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 non-sinusoidal current-phase dependence in YBCO dc-SQUIDs, which agree with the theoretical description of the system.

Keywords: Josephson Effect, High-Temperature Superconductivity, d-wave symmetry

It is well established 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 $\pi$.

The latter leads to a plethora of effects, like formation of Andreev bound states at surfaces and interfaces in certain crystallographic orientations. 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. Time-reversal symmetry can also be spontaneously violated and thus spontaneous currents generated. Another effect can be doubling of the Josephson frequency.

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 $2\pi$-periodic odd function, it can be expanded in a Fourier series. In the most cases only the first two harmonics give a significant contribution to the current:

$$I_S(\phi) = I_1^d \sin \phi - I_2^d \sin 2\phi$$

In Josephson systems of conventional superconductors the second harmonic will usually be negligible but in $dd$-junctions the second harmonic may dominate. If $I_2^d > I_1^d/2$ the equilibrium state is no longer $\phi = 0$ but becomes double degenerate at $\phi = \pm \arccos(I_1^d/2I_2^d) \to \pi/2$. 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 2nd harmonic in the current-phase relation (CPR) of a $dd$-junction was confirmed by Il’ichev et al. 3.

A non-sinusoidal CPR of the junctions will change the dynamics of a dc-SQUID. 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

$$I_s(\phi, \phi_x) = I_1^d \sin \phi - I_2^d \sin 2\phi + I_2^d \sin(\phi + \phi_x)$$

$$- I_2^d \sin 2(\phi + \phi_x)$$

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 (RSJ) approximation. By introducing $\delta(\phi, \phi_x) = \phi_2 - \phi_1$ and applying the same method as in 12 with the necessary generalizations, we obtain for the average voltage over the SQUID:

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

$$- I_1 \left( \phi + \frac{\delta}{2} \right) - I_2 \left( \phi - \frac{\delta}{2} \right)$$

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

$$\delta + \phi_x + \frac{\pi L}{\Phi_0} \left( I_2(\phi - \delta/2) - I_1(\phi + \delta/2) \right) = 0.$$
FIG. 1: The results of simulations of the \( I_c - \phi_0 \) and \( \bar{V} - \phi_x \) dependence for a dc-SQUID with \( I_{c1} = 1 \), \( I_{c2} = 0.1 \), \( I_{c1}' = 0.2 \) and \( I_{c2}' = 0.4 \) (arb. units). The different curves correspond to bias currents in the range \( I = I_{c1}' \) to \( I = 5I_{c1}' \). We assume \( L = 0 \) and \( G_1 = G_2 \).

Though \( I \) is only explicitly solvable in the limit \( L \to 0 \), 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 \( I \) and \( \bar{V} \) are shown in fig.1. The cusps in the critical current correspond to the points at which the global maximum in \( I \) switches from one local maximum to another \( \bar{V} \). Note the quasi-\( \Phi_0/2 \)-periodicity of the current isolines in the \( \bar{V} - \phi_x \) picture, reflecting the current-phase dependence \( I \), and their shift along the \( \Phi_x \)-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 \( \Phi_0 \)-periodicity is restored. Indeed, as the bias grows, one set of minima of the washboard potential, \( U = (\hbar/2e)[1 - I^T \cos \phi + (I^{TH}/2) \cos 2\phi - I \phi] \), disappears first unless the first harmonic \( I^T \) 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\(^\circ\) (0\(^\circ\) – 45\(^\circ\) GBJ). For more information on GBJs see for example reference \( I \).

The pattern was defined using E-Beam lithography and then transferred to a carbon mask employing a multi-step 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 \( \mu \)m, as has been reported elsewhere \( 20 \). In the SQUIDs under investigation the junctions are nominally 2 \( \mu \)m 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 4.2K.

The SQUID loops are 15×15 \( \mu \)m\(^2\). The numerically calculated inductance \( 21 \) is approximately 25 pH, yielding the factor \( \beta = 2\pi L_{Jc}/\Phi_0 \) between 0.5-2.

The SQUIDs were largely non-hysteretic with a resistance of about 2 \( \Omega \). 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 \( \mu \)m in all junctions, which means that the junctions are magnetically short. This is supported by the quasi-period of the pattern in fig.2 being close to the expected value \( \phi_0/2\lambda_J w \) \( 17 \). The differential conductance curves do not show any trace of a zero bias anomaly (ZBA), as is expected for 0\(^\circ\) – 45\(^\circ\) GBJs. ZBAs has 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 1\(^{st}\) and 2\(^{nd}\) harmonics of the critical current. This assumption is supported by the fairly small modulation depth (it is easy to see from equation \( 2 \) 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 equation \( 2 \), if we compensate for the residual background magnetic field and assume that we have a small excess current (of the order of a few \( \mu \)A) 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 nT is presented in fig.4 which shows the \( I_c \)-modulation of the SQUID enveloped by an anomalous Fraunhofer-pattern similar to what has been reported by other groups \( 22, 23 \) for 0\(^\circ\) – 45\(^\circ\) 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 \( \pi \)-loops in the junction interface \( 24 \).

Figure 4 shows the \( V - B \)-dependence of one of the
and the excess current of the junctions. The solid line represents the fitted expression. The fitting parameters are as follows: (a) \( I_{c1} = 9 \mu A, I_{c2} = 0.3 \mu A, I_{c3} = 3.7 \mu A \) and \( I_{c4} = 22.7 \mu A \) (b) \( I_{c1} = 7.8 \mu A, I_{c2} = 3.0 \mu A, I_{c3} = 5.3 \mu A \) and \( I_{c4} = 4.3 \mu A \). In both cases the fit has been adjusted with respect to the residual background field and the excess current of the junctions.

**FIG. 2:** Critical current as a function of magnetic field at 4.2K. The dashed box indicates the area plotted in fig. 3.

**FIG. 3:** Critical current as a function of applied magnetic field for two different SQUIDs that are nominally identical. 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 \( \Phi_0 \). 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. A deviation from the model occurs at \( V=100 \mu V \) where the minima and maxima switch. This is probably due to an LC-resonance in the SQUID. Taking \( L = 25 \mu H \), 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 \( \mu m \) wide, where the deviations from the usual sinusoidal CPR were not obvious from the 

Generally the nature of the transport through a GBJ will depend on its transmissivity \( D \). Il’ichev et al. 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 \( I_c^{2nd}/I_c \propto D \), a high-transmissivity GBJ is required in order to observe effects of the second harmonic. An estimate of the average transmissivity of our junctions would be \( \rho_{ab}/R_K \sim 10^{-2} \). Assuming \( l \), the mean free path, to be equal to 10 nm and a resistivity in the a-b plane \( \rho_{ab} \) equal to \( 10^{-4} \) \( \Omega \)cm. This is still too low to explain the strong 2\(^{nd}\) harmonic we observe. However, it is known

**FIG. 4:** Voltage modulation as a function of applied magnetic field for the SQUID whose \( I_c - B \) is shown in fig. 3. The pattern is again inversion symmetric. Note the sign change at 100 \( \mu V \) which we believe is due to a LC-resonance in the SQUID loop.
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 $I_c^1$ and $I_c^{11}$ can vary as much as ten 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 $I_c^{11}$ excludes a high value of $I_c^1$, since the 2nd harmonic usually dominates if the odd harmonics of the supercurrent are cancelled 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. tetracrytal $\pi$-SQUID experiments; the latter crucially depend on having one $\pi$-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 2nd 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.

To summarize, we have observed very pronounced 2nd 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 junctions and SQUIDs.

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