Phase dependence of the Josephson current in inhomogeneous high- T_c grain-boundary junctions

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We present an experimental and theoretical study of the current-phase relation $I_s(\varphi)$ for 45° grain boundaries in YBa₂Cu₃O_{7-x} films. A model of strongly inhomogeneous Josephson junctions, in which the presence

aries in YBa₂Cu₃O_{7-x} films. A model of strongly inhomogeneous Josephson junctions, in which the presence of randomly alternating current leads to a deviation of $I_s(\varphi)$ from the well-known $\sin(\varphi)$ dependence, has been used. This deviation decreases with the temperature T and is described by the formula $I_s(\varphi) = I_c(T)(\sin\varphi + \gamma(T)\sin 2\varphi)$. Using the developed model, the coefficient γ is calculated and its temperature dependence is found to be in good agreement with experiment. [S0163-1829(99)01417-4]

A fundamental property of a weak link of small size is the relationship between the total supercurrent I_s and the Josephson phase difference φ across a Josephson junction.^{1,2} This relationship has the general form $I_s(\varphi) = I_c f(\varphi)$ and is called the current-phase relation (CPR). For any kind of weak link the CPR has a period of 2π and is an odd function of the Josephson phase.² In particular, the simplest case can be realized for tunnel junctions between two conventional superconductors, where the CPR is given by the equation

$$I_{sT}(\varphi) = I_c \sin(\varphi). \tag{1}$$

Here, I_c is the critical current of the Josephson junction.¹

The deviation $\Delta I_s = I_s - I_{sT}$ from Eq. (1) has been theoretically predicted and experimentally observed in weak links with direct conductivity, i.e., point contacts and SNS (superconductor-normal-metal-superconductor) junctions.^{2,3} It has been shown that in all these cases the deviation of the CPR ΔI_s is *negative*, and the maximum of the supercurrent shifts towards larger φ . The situation is even more complicated in the case of a junction between two high-temperature superconductors.^{4–8} Recently, the observation of a nonsinusoidal CPR for 45° YBa₂Cu₃O_{7-x} grainboundary junctions (GBJ's) has been reported.⁸ In this work it has been observed that the CPR displays a positive deviation ΔI_s and the supercurrent reaches its maximum value for smaller φ . A number of experiments for most high- T_c cuprates is consistent with d-wave symmetry of the order parameter.^{5–7} In the frame of the *d*-wave scenario, strong deviations from the harmonic dependence have been predicted.⁴ However, the positive shift of $I_s(\varphi)$ has not been explained yet and there is an inconsistency between theory and experiment. A problem is that *d*-wave effects are more pronounced in GBJ's with large angles of misorientation, which, however, are strongly inhomogeneous,^{7,9,10} and a randomly distributed alternating current density is present.⁷

On the other hand, a similar highly inhomogeneous regime in a Josephson junction has been observed in the presence of pinned Abrikosov vortices penetrating the superconducting electrodes.^{11,12} A theory of the static and dynamic properties of strongly inhomogeneous Josephson junctions has been developed and has allowed the succesful description of the suppression of the critical current,¹¹ the decrease of the plasma frequency,¹³ and the position and magnitude of the flux flow step.¹⁴

In this paper we report on the measurements of the CPR's in 45° YBa₂Cu₃O_{7-x} GBJ's for different temperatures. We also derive the dependence $I_s(\varphi)$ of a Josephson junction in the presence of a randomly distributed alternating current density. It will be shown below that the predicted CPR is in good agreement with experiment.

We experimentally investigated the CPR by using a single junction interferometer circuit, where the weak link of interest is incorporated in a superconducting ring with a sufficiently small inductance L. In this method an external flux Φ_e is applied to a superconducting ring, and the phase difference $\varphi = 2 \pi \Phi / \Phi_0$ across a weak link is determined by a flux Φ penetrating into the ring.

In the absence of fluctuations and of a quasiparticle current the Josephson phase φ satisfies the general singlejunction interferometer equation

$$\varphi = \varphi_e - \beta f(\varphi), \tag{2}$$

where $\varphi_e = 2\pi \Phi_e / \Phi_0$ and $\beta = 2\pi L I_c / \Phi_0$ are the normalized external flux and critical current, respectively.¹⁵

When the condition $\beta < 1$ is valid one can obtain the CPR for the complete phase range. Thus by monitoring φ with a superconducting flux detector as a function of φ_e , the CPR $\beta f(\varphi)$ has been obtained.¹⁶ The application of this method is technically difficult because of the thin-film design and the required small values of the interferometer inductance. In this respect the method developed in Ref. 17 has a great advantage. The interferometer is coupled through a mutual inductance *M* to the tank circuit with a known impedance *Z*. In this arrangement the effective impedance Z_{eff} of the whole system depends on φ_e and the CPR can be obtained from the measurement of this dependence.¹⁷

For impedance measurements a driving radio-frequency current I_{rf} as well as a sweep current I_{dc} are supplied to the

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tank circuit. The external flux in the ring is generated by the total current I_{tot} flowing in the tank coil, i.e., $\varphi_e = 2\pi I_{\text{tot}}M/\Phi_0 = \varphi_{dc} + \varphi_{rf}$. It has been shown¹⁸ that in the underdamped regime, when the quasiparticle current is negligible and in the limit of a small signal ($\varphi_{rf} \ll 1$) the following equations are valid:

$$d\varphi = \frac{d\varphi_{dc}}{1 + \beta f'[\varphi(\varphi_{dc})]},$$

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

where k is a coupling coefficient and Q is the quality factor of the tank circuit. Here, $\alpha(\varphi_{dc})$ is the phase angle between the drive current I_{rf} and the tank voltage U. Making use of Eqs. (3) one can determine the CPR from the experimental data of $\alpha(\varphi_{dc})$.

All layers for the GBJ's were grown using the pulsed laser deposition technique on (100) oriented SrTiO₃ bicrystals with a [001] tilt angle of 45°. Two types of bicrystals were used: symmetric $(22.5^{\circ}/22.5^{\circ})$ and asymmetric $(19^{\circ}/26^{\circ})$ ones. Typical values for T_c and j_c (77 K) of the YBa₂Cu₃O_{7-x} films without artificial GB are 89 K and 2 $\times 10^6$ A/cm², respectively. Patterning of the layers was done using photolithography and Ar ion-beam etching. YBa₂Cu₃O_{7-x} layers were etched into 5×5 mm² or 4×4 mm² square-washer single junction interferometer structures. The washer holes had a side length of 50 μ m, leading to $L \approx 80$ pH. Layer thicknesses t = 70-100 nm and junction widths $W = 1-2.5\mu$ m were chosen to fulfill the condition $\beta < 1$ at the desired temperature.

Several tank circuits with inductances between 0.3 and 0.8 μ H and resonance frequencies between 15 and 35 MHz have been used for the measurements. The unloaded quality factor has been measured for all tank circuits at various temperatures and values 70 < Q < 150 were obtained. A driving rf current I_{rf} was supplied to the tank circuit and a voltage U across the circuit was measured using an amplifier at room temperature with a high input impedance. The amplitude and phase of the voltage were detected by a vector analyzer and recorded as a function of either the amplitude of I_{rf} or I_{dc} . The coupling coefficient k was extracted from the period ΔI_{dc} of the α - I_{dc} plot according to the relation $M\Delta I_{dc}$ $=\Phi_0$. The value of k was between 0.03 and 0.1. The interferometer was placed inside a five-layer magnetic shielding in a gas-flow cryostat at a temperature in the range of 4.2 < T < 90 K.

The examples of dependencies of the phase angle α on an external magnetic flux (dc current flowing in tank coil, I_{dc}) are shown in Fig. 1. The validity of a small signal limit and an underdamped regime were proven experimentally. First, from the $U(I_{rf})$ curves we defined the amplitude of I_{rf} generating a flux Φ_0 in the interferometer. Then, all measurements were carried out at the amplitude of I_{rf} corresponding to a flux $(0.05\cdots 0.1)\Phi_0$ ensuring the small signal limit. The underdamped regime has been confirmed by the measurement of both the resistance of a junction fabricated under identical condition and the tank circuit quality factor change in the presence of the interferometer ring. Our estimations



FIG. 1. The dependencies of phase angle α on a dc current I_{dc} flowing through the tank coil for various temperatures.

show that the quasiparticle current is negligible. The examples of the CPR's for two different temperatures, obtained from the $\alpha(\varphi_{dc})$ dependencies (Fig. 1), and Eqs. (3) are shown in Fig. 2. The curves are found to display a positive shift ΔI_s . Moreover, we obtain that this deviation decreases with temperature.

In the following, we present a theory of the CPR $I_s(\varphi)$ in a strongly inhomogeneous small Josephson junction. In these junctions the superconducting current density changes sign, and randomly depends on the coordinate 0 < x < W along the junction. Such a highly inhomogeneous regime can be obtained in the presence of pinned Abrikosov vortices penetrating the superconducting electrodes^{11,12} or in a nonuniform (meandering) grain-boundary Josephson junction between two *d*-wave superconductors.⁷ The alternating currentdensity fluctuations present in strongly inhomogeneous Josephson junctions can be characterized by an internal disordered static phase distribution $\varphi_V(x)$.¹⁴ Note here that a similar model has been used in Ref. 19.

We consider a small Josephson junction of a size $W \ll \lambda_J$, where the Josephson penetration depth $\lambda_J = \sqrt{(\hbar c^2 W t/16 \pi e \lambda_L I_c)}$ is mostly determined by the critical current I_c of a junction. Here, λ_L is the London penetration depth in superconductors.

The dependence of a static Josephson phase φ on x in the presence of inhomogeneities is determined by a general Ferrel-Prange equation:^{15,14}

$$\frac{d^2\varphi(x)}{dx^2} - \frac{1}{\lambda_{J0}^2} \sin[\varphi_V(x) + \varphi(x)] = 0, \qquad (4)$$



FIG. 2. The CPR's restored from the $\alpha(I_{dc})$ dependencies shown in Fig. 1. The fit of CPR's by Eq. (9) is shown by solid lines.

where $\lambda_{J0} = \sqrt{(\hbar c^2/16\pi e \lambda_L j_0)}$ and j_0 is the local critical current density. In inhomogeneous Josephson junctions the limit $\lambda_{J0} \ll \lambda_J$ is valid. The Josephson phase satisfies the boundary condition $\varphi|_{x=0} = \varphi_0$.

The superconducting current across the junction can be expressed as follows:

$$I_s = j_0 t \int_0^W dx \sin[\varphi_V(x) + \varphi(x)].$$
(5)

Taking into account a finite size W of the small junction we obtain

$$I_{s} = j_{0}t \int_{0}^{W} dx \left\{ \sin[\varphi_{0} + \varphi_{V}(x)] + \frac{1}{2\lambda_{J0}^{2}} \int_{0}^{x} dx_{1} \int_{0}^{x_{1}} dx_{2} \sin[2\varphi_{0} + \varphi_{V}(x) + \varphi(x_{2})] \right\}.$$
(6)

At this point it is convenient to introduce the complex variable $\eta(x)$ that randomly depends on the coordinate *x*, as follows:

$$\eta(x) = \int_0^x dy \exp i\varphi_V(y).$$
(7)

All characteristics of the inhomogeneous Josephson junction can be expressed with this variable. So, the critical current is given by

$$I_c = j_0 t |\eta(W)|. \tag{8}$$

Making use of the variable $\eta(x)$ we rewrite Eq. (6) in the form:

$$I_{S}(\varphi) = I_{c}(\sin \varphi + \gamma \sin 2\varphi),$$

$$\gamma = \frac{W^{2}}{2\lambda^{2}} \operatorname{Re} \int_{0}^{W} \frac{dx}{W} \frac{\eta(x)}{\eta(W)} \left[1 - \frac{\eta(x)}{\eta(W)} \right], \qquad (9)$$

where the Josephson phase $\varphi = \varphi_0 + \xi$ and the random angle ξ , given by the formula $e^{i\xi} = \eta(W)/|\eta(W)|$.

Equation (9) allows us to obtain the deviation of the CPR from Eq. (1) in the presence of inhomogeneities. We notice first that the CPR is determined by the behavior of the variable $\eta(x)$. In a homogeneous case when the disorder is absent the variable $\eta(x)$ is simply proportional to *x* and in the limit of a small Josephson junction $W \ll \lambda_J$ the parameter $\gamma = (1/12)(W/\lambda_J)^2$ is small. The deviation ΔI_s in this case is negligible.

A more complex scenario occurs in the inhomogeneous Josephson junctions in which the variable $\eta(x)$ randomly depends on x, and the particular value of γ and therefore ΔI_s are determined by the type of inhomogeneities present in a junction.

We stress here that even in strongly disordered GBJ's between two conventional (*s*-wave) superconductors, when the fluctuations of current density are large but do not change sign, the $\eta(x) \propto x$ and ΔI_s is negligible.

On the other hand, in a Josephson junction with periodically distributed alternating fluctuations of the current den-



FIG. 3. Experimental dependencies of the critical current I_c (circles) and parameter γ (squares) on the temperature for asymmetric (19°/26°, open circles and squares) and symmetric (22.5°/22.5°, solid circles and squares) samples.

sity, $\eta(x)$ is a periodic function x, $\eta(W)$ is always small and the deviation of the CPR is large and negative. It allows us to recover a result of Ref. 19 in which a Josephson junction in the presence of the alternating fluctuations of the current density with particular wave vectors has been considered.

In the presence of randomly distributed strong inhomogeneities, when the variable $\eta(x)$ is a random function x, we can express our results in terms of a generalized correlation radius ρ for the phase disorder $\varphi_V(x)$. This quantity is defined as

$$\rho = \int_0^W dx \langle \cos[\varphi_V(x) - \varphi_V(0)] \rangle. \tag{10}$$

In the case in which this correlation radius is small, $\rho \ll W$, we use the well established methods of random potential theory,²⁰ and the typical value of $\gamma(z = |\eta(W)|)$ is given by²¹

$$\gamma(z) = \left(\frac{W}{\lambda_J}\right)^2 \frac{1}{4} \left(\frac{2}{3} \pm \frac{\sqrt{\rho W}}{z} \pm \frac{\rho W}{3z^2}\right). \tag{11}$$

The parameter γ depends strongly on the particular value of $z = |\eta(W)|$ and can be large with respect to the homogeneous



FIG. 4. Experimental (circles) and theoretical (solid line) dependencies of parameter γ vs I_c for asymmetric (19°/26°, open circles) and symmetric (22.5°/22.5°, solid circles) samples.

case, when the value of $z \le \sqrt{\rho W}$. In our particular model the parameter γ changes sign from sample to sample and can be positive.

Using the developed theory we can explain all important features of experimentally observed CPR's. First, we show in Fig. 2, by solid lines, that the CPR's with the positive deviations ΔI_s are fitted very well by Eq. (9) with temperature dependent critical current I_c and a positive parameter γ . The temperature dependencies of both these quantities for two samples are shown in Fig. 3. Moreover, Eqs. (9) and (11) display an interesting scaling between γ and I_c . We obtain that $\gamma \propto I_c$, with a coefficient depending on the width of the junction and a particular distribution of inhomogeneities. This scaling is shown in Fig. 4. Note that the CPR's reported in Ref. 8 are fitted by Eq. (9) as well. As follows from the model, when current inhomogeneities rarely change sign or simply have a preferential direction, the critical current I_c is larger with respect to the "true" inhomogeneous case and the CPR approaches $sin(\varphi)$. This also has been observed experimentally. For two samples fabricated under the same conditions, I_c was one order of magnitude larger than for the samples described above. In this case the CPR's could be measured at T>60 K only, because below 60 K the parameter β was larger than one. For both samples at temperatures between 60 and 80 K, a nearly perfect sinusoidal CPR has been obtained.

In conclusion, we show that the observed positive deviation of the CPR and, moreover, its temperature dependence, can be explained consistently in the model of alternating fluctuations of the superconducting current density. The presence of these fluctuations in our case strongly supports the model of *d*-wave symmetry of the order parameter for YBa₂Cu₃O_{7-x} superconductors.

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