Nonsinusoidal Current-Phase Relationship of Grain Boundary Josephson Junctions in High-T_c Superconductors

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For various configurations of Josephson junctions incorporating superconductors with unconventional order parameter symmetry, such as most high-T_c cuprates, deviations from the standard sinusoidal current-phase dependence have been predicted. To this point, these deviations have never been observed experimentally. We have measured the current-phase relation of high-T_c Josephson junctions, namely, $YBa_2Cu_3O_{7-x}$ thin film bicrystals, comprising symmetric 45° [001] tilt grain boundaries. The current-phase relations of all junctions investigated were found to be extremely nonharmonic, in agreement with a $d_{x^2-y^2}$-wave dominated symmetry of the order parameter. [S0031-9007(98)06674-5]

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The current-phase relation (CPR) $f(\varphi)$ is a characteristic property of any weak link connecting two superconductors. It describes the dependence of the Cooper-pair current I_p on the phase difference φ of the order parameters of both superconducting electrodes. In a general form it is expressed as

$$I_p = I_c f(\varphi), \qquad -1 \leq f(\varphi) \leq 1\,, \tag{1}$$

I_c being the critical current of the weak link. It was shown by Josephson [1] that for ideal tunnel junctions between conventional superconductors the CPR is sinusoidal, i.e., $f(\varphi) = \sin(\varphi)$. This sinusoidal dependence has been confirmed experimentally numerous times for standard tunnel junctions between conventional superconductors [2].

Recently, it has been revealed that the order parameter of most high-T_c cuprates is unconventional, dominated by a $d_{x^2-y^2}$ symmetry component [3–5]. Because of the sign change of the order parameter associated with this symmetry, strong deviations from the standard sinusoidal dependence have been predicted for the current-phase relations of various configurations of Josephson junctions employing such unconventional superconductors [6–9]. In particular, nonharmonic and double-periodic current-phase relations are expected for junctions oriented nominally perpendicular to the $\langle 110 \rangle$ direction of one or of both electrodes, as well as for junctions for which the $\langle 110 \rangle$ direction of one of the electrodes is aligned with the $\langle 100 \rangle$ direction of the other, such as for 45° [001] tilt grain boundaries.

These predictions are highly unusual. Therefore, an experimental clarification of the CPR for high-T_c junctions for which deviations from a standard harmonic behavior are expected is desirable. Such experiments will further enhance the understanding of the influence of the order parameter symmetry on the properties of grain boundaries and high-T_c Josephson junctions. In addition, they will provide valuable information for the design and use of Josephson junction-based circuits, of which many characteristics directly depend on the CPR. However, to our knowledge, such experiments have not been carried out. All available data refer to Josephson junctions for which nominally sinusoidal current-phase relations are expected. The CPR was measured for weak links prepared by ion irradiation [10], for step-edge junctions [11,12], and also for 24° bicrystal grain boundaries [12]. In nearly all of these cases sinusoidal current-phase relations were found. Deviations from a sinusoidal dependence have been observed only for one step-edge junction, measured at 77 K [12]. These deviations can be explained by the influence of thermal noise [13].

For these reasons we have investigated the CPR of $YBa_2Cu_3O_{7-x}$ thin film bicrystals with symmetric 45° [001]-tilt grain boundaries, as sketched in Fig. 1(a). For these junctions, strong deviations from a sinusoidal CPR are anticipated.

Following a standard approach [14], the CPR was measured using a single-junction interferometer configuration in which the Josephson junction is part of a superconducting loop with a small inductance L. The phase difference φ across the junction is controlled by applying an external magnetic flux Φ_e penetrating the loop:

$$\varphi = \varphi_e - \beta f(\varphi) + \varphi_n + 2\pi m\,. \tag{2}$$

Here, $\varphi_e = 2\pi\Phi_e/\Phi_0$ is the external flux normalized to the flux quantum Φ_0 ($= 2.07 \times 10^{-15}$ Tm^2). The constant $\beta = 2\pi L I_c/\Phi_0$ is the normalized critical current, φ_n is a term accounting for the effective noise, and m is

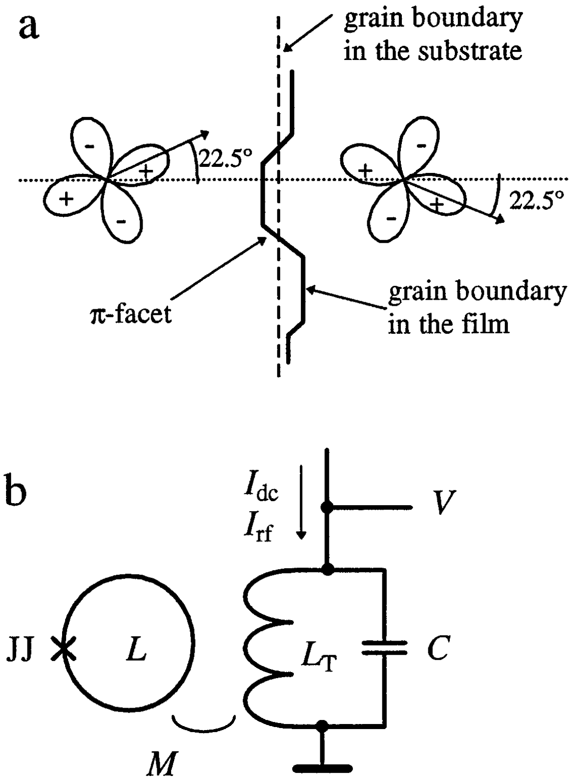

FIG. 1. (a) Schematic representation of a symmetric 45° [001]-tilt grain boundary junction in a $d_{x^2-y^2}$ superconductor. The boundary in the superconducting thin film is meandering, leading to the occurrence of π facets. (b) Schematic of the measurement setup. The Josephson junction is denoted by JJ, and C indicates the capacitance of the tank circuit. The other denotations are explained in the text.

an integer. As capacitive contributions to the loop current are insignificant at the measurement frequencies, the junction capacitance has been neglected. The quasiparticle current is also negligible, for reasons discussed below.

The superconducting loop is inductively coupled to a tank circuit with inductance L_T [see Fig. 1(b)]. This tank circuit is driven with a current I_{rf} at a frequency ω and a dc current I_{dc}. Thus, φ_e can be expressed as a sum of a dc and an rf component $\varphi_e = \varphi_{dc} + \varphi_{rf}$. In this arrangement, the effective impedance $Z_{eff}(\omega)$ of the loop-tank circuit combination is a function of φ_e. As shown by Rifkin and Deaver [14], the CPR can be obtained from this dependence, provided that $\varphi_{rf} \ll 1$. To obtain the CPR for the complete phase range $0 \leq \varphi \leq 2\pi$, the condition $\beta < 1$ has to be fulfilled in addition.

To enhance the accuracy of the measurement, we have adapted this common approach and retrieved the CPR from a measurement of the φ_{dc} dependence of the phase angle α between the drive current I_{rf} and the tank voltage V at the resonant frequency of the tank circuit ω_0. As described in Ref. [12], at ω_0, the $\alpha(\varphi_{dc})$ dependence is related to the derivative of the CPR $f'(\varphi) \equiv df(\varphi)/d\varphi$ in the following way:

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

Here k is the coupling factor between the tank inductance and the interferometer, $k^2 = M^2/(LL_T)$, where M is the mutual inductance [Fig. 1(b)]. Using Eq. (3), from the measured $\alpha(\varphi_{dc})$ dependence $f'(\varphi(\varphi_{dc}))$ is obtained. The CPR is restored by integrating $f'(\varphi(\varphi_{dc}))$ numerically, using the $d\varphi(\varphi_{dc})/d\varphi_{dc}$ dependence obtained from differentiating Eq. (2) with respect to φ_{dc}.

The samples investigated consisted of three bicrystalline $YBa_2Cu_3O_{7-x}$ films with a T_c ($R = 0$) of 88 K. The films, with thickness $t = 100$ nm, were deposited by standard pulsed laser deposition on (001)-oriented $SrTiO_3$ bicrystalline substrates [15] with symmetric [001]-tilt misorientation angles of 45° ± 2° and were subsequently patterned by Ar ion-beam etching into 8×8 mm^2 or 5×5 mm^2 square washer single-junction interferometer structures. The widths of the junctions were $b \approx 2$–3 μm. The washer holes had a side length of 50 μm, leading to $L \approx 80$ pH.

To minimize the influence of external noise, the samples were measured in superconducting and double magnetic shielding at a temperature of 4.2 K. The condition $\beta < 1$ for the investigated interferometers was confirmed experimentally from the character of its response versus φ_{dc} [12].

$YBa_2Cu_3O_{7-x}$ grain boundaries with a symmetric [001] tilt angle of 45° typically have a normal-state interface-resistivity $\rho_n > 1 \times 10^{-8}$ Ω cm^2, which we also measured for boundaries fabricated under identical conditions as the junctions used in the present experiments. In the configuration used, this ρ_n corresponds to normal-state resistances $R_n > 1$ Ω. Accordingly, the relaxation time of the interferometer $\tau = L/R$ is short ($\tau \ll 1/\omega_0$), and hence the quasiparticle current is negligible [12].

For the measurements of $\alpha(\varphi_{dc})$, two tank circuits with quality factor $Q = 120$ and inductance $L_T = 0.4$ μH, $\omega_0 = 30$ MHz, and $L_T = 0.73$ μH, $\omega_0 = 23$ MHz, respectively, were employed. The phase angle was recorded as a function of I_{dc} after amplification of the tank voltage by a high-impedance amplifier. The coupling coefficient k was determined from the period of the $\alpha(I_{dc})$ dependence. Values of $k = 0.072$ and $k = 0.054$ for the respective tank circuits are obtained. To ensure the validity of the small-signal limit, the measurements were carried out with $\varphi_{rf} < 0.15$.

A typical $\alpha(I_{dc})$ dependence is shown in Fig. 2. The corresponding CPR, depicted in Fig. 3, is clearly deviating from the standard sinusoidal behavior. Samples fabricated on different substrates and measured with both tank coils followed closely the same behavior. It is emphasized that the experimental setup employed and the procedure followed are identical to those used to measure the current-phase relations of step-edge junctions and thin-film bicrystals with a symmetric [001] tilt of 24°. For

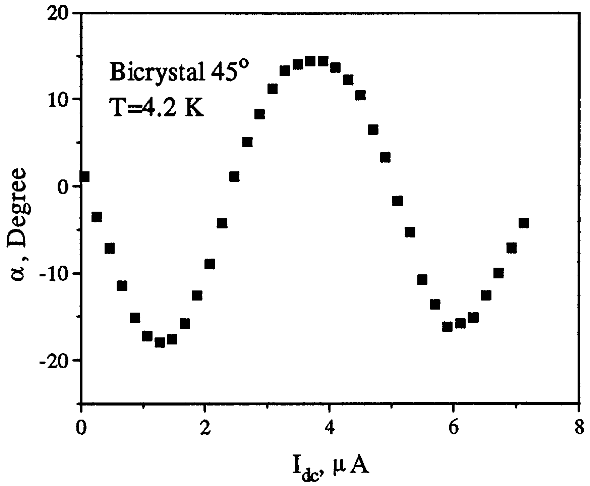

FIG. 2. Phase angle α between the driving current and the output voltage measured at 4.2 K as a function of the dc current I_{dc}, for an $YBa_2Cu_3O_{7-x}$ single junction interferometer circuit containing a symmetric 45° [001]-tilt grain boundary.

all of those samples nominally sinusoidal current-phase relations were observed, and all deviations of the apparent CPR from a sinusoidal one can be attributed to thermal noise [12,13].

The measured deviations from a sinusoidal dependence for the current-phase relations of these 45° bicrystals are startling. It is important to note that the effective Josephson penetration depth $\Lambda_{\mathrm{J}} = [\Phi_0/(4\pi\mu_0\langle j_c\rangle\lambda)]^{1/2} \approx 5~\mu$m is larger than the width of the junction b (narrow-junction limit). Here λ is the London penetration depth. Although several mechanisms are known to cause nonsinusoidal current-phase relations for narrow junctions fabricated from conventional superconductors, all of these mechanisms fail to account for the anomalous dependencies presented.

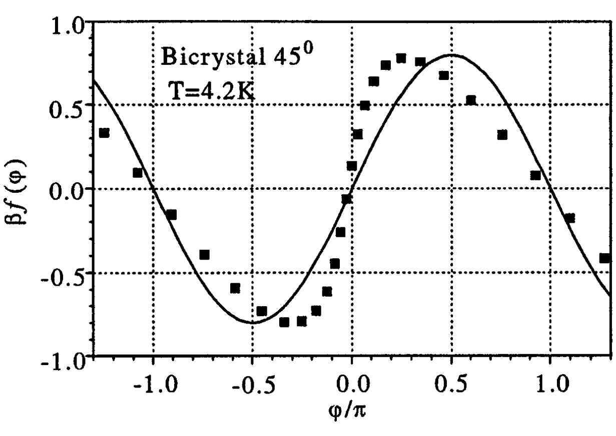

FIG. 3. The normalized current through the junction $\beta f(\varphi)$ as a function of the phase difference φ restored from the measured $\alpha(I_{\mathrm{dc}})$ as shown in Fig. 2. For comparison, the function $\beta\sin(\varphi)$ with $\beta = 0.8$ is plotted as a solid line.

First, one potential source of such deviations is thermal noise. To evaluate its influence we consider a sinusoidal CPR and calculate with Eq. (3) the $\alpha(\varphi_{\mathrm{dc}})$ dependence, assuming a thermally induced Gaussian spread $\rho(\varphi_n)$. With this, the value of $\tan\alpha(\varphi_{\mathrm{dc}})$ is given by [12]

$$\tan\alpha(\varphi_{\mathrm{dc}}) = k^2 Q\beta \times \int_{-\infty}^{\infty} \frac{\cos\varphi(\varphi_{\mathrm{e}},\varphi_n)}{1+\beta\cos\varphi(\varphi_{\mathrm{e}},\varphi_n)}\,\rho(\varphi_n)\,d\varphi_n\,. \tag{4}$$

Using Eq. (4), minor deviations of the CPR from a standard sinusoidal behavior are well described quantitatively for 24° boundaries measured at 77 K [12]. However, no realistic set of β and φ_n exists to account for the large deviations of the CPR observed for the 45° boundaries.

Second, for weak links with a high current density, current-induced suppression of the order parameter in the electrodes close to the weak link can give rise to nonharmonic current-phase relations [16]. It is unrealistic that this effect is the cause for the deviations presented here, as the intragrain critical current density exceeds $10^7~\mathrm{A/cm^2}$ at 4.2 K and is therefore much larger than the grain boundary $\langle j_c\rangle < 4000~\mathrm{A/cm^2}$ at the same temperature.

Third, several additional mechanisms, described in [2], lead to deviations from a harmonic CPR. In all these cases, the slope of $f(\varphi)$ at $\varphi = 0$ is smaller than at $\varphi = \pi$, which is in contrast to our results. Therefore, these mechanisms cannot explain the current-phase relations observed either.

On the other hand, as will be pointed out in the following, the measured CPR can be accounted for by the unconventional order parameter symmetry of $YBa_2Cu_3O_{7-x}$ and by the microstructural properties of the grain boundaries, in particular by their faceted nature [17,18]. Interestingly, due to the $d_{x^2-y^2}$-wave character of the order parameter, the faceting has a more significant influence on the electronic properties of boundaries with a misorientation close to 45° than on boundaries with considerably smaller misorientation angles [18]. This also concerns the CPR, as the 45° boundaries contain a higher density of facets that themselves show anomalous behavior. Two kinds of such anomalies, to be considered here, are described in the literature.

First, due to the sign difference of the adjacent lobes of the $d_{x^2-y^2}$-wave order parameter, many facets are biased with an additional π phase shift (π facets) [17–19]. These phase shifts give rise to unconventional junction properties, such as a spontaneous generation of magnetic flux in the grain boundary junction [19–21]. As described in Ref. [21], the local phase difference $\varphi(x)$ along the grain boundary ($0 < x < b$) can be written as

$$\varphi(x) = \xi(x) + \psi(x)\,, \tag{5}$$

where $\xi(x)$ is a rapidly alternating function accounting for the π phase shifts and for the spontaneously generated magnetic flux in the junction, and $\psi(x)$ is the remaining slowly varying phase difference reflecting the magnetic flux in the interferometer loop. For a narrow junction ($b < \Lambda_J$), ψ is independent of x. With Eq. (5), the time-independent sine-Gordon equation, describing the spatial dependence of the local phase difference over the junction, becomes

$$\Lambda_J^2 \frac{\partial^2 \xi(x)}{\partial x^2} = \frac{j_c(x)}{\langle j_c \rangle} \sin[\psi + \xi(x)]. \qquad (6)$$

The solution $\xi(x)$ of this equation, and thus the pattern of self-generated flux, depends on ψ. The redistribution of this flux by a change of ψ is expected to lead to remarkable deviations from a harmonic dependence for the CPR measured for the entire junction, also if the local CPR is nominally sinusoidal.

Second, it has been proposed [7,8] that the CPR of facets formed by the (110) and by the (100) planes of the adjacent grains is periodic with π and thus has a double periodicity as compared to the standard case. Transmission electron microscopy has revealed that 45° [001] tilt grain boundaries in $YBa_2Cu_3O_{7-x}$ tend to be composed for a considerable part of such facets [22]. For the whole junction, this leads to an anomalous CPR:

$$I = I_{c1} \sin\psi + I_{c2} \sin 2\psi, \qquad (7)$$

by which the observed CPR can be described.

In summary, the current-phase relations of grain boundary junctions with a misorientation of 45° were measured with a modified Rifkin-Deaver method. The CPRs were deduced from measurements of the phase angle between the rf drive current and the rf tank voltage. The current-phase relations of the Josephson junctions showed pronounced deviations from a harmonic behavior, which cannot be accounted for by thermal noise or by other standard mechanisms, but are attributed to the $d_{x^2-y^2}$-wave symmetry of the order parameter and the faceting of the grain boundaries.

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