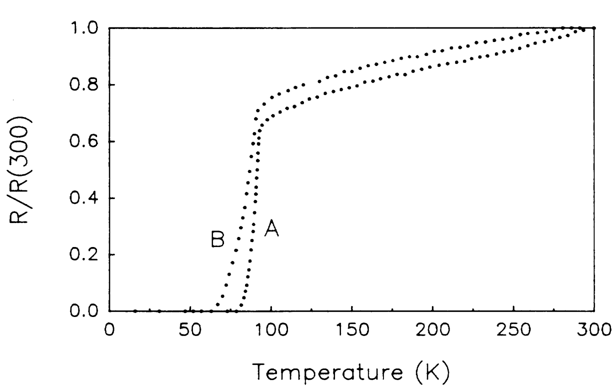

FIG. 1. Resistance-temperature relationships for two Y-Ba-Cu-O superconducting thin films (samples A and B) deposited on $SrTiO_3(110)$ substrate.

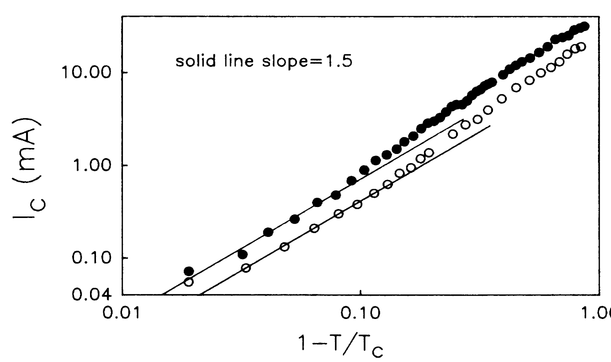

FIG. 3. Dependence of the critical current on $1-T/T_c$ for sample A (open circles) and sample B (solid circles).

of the form $(1-T/T_c)^{3/2}$; while at lower temperatures with $1-T/T_c$ values higher than about 0.3 the dependence changes its nature to approximately $(1-T/T_c)^2$. Apparently, Moriwaki *et al.* [21] could only observe an approximately square power dependence in their studies because their measurements were limited to $1-T/T_c$ values higher than about 0.1. These authors seem to have presumed that the same square power dependence holds at temperatures closer to T_c. Our data on the Y-Ba-Cu-O system show that this is not the case. Moriwaki *et al.* have concluded via a comparison of their results on La-Sr-Cu-O films with results for the case of Ba-Pb-Bi-O films [11] that the Josephson junctions have characteristically new features in the La-Sr-Cu-O films. They have also attributed the new features to the presence of layered structures in their films. Our observations bring out that in Y-Ba-Cu-O films (which also have layered structures) we, in fact, have a $\frac{3}{2}$ power dependence close to T_c, which is similar to the one observed for Ba-Pb-Bi-O films. It may be noted further from Fig. 3 that at temperatures very close to T_c (i.e., $1-T/T_c$ values smaller than 0.02) the dependence once again shows a departures from the $\frac{3}{2}$ power behavior. The $\frac{3}{2}$ power dependence at temperatures near T_c and departure from such behavior at temperatures very close to T_c has, in fact, been predicted to be the behavior of proximity junctions based on theoretical models which assume a BCS-like state for the superconductor. Thus it is possible that the grain boundaries in our films have a proximity junction character. Of course a non-BCS-like character of the superconducting state itself cannot be ruled out at the present state of understanding on the subject of the new oxide superconductors. It should be emphasized that the $\frac{3}{2}$ power dependence is predicted by the theoretical models for proximity junctions only at temperatures near T_c. [19] Hence the comparison sketched by Moriwaki *et al.* [21] between their observation of a square power dependence fitted to the data obtained over a broad range of $1-T/T_c$ values between 0.2 and 0.85 and the theoretical prediction of $\frac{3}{2}$ power dependence is not justified. It is of course a separate matter, and possibly an important one, to explore the origin of the approximately square power dependence observed in the films of La-Sr-Cu-O and Y-Ba-Cu-O at temperatures lower than about $0.8T_c$.

Josephson tunneling across a S-I-S′ (S: superconductor, I: insulator) junction has been theoretically studied by Ambegaokar and Baratoff (AB); [18] while tunneling across S-I-N-S′ (N: normal material) proximity sandwich has been investigated by de Gennes [19] and McMillan. [20] It should be emphasized that these models assume a BCS-like state for the superconductor and it is not clear whether these models would apply to the tunneling processes in the new oxide superconductors. Nevertheless, to make progress it is worthwhile to compare our results with the predictions of the above models. In order to see whether our films exhibit any features of S-I-S type tunneling (and if not, then to obtain the degree of departure) we sketch in Fig. 4 a comparison between our data and the prediction of AB model for the case of symmetric tunneling (dotted curve) which assumes a BCS-like state for the participating superconductors. Clearly, our data exhibit significant

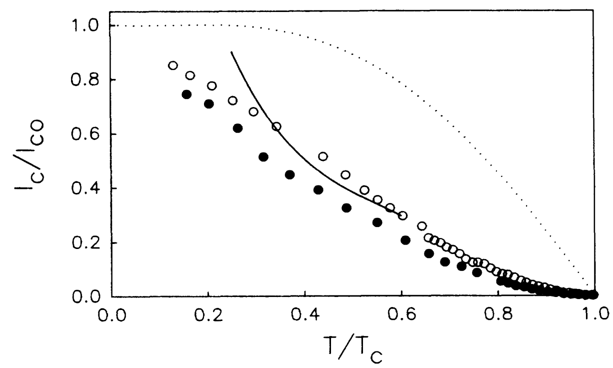

FIG. 4. Dependence of I_c/I_{c0} on T/T_c for sample A (open circles) and sample B (solid circles). Here I_{c0} is the extrapolated value of critical current at $T=0$ K. The dotted curve represents calculation based on the Ambegaokar and Baratoff model (Ref. 18) for S-I-S junction; while the solid line represents normalized result obtained for de Gennes's model for proximity junction (Ref. 19) for temperatures lower than $0.6T_c$.

departure from the AB curve over almost the entire temperature range, showing that the primary tunneling process in our films is not of S-I-S type and should be attributed to proximity junction tunneling (as mentioned earlier) and/or a weak-link type tunneling behavior. One may attempt to apply the AB model to a proximity sandwich by using the case of asymmetric (S-I-S′) tunneling discussed by AB with the use of an effective order parameter for S′ representing the N/S sandwich, however, this approach cannot be justified because of the intrinsic spatial variation of order parameter in such a structure, which is outside the scope of the AB model. [12,18] The model by de Gennes, [19] which studies the dc Josephson effect within the framework of Ginzberg-Landau theory via consideration of spatially dependent order parameters, is perhaps more applicable to the present situation. This model and its implications are discussed at length by Greenspoon and Smith [12] in the context of their experimental studies of proximity junctions in the Pb-PbO-Cu-Pb system. It is demonstrated that the key factor in the analysis is the leakage length K_n^{-1} given by

$$K_n^{-1}=(hv_{FN}l_N/6k_BT)^{1/2}=GT^{-1/2} , \tag{1}$$

where l_N is the intrinsic electron mean free path and v_{FN} is the average velocity of electrons on the Fermi surface of the normal (N) material forming the proximity junction. At low temperatures (T/T_c less than about 0.5) the predicted dependence has the following form:

$$I_{\max}(T)=\text{const}\times T^{-1/2}[\sinh(K_Nd_N)]^{-1} , \tag{2}$$

which is clearly different as compared to the $\frac{3}{2}$ power dependence predicted by the same model at temperatures close to T_c. As seen from Fig. 3 and discussed earlier we indeed observe a $\frac{3}{2}$ power dependence at temperatures near T_c and a different dependence at lower temperatures. Since the dependence given by Eq. (2) is divergent as T tends to 0 it cannot be expected to be valid at very low temperatures and it is reasonable to attempt a fit of our data to Eq. (2) over a limited range of T/T_c values between 0.25 to 0.6. We observed that for a reasonable range of Gd_N values between 0.01 and 1, one invariably obtains a curve with a concavity such as the one depicted by a typical curve shown by a solid line in Fig. 4 for the Gd_N value of 0.25. The experimental data clearly do not exhibit such a degree of concavity. It is possible that our films have some percentage of S-I-S junctions which obey the AB model and exhibit convexity in the temperature dependence of I_c/I_{c0}. The origin and location of such junctions is not clear at this moment. Another possibility which could lead to departure from the de Gennes or AB models is of course a non-BCS-like nature of the superconducting state itself. These issues can be clarified only after a better understanding of the mechanism of superconductivity in the new class of oxide superconductors is achieved.

The authors wish to thank J. M Rowell, W. L. McLean, and P. Lindenfeld for fruitful suggestions.

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