# Pair Tunneling from $c$-Axis $YBa_2Cu_3O_{7-x}$ to Pb: Evidence for $s$-Wave Component from Microwave Induced Steps

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(Received 13 November 1995)

In heavily twinned crystals or films of $YBa_2Cu_3O_{7-x}$ (YBCO), $d_{x^2-y^2}$ pairing symmetry is expected to cause the cancellation of first-order Josephson tunneling through a YBCO-Pb tunnel junction grown on an $a$-$b$ face; any residual tunneling is thus second order. As a result, microwaves at frequency $f$ are predicted to induce steps on the current-voltage characteristic at voltages that are multiples of $\frac{1}{2}(hf/2e)$. Experimentally, steps are observed only at multiples of $hf/2e$, suggesting that $s$-wave pairing is present in YBCO; however, the simultaneous presence of $d$-wave pairing is by no means ruled out.

PACS numbers: 74.50.+r, 74.72.Bk

The symmetry of the order parameter in the high transition temperature ($T_c$) oxide superconductors is the subject of ongoing debate. One school of thought proposes a $d_{x^2-y^2}$ symmetry (for example, [1,2]) while another proposes an anisotropic $s$-wave symmetry (for example, [3,4]). A series of experiments [5–8] has been performed on Josephson junctions made on the $a$-$b$ faces of $YBa_2Cu_3O_{7-x}$ (YBCO) crystals or films, with conventional low-$T_c$ superconductors as the counterelectrodes. These measurements provide evidence for a phase shift of $\pi$ between the order parameters measured by transport of pairs across weak links along two orthogonal directions, building a strong case for $d$-wave pairing. Sun *et al.* [9], however, performed extensive measurements of tunneling along the $c$ axis of YBCO and the alloys $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$, using an insulating barrier and Pb as the counterelectrode. At temperatures near 1 K the current-voltage ($I$-$V$) characteristics exhibited a Josephson supercurrent at zero voltage, and very small single-particle tunneling currents below the Pb gap. The application of a magnetic field parallel to the plane of the junction produced a well-defined Fraunhofer pattern in the critical current, and yielded values of the penetration depth $\lambda_{ab}(T)$ with a temperature dependence consistent with those obtained from earlier microwave measurements [10,11].

In a single crystal with tetragonal symmetry and purely $d_{x^2-y^2}$ pairing, the first-order pair tunneling currents cancel along the $c$-axis direction. Any pair tunneling would have to be second order, and the current thus very weak. In an untwinned single crystal of YBCO, however, the orthorhombic crystal symmetry leads to asymmetric $d$-wave pairing so that the first-order $c$-axis pair tunneling is not expected to vanish. The situation is different yet again in a heavily twinned single crystal or thin film. Here, the phase of the order parameter is maintained throughout the sample, but the amplitude variation along the $a$ and $b$ axes is expected to ensure that the net first-order tunneling current in the $c$-axis direction is zero. We note that any tunneling transitions in the $a$-$b$ directions would contribute only to second-order supercurrents; again, first-order components cancel by symmetry. Since the $c$-axis tunneling experiments demonstrating the existence of a nonzero Josephson supercurrent [9] were performed mostly on twinned samples, the issue of whether the tunneling is first or second order needs to be addressed.

Tanaka [12] showed that for pure $d$-wave to $s$-wave tunneling a nonzero supercurrent due to second-order processes can flow in the $c$ direction with a current-phase relation of the ($\pi$-periodic) form $I = I_0 \sin 2\delta$; $I_0$ is the critical current, and $\delta$ the difference between the phase of the $s$-wave order parameter and the phase of the $d$-wave order parameter averaged over the azimuthal angle. Setting $\hbar d\delta/dt = 2$ eV, we can write the Josephson tunneling current in the form

$$I(t) = I_0 \sin\left[2\pi(4e/h)\int_0^t V(t')dt'\right]. \qquad (1)$$

Equation (1) implies that applied microwaves of frequency $f$ will induce constant voltage steps at voltages $(n/2)f\Phi_0$, where $n$ is an integer and $\Phi_0 = h/2e$, as opposed to the conventional case, where the steps are at voltages $nf\Phi_0$. A similar prediction has been made by Zhang [13] for in-plane Josephson junctions between $d_{x^2-y^2}$ superconductors with their axes rotated to 45° to each other. In both cases, second-order tunneling is predicted to lead to microwave-induced Shapiro steps at one-half the voltage interval of first-order tunneling. We note that trapped flux or the presence of external magnetic fields may modify the height of integer steps but does not introduce half-integer steps [14]. However, caution in interpreting the experimental data is necessary. For example, if the microwave frequency coincides with a self-resonant frequency of the junction [14], odd-numbered steps can be suppressed or subharmonics introduced; if the junction parameters are within certain

0031-9007/96/76(12)/2161(4)$10.00  

TABLE I. Critical current, normal state resistance, and upper limit for second-order contribution of ten $c$-axis YBCO/Pb tunnel junctions.

| No. | Sample | $I_c$ ($\mu$A) | $R_N$ ($\Omega$) | $\alpha$ (%) |
|---|---|---|---|---|
| 1 | Detwinned crystal | 60 | 22.0 | 0.5 |
| 2 | Detwinned crystal | 80 | 13.0 | 0.5 |
| 3 | Detwinned crystal | 80 | 13.5 | 1.0 |
| 4 | Twinned crystal | 800 | 0.4 | 2.5 |
| 5 | Twinned crystal | 425 | 0.5 | 2.0 |
| 6 | Twinned crystal | 45 | 2.0 | 5.0 |
| 7 | $LaAlO_3$ | 190 | 0.025 | 1.5 |
| 8 | $LaAlO_3$ | 350 | 0.02 | 2.0 |
| 9 | $LaAlO_3$ | 460 | 0.045 | 1.5 |
| 10 | $LaAlO_3$ | 520 | 0.04 | 1.5 |

regimes [15], even first-order tunneling can produce half-integer steps. In this Letter we report experiments in which we irradiate a series of $c$-axis YBCO-insulator-Pb tunnel junctions with microwaves in order to distinguish between first-order and second-order tunneling.

Josephson junctions were fabricated at UCSD on the $a$-$b$ plane of both detwinned and twinned single crystals of YBCO ($>10^3$ twins per junction) and on heavily twinned $c$-axis epitaxial YBCO films ($>10^5$ twins per junction). Films were grown by pulsed laser ablation at Conductus, Inc. on yttrium stabilized zirconia (YSZ) substrates, and at UCB and UCSD on $LaAlO_3$ substrates. The transition temperatures were between 88 and 91 K. Details of the junction fabrication techniques have been published previously [9]. We investigated a total of 19 junctions, with typical areas of $225 \times 400$ $\mu m^2$. We measured the dependence of the critical current on a magnetic field applied parallel to the plane of the junction, and obtained $I$-$V$ characteristics in the presence of 2–18 GHz microwave fields. We coupled the microwaves into the junction via a dipole antenna at the end of a semirigid coaxial cable; the maximum input power was 20 mW. We obtained data at temperatures between 13 and 4.2 K at both UCSD and UCB in ambient magnetic fields below 1 $\mu$T. The transportation to UCB involved two additional thermal cyclings of the junctions and a resulting reduction in critical current, but the microwave data obtained in the two laboratories were not materially different.

The parameters of a representative set of 10 junctions are listed in Table I. The values of $I_0 R_N$ were typically 1 mV for junctions on untwinned crystals, 300 $\mu$V for twinned crystals, and 10 $\mu$V for thin films. At 1.35 K the resistance $R_g$ below the lead gap voltage $\Delta_{Pb}/e = 1.4$ mV was usually more than $10^4$ times the resistance $R_N$ above the gap. At 4.2 K, $R_g$ decreased to typically $(5-10)R_N$. Figure 1(a) shows a representative $I$-$V$ characteristic. This junction, like most in this study, exhibited self-resonant steps (not visible in the figure) [16] in nonzero magnetic fields, indicating that the junction forms a relatively high-$Q$ cavity. Figure 1(b) shows the dependence of $I_0$ on applied magnetic field, and provides evidence for a uniform tunnel barrier and the absence of trapped magnetic flux. All the junctions listed in Table I showed comparable behavior.

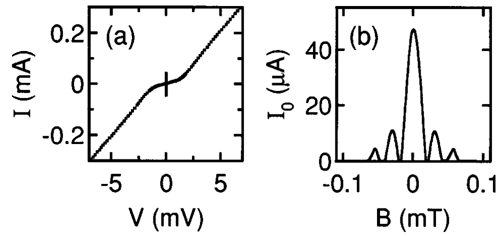


FIG. 1. Sample 1 (detwinned crystal) at 4.2 K: (a) $I$-$V$ characteristic, (b) $I_0$ vs magnetic field applied parallel to plane of junction.

Figure 2 is an oscilloscope trace of the $I$-$V$ characteristic of junction 5 at 4.2 K in a 10.2 GHz microwave field. The figure clearly shows steps at voltages $nf\Phi_0$ but none at $(n + 1/2)f\Phi_0$. The steps overlap strongly and even cross the voltage axis [15]. Since the junction was current biased, only some of the possible Shapiro steps can be traced out during each cycle of the current sweep. However, after many cycles (we used up to 1000 cycles for one picture), random switching ensured that all steps were included, even if their amplitude was much smaller than that of their neighbors. Like all data shown in the paper, Fig. 2 was obtained in nominally zero magnetic field; when we varied the field, the step amplitudes were modulated, but no additional steps appeared. We thus draw the qualitative conclusion that the $c$-axis supercurrents are *predominantly* of *first* order.

We now set a quantitative upper limit on the second-order contributions by investigating the step height $2I_n$ of the steps as a function of microwave power. In the limit $f_p \ll f$, where $f_p = (I_0/\Phi_0 C)^{1/2}$ is the Josephson plasma frequency and $C$ is the junction capacitance, the step height is well described by $I_n = I_0|J_n(\kappa V_m/f\Phi_0)|$,

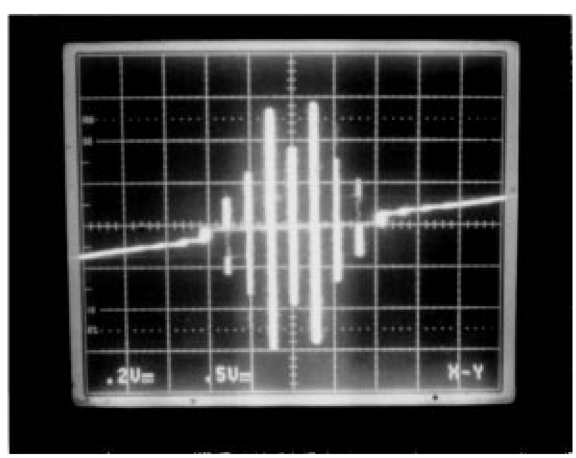


FIG. 2. $I$-$V$ characteristic of sample 5 (twinned crystal) at 4.2 K showing 10.2 GHz microwave-induced steps at multiples of $f\Phi_0$. Horizontal axis is 40 $\mu$V/division, vertical axis is 100 $\mu$A/division.

where $V_m$ is the microwave-induced voltage across the junction and $J_n$ is the $n$th-order Bessel function [14]. For first-order Josephson currents, $\kappa = 1$, while for second order one can easily show that $\kappa = 2$. We estimate that $f_p$ is between 1 and 3 GHz for our junctions. In the presence of both first- and second-order currents we may write the total Josephson current as $I = I_0[(1 - \alpha)\sin\delta + \alpha\sin(2\delta + \phi)]$; $\alpha = 0$ corresponds to purely first-order tunneling, and $\alpha = 1$ to purely second-order tunneling. The phase shift $\phi$ may be 0 or $\pi$, corresponding to an admixture $s \pm d$, or $\pi/2$ or $3\pi/2$, corresponding to an admixture ($s \pm id$). One can show that the height of the step at $V_n = nf\Phi_0$ is

$$I_n = I_0 \max[(1 - \alpha)J_n(V_m/f\Phi_0)\sin\delta_0 + \alpha J_{2n}(2V_m/f\Phi_0)\sin(2\delta_0 + \phi)], \quad (2)$$

where we have to maximize $I_n$ with respect to $\delta_0$. The results for $n = 0$, 1, and 2 are shown in Figs. 3(a)–3(c) for $\alpha = 0$ (solid lines), and for $\alpha = 0.2$ for the cases $s \pm d$ (dotted lines) and $s \pm id$ (dashed lines). For $\alpha = 0.2$ the results for the $s \pm d$ cases are virtually indistinguishable from $\alpha = 0$, whereas for the $s \pm id$ case the results differ significantly from $\alpha = 0$.

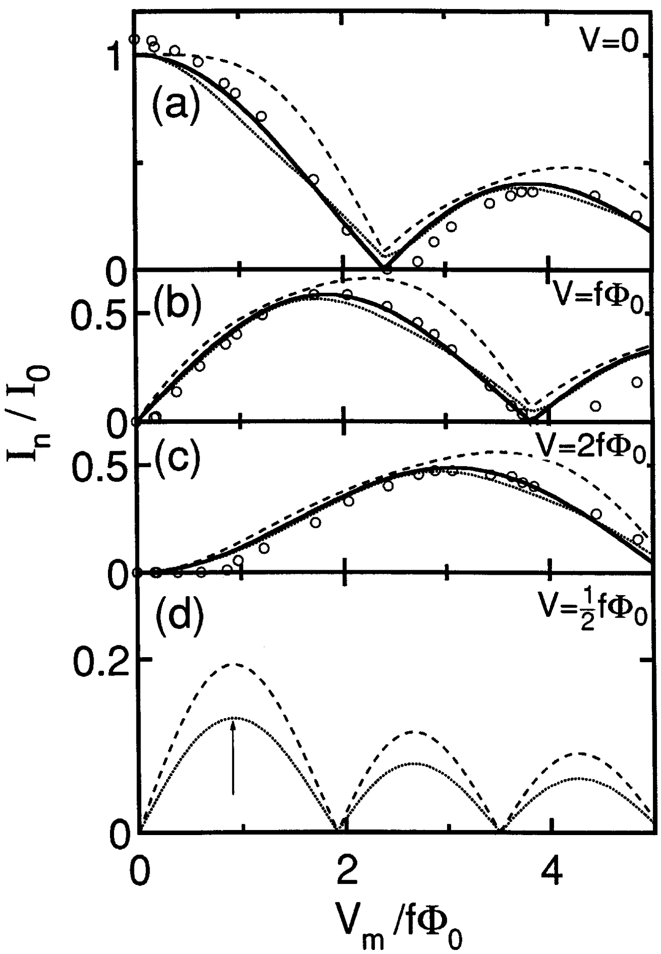


FIG. 3. Solid lines in (a)–(c) are computed step amplitudes for first-order tunneling ($\alpha = 0$), dotted lines are for $s \pm d$ with $\alpha = 0.2$, and dashed lines for are for $s \pm id$ with $\alpha = 0.2$. Open circles are measured step amplitudes $I_n$ at (a) $V = 0$, (b) $V = f\Phi_0$, (c) $V = 2f\Phi_0$ for sample 1 at 4.2 K and 11.5 GHz; data are fitted at first maximum of $J_1$. (d) Amplitude of steps at $V \pm \frac{1}{2}f\Phi_0$, with the same notation and $\alpha = 0.2$. Arrow indicates value of $V_m/f\Phi_0$ at which these steps have their first maximum.

The height of the steps at $V_n = (n + 1/2)f\Phi_0$ is given by

$$I_{n+1/2} = \alpha I_0 |J_{2n+1}(2V_m/f\Phi_0)|. \quad (3)$$

Figure 3(d) shows $I_{1/2}$ ($n = 0$) for $\alpha = 0.2$ for the $s \pm d$ case (dotted line) and $s \pm id$ case (dashed line). In Fig. 3(d) we assume that there are no subharmonic contributions from the first-order Josephson currents; this assumption is well satisfied in the limit $f_p \ll f$. Figure 3(d) shows that $I_{1/2}$ has a maximum when $V_m/f\Phi_0 \approx 0.9$; at this value of $V_m$, $I_1$ and $I_2$ are about $0.5I_0$ and $0.1I_0$, respectively. In Figs. 3(a)–3(c) we also show measured values of $I_0$, $I_1$, and $I_2$ for sample 1 at 4.2 K and 11.5 GHz. The data are fitted at the first maximum of $J_1$. We obtained similar data for all the other junctions. Given the somewhat different shape of the curves for the $s \pm id$ case, we can, at best, rule out $\alpha \gtrsim 0.2$. More important is the fact that the data are good fits with the Bessel functions $J_0$, $J_1$, and $J_2$, so that we are justified in using Eq. (3) to estimate $I_{n+1/2}$.

In order to impose a more stringent bound on the value of $\alpha$ we carefully examined our data for the appearance of Shapiro steps at values of microwave power predicted to produce the first maximum at $\pm\frac{1}{2}f\Phi_0$. Figures 4(a) and 4(b) show a set of microwave-induced steps for sample 1. On the expanded scale of Fig. 4(b) we would have been able to detect the step at $\pm\frac{1}{2}f\Phi_0$ if $I_{\pm1/2}$ had exceeded 0.2 $\mu$A. The absence of the steps enables us to put an upper limit on $\alpha$ of 0.5% for both the $s \pm d$ and $s \pm id$ cases. Figures 4(c) and 4(d) show a similar set of data for thin film sample 7. In this case, we would have detected the step at $\pm\frac{1}{2}f\Phi_0$ if $I_{\pm1/2}$ had exceeded 2 $\mu$A, leading to an upper limit of 1.5% on $\alpha$.

In the same manner we examined all the junctions for the $n = \pm1/2$ steps at a variety of frequencies and temperatures. At low temperatures we sometimes observed beating between the Shapiro steps and self-resonant modes, typically above 15 GHz for the crystals and above 10 GHz for the thin films. As a result, for junctions on single crystals we were able to establish the tightest margins on $\alpha$ at 4.2 K. For the thin film junctions, the subgap resistance at 4.2 K was low, typically 0.2 $\Omega$, and we obtained the best results at temperatures around 2 K. Our upper limits on $\alpha$ are listed in Table I.

Finally, we demonstrate that we can detect small amplitude steps in the presence of large adjacent steps. Sample 3 exhibited a self-resonance at about 12 GHz, and we were able to produce subharmonic steps for nearby microwave frequencies. Figures 4(e) and 4(f) show the $I$-$V$ characteristics at 1.4 K and 10.9 GHz. In Fig. 4(f) we can readily see the subharmonic step, which has a peak-to-peak height of about 4 $\mu$A, approximately 4%

of the adjacent $n = 0$ step. Had this step arisen from second-order Josephson currents we would have inferred a value of $\alpha$ of about 5%.

The values of $\alpha$ for the untwinned crystals, 0.5%, are the lowest. However, since the orthorhombicity of YBCO would lead us to expect first-order tunnel currents, these limits may not be the most significant. The lowest values of $\alpha$ on the twinned crystals, 2.0% and 2.5%, and on the very heavily twinned thin films grown on $LaAlO_3$, 1.5% and 2.0%, may well represent more significant upper limits. The simplest explanation for the presence of first-order tunneling is that the observed $c$-axis Josephson currents arise from a pairing state in YBCO that contains either an $s$-wave contribution in the bulk or has developed an $s$-wave component at the surface [17,18]. However, we cannot emphasize too strongly that this conclusion in no way precludes the simultaneous presence of a $d$-wave component, as is strongly indicated by phase-sensitive tunneling experiments along the $a$ and $b$ directions.

We thank Steve Kivelson, Dung-Hai Lee, Joe Orenstein, and Dan Rokhsar for valuable discussions, and Peng Xiong for technical assistance. This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098 (Berkeley), and by AFOSR Grant No. F4962 092 J70070 and NSF Grant No. DMR 91-13631 (San Diego). R. K. thanks the Deutsche Forschungsgemeinschaft for a fellowship.

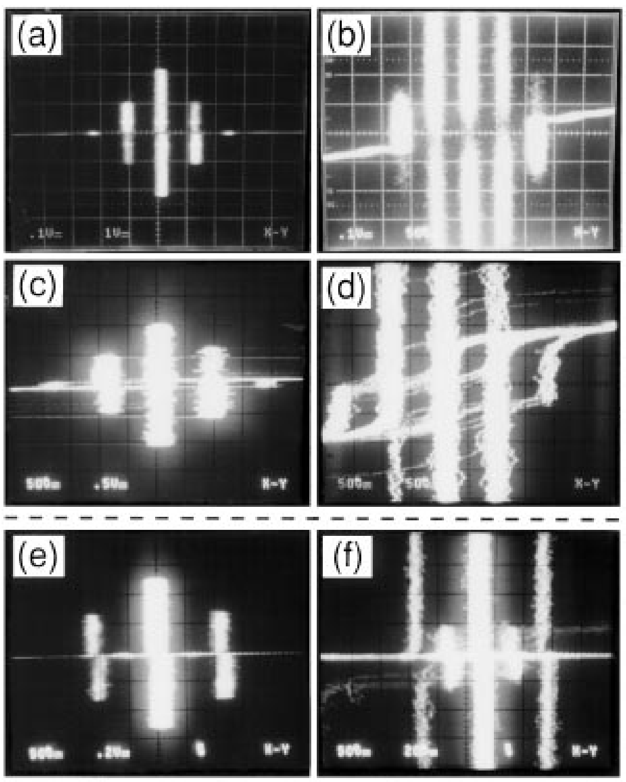


FIG. 4. (a)–(d) $I$-$V$ characteristics at microwave amplitude close to the value at which the steps at $\pm\frac{1}{2}f\Phi_0$ are predicted to be maxima. (a), (b) Sample 1 (detwinned crystal) at 4.2 K and 11.5 GHz: horizontal axes are 20 $\mu$V/division and vertical axes are (a) 20 $\mu$A/division and (b) 1 $\mu$A/division. (c), (d) Sample 7 (thin film) at 2.3 K and 4.4 GHz: horizontal axes are 5 $\mu$V/division and vertical axes are (c) 50 $\mu$A/division and (d) 5 $\mu$A/division. (e), (f) $I$-$V$ characteristics for sample 3 (detwinned crystal) at 1.4 K and 10.9 GHz showing substeps which arise at $\pm\frac{1}{2}f\Phi_0$ from interaction of microwaves with a junction resonance: Horizontal axes are 10 $\mu$V/division and vertical axes are (e) 20 $\mu$A/division and (f) 2 $\mu$A/division.

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