Proximity effect in $YBa_2Cu_3O_{7-\delta}/YBa_2(Cu_{1-x}Co_x)_3O_{7-\delta}/YBa_2Cu_3O_{7-\delta}$ junctions: From the clean limit to the dirty limit with pair breaking

L. Antognazza, S. J. Berkowitz, T. H. Geballe,* and K. Char
Conductus, Inc., Sunnyvale, California 94086
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We report on the proximity effect observed in $YBa_2Cu_3O_{7-\delta}/YBa_2(Cu_{1-x}Co_x)_3O_{7-\delta}/YBa_2Cu_3O_{7-\delta}$ junctions in an edge-junction geometry where $x=0.04$, 0.07, and 0.14. The temperature, thickness, and doping dependence of the critical current of the junctions can be well described by the proximity-effect theory, due to very little interface resistance. We observed a crossover from the clean limit to the dirty limit behavior where a pair-breaking scattering time $\tau_{pb}=6\times10^{-14}$ sec, consistent with short-range magnetic fluctuations in the barrier, was found.

We report a systematic study of superconductor-normal-superconductor (SNS) Josephson behavior involving the high-T_c cuprates. The junctions are found to crossover from the clean limit to the dirty limit where a new pair-breaking scattering appears. The SNS behavior is determined by the proximity effect and this depends upon behavior of Josephson pairs which leak into the barrier material which in turn provides information regarding the interaction potential in the barrier material. Different scattering mechanisms such as elastic scattering which maintains the pairing phase coherence, and inelastic or magnetic scattering which destroys it, can be studied through the proximity effect. In this paper we report the Josephson properties of high-T_c SNS junctions which can be analyzed within the framework of the same de Gennes' proximity theory[1] used for conventional superconductors. Our barrier layers are Co-doped YBCO, whose doping level can be easily controlled, since the interface between YBCO and Co-doped YBCO has been shown to be very clean.[2,3]

In the Josephson junctions which have been used recently[4–6] in important investigations of the wave-function symmetry of $YBa_2Cu_3O_{7-\delta}$ (YBCO), an ambiguity which has not been explicitly addressed is the excess resistance. The origin of the excess resistance is believed to be due to degraded YBCO as result of oxygen deficiency and disorder adjacent to the interface. In order to clarify the exact transport mechanism through such ill-defined YBCO interfaces, it is desirable to investigate Josephson coupling through interfaces that are well defined and can be controlled, as in the present study. Previous studies have been with elements such as Ag and Au,[7] as well as $PrBa_2Cu_3O_{7-\delta}$,[8] $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_{7-\delta}$,[9] $CaRuO_3$,[10] and $SrRuO_3$,[11] all of which, when measured, have had significant interface resistances on the order of 10^{-8}–10^{-7} Ωcm^2. The SNS junctions reported here have two or more orders of magnitude less interface resistance.

Our recent study of edge junctions[3] indicates the mismatch of thermal-expansion coefficients at the interface is an important factor in the excess interface resistance. A matching coefficient of expansion in the c direction can be achieved by using a related metallic layered structure. $PrBa_2Cu_3O_{7-\delta}$ (Ref. 8) and Pr-doped YBCO (Ref. 9) are, of course, such examples. These materials, unfortunately, have other types of disorder that result in barriers that are difficult to quantify. The Pr is believed to hybridize[12] with the oxygen in the CuO_2 planes thus removing carriers but leaving localized states on or adjacent to the CuO_2 planes. The residual transport in the CuO_2 planes of undoped $PrBa_2Cu_3O_{7-\delta}$ is by variable range hopping, an inelastic process which destroys phase coherence. The long-range proximity which is sometimes observed may involve the CuO chains and be very sensitive to small changes in oxygen concentration.[13] The use of Y to dope the barriers into the metallic degraded superconducting state $(Pr_{1-x}Y_x)Ba_2Cu_3O_{7-\delta}$ introduces an intrinsic disorder in the CuO_2 planes.

There are three other strategies for obtaining barriers with the favorable structural and coefficient of expansion features. One is to increase the carrier density and drive YBCO into the overdoped region; Ca-doped YBCO is an example of such overdoped YBCO. The proximity effect using Ca-doped YBCO will be described in a separate paper.[14] A second is to reduce the carrier concentration by cation substitution on lattice sites remote from the CuO_2 planes and thus minimize the introduction of disorder on the CuO_2 planes. This is the strategy employed here. A third way to dope YBCO and lower its T_c is to replace Cu atoms directly on the Cu-O_2 plane layers using, for example, Zn or Ni. It is known[15] that the increased scattering decreases T_c even through the carrier density remains basically the same.

Figure 1 shows the temperature dependence of an 1000-Å-thick epitaxial $YBa_2(Cu_{1-x}Co_x)_3O_{7-\delta}$ (Co-doped YBCO) film on a $LaAlO_3$ substrate deposited by laser ablation. At the low doping level of 4% ($x=0.04$) and 7% ($x=0.07$), the resistivity still extrapolates to near zero value at zero temperature with a slight curvature, suggesting that the increase in resistivity is due to reduction in carrier density rather than increased scattering. This temperature dependence of resistivity is almost identical to that found in Co-doped YBCO single crystals[16] and in oxygen-deficient YBCO single crystals.[17] For the highest

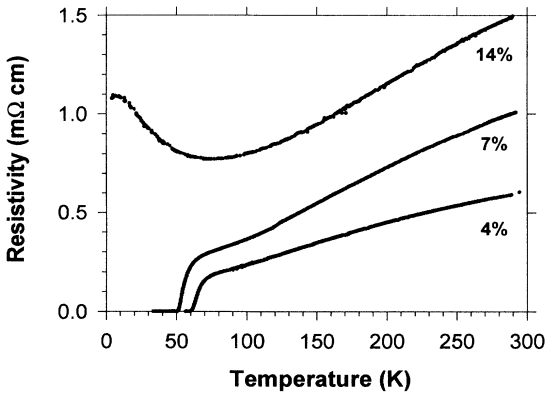

FIG. 1. Resistivity vs temperature curve of three epitaxial Co-doped YBCO films.

doping level of 14%, the high-temperature linear resistivity no longer extrapolates to zero value at zero temperature. For a given composition there is some dependence of T_c and the temperature dependence of the resistivity on the film-growth condition. We have chosen to work with the deposition conditions that generate the best x-ray-diffraction pattern, which also generate the highest resistivity slope for a given nominal composition.

Our junction process is described in detail elsewhere.[10] The thickness of both YBCO layers was between 1500 and 2000 Å, and the width of the junctions is 4 μm. All the junctions were made on $LaAlO_3$ substrates, and the edges were patterned in the twinned (100) or (010) direction. For each doping level and thickness, we have tested ten junctions on two different substrates. The variation of the critical currents was $\pm 25\%$ and the resistances had variations of $\pm 10\%$. For each doping level and thickness, we have selected a junction whose critical current value is in the middle of the distribution and measured its temperature dependence, as reported in Figs. 2, 3, and 4.

The conventional proximity effect given by de Gennes[1] predicts

$$I_c = I_0(1-t)^2 \frac{\kappa d}{\sinh(\kappa d)} = \frac{18.5\text{mV}}{R_n}(1-t)^2 \frac{\kappa d}{\sinh(\kappa d)} , \quad (1)$$

where $t = T/T_c$, $T_c = 88$ K, $I_0 = \pi \Delta_0^2 / 4 R_n k_B T_c = 18.5$ mV$/R_n$ in the case of BCS gap $2\Delta_0 = 3.52 k_B T_c$, d is the thickness of the barrier, and κ^{-1} is the decay length. Even though this relation was derived in the dirty limit near T_c, the thickness, temperature and doping dependence of our data can be surprisingly well described by Eq. (1) with very reasonable microscopic parameters. Since the electronic mismatch between YBCO and Co-doped YBCO is not significant, we used $(1-t)^2$ dependence, assuming continuous boundary conditions of the wave function and its derivative.[1]

The temperature dependence of the critical current and the resistance of 14% Co-doped junctions are shown in Fig. 2 for three different nominal thicknesses of 150, 300, and 450 Å. From the simple exponential relation between the critical current and the thickness of the barrier, $I_c \propto \exp(-\kappa d)$, the decay length κ^{-1} is plotted at several temperatures in the inset of Fig. 2. In order to find out whether the 14% Co-doped YBCO junctions are in the clean limit or in the dirty limit, $\kappa^{-1}(T)$ has been fitted with T^{-1} and $T^{-1/2}$ temperature dependences, since $\kappa^{-1} = v_F \hbar / 2\pi k_B T$ in the clean limit and $\kappa^{-1} = (D\hbar / 2\pi k_B T)^{1/2}$ in the dirty limit. Here v_F is the Fermi velocity and D is the diffusion constant. It is clear the 14% Co-doped YBCO is in the dirty limit as shown in the inset of Fig. 2.

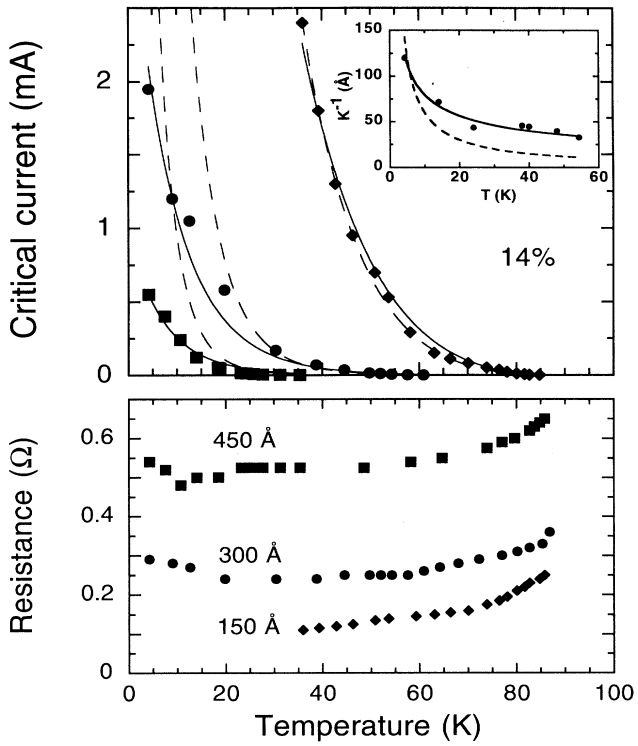

FIG. 2. Temperature dependence of critical currents and resistances of 14% Co-doped YBCO junctions with barrier thickness of 150, 300, and 450 Å. The solid lines are fits to Eq. (1) with a pair-breaking scattering rate of $\tau_{pb} = 6 \times 10^{-14}$ sec. The dotted lines are fits to Eq. (1) without a pair-breaking scattering. The inset shows the decay length κ^{-1} as a function of temperature for 14% Co-doped YBCO junctions. The solid line is a fit to $T^{-1/2}$ dependence and the dotted line is a fit to T^{-1} dependence.

In the dirty limit when the barrier material is superconducting with its transition at T_n, the decay length is expressed by

$$\kappa^{-1} = \left[D \frac{\hbar}{2\pi k_B T} \right]^{1/2} \left[1 + \frac{2}{\ln(T/T_n)} \right]^{1/2} , \quad (2)$$

where the diffusion constant $D = v_F 1/3$. Using $T_n = 0$, and the measured R_n right below T_c,[18] the I_c values in Fig. 2 have been fitted with L being the only parameter where the dimensionless L is defined by $\kappa d = L(T/T_c)^{1/2}$. The dotted lines are such fits with $L = 7.2$, 13, and 16.5, which yields the average diffusion constant $D = 4.1$ cm^2/sec. The scaling of L with the nominal bar-

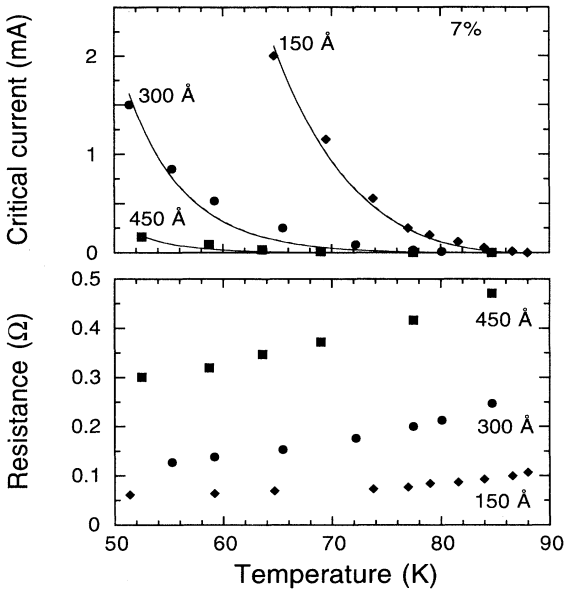

FIG. 3. Temperature dependence of critical currents and resistances of 7% Co-doped YBCO junctions with barrier thickness of 150, 300, and 450 Å. The solid lines are fits to a dirty limit and the overlapping dotted lines are fits to a clean limit.

rier thickness[19] or with the measured resistances is not very good and the fits are poor especially at low temperature. The problem is that the critical currents do not increase as fast as the Eq. (1) predicts at low temperatures, suggesting a cutoff scattering rate of superconducting pairs. This behavior has been observed in low-T_c system[20] when the barrier material has magnetic impurities

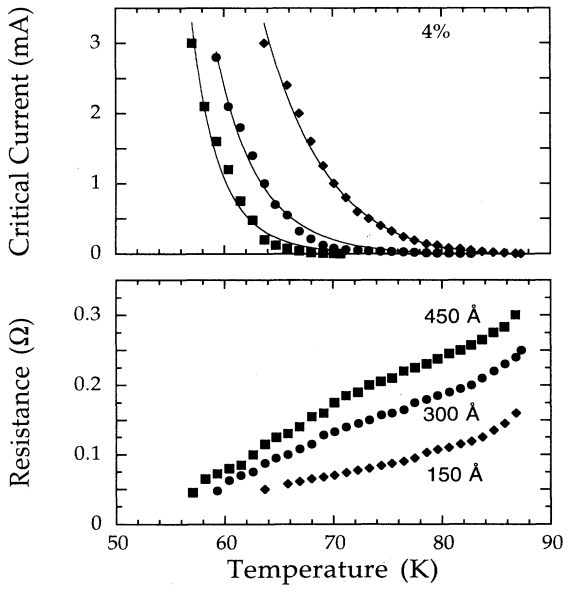

FIG. 4. Temperature dependence of critical currents and resistances of 4% Co-doped YBCO junctions with barrier thickness of 150, 300, and 450 Å. The solid lines are fits to a dirty limit and the overlapping dotted lines are fits to a clean limit.

which destroys superconducting pairs.

When there is a pair-breaking scattering in the barrier material, the lifetime of superconducting pairs inside the barrier is modified. The effective lifetime will be given[21] by

$$\frac{1}{\tau_{\text{eff}}}=\frac{1}{\tau}+\frac{1}{\tau_{\text{pb}}}=\frac{2\pi k_B T}{\hbar}+\frac{2\pi k_B T_{\text{pb}}}{\hbar}=\frac{2\pi k_B (T+T_{\text{pb}})}{\hbar}. \tag{3}$$

The net effect is that T is replaced by $T+T_{\text{pb}}$ in Eq. (2). The fitting to this new temperature with $T_{\text{pb}}=20$ K is shown as solid lines in Fig. 2, with $L=5.9$, 10.5, and 12.2, which yields the diffusion constant $D=6.6$ cm^2/sec with a better scaling with the nominal thickness and with the measured resistances. The data are fit nicely in the 14% Co-doped barrier with a pair-breaking scattering time $\tau_{\text{pb}}=\hbar/2\pi k_B T_{\text{pb}}=6\times10^{-14}$ sec. Since the junctions are well described by the dirty-limit formula, it also suggests that superconducting pairs still experience more weak elastic scattering than pair breaking. This is consistent with the fact that the resistivity of the barrier does not extrapolate to zero at zero temperature as shown in Fig. 1. The origins of the pair-breaking scattering is not obvious. It could simply be that the 42% (three times 14%) doping on the chains induces enough disorder to cause pair breaking by strong elastic scattering. Alternatively, the spin-glass state of underdoped nonsuperconducting YBCO can result in magnetic pair breaking on the CuO_2 planes. We rule out the possibility that segregated cobalt oxide may cause the pair breaking, since x-ray-absorption fine structure[22] (XAFS) on Co-doped YBCO did not show evidence of cobalt oxide segregation. We also point out that the slowdown of the critical current at low temperature is not due to the critical current saturation observed for large junctions of in-line geometries, since the critical current saturation is not expected for the overlap geometry[23] of our edge junctions. This is further confirmed by the good fit up to the 2.5-mA current level for the 150-Å-thick barrier junction and also by the fact that the 450-Å-thick barrier junction is not even in the large junction limit as its critical current is small.

The temperature dependence of critical currents and resistances of the 7 and 4 % Co-doped YBCO junctions are shown in Figs. 3 and 4, respectively. Initially, we analyzed the data in terms of dirty-limit formulas given by Eq. (2). The fits are shown in the figures as solid lines. For the 7% doping, $T_n=40$ K was used[24] for the fitting and L was found to be 10, 16.5, and 23 where

$$\kappa d=L\left[\frac{T}{T_c}\right]^{1/2}\left[1+\frac{2}{\ln(T/T_n)}\right]^{-1/2}.$$

For the 4% doping, L was found to be 12, 17.2, and 22.5 with $T_n=55$ K.[24] From the fits the average diffusion constants were found to be $D=2.2$ cm^2/sec for 7% and $D=2$ cm^2/sec for 4%. These values are unphysical when compared to $D=6.6$ cm^2/sec found for 14% Co-doped barrier, since the diffusion constant should increase as the doping level decreases, according to $D=v_F l/3$. This

suggests that the decay length of 7 and 4% Co-doped material is not determined by the diffusion constant, and thus that these materials are in the clean limit.

In the clean limit, according to the Ginzburg Landau theory, the decay length of a superconductor above its critical temperature T_n is given by[9] $\kappa^{-1}=\sqrt{2}\times 0.74\xi_0(T/T_n-1)^{-1/2}$, where the BCS coherence is given by $\xi_0=\hbar v_F/\pi\Delta$. The fits to this relation are shown as the dotted lines in Figs. 3 and 4. For 7% doping, L was found to be 4.8, 7.9, and 11 where $\kappa d=L(T/T_n-1)^{1/2}$ and $T_n=40$ K was used. In case of 4% doping, L was found to be 6.7, 9.6, and 12.6 with $T_n=55$ K. The fits are overlapping with the solid lines given by the dirty-limit formula. This is not surprising since it is easy to show that the Eq. (2) can be modified to $\kappa^{-1}\propto(T/T_n-1)^{-1/2}$ when T is near T_n, where even the dirty-limit formula is supposed to follow the Ginzburg-Landau relation. However, the average BCS coherence lengths we obtain from the fitting with the clean limits, 35 Å for 7% Co-doped YBCO and 28 Å for 4% Co-doped YBCO, are consistent with the BCS formula $\xi_0=\hbar v_F/\pi\Delta$. The small increase with doping follows from Fermi velocity, not decreasing as much as the critical temperature when the doping increases. We take the above analysis as strong evidence that the 4 and 7% Co-doped YBCO are indeed in the clean limit.

YBCO itself is known[25] to be in the clean limit. As the Co doping increases, the carrier density and the critical temperature decrease, increasing the BCS coherence length according to $\xi_0=\hbar v_F/\pi\Delta$, as long as Co-doped YBCO remains in the clean limit. As the doping level is further increased beyond 7%, the decreasing mean free path eventually becomes smaller than the increasing clean-limit coherence length, causing a crossover to the dirty limit. In this regime the coherence length decreases with increased doping and both elastic scattering and pair-breaking scattering are required to fit the data.

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*Also at the Dept. of Applied Physics, Stanford University, Stanford, CA 94305.

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[24]T_n was found lower than the superconducting transition temperature of a 1000-Å-thick bulk film measured without patterning. It is not unreasonable to assume that thinner films grown on the edges of YBCO patterned into a 4-μm-wide bridge may have a lower T_n, especially given the fact that T_n can change as much as 10 K even in a bulk film depending on the deposition conditions. A slightly different value may be used with reasonable fits to data. However the change in D value due to different T_n value does not affect the systematic conclusions we have found.
[25]See, for example, K. Kamáras *et al.*, Phys. Rev. Lett. **64**, 84 (1990).