Observation of Josephson Pair Tunneling between a High-T_c Cuprate ($YBa_2Cu_3O_{7-\delta}$) and a Conventional Superconductor (Pb)

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Josephson tunneling currents have been observed on low leakage Pb/insulator/$Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ *tunnel* junctions. The junctions are planar with tunneling into the c axis. We have studied these Josephson currents on junctions with resistances varying by more than 2 orders of magnitude. From the magnetic field modulation of the Josephson current, the low temperature value and the temperature dependence from 1.3 to 4.2 K of the c-axis penetration depth were measured. This observation of Josephson coupling along the c axis between a conventional superconductor (Pb) and a cuprate raises questions about the proposed d-wave nature of the superconducting order parameter in the cuprates.

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The symmetry of the superconducting order parameter in the high-T_c cuprates continues to be an unresolved issue. Theoretical arguments proposing a $d_{x^2-y^2}$ symmetry [1,2] have been put forth as have counter arguments [3,4] suggesting it to be an anisotropic s-wave symmetry. An increasing number of experiments have attempted to address this problem by trying to directly probe this symmetry using various techniques [5–9]. Two recent Josephson weak link experiments using different techniques have come to opposite conclusions [8,9]; one supporting the picture of a d-wave symmetry, the other concluding an s-wave symmetry. In this paper we report our results of studies of Josephson *tunneling* between a conventional superconductor (Pb) and a series of high-T_c cuprates ($YBa_2Cu_3O_{7-\delta}$ and the alloys $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$). Using a well characterized tunnel junction [as opposed to a superconductor-normal-superconductor (SNS) or weak link] between Pb and the cuprates, we have reproducibly observed well defined Josephson coupling for tunneling along the c axis. In addition, using the magnetic field variation of the Josephson current $I_c(B)$ we have measured the penetration depth in the c axis (usually labeled λ_{ab} because it entails screening currents in the a-b plane). We have measured the absolute value of λ_{ab} and the temperature dependence over a limited low temperature range (1.3 K$\rightarrow$4.2 K). We argue that these results are difficult to understand in the context of a d-wave symmetry.

The crystals were grown in a fashion described previously [10]. They were then annealed in oxygen for a period of $\sim$1 week. Without this anneal, the superconducting properties were inferior, and the results were not reproducible. The junctions are fabricated on the broad surface perpendicular to the c axis of single crystals of $YBa_2Cu_3O_{7-\delta}$ and $Pr_xY_{1-x}Ba_2Cu_3O_7$ utilizing a natural insulating barrier. We chose single crystals that have large, smooth areas (as determined with a Nemarsky microscope) and that are free of steps larger than 25 Å. For the most part, we used a Br_2 etch technique to clean the single crystal surface [11] and a 1-μm-thick Pb film was then evaporated through a shadow mask.

Early in these studies, in a junction made on a crystal with 20% Pr doping, with resistance of 22.5 Ω, a Josephson current of 1.75 μA was observed. The junction showed very low leakage for voltages less than the Pb energy gap Δ_{Pb}. A hysteretic I-V with a clear Pb gap was observed, as were the Fiske ac Josephson resonance modes upon applying a magnetic field. The Josephson current disappeared at a magnetic field of 0.3 G and only the central lobe of the expected Fraunhofer pattern was observed due to the fact that for high resistance junctions, the Josephson effect is noise limited [12]. This observation was reproduced on another superconductor-insulator-superconductor (SIS) junction of resistance 12.5 Ω made on a pure $YBa_2Cu_3O_{7-\delta}$ single crystal.

In an effort to reduce the junction resistance, we adopted the technique of using Ag as a diffusion barrier, since we know that the junction resistance increases *significantly* through prolonged diffusion of the counterelectrode Pb into $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ crystals and subsequent oxidation. By evaporating about 10 Å of Ag on the crystal after the Br_2 etch through a shadow mask, and then without breaking vacuum, evaporating the counterelectrode Pb over the Ag stripe through the same mask, we were able to reduce the junction resistance by 2 orders of magnitude. The quality of the junctions is not affected by the thin Ag layer, as demonstrated by the observation of the full opening of the Pb gap, the hysteretic I-V characteristics, and the high Q Fiske modes. The Ag is too thin to affect the superconducting properties of Pb, as proved by the appearance of the Josephson current (Fig. 2, inset) at the bulk Pb T_c of 7.19 K. The picture we have at the junction is that the Ag layer sits atop the natural insulating barrier formed on the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ surface. While we do not have any direct evidence, we believe the Ag layer is of uniform thickness because of its effectiveness as a diffusion barrier at a thickness of only 10 Å. We have fabricated twenty such junctions to date and the results shown here are typical and representative.

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In Fig. 1 we show the I-V characteristics of a $YBa_2Cu_3O_{7-\delta}$/Pb tunnel junction at 1.3 K. The inset illustrates the geometry of the junctions studied. At higher voltages this junction has very similar characteristics to those reported earlier [11,13]. The "gaplike" structure and linear background conductance associated with the cuprate are reproduced. We focus here on the low voltage regime. The superconducting Pb energy gap (Δ_{Pb}) is clearly visible at a voltage $V \approx 1.4$ meV. Tunneling is the only conduction mechanism as is evidenced by the extremely low (< 1 part in 10^4) leakage current below the Pb gap. All of this (as well as the clear observation of the Pb phonons of the correct strength and energy) is convincing evidence that elastic tunneling (where transverse momentum of the electrons is conserved) is the conduction mechanism. The Josephson current at $V=0$ is then a tunneling current which couples the pair wave function in the Pb to that in the $YBa_2Cu_3O_{7-\delta}$ via an elastic, single step, transverse momentum conserving tunneling process. This observation of Josephson coupling between the pair states is difficult to understand if the $YBa_2Cu_3O_{7-\delta}$ is a superconductor with strictly $d_{x^2-y^2}$ symmetry as it should be orthogonal to the s-wave symmetry of the Pb.

$YBa_2Cu_3O_{7-\delta}$ is orthorhombic and not tetragonal and so perhaps this coupling is due to the in-plane distortion of the possible $d_{x^2-y^2}$ symmetry. We do not think this is an explanation as the crystals studied in this work are heavily twinned (10^3-10^4 twins in the area of the junction) and the average over all these twins should yield a zero critical current. The etching process used during junction preparation eventually leaves etch pits in the junction area. We have studied the etch rates and have varied the amount etched from ≈ 100 Å up to several thousand angstroms. After studying the surface morphology under a Nemarsky microscope, and with various degrees of etching, we have made a variety of junctions from those with $\gg 99\%$ of its area perpendicular to the c axis (no etch pits or steps visible) to those of $\geq 95\%$ for a heavy density of pits (or $\leq 5\%$ of the junction area being the walls of the pits). We realize that the presence of the etch pits can give rise to tunneling channels along the a and b directions but in the course of this work we have not observed any systematic variation of the Josephson coupling due to the density of the etch pits. In fact, because of the long range phase coherence of a superconductor, it is not even clear whether it matters if there are tunneling channels in the a-b plane. A strictly d-wave superconductor would still result in no Josephson coupling to the Pb.

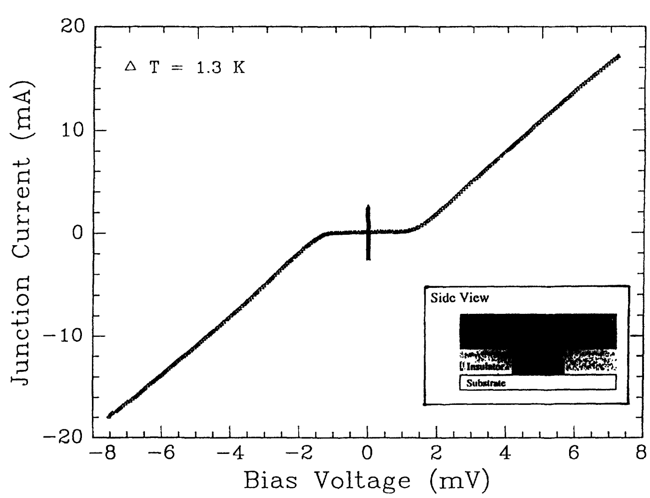

FIG. 1. Typical I-V characteristics for the Pb/$YBa_2Cu_3O_{7-\delta}$ tunnel junctions studied in this work. The Josephson pair tunneling current is at zero bias. The Pb energy gap is at ± 1.4 mV. Inset: Geometry of the tunnel structure.

In Fig. 2 we show the variation of the critical current with applied magnetic field B in the plane of the junction $I_c(B)$ for a $YBa_2Cu_3O_{7-\delta}$/Pb junction at 1.3 K. The familiar Fraunhofer pattern is observed in this case. This is typical and similar curves have been seen for both pure $YBa_2Cu_3O_{7-\delta}$ and $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples. The systematic behavior of the $I_c(B)$ patterns gives us additional confidence in the results reported in this paper. In addition, at the nodes of the $I_c(B)$ pattern (where $I_c \to 0$) the I-V characteristics reveal Fiske [14] resonances at finite voltage. These are resonances of the ac Josephson effect with the cavity defined by the junction geometry. These resonances are very sharp (instrumentally limited) and so have a very high Q suggesting very little damping. This also gives us confidence in the integrity of the tunnel junction and very low losses implying superconducting screening up to the surfaces (low surface losses).

In the inset of Fig. 2 we show the variation of I_c as a function of T. As expected, I_c disappears at the T_c of Pb and the behavior of this temperature dependence again is evidence for "conventional" Josephson behavior. This behavior follows that expected for a Josephson junction of two dissimilar superconductors. The $I_c(B)$ pattern of Fig. 2 shows that the Josephson current is uniformly distributed over the junction area, and we can, from the periodicity of the pattern, extract a measurement of the penetration depth λ_{ab} in the cuprate. Each of the nodes corresponds to an integral number of flux quanta enclosed in the geometry defined by the junction width W in one direction and the sum of the penetration depths in Pb and $YBa_2Cu_3O_{7-\delta}$ as well as the barrier thickness in the other direction. The critical current I_c is given by $I_c = I_0 |(\sin\pi\phi/\phi_0)/(\pi\phi/\phi_0)|$, where I_0 is the maximum Josephson current, ϕ_0 is the flux quantum, and ϕ is the flux enclosed in the area $[W(\lambda_{Pb}+\lambda_{ab}+t)]$. The barrier thickness t is ≈ 20 Å; the junction width W is 0.225 mm as defined by a shadow mask. We independently measured λ_{Pb} using this same Josephson technique $I_c(B)$ on a Pb-insulator-Pb junction. We can then extract an accurate measure of the penetration depth in the cuprates by measuring $\lambda_{Pb}(T)$ and subtracting this from our mea-

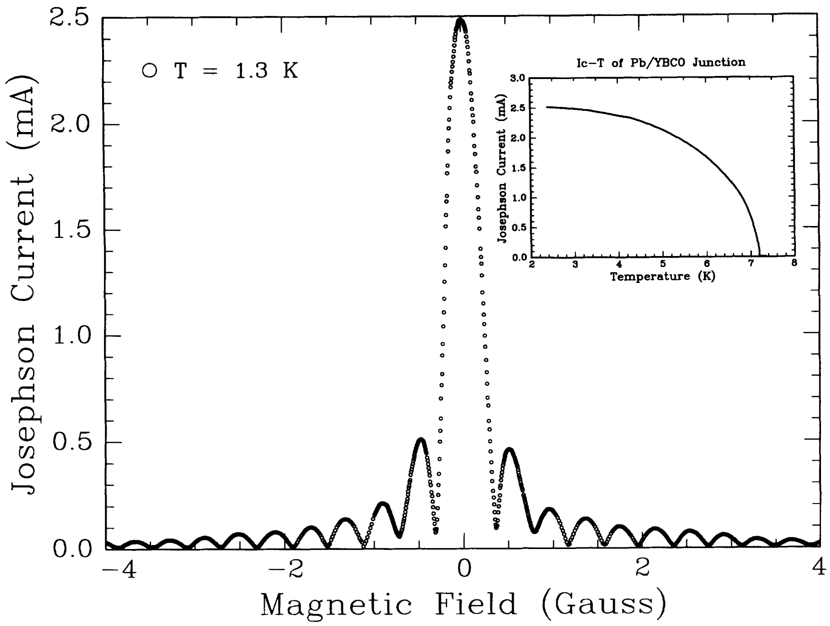

FIG. 2. $I_c(B)$ for a Pb/$YBa_2Cu_3O_{7-\delta}$ Josephson tunnel junction. Inset: $I_c(T)$ for $B=0$ for this junction.

sured value of $\lambda_{ab}+\lambda_{Pb}+t$. The results of this analysis are shown in Fig. 3 where we plot $\lambda_{ab}(T)$. The order of magnitude of the temperature dependence is in agreement with previous measurements [15,16]. It has been argued that in "clean" samples the temperature dependence of $\Delta\lambda(T)$ follows a linear T dependence [16] and crosses over to T^2 as a result of increased scattering. We choose not to "interpret" our data in terms of a T or T^2 dependence but simply present it here. In an attempt to explore the effects of scattering, we have compared $\lambda(T)$ of $YBa_2Cu_3O_{7-\delta}$ [Fig. 3(a)] with that of the alloy $Y_{0.9}Pr_{0.1}Ba_2Cu_3O_{7-\delta}$ [Fig. 3(b)]. We see very little difference between the cases of $x=0$ and $x=0.1$ except in the I_cR product discussed below. $\lambda(0)$ is the same to within ≈ 20 Å and the temperature dependences are almost identical. We see no real differences in the values determined for λ. In the samples where we have performed extensive $I_c(B)$ measurements we see remarkable reproducibility with a variation of only about 20 Å.

The observation of Josephson coupling between Pb and the cuprate raises questions about whether this excludes the possibility of a $d_{x^2-y^2}$ symmetry order parameter in $YBa_2Cu_3O_{7-\delta}$ and the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ alloys. An important quantity in Josephson tunnel studies is the I_cR product. For conventional SIS tunnel junctions, Ambegaokar and Baratoff [17] showed that at zero temperature the product $I_cR=\pi\Delta/2e$ for BCS superconductors with energy gaps Δ on each side of the junction. It has also been shown that strong coupling reduces this product in the case of Pb by $\approx 20\%$. For a Josephson junction with dissimilar superconductors SIS; the zero temperature I_cR product in the weak coupling limit is given by

$$I_cR=\frac{\pi\Delta_1\Delta_2}{\Delta_1+\Delta_2}K\left(\left|\frac{\Delta_1-\Delta_2}{\Delta_1+\Delta_2}\right|\right),$$

where K is a complete elliptic integral [18]. If we choose for Pb the measured energy gap of $\Delta_{Pb}=1.4$ meV and for the $YBa_2Cu_3O_{7-\delta}$ a BCS $(2\Delta/kT_c)$ energy gap of 14.0 meV, we obtain an Ambegaokar-Baratoff weak coupling value of $I_cR=8.0$ meV. Strong coupling corrections will

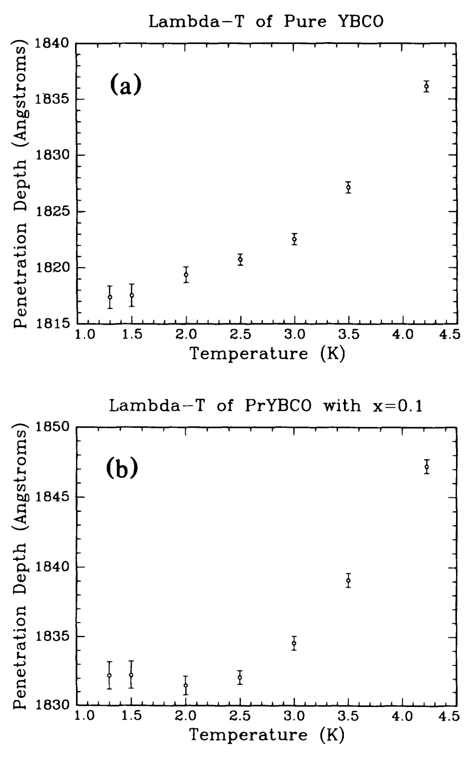

FIG. 3. (a) $\lambda_{ab}(T)$ determined by techniques described in the text for $YBa_2Cu_3O_{7-\delta}$. (b) $\lambda_{ab}(T)$ for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ with $x=0.1$. Note the similarity with (a).

renormalize this value lower (at least 20%), but we do not have a good estimate of the strong coupling correction to $YBa_2Cu_3O_{7-\delta}$. In the measurements over the series of junctions in this work, we have determined I_cR values from ~0.3 mV up to 0.9 mV, the highest values in the case of pure $YBa_2Cu_3O_{7-\delta}$ and then systematically decreasing with increasing Pr doping. While there is some scatter in the variation of I_cR with Pr concentration it is quite clear that it is maximized for the clean limit $x=0$. We emphasize this point because it illustrates a behavior which is counter to that expected if the explanation for this Josephson current is based on elastic scattering in the cuprate. It could be argued that elastic scattering might break the $d_{x^2-y^2}$ symmetry and allow a small coupling of the pair wave function from Pb. However, with increasing scattering [13] the trend is in the opposite direction; the I_cR product decreases. Nevertheless, it is a concern that for $YBa_2Cu_3O_{7-\delta}$, the highest I_cR product we have measured is 0.9 meV and the expected value for BCS superconductors is substantially higher. We note, however, that our tunneling measurements have *consistently* shown $YBa_2Cu_3O_{7-\delta}$ to be gapless for tunneling into the c axis [13] and so applying the Ambegaokar-Baratoff limit for I_cR is most likely incorrect. With gapless superconductivity we would expect a substantially reduced I_cR product and we are not aware of any extensive studies, either theoretical or experimental, to investigate this limit.

Because of the measurably high quality of the tunnel junctions fabricated, the systematics of the $I_c(B)$ pattern, the high Q of the Fiske modes, and the conventional behavior of $I_c(T)$, we argue that these junctions allow a critical probe of the pair state in the cuprate. Unlike SNS, step edge, weak link, and the various other nontunneling Josephson devices, the pair transfer across the tunnel barrier is elastic, with transverse momentum conserved. If the cuprate superconducting order parameter is of $d_{x^2-y^2}$ symmetry, the finite Josephson current into the s-wave superconductor Pb must be explained.

In summary, we have fabricated a substantial number of Josephson tunnel junctions between the conventional superconductor Pb and the high-T_c $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$. The Josephson current has many aspects of a conventional SIS′ tunnel junction (where S′ can be a different superconductor than S). The $I_c(B)$ Fraunhofer pattern is observed as is the ac effect. From the $I_c(B)$ dependences, we have measured the penetration depth $\lambda_{ab}(T)$ to an accuracy of a few angstroms. We see a temperature dependence consistent with earlier microwave measurements for both "clean" $YBa_2Cu_3O_{7-\delta}$ and "dirty" $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$. We find the results difficult to resolve in the sample framework of pair tunneling from an s to a d symmetry superconductor.

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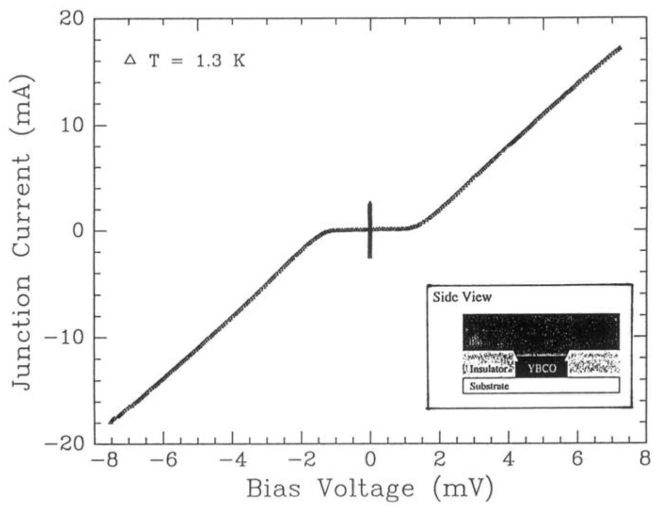

FIG. 1. Typical *I*-*V* characteristics for the Pb/$YBa_2Cu_3O_{7-\delta}$ tunnel junctions studied in this work. The Josephson pair tunneling current is at zero bias. The Pb energy gap is at ±1.4 mV. Inset: Geometry of the tunnel structure.