Anomalous periodicity of the current-phase relationship of grain-boundary Josephson junctions in high-T_c superconductors

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The current-phase relation (CPR) for asymmetric 45° Josephson junctions between two d-wave superconductors has been predicted to exhibit an anomalous periodicity. We have used the single-junction interferometer to investigate the CPR for these kinds of junctions in $YBa_2Cu_3O_{7-x}$ thin films. A remarkable amplitude of the π-periodical component of the CPR has been experimentally found, providing an additional source of evidence for the d-wave symmetry of the pairing state of the cuprates. [S0163-1829(99)05629-5]

A number of experimental results confirm $d_{x^2-y^2}$-wave symmetry of the pairing state of high-temperature superconductors.[1] An unconventional pairing state requires the existence of zeros of the order parameter in certain directions in momentum space. Thermodynamic and spectroscopic measurements do indeed suggest their existence, but by themselves they do not exclude conventional s-wave pairing with nodes.[1] Direct evidence for the d-wave pairing state is provided by phase-sensitive experiments, which are based on the Josephson effect.[2] Quite generally, the current-phase relationship (CPR) of a Josephson junction, $I(\varphi)$ is an odd periodic function of φ with a period 2π.[3]. Therefore $I(\varphi)$ can be expanded in a Fourier series

$$I(\varphi)=I_1 \sin\varphi+I_2 \sin 2\varphi+\cdots. \tag{1}$$

In the tunnel limit we can restrict ourselves to the first two terms in Eq. (1). Since the order parameter is bound to the crystal lattice, $I(\varphi)$ of a weak link depends on the orientation of the d-wave electrodes with respect to their boundary. The existing phase-sensitive experiments exploit possible sign changes of I_1 between different geometries.[2] In this work we present a phase-sensitive experimental test of the pairing state symmetry of cuprates. Namely, in certain geometries, the I_1 term should vanish by symmetry. In such cases, the CPR should exhibit an anomalous periodicity.

Let us analyze the angular dependence of $I_{1,2}$ in a junction between two macroscopically tetragonal d-wave superconductors. As emphasized in Ref. 4, also heavily twinned orthorhombic materials such as $YBa_2Cu_3O_{7-x}$ belong to this class, if the twin boundaries have odd symmetry. We consider an ideally flat interface between two superconducting electrodes. Let θ_1 (θ_2) denote the angle between the normal to the grain boundary and the a axis in electrode 1 (2), see Fig. 1. If we only keep the lowest-order angular harmonics, the symmetry of the problem dictates that[4]

$$I_1=I_c \cos 2\theta_1 \cos 2\theta_2+I_s \sin 2\theta_1 \sin 2\theta_2. \tag{2}$$

The coefficients I_c, I_s are functions of the barrier strength, temperature T, etc. The I_2 term results from higher-order tunneling processes and we neglect its weak angular dependence. It is seen from Eq. (2) that the criterion for the observation of an anomalous period of the CPR, $I_1=0$, is realized for an asymmetric 45° junction, i.e., a junction with $\theta_1=45°$ and $\theta_2=0$.

The I_2 term is also present in weak links based on conventional s-wave superconductors but for all known types of weak links $|I_2/I_1|<1$. For instance, for a tunnel junction $|I_2/I_1|\ll 1$. For a superconductor–normal-metal–superconductor (SNS) junction, $I\propto\sin\varphi/2$ at $T=0$,[5] and the Fourier expansion of Eq. (1) leads to $I_2/I_1=-2/5$. Therefore a possible experimental observation of $|I_2/I_1|\gg 1$ in an asymmetric 45° junction provides direct evidence of d-wave symmetry of the pairing state in the cuprates.

We have investigated the CPR of $YBa_2Cu_3O_{7-x}$ thin-film bicrystals with asymmetric 45° [001]-tilt grain boundaries as sketched in Fig. 1, using a single-junction interferometer configuration in which the Josephson junction is inserted into a superconducting loop with a small inductance L. In a stationary state without fluctuations, the phase difference φ across the junction is controlled by applying an external magnetic flux Φ_e penetrating the loop: $\varphi=\varphi_e-\beta f(\varphi)$. Here $\varphi_e=2\pi\Phi_e/\Phi_0$; $\Phi_0=2.07\times10^{-15}$ Tm^2 is the flux quantum; $f(\varphi)=I(\varphi)/I_0$ is the CPR normalized to the maximal Josephson current I_0, and $\beta=2\pi LI_0/\Phi_0$ is the normalized

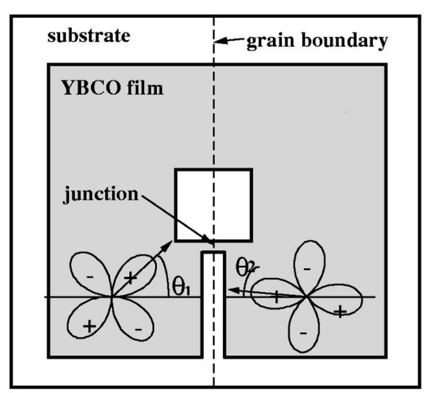

FIG. 1. Washer-shaped interferometer with one short Josephson junction (not in scale). Dimensions are given in the text.

critical current. In order to obtain the CPR for the complete phase range $-\pi \leqslant \varphi \leqslant \pi$ the condition $\beta < 1$ has to be fulfilled, because for $\beta > 1$ the curve $\varphi(\varphi_e)$ becomes multivalued. Following Ref. 3, we express the effective inductance of the interferometer using the derivative f' with respect to φ as $L_{int} = L[1 + 1/\beta f'(\varphi)]$. The inductance can be probed by coupling the interferometer to a tank circuit with inductance L_T, quality factor Q, and resonance frequency ω_0 through the mutual inductance M.[8] External flux in the interferometer is produced by a current $I_{dc} + I_{rf}$ in the tank coil and can be expressed as $\varphi_e = 2\pi(I_{dc} + I_{rf})M/\Phi_0 = \varphi_{dc} + \varphi_{rf}$, where $M^2 = k^2 L L_T$ with k a coupling coefficient. Taking into account the quasiparticle current in the presence of a voltage V across the junction the phase difference is given by the relation $\varphi = \varphi_{dc} + \varphi_{rf} - \beta f(\varphi) - 2\pi\tau(\varphi)V/\Phi_0$, where $\tau(\varphi) = L/R_J(\varphi)$ with $R_J(\varphi)$ the resistance of the junction. In the small-signal limit $\varphi_{rf} \ll 1$ and in the adiabatic case $\omega\tau \ll 1$, keeping only the first-order terms, the effective inductance L_{eff} of the tank circuit-interferometer system is

$$L_{eff} = L_T\left(1 - k^2 \frac{L}{L_{int}}\right) = L_T\left(1 - \frac{k^2 \beta f'(\varphi)}{1 + \beta f'(\varphi)}\right).$$

Thus the phase angle α between the driving current and the tank voltage U at the resonance frequency of the tank circuit ω_0 is

$$\tan\alpha(\varphi) = \frac{k^2 Q \beta f'(\varphi)}{1 + \beta f'(\varphi)}. \qquad (3)$$

Using the relation $[1 + \beta f'(\varphi)]d\varphi = d\varphi_{dc}$ which is valid for $\varphi_{rf} \ll 1$ and $\omega\tau \ll 1$, one can find the CPR from Eq. (3) by numerical integration.

The advantage of the CPR measurement of an asymmetric 45° junction with respect to the by-now standard phase-sensitive tests of pairing symmetry based on the angular dependence of I_1 is twofold. First, it avoids the complications of the analysis of experiments caused by the presence of the term I_s.[4] Second, flux trapped in the interferometer washer (see Fig. 1) does not invalidate the conclusions about the ratio $|I_2/I_1|$ and hence about the pairing symmetry, which is not the case in standard phase-sensitive tests of the *d*-wave symmetry of the pairing state.[9]

The films of 100-nm thickness were fabricated using standard pulsed laser deposition on (001) oriented $SrTiO_3$ bicrystalline substrates with asymmetric [001] tilt misorientation angles of 45°±1°. The films were subsequently patterned by Ar ion-beam etching into 4×4-mm^2 square washer single-junction interferometer structures (Fig. 1). The widths of the junctions were 1–2 μm. The square washer holes had a side length of 50 μm. This geometry of the interferometer gives $L \approx 80$ *p*H. The resistance of a similar single junction (without interferometer loop) was measured directly and $R_J > 1$ Ω was found. Therefore the condition for the adiabatic limit $\omega\tau \ll 1$ is satisfied. For measurements of $\alpha(\varphi_{dc})$, several tank circuits with inductances 0.2–0.8 μH and resonance frequencies 16–35 MHz have been used. The unloaded quality factor of the tank circuits $70 < Q < 150$ has been measured at various temperatures. The coupling factor k was determined from the period ΔI_{dc} of $\alpha(I_{dc})$ using $M\Delta I_{dc} = \Phi_0$. Its value varied between 0.03 and 0.09. The amplitude of I_{rf} was set to produce a flux in the interferometer smaller than 0.1 Φ_0 to ensure the small-signal limit.

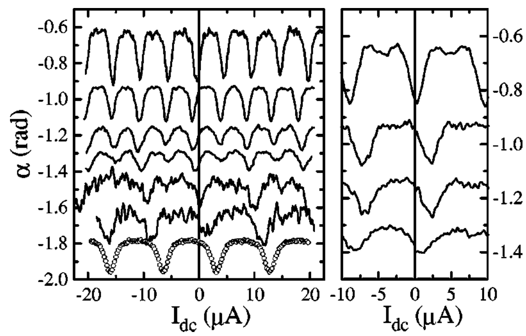

FIG. 2. Left panel: Phase angle between the driving current and the output voltage measured for sample No. 1 at different temperatures as a function of the dc current I_{dc}. The curves are shifted along the *y* axis and the data for $T = 30$ and 40 K are multiplied by factor 4 for clarity. From top to bottom, the data correspond to $T = 4.2$, 10, 15, 20, 30, and 40 K. The data measured for 36° bicrystals ($\theta_1 \approx 36°, \theta_2 \approx 0$) at $T = 40$ K in the same washer geometry are shown for comparison (open circles). Right panel: The same for sample No. 3. From top to bottom, the data correspond to $T = 4.2$, 10, 15, and 20 K.

The measurements have been performed in a gas-flow cryostat with a five-layer magnetic shielding in the temperature range $4.2 \leqslant T < 90$ K. The experimental setup was calibrated by measuring interferometers of the same size with 24° and 36° grain boundaries. We have studied six samples, out of which for four samples the π-periodic component of $I(\varphi)$ was experimentally observed. At low temperatures for two samples (Nos. 1 and 2) the value of I_2 is larger than I_1. For sample Nos. 3 and 4 I_2 is approximately 10–20 % of I_1 and for sample Nos. 5 and 6 I_2 is negligible. As an example we plot the phase angle α as a function of the dc current I_{dc} for sample Nos. 1 and 3 (Fig. 2). The behavior of sample No. 1 at low temperatures is defined by the π periodic component of $I(\varphi)$. The curves for sample No. 3 are 2π-periodic, nevertheless for the curve at $T = 4.2$ K the local minima clearly show the presence of a π-periodic component.

In order to determine the CPR we assume that the period of $\alpha(I_{dc})$ at $T = 40$ K and $\Delta I_{dc} = 9.6$ μA, corresponds to $\Delta\varphi_{dc} = 2\pi$. We take $\varphi_{dc} = 0$ at a maximum or minimum of α. This is necessary in order to satisfy $I(\varphi = 0) = 0$, as required by general principles.[3] The experimentally observed shift of the first extreme of $\alpha(I_{dc})$ from $I_{dc} = 0$ (Fig. 2) can be due to flux trapped in the interferometer washer. Most probably, this flux resides in the long junction originated by the grain boundary crossing the washer of the interferometer. This long junction does not play an active role because the Josephson penetration depth is much smaller than the junction length, and external fields produced by I_{dc} are smaller than the first critical field. Nevertheless, the long junction sets the phase difference for $I_{dc} = 0$ at the small junction.

In Fig. 3, we show the CPR determined from the data in Fig. 2. For all curves we have performed a minimal necessary shift consistent with $I(\varphi = 0) = 0$. Thus we have assumed that at $\varphi_{dc} = 0$ a minimum of $\alpha(\varphi_{dc})$ is realized. For an interferometer with a conventional *s*-wave weak link (and also for the 36° junction), at $\varphi_{dc} = 0$ one gets a maximum of

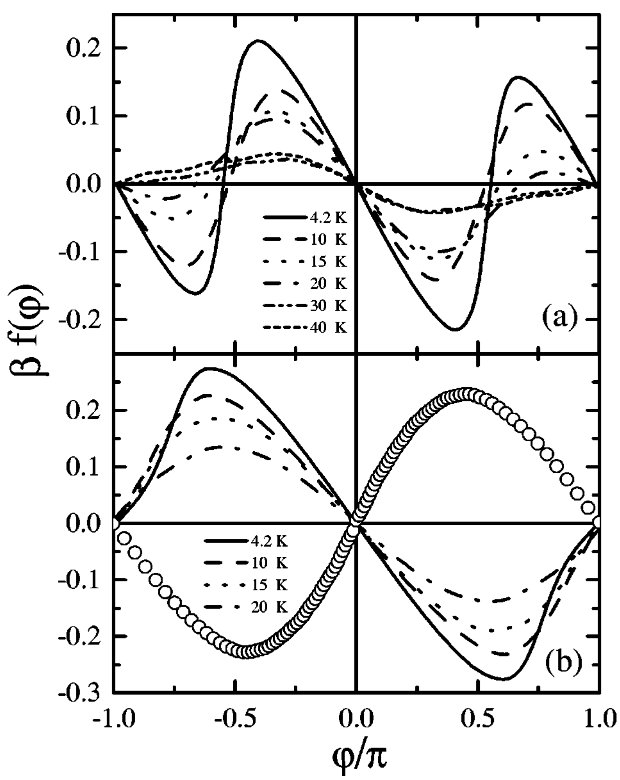

FIG. 3. (a) Josephson current through the junction for sample No. 1 as a function of the phase difference φ, determined from the data in Fig. 2. The scattering of $\alpha(\varphi)$ values was reduced by folding the data back to the interval $\langle 0,\pi \rangle$ and taking the average. Here, the symmetry $\alpha(\varphi)=\alpha(-\varphi)$ was assumed. (b) The same for sample No. 3. The data for the asymmetric 36° bicrystal at $T=40$ K (open circles) are also shown.

$\alpha(\varphi_{dc})$. Note that the minimum of $\alpha(\varphi_{dc})$ at $\varphi_{dc}=0$ implies a paramagnetic response of the interferometer in the limit of small applied fields.

The amplitude of the π-periodic component of the CPR decreases drastically with increasing temperature, and at $T=40$ K its contribution is negligible for all samples. The temperature dependence of I_1 and I_2 could be determinated with acceptable accuracy for sample No. 1 only. With decreasing T, $|I_2|$ grows monotonically down to $T=4.2$ K, while the I_1 component exhibits only a weak temperature dependence (Fig. 4).

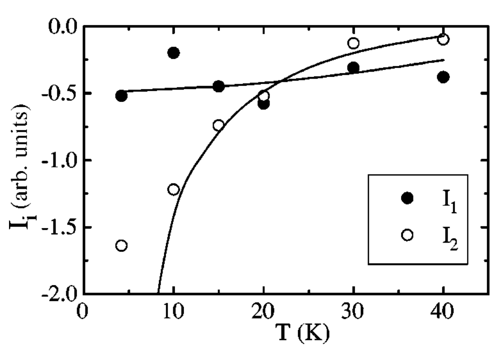

FIG. 4. Temperature dependence of the Fourier expansion coefficients $I_{1,2}$ determined from the experimental data in Fig. 3(a). Solid lines are the Fourier expansion coefficients for the numerical data in Fig. 5.

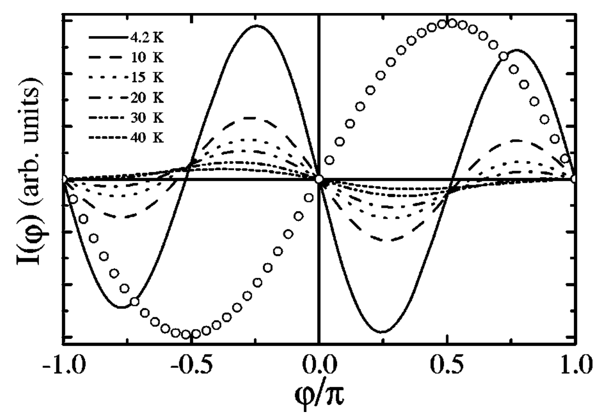

FIG. 5. $I(\varphi)$ calculated according to Eq. (64) of Ref. 11 for a junction with $\theta_1=45.5°$, $\theta_2=0$, $\lambda d=1.5$, $\kappa=0.5$, and $T_c=60$ K. $I(\varphi)$ at $T=40$ K for the 36° bicrystal (open circles) was calculated with the same parameters except for $\theta_1=36°$.

Our experimental results can be understood as follows. It is well known that the microstructural properties of the grain boundaries, especialy 45° boundaries, are defined by their faceted nature. Faceting is an intrinsic property of the grain boundaries,[6,7] and, due to d-wave symmetry of the order parameter, the properties of the junctions strongly depend on the particular distribution of the facets. Small deviations from the ideal geometry of the asymmetric 45° junction lead to a finite value of I_1. Thus for nearly ideal junctions $|I_2/I_1|\gg 1$ at $T\to 0$. The region $T\sim T_c$ can be analyzed quite generally within the Ginzburg-Landau theory. Let the electrodes be described by the (macroscopic) order parameters $\Delta_{1,2}=|\Delta|e^{i\varphi_{1,2}}$. Then the phase-dependent part of the energy of the junction is $E=a[\Delta_1\Delta_2^\star+\text{H.c.}]+b[(\Delta_1\Delta_2^\star)^2+\text{H.c.}]+\cdots$ where $a,b,\ldots$ depend weakly on T.[10] Thus for T close to T_c we estimate $I_1\propto|\Delta|^2\propto(T_c-T)$ and $I_2\propto|\Delta|^4\propto(T_c-T)^2$, leading to $|I_2/I_1|\ll 1$. With increasing deviations from ideal geometry $|I_2/I_1|$ decreases. For large enough deviations, negligible values of $|I_2|$ are expected. These expectations are qualitatively consistent with the experimental data (see also Fig. 4).

So far, our discussion was based solely on symmetry arguments. Let us attempt a more quantitative analysis of our data now. Two different microscopic pictures of asymmetric 45° Josephson junctions between d-wave superconductors have been considered in the literature. The first picture assumes a microscopically tetragonal material and an ideally flat interface.[10–12] Within this picture, only sample No. 1 can be analyzed. Sample No. 2 had $I_0(T=1.5\ \text{K})\cong 10^{-2}\mu$A. At this temperature only the π-periodic component of $I(\varphi)$ was observed. At higher temperatures I_0 was not measurable. $I(\varphi)$ for sample No. 1 calculated according to the model of Ref. 11 is shown in Fig. 5. The experimental data can be fitted within a relatively broad range of barrier heights. However, if we require the $I(\varphi)$ relation of the 36° junction to be fitted by the same (or smaller) barrier height as for the 45° junction, we conclude the barrier of the 45° junction to be rather low.[14] The dependence of $I(\varphi)$ on T requires a choice of $T_c\approx 60$ K in the non-self-consistent theory of Ref. 11. The reduction from the bulk $T_c=90$ K is probably due to a combined effect of surface degradation and order-parameter suppression at the sample surface. The temperature dependence of the ratio of the π and 2π periodic components in $I(\varphi)$ is seen to be in qualitative agreement with experimental data in Fig. 3(a). This is explicitly demonstrated in Fig. 4 where we compare the experimentally obtained $I_{1,2}$ with the results of the Fourier analysis of the curves in Fig. 5. The divergence of I_2 as $T\rightarrow 0$ is an artifact of the ideal junction geometry assumed in Ref. 11. If a finite roughness of the interface is taken into account, this divergence is cut off and the experimental data in Fig. 4 do indeed resemble theoretical predictions for a rough interface.[12] However, the non-self-consistent theory of Ref. 11 is unable to explain the experimentally observed steep CPR close to the minima of the junction energy [see Fig. 3(a)].

In a different approach a heavily meandering interface with $\theta_i=\theta_i(x)$ is assumed. Now, the critical current density $j_c(x)$ is a random function with a typical amplitude $\langle|j_c(x)|\rangle\sim j_c$. If the average critical current along the junction $\langle j_c\rangle < j_c$, a remarkable π-periodic component is present in the CPR. The relation $|I_2/I_1|$ depends on the distribution of $j_c(x)$ and can be much larger than one for $\langle j_c\rangle \ll j_c$.[15,16] This model qualitative explains the obtained results for all samples, however for a quantitative comparison the actual microscopic distribution $j_c(x)$ should be known. Note that also within the picture of Refs. 15 and 16 the *d*-wave symmetry of the pairing state is crucial, otherwise the condition $\langle j_c\rangle \ll j_c$ is difficult to satisfy.

Our present understanding of $I(\varphi)$ in the asymmetric 45° junction is only qualitative. We cannot say whether the remarkable amplitude of the π-periodic component of $I(\varphi)$ is dominated by the microscopically flat regions,[13] or due to the spatial inhomogeneity of the junction. This issue requires further study.

In conclusion, we have measured the magnetic-field response of a single-junction interferometer based on asymmetric 45° grain-boundary junctions in $YBa_2Cu_3O_{7-x}$ thin films. A large π-periodic component of $I(\varphi)$ has been experimentally found, which is in agreement with theoretical predictions for $d_{x^2-y^2}$-wave superconductors. Hence our results provide an additional source of evidence for the *d*-wave symmetry of the pairing state in the cuprates.

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