Dynamical Effects of an Unconventional Current-Phase Relation in YBCO dc SQUIDs

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The predominant d-wave pairing symmetry in high-temperature superconductors allows for a variety of current-phase relations in Josephson junctions, which is to a certain degree fabrication controlled. In this Letter, we report on direct experimental observations of the effects of a nonsinusoidal current-phase dependence in YBCO dc SQUIDs, which agree with the theoretical description of the system.

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It is well established [1] that the wave function of a Cooper pair in most cuprate high-temperature superconductors (HTS) has a d-wave symmetry. Its qualitative distinction from, e.g., the anisotropic s-wave case is that the order parameter changes sign in certain directions, which can be interpreted as an *intrinsic* difference in the superconducting phase between the lobes equal to π.

The latter leads to a plethora of effects, such as formation of Andreev bound states at surfaces and interfaces in certain crystallographic orientations [2–4]. The current-phase dependence $I_S(\phi)$ in Josephson junctions formed by dd junctions, as well as by sd junctions comprised of a cuprate and a conventional superconductor, depends both on the spatial orientation of the d-wave order parameter with respect to the interface, and on the quality of the latter [5–9]. Time-reversal symmetry can also be spontaneously violated and thus spontaneous currents generated [10–12]. Another effect can be doubling of the Josephson frequency [6,13,14].

In this Letter, we report on experimental observations of strong effects of an unconventional current-phase relation on the dynamics of two dd junctions integrated into a superconducting interference device (SQUID) configuration.

Since $I_S(\phi)$ must be a 2π-periodic odd function, it can be expanded in a Fourier series. In most cases, only the first two harmonics give a significant contribution to the current:

$$I_S(\phi) = I_c^{\mathrm{I}} \sin\phi - I_c^{\mathrm{II}} \sin 2\phi. \tag{1}$$

In Josephson systems of conventional superconductors, the second harmonic will usually be negligible [15] but in dd junctions the second harmonic may dominate. If $I_c^{\mathrm{II}} > I_c^{\mathrm{I}}/2$, the equilibrium state is no longer $\phi = 0$ but becomes double degenerate at $\phi = \pm \arccos(I_c^{\mathrm{I}}/2I_c^{\mathrm{II}}) \rightarrow \pm\pi/2$. The system can then spontaneously break time-reversal symmetry by choosing either state. Spontaneous currents as well as fluxes can be generated in this state. The potential will have the shape of a double well, and there are reasons to believe that it will be possible to observe quantum coherence in this system. The presence of a second harmonic in the current-phase relation (CPR) of a dd junction was confirmed by Il'ichev *et al.* [8].

A nonsinusoidal CPR of the junctions will change the dynamics of a dc SQUID [16]. Regarding the junctions as magnetically small, the supercurrent through the SQUID in the presence of an external flux $\Phi_x \equiv \Phi_0 \cdot (\phi_x/2\pi)$ can be written as

$$I_s(\phi, \phi_x) = I_{c1}^{\mathrm{I}} \sin\phi - I_{c1}^{\mathrm{II}} \sin(2\phi) + I_{c2}^{\mathrm{I}} \sin(\phi + \phi_x) - I_{c2}^{\mathrm{II}} \sin 2(\phi + \phi_x). \tag{2}$$

The critical current through the SQUID is given by the usual expression $I_c(\phi_x) = \max_\phi I_s(\phi, \phi_x)$. The time-averaged voltage over the SQUID in the resistive regime is readily obtained in the resistively shunted junction approximation. By introducing $\delta[\phi, \phi_x] = \phi_2 - \phi_1$ and applying the same method as in [17] with the necessary generalizations, we obtain the following for the average voltage over the SQUID:

$$\overline{V}^{-1} = \frac{G_1 + G_2}{2\pi} \int_{-\pi}^{\pi} d\phi \left[I - (G_1 - G_2)\frac{\hbar}{2e}\frac{d\delta}{dt} - I_1\left(\phi + \frac{\delta}{2}\right) - I_2\left(\phi - \frac{\delta}{2}\right)\right]^{-1}. \tag{3}$$

Here $G_{1,2}$ are the normal conductances of the junctions, and

$$\delta + \phi_x + \frac{\pi L}{\Phi_0}[I_2(\phi - \delta/2) - I_1(\phi + \delta/2)] = 0 \tag{4}$$

gives the difference, δ, in phase drops across each junction. In deriving (3) and (4), we have assumed that the inductance L is equally divided between the SQUID arms. We have also neglected the spontaneous magnetic fluxes in the dd junctions, due to their small amplitude [11,18]. Though (4) is only explicitly solvable in the limit

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$L \to 0$, it always yields $\delta[-\phi, -\phi_x] = -\delta[\phi, \phi_x]$. This means that the usual inversion symmetry is retained.

The results of numerical calculations based on (2) and (3) are shown in Fig. 1. The cusps in the critical current correspond to the points at which the global maximum in (1) switches from one local maximum to another [16]. Note the quasi-$\Phi_0/2$ periodicity of the current isolines in the $\overline{V} - \phi_x$ picture, reflecting the current-phase dependence (1), and their shift along the Φ_x axis, which depends on the sign of the bias current (as it must to maintain the central symmetry with respect to the origin). The shift does not depend on the magnitude of the current since we neglect the self-inductance. For large biases, the Φ_0 periodicity is restored. Indeed, as the bias grows, one set of minima of the washboard potential, $U = (h/2e)[-I^{\mathrm{I}}\cos\phi + (I^{\mathrm{II}}/2)\cos 2\phi - I\phi]$, disappears first unless the first harmonic I^{I} is *exactly* zero.

We have fabricated and studied a large number of dc SQUIDs. The samples were fabricated from 250 nm thick YBCO films deposited on $SrTiO_3$ bicrystals. The grain-boundary junctions (GBJs) are of the asymmetric [001]-tilt type with the misorientation angle of 45° (0°–45° GBJ). For more information on GBJs see, for example, Ref. [19].

The pattern was defined using *E*-beam lithography and then transferred to a carbon mask employing a multistep process. Finally, the YBCO is etched through the mask using ion milling. This scheme allows us to fabricate high-quality bicrystal junctions as narrow as 0.2 μm, as has been reported elsewhere [20]. In the SQUIDs under investigation, the junctions are nominally 2 μm wide; hence, the fabrication-induced damage of the junctions is small.

The measurements were done in an EMC-protected environment using a magnetically shielded LHe cryostat. However, the magnetic shielding is imperfect, as is evident from the fact that the expected zero-field response of our SQUIDs is not exactly at zero. The measuring electronics is carefully filtered and battery powered whenever possible. In order to measure the dependence of the critical current on the applied field, we used a voltage discriminator combined with a sample-and-hold circuit. All measurements reported here were performed at 4.2 K.

The SQUID loops are (15×15) μm^2. The numerically calculated inductance [21] is approximately 25 pH, yielding the factor $\beta = 2\pi L I_c/\Phi_0$ between 0.5–2.

The SQUIDs were largely nonhysteretic with a resistance of about 2 Ω. The measured critical current varies from sample to sample but is in the range of tens of microamperes giving a current density of the order of $J_c = 10^3$ A/cm^2. The estimated Josephson penetration length $\lambda_J = \Phi_0/\sqrt{4\pi\mu_0 j_c \lambda_L}$ is approximately 2 μm in all junctions, which means that the junctions are magnetically short. This is supported by the quasiperiod of the pattern in Fig. 2 being close to the expected value $\phi_0/2\lambda_L w$ [17]. The differential conductance curves do not show any trace of a zero bias anomaly (ZBA), as is expected for 0°–45° GBJs. ZBAs have been observed by other groups in GBJs with other orientations [2].

The critical current is plotted as a function of applied magnetic field for two SQUIDs in Fig. 3. The result is in qualitative agreement with theory if we assume that the SQUID junctions have different ratios of the first and second harmonics of the critical current. This assumption

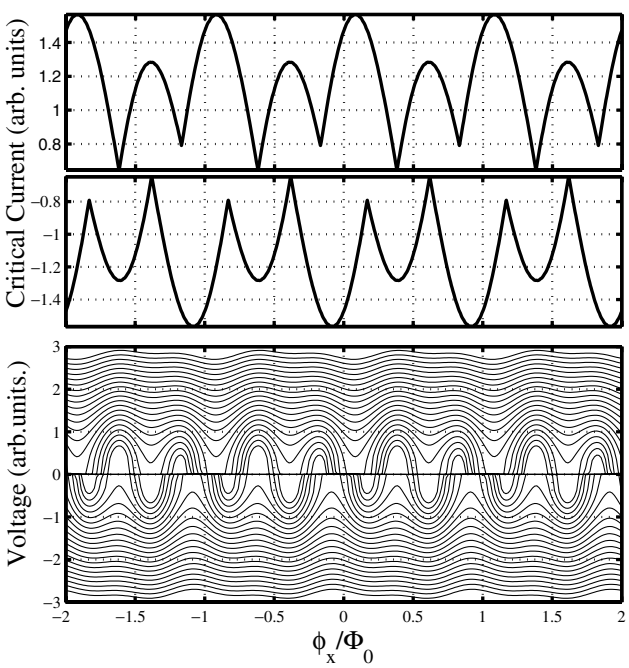

FIG. 1. The results of simulations of the $I_c - \phi_x$ and $\bar{V} - \phi_x$ dependence for a dc SQUID with $I_{c1}^{\mathrm{I}} = 1$, $I_{c2}^{\mathrm{I}} = 0.1$, $I_{c1}^{\mathrm{II}} = 0.2$ and $I_{c2}^{\mathrm{II}} = 0.4$ (arbitrary units). The different curves correspond to bias currents in the range $I = I_{c1}^{\mathrm{I}}$ to $I = 5I_{c1}^{\mathrm{I}}$. We assume $L = 0$ and $G_1 = G_2$.

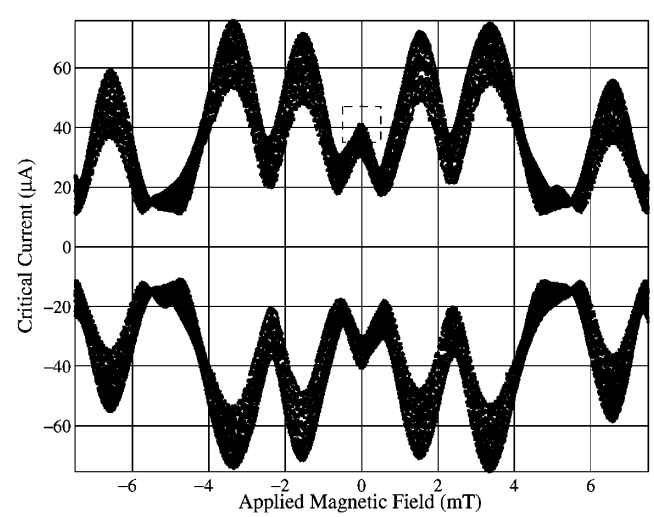

FIG. 2. Critical current as a function of magnetic field at 4.2 K. The dashed box indicates the area plotted in Fig. 3(a).

is supported by the fairly small modulation depth [it is easy to see from Eq. (2) that I_c would go exactly to zero in a SQUID with junctions of identical I_{c2}/I_{c1}].

We can fit the data to Eq. (2), if we compensate for the residual background magnetic field and assume that we have a small excess current (of the order of a few μA) in the junctions. The fitting parameters again confirm that there is a large asymmetry between the arms of the SQUIDs. Note that the model does not consider the flux penetration into the junctions,

The result for fields of the order of mT is presented in Fig. 2, which shows the I_c modulation of the SQUID enveloped by an anomalous Fraunhofer pattern quite similar to what has been reported by other groups [22,23] for 0°–45° GBJs. Note the inversion symmetry of the pattern with respect to the origin. That the global maximum is not in the center can be explained in several ways; it has been shown, for example, that this could be due to the presence of so-called π loops in the junction interface [24].

Figure 4 shows the V-B dependence of one of the SQUIDs. The pattern is again field inversion symmetric. The overall structure is the same as in the model dependence of Fig. 1, but there is also an additional shift due to self-field effects, which depends on the magnitude of the bias current and corresponds (at maximum) to a flux $\sim 0.1\Phi_0$. In a beautiful experiment, a similar dependence was recently observed by Baselmans *et al.* in a Nb-Ag-Nb SNS junction where current injectors were used to change the occupation of current-carrying states in the normal region [25]. A deviation from the model occurs at $\overline{V} = 100\ \mu$V where the minima and maxima switch. This is probably due to an LC resonance in the SQUID. Taking $L = 25\ p$H, this would require $C = 0.8\ pF$, which agrees with our measurements on single junctions

Remarkably, the observed offset of the V-B characteristics with respect to the *direction* of the bias current appears to be a much more robust manifestation of the presence of a second harmonic of the Josephson current than the shape of the $I_c - B$ curves itself. We observed the shift even in SQUIDs with the smallest junctions down to 0.5 μm wide, where the deviations from the usual sinusoidal CPR were not obvious from the $I_c - B$ dependence.

Generally, the nature of the transport through a GBJ will depend on its transmissivity D. Il'ichev *et al.* [8] have reported values of D as high as 0.3 in symmetric (22.5°–22.5°) dd junctions as opposed to the usual estimate for a GBJ, $D \sim 10^{-5}$–10^{-2}. Since usually $I_c^{\mathrm{II}}/I_c^{\mathrm{I}} \propto D$, a high-transmissivity GBJ is required in order

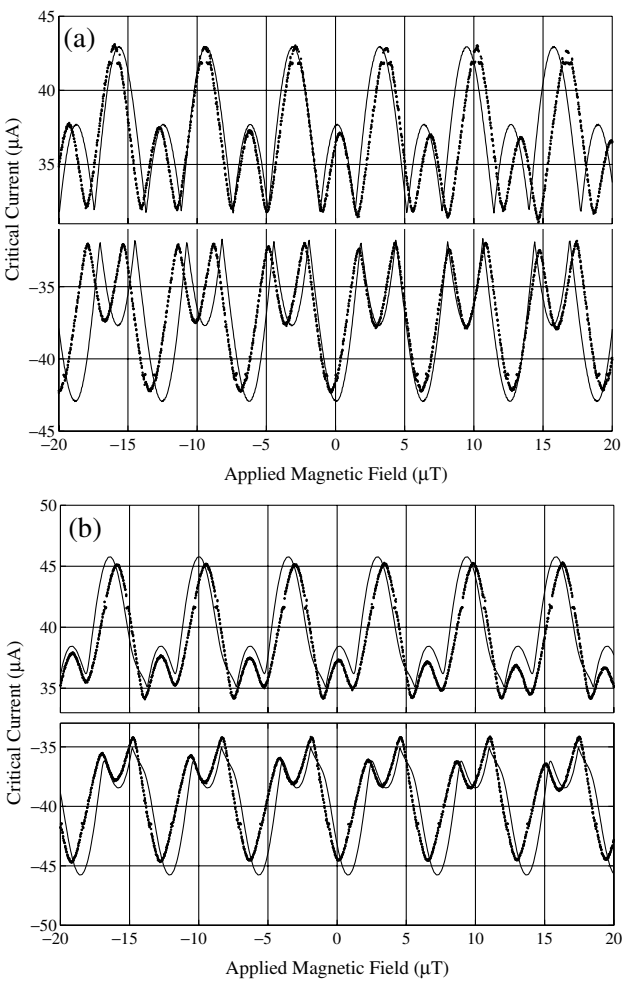

FIG. 3. Critical current as a function of applied magnetic field for two different SQUIDs that are nominally identical. The solid line represents the fitted expression. The fitting parameters are as follows: (a) $I_{c1}^{\mathrm{I}} = 9\ \mu$A, $I_{c2}^{\mathrm{I}} = 0.3\ \mu$A, $I_{c1}^{\mathrm{II}} = 3.7\ \mu$A, and $I_{c2}^{\mathrm{II}} = 22.7\ \mu$A; (b) $I_{c1}^{\mathrm{I}} = 7.8\ \mu$A, $I_{c2}^{\mathrm{I}} = 3.0\ \mu$A, $I_{c1}^{\mathrm{II}} = 5.3\ \mu$A, and $I_{c2}^{\mathrm{II}} = 4.3\ \mu$A. In both cases, the fit has been adjusted with respect to the residual background field and the excess current of the junctions.

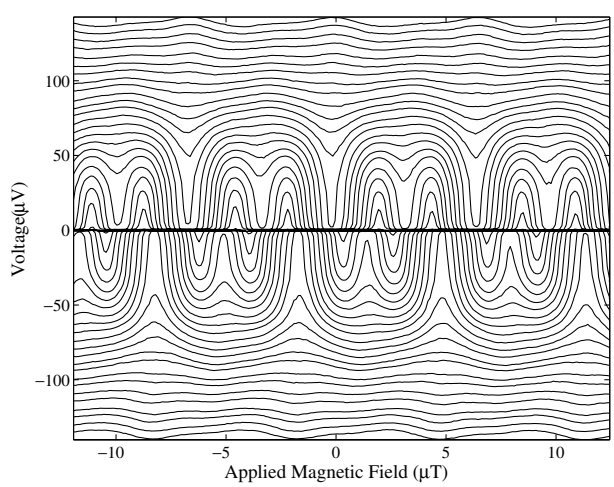

FIG. 4. Voltage modulation as a function of applied magnetic field for the SQUID whose $I_c - B$ is shown in Fig. 3(a). The pattern is again inversion symmetric. Note the sign change at 100 μV, which we believe is due to a LC resonance in the SQUID loop.

to observe effects of the second harmonic. An estimate of the *average* transmissivity of our junctions would be $\rho_{ab} l/R_N A \sim 10^{-2}$ [26] assuming l, the mean-free path, to be equal to 10 nm and a resistivity in the a-b plane ρ_{ab} equal to 10^{-4} Ω cm. This is still too low to explain the strong second harmonic we observe. However, it is known from, e.g., TEM studies [19], that the grain-boundary is far from uniform; the properties can significantly vary depending on the local properties of the interface, effects such as oxygen diffusion out of the GB, etc., which are difficult to control. It is therefore reasonable to assume that there are many parallel transport channels through the GB [27,28]. Channels with high-transmissivity dominate the transport and might have $D \sim 0.1$ even though the *average* transmissivity is much lower. This is also consistent with the fact that most of our SQUIDs seem to be highly asymmetrical which is to be expected if the distribution of channels is random. The ratios of I_c^{I} and I_c^{II} can vary as much as 10 times between two junctions in the same SQUID, even though the fluctuations of the *total* I_c from sample to sample are much smaller. It is also clear from general considerations that a high value of I_c^{II} *excludes* a high value of I_c^{I}, since the second harmonic usually dominates if the odd harmonics of the supercurrent are canceled by symmetry [29].

Recent studies of 0°–45° GBJs have demonstrated that the SQUID dynamics can be altered by the d-wave order parameter in YBCO [30]. It is, however, important to point out that our results *do not* directly relate to, e.g., tetracrystal π-SQUID experiments; the latter crucially depend on having one π junction with negative critical current, but still only the first harmonic present in $I_c(\phi)$. Our SQUIDs have a conventional geometry, but unconventional current-phase relations.

One explanation for the pronounced effects of the second harmonic could be that relatively large sections of the interface are highly transparent and have a low degree of disorder. This in turn could be related to our fabrication scheme which seems to preserve the integrity of the barrier. This makes feasible their applicability in the quantum regime and supports our expectations that quantum coherence can be observed in this kind of structures.

In summary, we have observed a very pronounced second harmonic in the current-phase relation of a "conventional" YBCO dc SQUID with 0°–45° grain-boundary junctions. It has strongly influenced the SQUID dynamics. All details of the SQUID behavior were explained within a simple model of a dd junction with relatively high transparency. We believe that these effects are important for better understanding of HTS Josephson junction and SQUIDs.

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