Use of Tricrystal Junctions to Probe the Pairing State Symmetry of $YBa_2Cu_3O_{7-\delta}$

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We propose a new method of probing superconducting pairing state symmetry, using tricrystal devices analogous to s-d corner junctions. Two central peaks are observed in the field-modulated critical currents of frustrated $YBa_2Cu_3O_{7-\delta}$ tricrystal junctions operating in the short junction limit, consistent with $d_{x^2-y^2}$ pairing symmetry, while a single peak is observed for unfrustrated tricrystal devices. A single peak is also observed for frustrated tricrystal junctions operating in the long junction limit, and it is shown theoretically that s- and d-wave superconductors behave the same in this limit.

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A determination of the pairing state symmetries of high-T_c superconductors has emerged as a paramount problem of condensed matter physics. Most low-T_c superconductors have s-wave pairing symmetry. In strongly correlated electron systems, however, the s channel can be blocked by the strong on-site Coulomb repulsion, leaving open the possibility of higher angular momentum pairing states, such as a $d_{x^2-y^2}$-symmetric pairing state [1] or a $d_{x^2-y^2} + i\alpha d_{xy}$ state [2]. A complex mixture of s- and d-symmetric order parameters ($s + id$ state) has also been proposed [3], and is believed to be energetically preferred to a real mixture if the structure and electronic properties have tetragonal symmetry. However, a real mixture ($s + d$ state) is possible for a system with orthorhombic symmetry.

Most experiments that directly probe the pairing state symmetry of $YBa_2Cu_3O_{7-\delta}$ (YBCO) using Josephson devices are consistent with $d_{x^2-y^2}$-symmetric pairing. Field-modulation measurements of corner SQUID's and junctions between YBCO and an s-wave superconductor provide evidence for a π phase shift in the Josephson coupling energy [4,5]. Odd multiples of half a flux quantum have been measured, using a scanning SQUID microscope, in frustrated tricrystal rings [6]. More recently, careful SQUID microscopy experiments on thin film YBCO-Ag-Pb SQUID's [7] provide compelling evidence for a time-reversal-invariant order parameter, with less than 5% contribution from any s-wave-symmetric imaginary component. However, it should be noted that all of the SQUID microscopy experiments performed thus far are insensitive to a *real* combination of large d-wave and small s-wave order parameters ($d + \alpha s$ pairing).

The issue remains controversial, partly because other experiments appear to support s-wave pairing, and partly because the interpretations of some experiments supporting d-wave pairing symmetry have been called into question. Recently, Klemm [8] suggested that singular demagnetization effects at the YBCO corners could mimic the π phase shift, originally attributed to d-wave pairing symmetry, observed in corner SQUID's and junctions [4,5]. The observation of Josephson supercurrents along the c direction in YBCO-Pb superconductor-insulator-superconductor (SIS) tunnel junctions [9] and across 45° tilt boundary junctions [10] suggest the existence of an s-wave component in YBCO. Experiments measuring the supercurrent between a 45° misoriented YBCO grain and the surrounding film [11] have also been interpreted as supporting s-wave pairing in YBCO. However, it has been argued [12] that these experiments are also consistent with $d_{x^2-y^2}$ pairing symmetry due to the formation of Josephson vortices.

The experiments reported here are field-modulated critical current measurements of tricrystal devices analogous to s-d corner junctions. We investigated both frustrated tricrystal junctions and unfrustrated tricrystal devices. We observed pronounced double peaks in the field-modulated critical currents of frustrated tricrystal junctions operating in the short junction limit, while we observed a single peak for unfrustrated tricrystal junctions. We believe these observations provide convincing evidence that the behavior results from the intrinsic properties of YBCO, and not from corner demagnetization effects [8]. Unlike the SQUID microscopy experiments [6,7], our experiments can detect either a complex or a real mixture of s- and d-wave order parameters. In addition, we investigated both short and long frustrated tricrystal junctions, finding that the behaviors are dramatically different in these two limits.

An example of a frustrated tricrystal junction is illustrated in Fig. 1, where the boundaries crossed by the microbridge, A and C, limit the critical current. Boundary B is much longer than the other two boundaries, so the current flowing across it is much less than its critical current. The phase across boundary B thus minimizes its coupling energy and introduces any phase shift resulting from unusual pairing state symmetry. This device is somewhat analogous to an s-d corner junction [4]. If the pairing symmetry is $d_{x^2-y^2}$, then either one boundary (A, B, or C) acts as a π junction and the other two act as 0 junctions or all three junctions act as π junctions. More generally, if the order parameter has the form $\Delta = \Delta_s + i\Delta_d$, then the current-phase relation for any given junction will have the form $I = I_0 \sin(\varphi \pm n\delta)$ $(n = 0, 1)$. The phase shift $\delta = 2\tan^{-1}|\Delta_d/\Delta_s|$ [13] reduces to $\delta = \pi$ for pure $d_{x^2-y^2}$ pairing symmetry ($\Delta_s = 0$) and $\delta = 0$ for pure

s-wave symmetry ($\Delta_d = 0$). Taking the lengths to be $L_A = (L/2)(1 - \gamma)$ and $L_C = (L/2)(1 + \gamma)$, where γ represents the junction asymmetry, assuming uniform critical current density J_0, and assuming that the field penetrates areas A_B and A_{AC} along boundaries B and AC, respectively ($\sim 2\lambda_L\lambda_J$ and $\sim 2\lambda_L L$, respectively, if we neglect flux focusing), a straightforward calculation yields the following expression for the critical current:

$$I_c(B,\gamma,\beta) = \frac{I_0}{\pi|\Phi/\Phi_0|}\left\{1 - \cos\left[(1+\beta)\pi\frac{\Phi}{\Phi_0} \pm \delta\right]\cos\left[\pi\frac{\Phi}{\Phi_0}\right] - 2\sin\left[\left(1+\frac{\beta}{2}\right)\pi\frac{\Phi}{\Phi_0} \pm \frac{\delta}{2}\right]\sin\left[\frac{\beta}{2}\pi\frac{\Phi}{\Phi_0} \pm \frac{\delta}{2}\right]\cos\left[\gamma\pi\frac{\Phi}{\Phi_0}\right]\right\}^{1/2}, \tag{1}$$

where $I_0 = J_0 tL$, $\Phi = A_{AC}B$, and $\beta = A_B/A_{AC}$. If we assume that $A_B = 0$ and $\delta = \pi$, then Eq. (1) reduces the expressions given in Ref. [4] for YBCO-Pb corner junctions. Figure 2 shows plots of critical current vs field, as predicted by Eq. (1), for several values of δ (choosing the minus sign for δ). The plots enable one to distinguish readily between mostly $d_{x^2-y^2}$ and s-wave pairing symmetries, and are asymmetric for $s + id$ states. Choosing a plus sign for δ would invert this asymmetry, which is related to the direction of spontaneous circulating current. On the other hand, if the order parameter is a real combination of s- and d-wave components, then the field-modulated critical current will be a symmetric linear superposition of s- and d-wave interference patterns.

The YBCO films used in our experiments were deposited by laser ablation onto $SrTiO_3$ tricrystal substrates with nominal misorientation angles of 30° [14]. Prior to patterning, the transition temperatures of the films were generally ~90 K. The films were then photolithographically patterned into devices of the type illustrated in Fig. 1. Bicrystal control devices were also fabricated, and were always observed to exhibit a single central peak in the field-modulated critical current. One frustrated tricrystal device, no. 1, had a microbridge width $L = 3$ μm, a thickness $t = 80$ nm, and less than 10%–15% geometrical asymmetry between the junctions formed by boundaries A and C. The device had a $T_c^{\rm onset}$ of 85 K, and had an extended "foot" in its temperature-dependent resistance below about 75 K, ultimately reaching zero resistance at 20 K. The maximum critical current density at 10 K was 2.1×10^4 A/cm^2. The Josephson penetration length is given by $\lambda_J^2 \cong \Phi_0/\mu_0 J_c t$ for a film whose thickness t is less than the London penetration length $\lambda_L \cong 140$ nm. For device 1, we estimate that $\lambda_J = 10.4$ μm $\sim 3.5L$, so this device is well into the short junction limit.

The current-voltage (I-V) characteristics were generally resistively-shunted-junction-(RSJ) like with little rounding. The critical currents were defined using a ~5 μV criterion, although the exact value of the voltage criterion was not critical due to the sharply nonlinear I-V curves. Figure 3(a) shows a plot of the average $I_c = \{|I_c^+| + |I_c^-|\}/2$

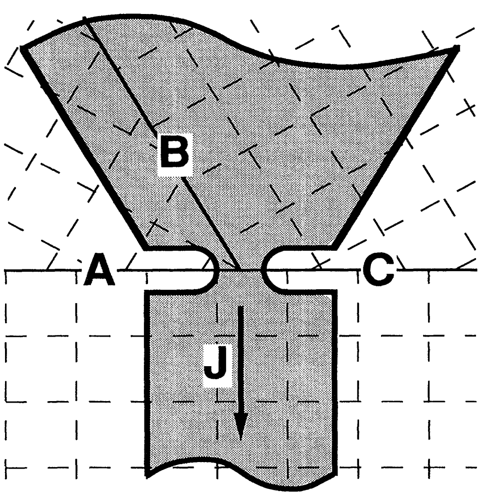

FIG. 1. Frustrated tricrystal microbridge. The misorientation angles are nominally 30° and the angle between boundaries A and B is 60° for the tricrystal illustrated. The unfrustrated tricrystal device had a 90° angle between boundaries A and B, and had misorientation angles of 19°, 38°, and 19° across boundaries A, B and C, respectively.

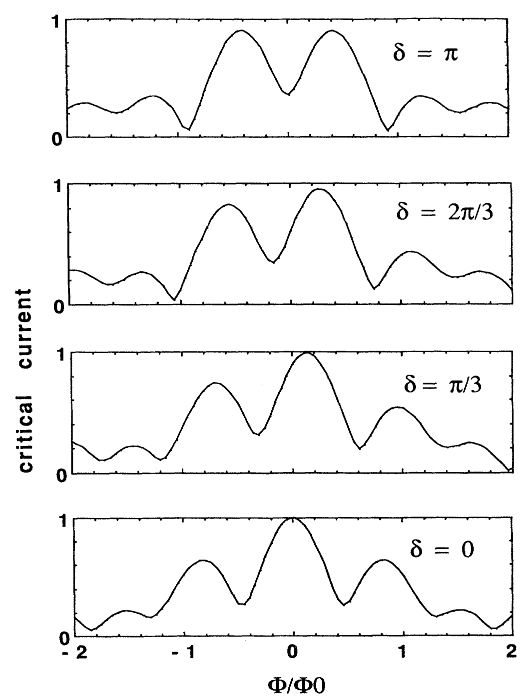

FIG. 2. Theoretical plots of critical current vs magnetic flux $\Phi = A_{AC}B$, using Eq. (1) and assuming $\beta = 1.3$ and $\gamma = 0.35$ for several values of $\delta = 2\tan^{-1}(\Delta_d/\Delta_s)$.

of the positive and negative critical currents as a function of field for frustrated tricrystal junction 1 at 10 K. The data in Fig. 3(a) exhibit two central peaks separated by a pronounced minimum at zero field, consistent with predominantly $d_{x^2-y^2}$ pairing symmetry. We have consistently observed this same overall behavior in device 1 during several cool downs over a two-week period, although variations in the observed slight asymmetry and field offset suggest that these effects are mainly due to flux trapping. We estimate the residual field due to imperfect shielding of the earth's field to be less than 10 mG. We also observed double peaks in other devices that operated marginally in the short junction region ($L \lesssim \lambda_J$), although the minimum between the two central peaks was far less pronounced in those devices. Note that the two peak heights in Fig. 3(a) are nearly equal, suggesting that there is not a complex mixture of s- and d-wave components.

We also measured the field-dependent critical current of a 2.6 μm wide unfrustrated tricrystal junction with a Josephson penetration length of about 6.7 μm. The angle between boundaries A and B was 90° for this device and the misorientation angles were 19°, 38°, and 19° across boundaries A, B and C, respectively. The measured field-dependent critical current at 10 K is shown in Fig. 3(b), and exhibits a single central peak. We believe these results provide convincing evidence that the observed behavior is a direct consequence of the intrinsic properties of YBCO and is not an artifact due to corner demagnetization effects [8].

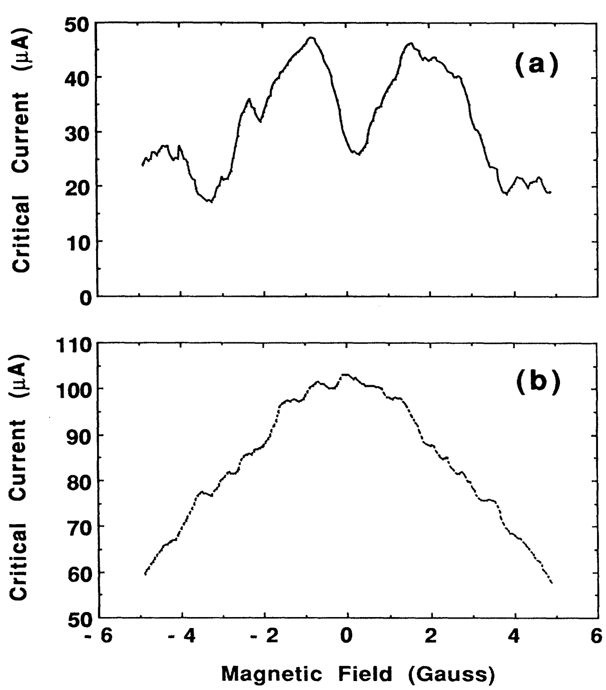

FIG. 3. (a) Critical current vs field for frustrated tricrystal device 1 at 10 K. (b) Critical current vs field at 10 K for a 2.6 μm wide unfrustrated tricrystal junction with $\lambda_J = 6.7$ μm.

The nonvanishing critical current at zero field in Fig. 3(a) can at least partly be attributed to critical current asymmetries between the two halves of the "0-π junction." Another possible explanation is that there might be a real combination of large d-wave and small s-wave components. If this were the case, then the s-channel supercurrents ($I_s \sin\varphi$) would have zero phase shift while the d-channel supercurrents ($\pm I_d \sin\varphi$) would have phase shifts of zero and π across the two boundaries. Thus, the supercurrents flowing through the two boundaries would be $(I_d + I_s)\sin\varphi$ and $-(I_d - I_s)\sin\varphi$. This intrinsic asymmetry would result in a zero-field minimum given by $I_c(B = 0)/I_{\max} \sim I_s/I_d$. One possible $d + \alpha s$ state is an "orthorhombic d-wave"-symmetric pairing state, where the gap, within each twin domain (taking $a \sim b$) has the form [3] $\Delta(\mathbf{k}) = \Delta_0[(1 \pm \alpha)\cos k_x a - (1 \mp \alpha)\cos k_y a] = \Delta_0[\cos k_x a - \cos k_y a] \pm \alpha\Delta_0[\cos k_x a + \cos k_y a]$, where the first term $d_{x^2-y^2}$ symmetry and the second term has extended s-wave symmetry. It has recently been found that the London penetration lengths λ_a and λ_b in untwinned YBCO crystals are highly anisotropic, with ratios λ_a/λ_b ranging from 1.3 to 3 having been reported [15], lending support to the notion of a distorted d-wave symmetry.

In the tricrystal ring experiments [6], the current flowing through the three junctions is much less than their critical currents. Assuming $d + \alpha s$ pairing symmetry, the coupling energy $-(I_s \pm I_d)\Phi_0 \cos\varphi$ associated with each boundary is minimized when the phase φ across the junction is either zero or π. The magnetic flux through the ring will thus be exactly half a flux quantum in the lowest energy state (assuming $I_d \gg I_s$), so the ring experiments cannot detect a small real s component. In addition, the finite misorientation angles of tricrystal junctions might make them more sensitive to the relative contribution of a small real s component than YBCO-Pb corner junctions [4], since the d-channel critical current I_d would be expected to decrease more rapidly with increasing misorientation angle than the s-channel critical current I_s.

Another frustrated tricrystal device, no. 2, consisted of a 75 nm thick YBCO film patterned into a 40 μm wide microbridge, with essentially identical widths for junctions A and C. The T_c ($R = 0$) of device 2 was higher ($= 58$ K) than that of device 1, and the maximum critical current was 1.4 mA at 10 K. The Josephson penetration length λ_J was about 7 μm, so this device was a long junction, with $L \gg \lambda_J$. Figure 4 shows the field-dependent critical current, in which only a single central peak is observed. We have consistently observed a single peak in all devices operating in the long junction limit.

Xu, Miller, and Ting [16] theoretically studied a long Josephson junction with a π discontinuity in the coupling energy at the center, in order to determine the expected behavior for a $d_{x^2-y^2}$ superconductor in the limit $L \gg \lambda_J$. Combining the dc Josephson relation with one of Maxwell's equations yields the sine-Gordon equation $\partial^2\varphi/\partial x^2 = \pm\lambda_J^{-2}\sin\varphi(x)$, where opposite signs are se-

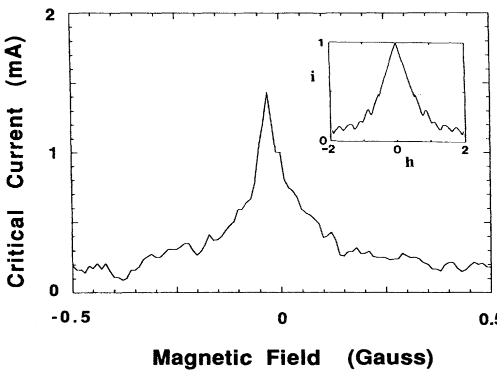

FIG. 4. Field-dependent critical current for tricrystal device 2 at 10 K. Inset: theoretical plot of I_c/I_0 vs H/H_0 for an s-d corner junction or d-wave tricrystal junction in the long junction limit $L \gg \lambda_J$.

lected for the left- ($x < 0$) and right- ($x > 0$) hand sides of the discontinuity. Following the pioneering work of Owen and Scalapino [17], the solutions are found to contain a π vortex that can be regarded as an intrinsic π phase shift between the left- and right-hand sides of the discontinuity. Xu, Miller, and Ting [16] also calculated the free energies with (F_ν) and without (F_0) the π vortex and found that $F_\nu < F_0$, when $L > 2\lambda_J$, showing that the vortex state is favorable for long junctions. The π vortex has little effect on the zero-field supercurrent, since its positive and negative contributions exactly cancel out and are localized near the center, while the bias current is confined near the edges. The theoretical critical current will therefore be a maximum at zero field, as indicated by the calculated field-dependent critical current shown in the inset of Fig. 4.

We believe it pertinent to comment on the measurements [11] of the supercurrent between a 45° misoriented hexagonal YBCO grain and the surrounding film. The critical current was found to depend linearly on the number of coupled faces in the hexagon, showing no evidence for the interference effects one might naïvely expect for $d_{x^2-y^2}$ symmetry. Using the expression $\lambda_J^2 = \Phi_0/\mu_0 J_c \lambda_L$ and using the reported value of J_c of $\sim 2 \times 10^3$ A/cm^2, λ_J is estimated to be 24 μm. The reported length of each face of the hexagons, on the other hand, was 50–500 μm—well into the long junction region. We believe that our results for long junctions underscore the arguments by Millis [12], and strongly suggest that no interference effect would be expected for $d_{x^2-y^2}$-wave superconducting films.

In summary, we have implemented a new method of probing the pairing state symmetry of YBCO films, by measuring the field-modulated critical currents of tricrystal junctions. The observation of double peaks in the field-modulated critical currents of short frustrated tricrystal junctions is consistent with predominantly $d_{x^2-y^2}$ pairing symmetry, although a real s component cannot be ruled out. Only a single central peak is observed for an unfrustrated tricrystal device, thus allowing us to rule out corner demagnetization effects. We find it nearly impossible to distinguish between s-wave and d-wave superconductors using long junctions. We believe that this will also be true for other experiments that utilize interference effects in Josephson devices as probes of pairing state symmetry.

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