## Proximity Effect in  $YBa_2Cu_3O_7/Y_0.6Pr_{0.4}Ba_2Cu_3O_7/YBa_2Cu_3O_7$  Junctions

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We report critical-current measurements in all high- $T_c$  superconducting-normal-superconducting junctions using Y<sub>0.6</sub>Pr<sub>0.4</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (with  $T_{cN}$  =40 K) as the normal metal. Above  $T_{cN}$ , we find a clear exponential dependence of  $I_c$  on the thickness of the barrier which is characteristic of the proximity effect. The order-parameter decay length is about 120 Å for  $T > 60$  K, and it diverges as  $T_{cN}$  is approached. We estimate that  $\xi_0$  for this material is 80  $\pm$  25 Å.

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The existence of the proximity effect at high temperatures characteristic of high- $T_c$  materials has not been as yet convincingly demonstrated. The existence or nonexistence of this effect is important, both for establishing the validity of the conventional description of superconductivity in high- $T_c$  materials and as a route for fabrication of practical Josephson junctions. Much work has been expended on  $S/N$  structures, where S is a high- $T_c$ superconductor and  $N$  is a normal metal (Ag, Au) [1,2]. To demonstrate the tunneling of Cooper pairs through the normal metal, another low- $T_c$  superconductor is deposited on top of the  $S/N$  to create an SNS sandwich. Using a low- $T_c$  superconductor as a counterelectrode limits the maximum temperature at which the effect can be observed to a few K. In order to check whether the effect exists at all temperatures pertinent to high- $T_c$  materials, one needs an SNS sandwich with both superconductors of the high- $T_c$  variety. Deposition of a high- $T_c$  material on top of an  $S/N$  structure requires temperatures of  $650-750$  °C, where the noble metal diffuses rapidly into the superconductor, creating a barrier layer whose dimensions and composition are rather uncertain. Additionally, one cannot rule out the presence of direct microshorts of superconducting material. Although we were able to demonstrate the Josephson effect in such structures [3], a systematic study of the superconducting properties of the SNS sandwich containing a noble metal as a function of its thickness is extremely difficult. Thus, the existence of the proximity effect cannot be unambiguously proven with noble-metal normal barriers.

To find another suitable material, one should take into account the basic boundary conditions at the  $S/N$  interface. In the dirty limit, these boundary conditions are expressed as [4]

$$
\frac{\Delta_S}{\Delta_N} = \frac{(N_0 V)_S}{(N_0 V)_N}, \quad D_S \left(\frac{d\Delta}{dx}\right)_S = D_N \left(\frac{d\Delta}{dx}\right)_N. \tag{1}
$$

Here  $N_0$  is the density of states,  $\Delta$  is the pair potential, V is the Bardeen-Cooper-Schrieffer electron-electron interaction, and  $D$  is the diffusion coefficient. The first part of Eq. (1) shows that in order for  $\Delta_S$  to be as close as possible to  $\Delta_N$  at the interface, one should use a normal metal with a nonvanishing attractive interaction. The second part of Eq. (1) relates the spatial derivatives of the pair potential on both sides of the interface. A large value of this derivative on the  $S$  side implies a strong depression of the pair potential at the interface. It is easy to see that this derivative is of the order  $D_N\Delta/D_S\xi_S$ , where  $\xi_S$  is the coherence length in S. In the high- $T_c$ materials, with their small  $\xi_S$ , this derivative tends to be quite large, indicating that in this case the pair potential is indeed strongly depressed, particularly when the normal material is a good conductor with large  $D_N$ . For that reason, Deutscher and Simon [5] suggested that one can compensate this depression somewhat by using a normal material having a density of states lower than that of the superconductor, i.e.,  $D_N < D_S$ . Our choice of a material fulfilling these requirements was  $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ . First, it was experimentally demonstrated that even at 700-800 °C, the interdiffusion of  $Y_x Pr_{1-x} BCO$  and YBCO takes place on a scale not larger than one unit cell [6]. Thus, normal layers with well-defined dimensions can be created. Second,  $Y_{0.6}Pr_{0.4}BCO$  is a superconductor with  $T_{cN}$  of 40 K, so the strength of the pairing interaction should be comparable to that of YBCO. Finally, full substitution of Pr for Y turns the material into a semiconductor, which implies that the density of states is indeed lower than in YBCO.

The experiments were performed on a series of  $YBCO/Y_{0.6}Pr_{0.4}BCO/YBCO$  edge junctions, differing only by  $d_N$ , the thickness of the Y<sub>0.6</sub>Pr<sub>0.4</sub>BCO layer. A cross section of such a junction is shown schematically in the inset of Fig. 1. We established that both YBCO and  $Y_{0.6}Pr_{0.4}BCO$  grow epitaxially on the (100) SrTiO<sub>3</sub> substrate, with the  $c$  axis up. Thus, the junctions link the two YBCO electrodes along the a-b plane. It is well known that the surface of thin YBCO film is degraded as soon as it is removed from the growth chamber. If an N material is deposited onto such a degraded surface, there will be a large, sample-dependent effect on the supercurrent through the  $S/N$  interface, due to the presence of



FIG. 1. Critical current vs temperature for an SNS junction with  $d_N = 1250$  Å. Inset: The cross section of the edge junction.

a surface barrier. To prevent that, the junctions were prepared in an in situ laser ablation deposition process, using multiple targets and externally manipulated stainless-steel contact masks. Our basic film deposition setup has been described previously [7]. The in situ technique of junction preparation was developed in conjunction with our ongoing work on the fabrication of all high- $T_c$  Josephson junctions [8,9]. In order to keep the quality and deposition rates of the YBCO and  $Y_{0.6}$ - Pr $_{0.4}$ BCO exactly the same in all the junctions, the whole series of junctions was prepared in the same deposition run.

We now describe the deposition sequence, referring to the cross section in Fig. 1. First, a  $1-\mu m$  YBCO film is deposited on half the substrate, while the other half is covered by a stainless-steel contact mask. Then, using external manipulators, another mask is placed parallel to the first one, covering part of the deposited YBCO film, so that only a strip of the film near the junction is exposed. The target holder is rotated to bring the  $SrTiO<sub>3</sub>$ target into the laser beam, and a  $1-\mu$ m-thick strip of insulator is deposited. Next, both masks are removed and a third mask is put in place. This mask covers the whole substrate except for a narrow slit which runs along the line of the junction. The slit has the exact width of the substrate, thus exposing only the junction region. We deposit  $Y_{0.6}Pr_{0.4}BCO$  using a third target through this slit. Once the thickness is sufficient to create a 250-A barrier across the edge of the YBCO film, the mask is shifted sideways by 1 mm, and another 250 Å are deposited. Note that as the slit has the exact width of the substrate, each time this mask is shifted, another section of the substrate is covered. As the slit moves across the substrate, it leaves behind a sequence of barriers, each one thicker by 250 A than its predecessor. After the whole substrate is covered, this mask is removed and the first mask is brought back to cover the other half of the substrate, so that an overlaying film of YBCO, identical to the bottom one, can be deposited. The substrate temperature during the deposition is  $730^{\circ}$ C and the oxygen pressure is 200 mTorr. For each junction,  $d_N$  was determined by the number of laser pulses hitting the  $Y_{0.6}Pr_{0.4}BCO$  target. Since the barrier grows on an edge of a YBCO film, its thickness must be calibrated. This was done by growing two films simultaneously: one on a substrate perpendicular to the beam of evaporated material and the other parallel to it (the substrate parallel with the beam has the same orientation as the edge of the YBCO). By comparing the thickness of these two films, the ratio of the barrier thickness to the (known) film thickness is established, 0.3 in our case. The finished structure was patterned into microbridges 200  $\mu$ m long and 25  $\mu$ m wide, with each microbridge containing one junction. The patterning was done using deep-uv photolithography with PMMA photoresist, followed by a dilute phosphoric acid etch. The thickness of the junction,  $d_N$ , varied between 250 and 1250 A.

In Fig. 1, we show the critical current  $I_c$  versus temperature for a 1250-A-thick junction. The solid line is a fit by a  $(T - T_c)^2$  dependence, characteristic of an SNS sandwich near  $T_c$  [10]. It is seen that  $I_c$  starts to increase rapidly below about 45 K. This is due to the incipient superconducting transition of the barrier material at  $T_{cN}$ . We point out that the measured  $T_{cN}$  ( $R=0$ ) for a Y<sub>0.6</sub>- $Pr_{0.4}$ BCO film was 40 K, which is lower by about 5 K than the temperature at which  $I_c$  starts to increase. We show below that this difference is due to the divergence of the decay length in the normal metal,  $K^{-1}$ , as  $T_{cN}$  is approached. The observation of two distinct transitions, one at  $T_c$  and the other near  $T_{cN}$ , shows that the barrier is made of a homogeneous material having its own distinct  $T_{cN}$ . In particular, this observation is important in order to rule out the possibility that the supercurrent in the junctions flow through YBCO microshorts.  $A$  priori, our choice of this particular barrier material was based on the almost total absence of interdiffusion between YBCO and PrBCO or  $Y_x Pr_{1-x} BCO$  [6], which allowed the study of their superlattices with a resolution of one unit cell [11] without creating a short. Thus, the possibility that a short will appear through a barrier hundreds of A thick is truly remote. Finally, in the case of microshorts the supercurrent flows through a percolative network, and one would therefore expect the dependence of  $I_c$  on the barrier thickness to be a power law. Instead, we show below that this dependence in our junctions is exponential, which is characteristic of the proximity effect.

The critical current of several junctions versus temperature is shown in Fig. 2. The strong dependence of  $I_c$  on  $d_N$  is evident. At first glance, it appears that  $T_c$  is shifted downwards for thicker junctions (the typical  $T_c$  of our YBCO films is 90 K). However, an extrapolation to  $I_c = 0$  shows that within experimental error  $T_c$  is the same for all the junctions. The apparent shift of  $T_c$  arises from 3039



FIG. 2. Critical current vs temperature for several junctions. Sets of data are labeled by  $d_N$ .

the fact that once  $I_c$  becomes less than about 10  $\mu$ A, the  $I-V$  characteristics show a finite resistance. We attribute this effect to thermal fluctuations. As  $I_c$  gets smaller, thermal fluctuations will disrupt the superconducting order. The minimum value of  $I_c$  which can still be measured is on the order of  $2kT/\Phi_0$  and its value is  $\sim$ 1  $\mu$ A at 77 K. Hence, thicker junctions, having smaller  $I_c$ , would remain in this resistive state at lower temperature than thin junctions and their  $T_c$  would appear lower.

For an SNS junction, the theory of the proximity effect predicts  $I_c$  to have the form [10]

$$
I_c \propto (T - T_c)^2 e^{-K d_N} \tag{2}
$$

Here  $K^{-1}$  is the decay length, and  $Kd_N \gg 1$ . A plot of  $\ln I_c$  against  $d_N$  at constant T should be a straight line with a slope of  $-K$ . A series of such plots is shown in Fig. 3, confirming that the thickness dependence of  $I_c$  is indeed exponential. In Fig. 4, we plot the temperature dependence of  $K^{-1}$ . It is observed that the decay length diverges as  $T_{cN}$  is approached from above. For a dirty metal with  $T_{cN} = 0$ , we can identify  $K^{-1}$  with the coherence length in the normal states,  $\xi_N$ , where  $\xi_N$  $= (\hbar D_N/2\pi kT)^{1/2}$ . When  $T_{cN}$  is finite, K is given by the smallest root of the expression [4]

$$
\ln(T/T_{cN}) = \Psi(\frac{1}{2} - \frac{1}{2}\xi_n^2 K^2) - \Psi(\frac{1}{2}).
$$
 (3)

Here  $\Psi$  is the logarithmic derivative of the  $\Gamma$  function. The calculated value of the solution of Eq. (3) at each temperature gives  $K\xi_N$ . Dividing by the measured K, we extracted the values of  $\xi_N$ . We found  $\xi_N = 50 \pm 4$  Å, independent of temperature. This is inconsistent with the dirty-limit form of  $\xi_N$ , which should scale as  $T^{-1/2}$ . A further check of the consistency of the analysis can be made by comparing the mean free path  $l_N$  and  $\xi_N$ . To calculate  $l_N$ , we first write  $D_N = \frac{1}{3} v_{FN} l_N$ , and use our measurements of the conductivity of  $Y_{0.6}Pr_{0.4}BCO$  to



FIG. 3. Thickness dependence of  $I_c$  at three temperatures. Solid lines are fits by an exponential dependence.

estimate  $v_{FN}$ . Below 100 K, the conductivity is  $\sim$  10<sup>3</sup> ( $\Omega$  cm)<sup>-1</sup>, which is an order of magnitude smaller than that of YBCO. Assuming that the decrease of the conductivity relative to YBCO results mainly from a decrease in the carrier density, we obtain  $v_{FN} \sim 1.7 \times 10^7$ cm/sec, which is about half of  $v_F$  for YBCO [12]. Putting this number into the expression for  $\xi_N$ , we get at 70 K  $l_N$  = 260 Å, i.e.,  $\sim$  5 $\xi_N$ . Thus, our material *cannot* be described by the dirty-limit formulas [13]. We now turn to the case of the clean limit.

In the clean limit of the de Gennes theory, there is no expression analogous to Eq. (3) relating the decay length to temperature for  $T_{cN} \neq 0$ . Instead, we shall use the Ginzburg-Landau theory, which is more general than Eq. (3). The clean-limit expression [14] for  $\xi$  in the superconducting phase near  $T_{cN}$  is  $\xi = 0.74\xi_0(1 - T/T_{cN})^{-1/2}$ .



FIG. 4. Temperature dependence of the decay length in  $Y_{0.6}Pr_{0.4}BaCuO$ . The solid line is a fit by the Ginzburg-Landau form near  $T_{cN}$ .

It can be shown that  $K^{-1}$  in the normal phase has the same form as  $\xi$ , except that the numerical coefficient is larger by  $\sqrt{2}$ . The solid line in Fig. 4 shows the fit by this form near  $T_{cN}$ . From the fit, we find  $\xi_0=80\pm25$  Å. The larger error bars reflect the much smaller temperature range over which the fit is made. Since the value of  $\xi_0$  is the low-temperature limit, we can compare it to the decay lengths measured for supercurrents flowing in the a-b plane of junctions made of materials similar to ours. These values are 100 A for PrBCO [15] at 10 K and 50 Å for Nb-doped SrTiO<sub>3</sub> [16] at 4.2 K. All these seem to agree quite well, although our material is metallic while the other two are semiconducting. It is also interesting to compare the measured value of  $\xi_0$  for our material with that obtained from the BCS formula  $\xi_0=0.18(hv_{FN})$  $kT_c$ ). Using  $v_{FN}$  estimated above, we calculate  $\xi_0=55$ A. This value agrees with the experimental one within 50%. We carried out a similar comparison for some well-known superconductors such as Al, Sn, In, Pb, and Nb. The Fermi velocities were obtained using the calculated  $k_F$  divided by the specific-heat effective mass [17]. Experimental values of  $\xi_0$  were taken from Ref. [18]. The spread between the calculated and the experimental values [18] ranged from 20% for Al up to a factor of 2 for Sn, Pb, and Nb. It seems therefore that this rough estimate of  $\xi_0$  is as good for our material as it is for the well-known low- $T_c$  superconductors. In conclusion, we have demonstrated the existence of the proximity effect in SNS junctions made exclusively of high- $T_c$  materials. The effect was observed over a wide range of temperatures relevant, among other things, to the development of high- $T_c$  Josephson junctions [8,9].

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FIG. 1. Critical current vs temperature for an SNS junction with  $d_N = 1250$  Å. Inset: The cross section of the edge junction.