Degenerate Ground State in a Mesoscopic YBa$_2$Cu$_3$O$_{7-x}$ Grain Boundary Josephson Junction

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We have measured the current-phase relationship $I(\varphi)$ of symmetric 45$^\circ$ YBa$_2$Cu$_3$O$_{7-x}$ grain boundary Josephson junctions. Substantial deviations of the Josephson current from conventional tunnel-junction behavior have been observed: (i) The critical current exhibits, as a function of temperature $T$, a local minimum at a temperature $T^*$. (ii) At $T = T^*$, the first harmonic of $I(\varphi)$ changes sign. (iii) For $T < T^*$, the second harmonic of $I(\varphi)$ is comparable to the first harmonic, and (iv) the ground state of the junction becomes degenerate. The results are in good agreement with a microscopic model of Josephson junctions between $d$-wave superconductors.

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The most important phenomenological difference between the high-$T_c$ cuprates and conventional superconductors regards the orbital symmetry of the superconducting order parameter. In the cuprates the pair potential changes sign depending on the direction in momentum space according to $[1,2] \Delta(q) = \Delta_0 \cos(\varphi - \theta)$, where $\varphi$ is the angle between the wave vector and the (laboratory) x axis, while $\theta$ is the angle between the Cu-Cu bond direction of the superconductor and the x axis. This unconventional $d$-wave symmetry was predicted [3] and experimentally confirmed [1,2] to be directly measurable in the Josephson effect between a high $T_c$ and a conventional superconductor. Another consequence of the $d$-wave symmetry is that midgap states (MGS) with energy $\epsilon = 0$ should form on the free surface of a $d$-wave superconductor if $\Delta(q)$ has opposite signs on incident and reflected electronic trajectories [4]. The MGS density must be maximal for (110)-like surfaces and this prediction has in fact been confirmed by STM microscopy on YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) single crystals [5] which revealed the MGS contribution to the YBCO tunneling density of states. The presence of the MGS is expected to influence in a spectacular way also the Josephson effect in junctions between $d$-wave superconductors with different crystallographic orientations. Yet no clear manifestation of the MGS in the Josephson effect in such junctions has been observed so far, which is a challenge for the concept of $d$-wave superconductivity in the cuprates.

Moreover, due to possible applications in quantum computing [6,7], there is substantial interest in Josephson junctions and circuits with a doubly degenerate ground state. Such a state was predicted in an asymmetric 45$^\circ$ junction ($\theta_1 = 0^\circ$ and $\theta_2 = 45^\circ$, the angles $\theta_1, \theta_2$ are defined in Fig. 1), since odd harmonics of the Josephson current $I(\varphi) = \sum_n I_n \sin n \varphi$ are suppressed by symmetry [8,9]. The current-phase relation observed in Ref. [10] indeed showed a substantial contribution of the second harmonic $I_2$. However, there is a finite supercurrent flowing along the interface in the ground state of asymmetric 45$^\circ$ junctions [9]. Therefore they do not lead to completely quiet qubits in the sense of Ref. [6].

Motivated by the search for both, the MGS in high-$T_c$ Josephson junctions and a quiet qubit, we have studied symmetric 45$^\circ$ junctions (i.e., junctions with $\theta_1 = -\theta_2 = 22.5^\circ$). In this paper we report the first direct observation of several effects exclusive to such junctions: temperature controlled sign change of the first harmonic of the Josephson current, a nonmonotonic temperature dependence of the critical current, and the development of a doubly degenerate ground state of the system.

Let us start with a description of the theoretical predictions for Josephson junctions between $d$-wave superconductors. In symmetric short junctions the Josephson current density is conveniently described in terms of the Andreev levels in the junction as [11]

$$j(\varphi) = \frac{k_F}{\Phi_0 d} \sum_n \int_{-\pi/2}^{\pi/2} d\theta \cos \theta f(\epsilon_n(\varphi, \theta)) \frac{\partial \epsilon_n(\varphi, \theta)}{\partial \varphi},$$

where $\varphi$ is the superconducting phase difference between the banks, $\Phi_0$ is the magnetic flux quantum, $k_F$ is the Fermi momentum, $d$ is the average separation of the CuO$_2$ planes, $f(\epsilon)$ is the Fermi distribution function, and $\epsilon_n(\varphi, \theta)$ is the energy of the nth Andreev level for an electron incident on the junction at an angle $\theta$ with respect to the boundary normal. At a given $\theta$ there exist only two Andreev levels with energies $\pm \epsilon(\varphi, \theta)$. 
The nature of the Andreev levels changes with the impact angle $\vartheta$: (i) For $22.5^\circ < |\vartheta| < 67.5^\circ$, MGS are formed at $\varphi = 0$ whose energy is split by a finite phase difference $\varphi$ across the contact. In this range of impact angles $\epsilon(\varphi)$ can be qualitatively described by $\epsilon_{\text{MGS}}(\varphi) = \Delta(\pi/4) \sin(\varphi/2) \sqrt{D(\pi/4)}$, where $0 \leq D(\vartheta) \leq 1$ is the angle-dependent barrier transparency [12]. (ii) For $|\vartheta| < 22.5^\circ$ and $67.5^\circ < |\vartheta| < 90^\circ$, no MGS are formed at $\varphi = 0$ and the Andreev levels resemble those formed in a Josephson junction between s-wave superconductors. In a symmetric junction the dimensionless units $\varphi_{\text{dc}} = 2\pi \Phi_{\text{dc}}/\Phi_0$. The phase difference across the junction $\varphi$ was calculated from the $\alpha(\varphi_{\text{dc}})$ data using the coupling coefficient between the rf SQUID and the tank coil $k^2 = 2.6 \times 10^{-3}$ and $3.6 \times 10^{-3}$ for samples No. 1 and 2, respectively. After inverting the $I(\varphi) = I(\varphi_{\text{dc}})$ function, $I(\varphi)$ can be obtained from $\beta f(\varphi) = \varphi_{\text{dc}}(\varphi) - \varphi$, where $f(\varphi) = I(\varphi)/I_c$, $\beta = 2\pi L I_c/\Phi_0$, and $L = 80$ pH is the inductance of the rf SQUID. The details of the experimental method are given elsewhere [15].

The measured $\alpha(\varphi_{\text{dc}})$ curves shown in Fig. 2 exhibit local minima at low $T$ when $\varphi_{\text{dc}}$ is a multiple of $2\pi$. This is qualitatively different from what has been observed before for 45° symmetric grain boundary Josephson junctions on samples with $w_s > 1 \mu$m where no such minima were found [16]. We believe that the difference is caused by the existence of the bicrystal boundary defects with a typical

![FIG. 1. Schematic picture of the rf SQUID.](image-url)
The Josephson current calculated from the measured \( \alpha(\varphi_{dc}) \) data is shown in Fig. 3. Note the anomalous form of \( I(\varphi) \) at low temperatures. The anomalies in sample No. 2 are much more pronounced than in sample No. 1. We believe this is a combined effect of smaller junction cross sections and higher junction quality, as evidenced by the much smaller values of \( I_c \) in sample No. 2. In Fig. 4 we plot the first two harmonics \( I_1 \) and \( I_2 \) for the sample No. 2. The most striking result is that for \( T^* \approx 12 \) K, \( I_1 \) changes sign. In the same temperature region where \( I_1 \) starts to exhibit a downturn, the value of \( |I_2| \) rises from the negligible high-\( T \) values to values comparable to \( |I_1| \) at low \( T \), suggesting a common origin of both phenomena. Furthermore, Fig. 4 shows that close to \( T^* \), there is a local minimum of the critical current \( I_c \) as a function of \( T \), which is associated with the sign change of \( I_1 \). These results are in a qualitative agreement with theoretical predictions for \( I(\varphi) \) of 45° junctions with ideally flat interfaces [8,17].

We can reconstruct the free energy \( F \) of the junction as a function of \( \varphi \) from \( F(\varphi) = (\Phi_0/2\pi) \int_0^\infty d\phi I(\phi) \). The result is shown in Fig. 5. Note that for \( T \leq 15 \) K, the free energy minimum of the sample No. 2 moves away from \( \varphi = 0 \), and the \( F(\varphi) \) curve exhibits two degenerate minima at \( \varphi = \pm \varphi_0 \), as observed previously in Ref. [10] on asymmetric 45° junctions; see Fig. 5.

In Fig. 4 we compare the experimental data with a theoretical treatment based on the quasiclassical Eilenberger equations which was introduced in the \( s \)-wave case in Ref. [18] and will be described in detail elsewhere. Within our approach the junction is described by two phenomenological parameters, the junction transparency \( D \) and the roughness parameter \( 0 < \rho < \infty \). The temperature dependence of \( I_1, I_2, \) and \( I_c \) is fit well by our theory with \( D = 0.3 \) and \( \rho = 0.3 \). The theoretical \( I_c \) is reported in

![Image](https://example.com/image1.png)

**Fig. 2.** The phase angle \( \alpha \) as a function of \( \varphi_{dc} \) measured at different temperatures for samples No. 1 (a) and No. 2 (b). From top to bottom, the data correspond to (a) \( T = 30, 20, 15, 10, 4.2, \) and 1.8 K and (b) \( T = 35, 30, 25, 20, 15, 11, 10, 5, \) and 1.6 K. The data are vertically shifted for clarity.

![Image](https://example.com/image2.png)

**Fig. 3.** \( I(\varphi) \) for samples No. 1 (a) and No. 2 (b). From top to bottom at \( \varphi / \pi = 0.5 \), the data correspond to (a) \( T = 30, 20, 15, 10, 4.2, \) and 1.8 K and (b) \( T = 20, 25, 30, 35, 15, 11, 10, 5, \) and 1.6 K. The data in Fig. 3b corresponding to \( T = 20, 25, 30, \) and 35 K were multiplied by a factor 4 for clarity.

![Image](https://example.com/image3.png)

**Fig. 4.** The critical current \( I_c \) (triangles) and the harmonic components \( I_1 \) (squares) and \( I_2 \) (circles) of the Josephson current as a function of temperature for sample No. 2. The figure is obtained by the Fourier analysis of \( I(\varphi) \) shown in Fig. 3b. Inset: Theoretical prediction for the temperature dependence of \( I_c, I_1, \) and \( I_2 \) for a junction with \( D = 0.3 \) and \( \rho = 0.3 \). The current densities are plotted in units of the Landau critical current density; the temperature is in units of \( T_c \).
The zero of energy has been set so that \( F(0) = 0 \). (a) Asymmetric 45° grain boundary (Ref. [10]). Top to bottom curves correspond to \( T = 30, 20, 15, 10, \) and \( 4.2 \) K, respectively. (b) Symmetric 45° grain boundary (present work). Top to bottom curves correspond to \( T = 20, 15, 11, 10, 5, \) and \( 1.6 \) K, respectively.

In conclusion, we have found that symmetric 45° junctions exhibit doubly degenerate ground states. Therefore they are of potential use in superconducting qubit fabrication. The qubits based on asymmetric 45° junctions are not quiet [6], since there exists spontaneously generated flux along the interface [21] which should change sign between the two different ground states of the junction. The qubits based on symmetric junctions might be close to being completely quiet, since their spontaneously generated flux is unmeasurably small [21]. The key technological question is how to increase the energy barrier between the two degenerate minima at \( \varphi = \pm \varphi_0 \). An interesting possibility seems to be to increase the barrier transparency by an appropriate doping of the grain boundary [22].

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