## Tunneling characteristics of internal Josephson junctions in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> –  $\delta$  thin films

W. S. Brocklesby, Don Monroe, M. Hong, S. H. Liou, J. Kwo, and G. J. Fisanick AT&T Bell Laboratories, Murray Hill, New Jersey 07974

> P. M. Mankiewich and R. E. Howard AT& T Bell Laboratories, Holmdel, New Jersey 07733 (Received 5 August 1988)

We have measured detailed I-V characteristics of small areas of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-8</sub> films, which exhibit structure associated with individual Josephson junctions within the film. Both hysteretic and nonhysteretic junctions are seen. A simple model relating the voltage steps to the junction properties is developed. The temperature dependence of  $I_c$  for the hysteretic junctions fits that predicted for quasiparticle tunneling junctions between conventional superconductors.

It is widely believed that the current-carrying properties of both bulk and thin films of the high- $T_c$  superconductor  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  are determined by Josephson-coupled links between grains of "pure" material. A number of experiments have shown the presence of these links in bulk material<sup>1,2</sup> and in thin films.<sup>3,4</sup> At least one of these experiments<sup>4</sup> has shown that in an epitaxial film, junction behavior and quasiparticle tunneling are observed. One of the major problems of studying both bulk ceramic and thin films is that the observed characteristics are the properties of large arrays of junctions, and provide little detail about the individual junctions. (In this paper we include under the name "junction" all types of weak links, whether shunted by tunneling or by normal resistances.) In an effort to look in some detail at these individual junctions, we have measured detailed I-V characteristics of small areas of thin films, whose size is comparable with the scale of microstructure within the film. We see a large number of discrete structures which we ascribe to individual junctions intrinsic to the film morphology. A wide range of junction behavior is observed, indicating the presence of both resistively shunted junctions and tunnel junctions. We have also studied the temperature dependence of the critical current,  $I_c$ , for some of these junctions. Several of the junctions show a temperature dependence which is consistent with Ambegaokar and Baratoff's theory of the variation of critical current with temperature in a tunnel junction between conventional BCS superconductors.<sup>5</sup> In this paper we describe some of the different junction behavior observed, and then try to establish relationships between these characteristics and the microscopic parameters of the junctions. We end by discussing the temperature dependence of junction critical currents.

The films used in these experiments were prepared by the mins used in these experiments were prepared by both sputtering  $6$  and evaporation.<sup>7</sup> Film thicknesse ranged from 1500 Å to 1  $\mu$ m. Substrates used were  $\langle 100 \rangle$ -oriented SrTiO<sub>3</sub> single crystals. Contacts, made using silver-paint or indium, had typical resistances of tens of ohms.

The first sample we discuss was grown by evaporation from multiple targets,<sup>7</sup> and designated  $A$ . Its thickness is  $\sim$ 0.7  $\mu$ m. Its microstructure consists of patches of irregular but relatively smooth material together with needlelike areas of length  $\sim$  10  $\mu$ m, whose long axes are oriented along the [100] axes of the substrate. Both these regions are YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> -  $_{\delta}$ , in different crystal orientations. The smoother patches are known to have the  $c$  axis perpendicular to the film plane. This sample has a superconducting transition onset at 89 K, and has a width (10%-90%) of 10 K.

After patterning the film into a  $30 \times 500$ - $\mu$ m<sup>2</sup> bridge by cw laser ablation, $8$  we observed the distinctive structure shown in Fig. 1. This figure shows the differential resistance,  $dV/dI$  (measured at 45 Hz) vs dc bias current I at a temperature of 57 K for sample  $A$ . In this range of bias current, we observe two discrete steps, the first of which is preceded by a large, sharp peak. This bias current at which these steps occur could be shifted by  $\sim$  10% by applying magnetic fields of a few gauss. Similar discrete steps in resistance were seen in several other samples, made by either sputtering or evaporation. The peaks that precede the steps in resistance are seen in many of the samples; in all cases the relative height of peak to plateau increases as temperature is decreased. This agrees qualitatively with theories of the effect of thermal fluctuations on junction characteristics<sup>9</sup> as discussed below.

A sputtered sample, <sup>6</sup> designated  $B$ , showed a wider



FIG. 1. Resistance vs dc bias for evaporated sample A. Sample temperature was 57 K.

variety of structure in the  $I-V$  characteristic. This film is nominally YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-6</sub>, and underwent an anneal for  $\sim$ 5 min at a temperature of 1045°C to form the superconducting phase. Its thickness is  $\sim$ 1  $\mu$ m. It consists of large rectangular platelets of typical size  $200 \times 50 \mu m^2$ , oriented with their side along two perpendicular directions, covering  $\sim$  20%-30% of the surface. These platelets have their  $c$  axis perpendicular to the plane of the film. The rest of the film contains both  $YBa_2Cu_3O_{7-\delta}$ and various nonsuperconducting oxides. Energy dispersive x-ray  $(EDAX)$  analysis of the platelets<sup>10</sup> showed that about 13 at. % of strontium had diffused into the film from the substrate during the high-temperature anneal. The sample was narrowed by scribing to a width of 500  $\mu$ m. Its resistance showed a semiconductorlike increase with cooling, and the onset of superconductivity at  $\sim$ 70 K. The platelet morphology was limited to a region of about 1 mm<sup>2</sup>, and contacts and scratches were made to isolate this region. All measurements were made with a twoprobe geometry and show a finite background resistance, which varies slowly with bias.

The  $I-V$  characteristics of this film at temperatures below  $\sim$ 35 K were studied in some detail. At least 30 discrete features could be seen in the I-V characteristic between 0 and 3 mA dc bias. A portion of the I-V characteristic containing a hysteretic feature is shown in Fig. 2, recorded using a digital oscilloscope. The voltage step is  $\sim$  1 mV. Other loops show different voltage steps, varying from 2 mV to 2  $\mu$ V.

The switching between the branches of the  $I-V$  curve followed Poisson statistics, with average switching times as long as <sup>1</sup> sec. These times were substantially reduced when high-frequency noise was added to the dc current drive. It is possible that even when no noise was intentionally added, external rather than thermal noise limited the extent of the loops, since no special filtering of the bias was performed.

Hysteresis loops with a very different character were observed in this sample at high  $(-3 \text{ mA})$  current densities. These loops were much more stable, and had a much larger extent in bias current. Their extent was also very dependent on the size and frequency of the ac current drive used for their observation. These loops are caused by Joule heating of the film as a whole, as has been ob-<br>served in the case of superconducting microbridges.<sup>11</sup> The served in the case of superconducting microbridges.<sup>11</sup> The qualitative differences in the properties of the hysteresis loops at low and high currents leads us to believe that hysteresis effects at low currents are intrinsic to the junctions. It is conceivable, however, that the thermal time relevant to individual junctions could be faster than that for a collective heating phenomenon.

Nonhysteretic junction characteristics like those in film A were also seen in this sample. Both voltage and  $dV/dI$ versus bias current for one of these is shown in Fig. 3. In this case the changes in resistance can be seen clearly in the  $I-V$  characteristic, and the R vs I curve resembles that for sample A.

The junction characteristics we observe are qualitatively similar to those of conventional superconductors. Although several exotic mechanisms have been proposed for high- $T_c$  superconductivity, the analysis in this paper has been performed using conventional junction theory. The form of the I-V characteristic of a conventional Josephson junction depends on whether the lowest-impedance shunting path is resistive or capacitative at the Josephson frequency. The  $I-V$  characteristics of junctions whose shunt resistance dominates the impedance are similar to Fig. 3, a smooth, nonhysteretic curve. Junctions whose impedance is dominated by capacitance show a hysteretic characteristic, similar to that shown in Fig. 2. (It must be noted that the characteristics shown in the figures correspond to junctions in series with a large resistance.) In conventional superconductors, hysteretic I-V characteristics are often observed if the shunting current path is quasiparticle tunneling, because the junction geometry that gives a large tunneling matrix element also gives a large junction capacitance.

To relate the characteristics of the junctions to the observed I-V characteristics, we need to model the effect of the rest of the film. The equivalent circuit for a connection of series and parallel junctions shown in Fig. 4 seems



FIG. 2. Detail from the  $I-V$  characteristic of sample  $B$  showing the hysteretic switching of a junction. The bias current was a 100 Hz triangle wave. In the two cycles shown, the switch between branches occurred at different points. Sample temperature was 4.2 K (Ref. 16).



FIG. 3. Nonhysteretic junction in sample B. Sample temperature was 14.3 K.



FIG. 4. Equivalent circuit for a single junction within a film.

to be quite general. Here  $R_{S1}$  is the resistance in series with the junction and  $R_{S2}$  represents the shunting resistance of the rest of the film. In general there will also be a resistance in series with the whole circuit, but this resistance is not relevant to the junction behavior.

If the critical current densities for different junctions are similar, the switching of one junction may increase the current in other junctions so that they also switch to the normal state. In this case, all the switching junctions should be included in the single effective "junction" of the equivalent circuit (note that the choice of current path does not depend on a coherent phase relationship between different junctions). In this model, if the effective junction develops a voltage drop  $\delta V_1$ , the voltage step observed across the voltage probes is reduced:

$$
\delta V = \frac{R_{S2}}{R_{S2} + R_{S1}} \delta V_1.
$$
 (1)

In BCS theory,  $12$  the voltage developed across a tunnel junction in the normal state just above  $I_c$  is  $2\Delta(T)/e$ , where  $\Delta(T)$  is the superconducting energy gap. The size of the voltage step observed in this experiment gives only a lower limit on  $\Delta(T)$  in this material.

The apparent value of the critical current is increased by the same factor:

$$
I_{c1} = \frac{R_{S2}}{R_{S2} + R_{S1}} I_{\text{meas}} ,
$$
 (2)

where  $I_{\text{meas}}$  is the measured bias current for the whole film at which the voltage jump occurs.

If the voltage drop  $\delta V_1$  may be no larger than  $2\Delta(T)/e$ , this model allows us to put a lower limit on the total critical current of the junctions comprising the effective junction

$$
I_{c1} \ge I_{c1, \text{meas}} \frac{\delta V}{2\Delta(T)/e} \,. \tag{3}
$$

For the junction shown in Fig. 2 we estimate, using BCS<br>theory for  $2\Delta(-27 \text{ mV}$  for  $T_c = 90 \text{ K}$ ), that  $I_c > 40 \mu\text{A}$ .

The temperature dependence of the critical current of two hysteretic junctions in sample  $B$  is shown in Fig. 5. The data are shown as dots. The solid line is a fit to the theory of Ambegaokar and Baratoff<sup>5</sup> for tunnel junctions, which states that

$$
\frac{I_c(T)}{I_c(0)} = \frac{\Delta(T)}{\Delta(0)} \tanh \frac{\Delta(T)}{2ekT} .
$$
 (4)

The values of  $\Delta(T)$  for BCS have been tabulated by Mühlschlegel.<sup>13</sup> The fit has two free parameters: the low-temperature critical current [scaled by the relation in



FIG. 5. Temperature dependence of the "critical current," that is, the bias at which features in the  $I-V$  characteristic are observed. Dots are measured current for two of the hysteretic junctions in sample  $B$ , while the solid lines are fits to the Ambegaokar-Baratoff theory (using weak-coupling BCS temperature dependence for the gap). Critical temperatures used in the fits were  $42.5$  K for  $(a)$ , and  $39.5$  K for  $(b)$ .

Eq. (2)], and the  $T_c$  of the superconductor, which is found to be  $\sim$  40 K for both of these junctions. This is reasonable in the light of the amount of Sr in areas of this film, which is likely to lower the  $T_c$  of these areas. The film is also likely to have lost oxygen over its lifetime. These data show that the shunting current path across the junctions is due to quasiparticle tunneling, in a fashion very similar to that seen in conventional superconductors.

In making this fit, we have assumed that the fraction of current passing through the rest of the film just below  $I_c$  is the same at all temperatures. This is true only if all the junctions have the same form for  $I_c$  vs  $T$ . Some of the junctions have different temperature dependences on  $I_c$ , but because the resistance  $R_{S2}$  represents the combination of a large number of junctions, any change in the current path should be a small effect. Indeed, the smoothness of the curves of  $I_c$  vs  $T$ , suggests that no radical changes of the current path are occurring, and there are no abrupt changes in the normal resistivity of other sections of the film.

As the temperature of the film was increased, the characteristics of both of the junctions whose critical current versus temperature is shown in Fig. 5 changed from hysteretic to nonhysteretic, and finally were so reduced in amplitude and broadened that they were indistinguishable from the background. For this reason we could not follow the critical currents down to zero current. Similar evolution of the junction characteristics has also been seen in granular lead<sup>14</sup> and  $BaPb_xB_1-xO_3$ ,<sup>15</sup> where tunnel junctions are observed at grain boundaries. For  $BaPb_xB_{1-x}O_3$  the effect was ascribed to the particular parabolic shape of the potential barrier at the grain boundary. Such a specialized description does not appear necessary, however, as we now discuss.

In general both branches of the hysteresis curve have a finite lifetime at any nonzero temperature because of thermal fiuctuations. As the ratio of the Josephson coupling energy to  $kT$ ,

$$
\gamma \equiv \frac{\hbar I_c(T)}{ekT} \,,\tag{5}
$$

decreases, the switching time between states must decrease, so a curve that looks hysteretic at low temperatures will look nonhysteretic when thermal fluctuations reduce the time for switching below the observation time. Thus, both the direct effect of temperature on  $\gamma$ , as well as the reduction of critical current with  $T$ , will tend to wash out the structure.

Additional evidence for the presence of quasiparticle tunnel junctions comes from the very small increase in resistance in the normal state just above  $I_c$  seen for some junctions. In a resistively shunted junction, the voltage step would correspond to the replacement of the superconducting junction by a nonzero resistance, which would contribute even more to the voltage drop as the current increased. In contrast, a tunnel junction just above threshold should have a small differential resistance. Our observations show that both tunnel junctions and resistively shunted junctions are present in these films.

We cannot determine from these data the normal resistance path for the nonhysteretic junctions seen in film A. However, the fact that the normal resistance path is much larger than the impedance of the junction capacitance at the Josephson frequency suggests the formation of proximity-coupled Josephson junctions with wide barriers.

In summary, we have seen structure in the  $I-V$  characteristics of narrowed films of  $YBa_2Cu_3O_{7-\delta}$  which can be ascribed to discrete Josephson junctions. Both hysteretic and nonhysteretic behavior is observed. The effects of thermal fluctuations are seen to smear out junction characteristics in a manner qualitatively consistent with theory. The consistency of the temperature dependence of  $I_c$  with Ambegaokar-Baratoff theory, as well as the small increase in resistance above  $I_c$  seen, shows that the shunting current path in some of the hysteretic junctions can be ascribed to quasiparticle tunneling. Clearly, a precise measurement of detailed parameters requires the isolation of single junctions. However, while single junctions remain difficult to fabricate, these intrinsic junctions provide a unique opportunity to study the physics of the Josephson effect in  $YBa_2Cu_3O_{7-\delta}$ , and to study the relationship between growth morphology and film behavior.

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