Evidence for $d_{x^2-y^2}$ Pairing from the Magnetic Field Modulation of $YBa_2Cu_3O_7$-Pb Josephson Junctions

D. A. Wollman, D. J. Van Harlingen, J. Giapintzakis, and D. M. Ginsberg
Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, 1110 West Green Street, Urbana, Illinois 61801

(Received 20 June 1994)

We have measured the magnetic field modulation of the critical current of YBCO-Au-Pb Josephson junctions fabricated on the *a* and *b* edges and straddling the *a*-*b* corners of YBCO single crystals. In corner junctions, the critical current drops sharply at zero applied field and increases as the field is increased in either direction, in sharp contrast to the Fraunhofer diffraction patterns observed for single edge junctions. This behavior signifies a sign change of the order parameter between the *a* and *b* directions, strong direct evidence that the superconducting pairing state of YBCO has $d_{x^2-y^2}$ symmetry.

PACS numbers: 74.50.+r, 74.20.Mn, 74.72.Bk

A growing list of theoretical calculations and experiments now suggests that the high temperature cuprate superconductors exhibit an unconventional pairing state. Much attention has been focused on the $d_{x^2-y^2}$ pairing state since a phenomenological model incorporating antiferromagnetic spin fluctuation coupling in this channel has successfully explained many of the normal state and superconducting properties of the cuprates [1]. Alternate models argue for an anisotropic *s*-wave state [2], or complex mixtures of *s* and $d_{x^2-y^2}$ [3] or $d_{x^2-y^2}$ and d_{xy}, the latter of which has been shown to correspond to an anyon state [4]. There is substantial experimental evidence from angle-resolved measurements by photoemission [5], Raman scattering [6], and point-contact tunneling [7] that the magnitude of the order parameter is anisotropic with fourfold rotation symmetry in the *a*-*b* plane. However, these results are in general agreement with all of the proposed models which differ only in the extent of the magnitude anisotropy. Measurements of the low temperature penetration depth in the best YBCO crystals show a linear dependence [8], characteristic of line nodes in the order parameter that are only predicted for the *d*-wave state. Nonetheless, because impurities can either obscure or mimic the presence of nodes, it is difficult to determine unambiguously the symmetry of the pairing state from measurements of the order parameter magnitude alone.

A more definitive approach for distinguishing candidate states is to probe the phase of order parameter. Previously, we presented measurements of the phase coherence in bimetallic YBCO-Pb dc SQUIDs that exhibited a sign change of the order parameter between the *a* and *b* directions, providing strong evidence for the $d_{x^2-y^2}$ pairing state [9]. Recently, this result has been reproduced in YBCO-Pb SQUIDs [10], and supported by direct SQUID magnetometer measurements of the spontaneous circulating currents predicted for the *d*-wave state [11,12] and indirect measurements of their averaged effect in the paramagnetic Meissner (Wohlleben) effect [13–15]. Although these SQUID results are rather compelling, it is necessary to account for asymmetries in the SQUIDs and for the possibility of magnetic flux trapping near the SQUID loop in order to verify the result.

In this Letter, we present an alternative approach to determining the order parameter symmetry from measurements of the magnetic field modulation of the critical current of single YBCO-Pb Josephson junctions. We compare diffraction patterns obtained on junctions fabricated on *a* and *b* edges to junctions straddling the *a*-*b* corners of YBCO single crystals. This experiment has several distinct advantages over the SQUID experiments and elucidates the role of phase coherence and flux trapping in tunneling between conventional and unconventional superconductors. In agreement with the SQUID measurements, we find that the order parameter of YBCO has a phase shift of π between the *a* and *b* directions as required for $d_{x^2-y^2}$ symmetry. A preliminary discussion of single junction tunneling was given in our previous paper [9] and was used to interpret tunneling into step edges on *c*-axis oriented YBCO thin films [16] and in YBCO grain boundary junctions [17] as evidence for *d*-wave pairing.

The effects of an applied magnetic field on the critical current of Josephson tunnel junctions have been extensively studied. The magnetic field penetrating through the barrier region transverse to the tunneling direction creates a phase gradient across the width of the junction, resulting in a variation of the local current density and a reduction in the total critical current. For a rectangular junction of area A, width w, magnetic barrier thickness t, and uniform critical current density J_0 low enough that self-field effects are negligible, the critical current has the usual Fraunhofer diffraction form:

$$I_c(\Phi) = J_0 A \left| \frac{\sin(\pi\Phi/\Phi_0)}{\pi\Phi/\Phi_0} \right|, \tag{1}$$

where $\Phi = Bwt$ is the magnetic flux through the junction for applied magnetic field B. The geometry and expected flux modulation are shown in Fig. 1(a).

To test the symmetry of the pairing state of the YBCO crystal, we measure the critical current of a junction fabricated on the corner of the crystal as shown in

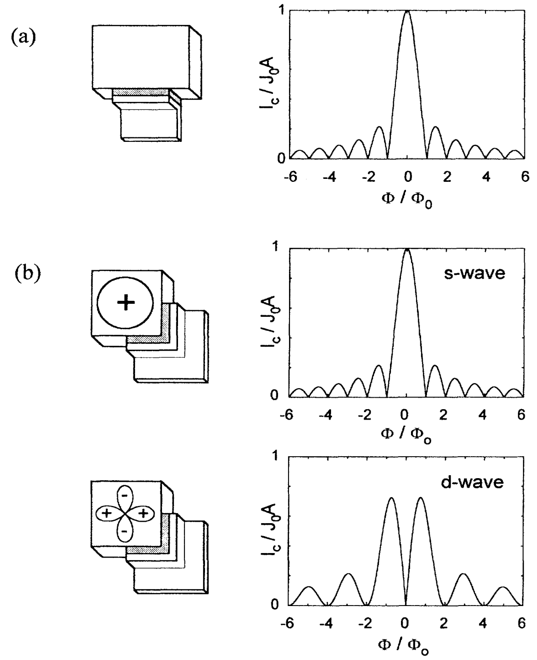

FIG. 1. (a) Fraunhofer diffraction pattern for the critical current vs magnetic flux threading the junction barrier that is characteristic of a single Josephson tunnel junction. (b) Diffraction patterns expected for a single corner junction with s-wave (isotropic or anisotropic) and $d_{x^2-y^2}$ pairing symmetry.

Fig. 1(b). In this geometry, part of the tunneling is into the a-c face of the crystal and part is into the b-c face. A magnetic field applied along the c axis penetrates through each segment of the junction barrier. For an s-wave material with either an isotropic or anisotropic order parameter magnitude, each face would see the same phase and the critical current would have the usual Fraunhofer diffraction pattern. However, for a d-wave superconductor, the order parameters in the a and b directions would be of opposite sign, modifying the single junction diffraction pattern. In a symmetric corner junction (equal junction geometries on the a and b faces) and uniform critical current density, the critical current modulates according to

$$I_c(\Phi) = J_0 A \left| \frac{\sin^2(\pi\Phi/2\Phi_0)}{\pi\Phi/2\Phi_0} \right| . \tag{2}$$

At zero applied field, the current through the two orthogonal faces cancel exactly and the critical current vanishes. Applying a field in either direction increases the critical current, with a maximum value at $\Phi/\Phi_0 = 0.74$ of about 72% of the maximum current for an s-wave junction. The critical current again vanishes when an integer number of flux quanta thread each half of the junction separately, giving a flux modulation period twice that for the edge junction Fraunhofer diffraction pattern. The modulation versus applied flux threading the junction for the s-wave and d-wave cases is shown in Fig. 1(b).

We have measured the single junction modulation characteristics of Josephson junctions fabricated on the edges and corners of YBCO single crystals. A typical sample is shown in Fig. 2. The junctions are formed by depositing Pb thin films onto the a-c and b-c faces of a crystal which had been previously coated with a Au overlayer; a drop of polyamide adheres the crystal to the substrate, providing a smooth ramp from the substrate surface to the crystal face so that the Pb film is continuous. The YBCO-Au-Pb junctions exhibit resistively shunted junction (RSJ) current-voltage characteristics. Typical dimensions are 100 μm wide × 20 μm (the crystal thickness), with critical currents at temperature 2 K of 20–100 μA and resistances of 200–500 mΩ. For these critical current densities (1–5 A/cm^2), the Josephson penetration depth is much greater than 100 μm so the junction may be considered to be in the small junction limit. We determine the critical current by ramping up the bias current until a resistance appears. For very low critical currents (<5 μA), it is necessary instead to measure the flux-induced modulation of the dynamic resistance and deduce a value for the critical current. A magnetic field oriented perpendicular to the a-b plane of the crystal is applied with a Helmholtz coil. Typically, the magnetic coupling area is about 2×10^{-7} cm^2 so that a local magnetic field of order 1 G is required to put a single flux quantum of magnetic flux through the junction; the applied field required is somewhat lower due to flux focusing by the superconducting crystal and films. External magnetic fields are shielded to less than 1 mG with a room-temperature Mumetal shield and a superconducting Pb foil can. Measurements are made in the temperature range 1.5–4.2 K.

FIG. 2. Electron micrograph of a YBCO single crystal with YBCO-Au-Pb Josephson junctions fabricated on the a-b corner and along the a and b edges.

The magnetic field modulation of a single junction on the edge face of a crystal is shown in Fig. 3(a). The critical current exhibits a Fraunhofer-like diffraction pattern with a maximum at zero applied field. In contrast, the modulation for a corner junction on the same crystal plotted in Fig. 3(b) is strikingly different, exhibiting a pronounced dip near zero applied field as expected for the d-wave case. The flux periodicity of the corner junction is also different, with all peaks being roughly the same width, whereas the edge junction shows a central peak with a width approximately twice that of the side lobes, as expected for the Fraunhofer pattern. We have consistently observed this behavior in approximately 10 junctions of each type. Because of uncertainties in the effective junction area and the flux-focusing effects, we are not able to determine the actual amount of magnetic flux threading the tunnel junctions. As a result, we cannot determine if the separation of minima in the corner junctions is twice that in the edge devices as predicted for d-wave pairing.

Another example of a corner junction is shown in Fig. 4(a). Here, we plot the current-voltage characteristic for two values of magnetic flux, showing an increase in I_c as the field is increased. The flux modulation patterns for both polarities of bias current are plotted in Fig. 4(b). Again, there is a sharp dip in the critical current at zero field, falling to less than 25% of its peak value and to only about 15% of what we estimate the critical current would be for an s-wave junction with the same critical current density and area. At higher fields, the critical current decays sharply and exhibits several side lobes. The detailed shape of the diffraction pattern is largely governed by the asymmetry of the corner junction. As an example, we consider a junction with uniform current density and barrier thickness and widths w_a and w_b of the junction segments on the two faces. The critical current for a d-wave crystal then depends on the relative asymmetry parameter $\gamma = (w_a - w_b)/(w_a + w_b)$ according to

$$I_c(\Phi,\gamma) = J_0 A \left| \frac{\sqrt{\sin^2(\gamma\pi\Phi/\Phi_0) + [\cos(\pi\Phi/\Phi_0) - \cos(\gamma\pi\Phi/\Phi_0)]^2}}{\pi(\Phi/\Phi_0)} \right| . \tag{3}$$

In particular, the cancellation of the critical current at zero flux will not be complete if the widths are unequal; the residual critical current at zero field is a fraction γ of the peak current expected for a uniform s-wave junction. Although I_c is increased at zero field by asymmetry, the mirror symmetry with respect to applied field is maintained. In Fig. 4(b), the dashed curve is the critical current calculated from this expression for a junction asymmetry $\gamma = 0.15$, which fits the observed dip in the critical current at zero field. Note that the shape of the side lobes is also reasonably modeled; in particular, the suppression of the first and fourth side lobes is reproduced.

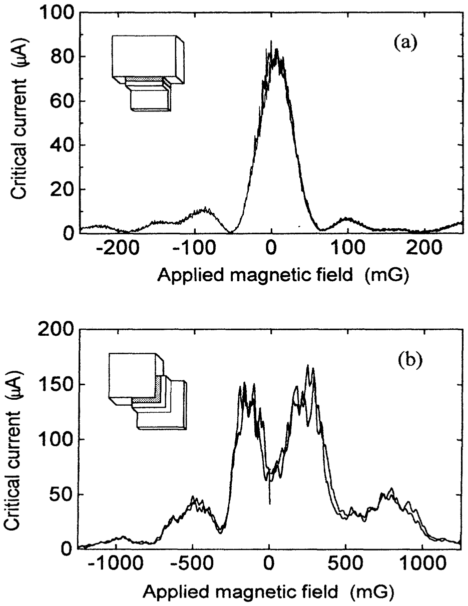

FIG. 3. Measured critical current vs applied magnetic field for (a) an edge junction and (b) a corner junction straddling the a and b faces. The dip at zero field in the corner junction is evidence for $d_{x^2-y^2}$ pairing symmetry in the YBCO.

Single junction measurements have several distinct advantages over the dc SQUID measurements reported previously [9]. First, the smaller junction area makes the magnetic field scale for coupling a flux quantum to the junction much larger (×100), rendering residual fields relatively unimportant and reducing the effect of trapped

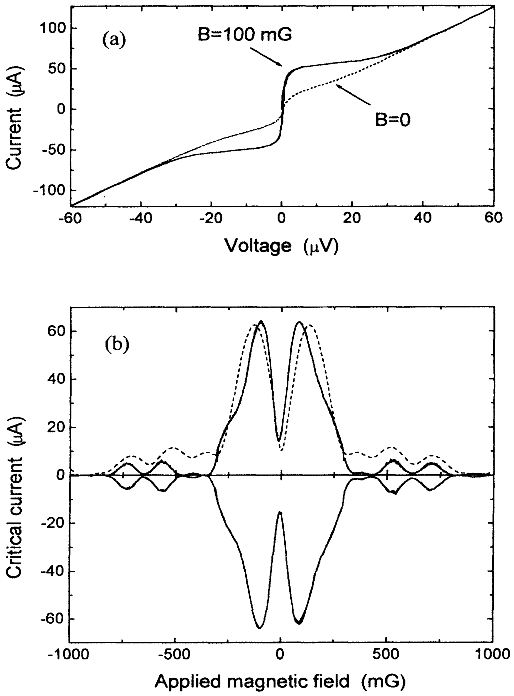

FIG. 4. (a) Current vs voltage for a corner junction at applied magnetic fields of 0 and 100 mG, showing the increase in the critical current for finite fields. (b) Critical current vs applied field of a corner junction for both bias current polarities. The dashed curve is calculated for a $d_{x^2-y^2}$ pairing state in the YBCO assuming an asymmetry of 15% in the junction areas on the a and b faces.

magnetic flux, which we find to be significantly less prevalent in these experiments. Second, junction asymmetry is less critical, excluding the need for extrapolation to zero bias current. Perhaps the most significant advantage is that the shape of the diffraction pattern indicates where the absolute zero for magnetic flux in the junction occurs. For both the s-wave and d-wave cases, the diffraction pattern is symmetric around zero field in the absence of trapped magnetic vortices near the junction. The effect of trapped flux is to skew the diffraction pattern significantly, destroying the mirror symmetry about zero flux, as exhibited by the corner junction shown in Fig. 5(a). To illustrate the effects of flux trapping, we have calculated the modulation of the critical current with field for junctions with a magnetic vortex trapped near the junction. We model the vortex by assuming a nonuniform magnetic field through the junction barrier, consisting of the uniform applied field plus a Gaussian-distributed flux contribution localized in the junction. For both the s-wave and d-wave cases, the diffraction pattern is significantly distorted for all vortex positions in the short junction regime, giving a clear signature when a trapped vortex is present. In Fig. 5(b), we show the critical current modulation for a corner junction on a d-wave crystal with a vortex of width 10% of the junction width and total integrated flux, $\Phi_0/2$, located halfway between the corner and one edge of the junction; the diffraction pattern closely models the experimental pattern of Fig. 5(a). The key point is that for the single junction measurements, symmetry in the diffraction pattern is a strong indication that there is no flux trapping in the junction. This is in sharp contrast to the SQUID measurements in which trapped flux only shifts the periodic flux modulation and is virtually indistinguishable from an intrinsic phase shift. Finally, we emphasize that the symmetrical dip observed in the critical current is not easily explained nor mimicked by any other mechanism; to our knowledge such behavior has rarely if ever been reported in the extensive literature on diffraction patterns in single Josephson junctions.

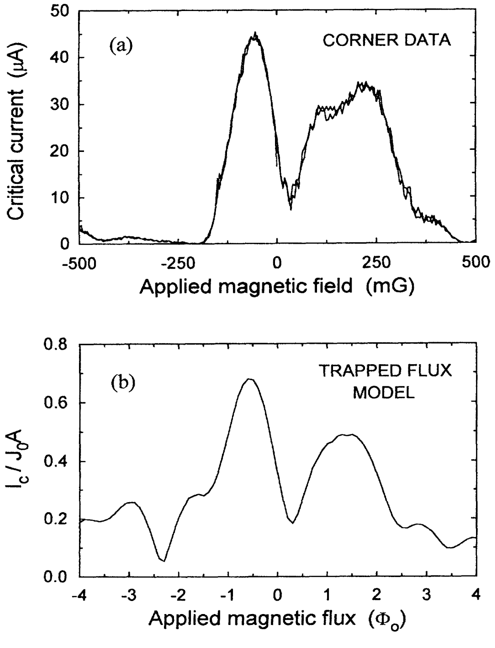

FIG. 5. (a) Measured critical current modulation with applied field for a corner junction showing asymmetry arising from trapped magnetic flux near the junction. (b) Calculated modulation pattern for a vortex located halfway between the corner and edge of a corner Josephson junction, assuming a d-wave pairing state.

In summary, we find strong evidence that YBCO exhibits a $d_{x^2-y^2}$ pairing state from the modulation of the critical current with applied magnetic field. The primary indication is a cusplike suppression of the critical current near zero applied field in junctions with tunneling partially into the *a* and *b* directions at the corner of the crystal. Junctions on the *a* or *b* edges exhibit conventional Fraunhofer diffraction patterns as expected. These measurements are largely impervious to junction asymmetries and trapped magnetic flux in the junction.

We acknowledge valuable discussions with Tony Leggett and David Pines. This work was supported by the NSF Program of the Frederick Seitz Materials Research Laboratory under Grant No. NSF-DMR89-20538. J. G. was supported by the NSF Science and Technology Center for Superconductivity under Grant No. NSF-DMR91-20000, and we have benefited from interactions with the STCS. We also acknowledge extensive use of the facilities of the MRL Microfabrication Laboratory.

---

[1] P. Monthoux, A. Balatsky, and D. Pines, Phys. Rev. B **46**, 14 803 (1992).
[2] S. Chakravarty *et al.*, Science **261**, 337 (1993).
[3] G. Kotliar, Phys. Rev. B **37**, 3664 (1988); Q. P. Li, B. E. Koltenbach, and R. Joynt, Phys. Rev. B **48**, 437 (1993).
[4] D. S. Rokhsar, Phys. Rev. Lett. **70**, 493 (1993); R. B. Laughlin (to be published).
[5] Z.-X. Shen *et al.*, Phys. Rev. Lett. **70**, 1553 (1993).
[6] T. P. Devereaux *et al.*, Phys. Rev. Lett. **72**, 396 (1994).
[7] J. Kane, Q. Chen, K.-W. Ng, and H.-J. Tao, Phys. Rev. Lett. **72**, 128 (1994).
[8] W. N. Hardy *et al.*, Phys. Rev. Lett. **70**, 3999 (1993).
[9] D. A. Wollman, D. J. Van Harlingen, W. C. Lee, D. M. Ginsberg, and A. J. Leggett, Phys. Rev. Lett. **71**, 2134 (1993).
[10] D. A. Brawner and H. R. Ott, Phys. Rev. B **50**, 6530 (1994).
[11] C. C. Tsuei *et al.*, Phys. Rev. Lett. **73**, 593 (1994).
[12] A. Mathia *et al.* (to be published).
[13] W. Braunisch *et al.*, Phys. Rev. Lett. **68**, 1908 (1992); Phys. Rev. B **48**, 4030 (1993).
[14] M. Sigrist and T. M. Rice, J. Phys. Soc. Jpn. **61**, 4283 (1992).
[15] D. Domínguez, E. A. Jagle, and C. A. Balseiro, Phys. Rev. Lett. **72**, 2773 (1994).
[16] I. Iguchi and Z. Wen, Phys. Rev. B **49**, 12 388 (1994).
[17] J. H. Miller *et al.* (to be published).