Josephson Coupling in Untwinned YBa$_2$Cu$_3$O$_{7-x}$/Nb d-wave Junctions

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Abstract. Theory predicts that d-wave superconductivity induces a significant second harmonic $J_2$ in the Josephson current, as a result of zero-energy Andreev states (ZES) formed at the junction interface. Consequently, anomalies such as half-integer Shapiro steps should be observed. Both ZES formation and $J_2$ are expected to be highly anisotropic as we change the tunneling orientation in the $ab$ plane reaching their maximum for tunneling close to [110] direction and their minimum for [100] or [010] directions. We performed experiments on junctions between untwinned d-wave YBa$_2$Cu$_3$O$_{7-x}$ and Nb and found clear evidence of ZES formation for all 72 different tunneling directions in the $ab$ plane investigated. However, in contrast to the theoretical predictions, we found no trace of half-integer Shapiro steps. That suggests $J_2$ is insignificantly small compared to the first Josephson harmonic for all of the orientations. We believe that microscopic scale roughness, or diffusive reflection at a scale that is much smaller than the Fermi wavelength, dramatically suppresses $J_2$ due to scattering processes. Our findings suggest that YBa$_2$Cu$_3$O$_{7-x}$/Nb d-wave junctions have a purely sinusoidal current-phase relation which is essential to take into consideration for their implementation as qubits or π-junctions in digital circuits.

1. Introduction

d-wave junctions formed between two superconductors of which at least one is a d-wave superconductor are interesting candidates for the implementation of superconducting qubits in quantum computation [1] or π-junctions in Josephson (low-dissipative) digital circuits [2]. In addition, arrays of d-wave junctions are of interest as model systems for studying magnetic phenomena—including frustration effects—in Ising antiferromagnets [3]. Moreover, d-wave junctions are among the most reliable tools to investigate the unconventional superconducting order parameter in these materials [4, 5]. The physics of d-wave junctions, however, is not fully understood. First, a key element, namely the knowledge of the current-phase relation (CPR) of the Josephson current remains unsettled [6]. It has been predicted [7-11] that zero-energy Andreev states (ZES) formed at the d-wave junctions interface induce a second harmonic Josephson current $J_2$ in the CPR. $J_2$ is an important parameter, as a superconducting qubit based on $J_2$ will have an operating point intrinsically stable and protected against the environmental noise, which will reduce decoherence [12]. Secondly, within the

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same theoretical framework, \( d \)-wave is expected to induce ZES at junctions interface [5], anomalies in the Josephson effect [4] and, a ZES-induced Josephson current. Whereas the first two effects are well established experimentally, the existence of \( J_2 \) remains unconfirmed. In this paper (a more detailed description will be published elsewhere [13]) we address this controversy for Josephson junctions made between predominantly \( d \)-wave \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \) and Nb. First we provide evidence for the formation of ZES and we look for the existence of \( J_2 \). If it occurs, this second harmonic component is expected [7-11] to be highly anisotropic as we change the tunneling orientation in the \( ab \) plane reaching its maximum for tunneling close to \([110]\) direction and its minimum for \([100]\) or \([010]\) directions.

\( J_2 \) is expected [7-11] to produce a significant deviation from the standard sinusoidal CPR of the Josephson current density \( J_\ell \) [6]

\[
J_\ell(q) = J_1 + J_2 = Jc_1 \sin(q) + Jc_2 \sin(2q)
\]

(1).

Here \( q \) is the phase difference across the junction. For a purely \( d \)-wave order parameter as we increase \( \theta \) (the angle in the \( ab \) plane between the normal to the junction interface and the \([100]\) crystal axis) starting from 0, \( J_2 \) is expected [10] to increase monotonically up to \( \theta = 45^\circ \) which corresponds to tunneling into the \([110]\) direction. It then should decrease monotonically as we further increase \( \theta \) from \( 45^\circ \) to \( 90^\circ \), corresponding to tunneling into the \([010]\) direction. In particular, for tunneling close to the \([110]\) direction, where \( J_1 \) vanishes due to the nodes of the \( d \)-wave order parameter, \( J_2 \) will dominate the CPR [7-11], [14-17]. So far, there have been experimental reports consistent with the presence of a finite second harmonic [18-19] in various types of twinned \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \) junctions but in none of these cases has the formation of ZES at the junction interface been confirmed. Therefore, its presence cannot be attributed to ZES formation, while there are other alternative mechanisms that may generate it [6].

2. Quasiparticle conductance measurements

We prepared thin film ramp-edge junctions between 170-nm untwinned \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \) and 150-nm Nb using a 30-nm Au barrier. The use of untwinned \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \) thin films is especially important because otherwise \( J_2 \) may be strongly suppressed due to excessive diffusive scattering [9] at the twin boundaries. Also, \( J_2 \) may be averaged out for a badly defined nodal orientation in a twinned film. The junctions are fabricated on the same chip, and the angle \( \theta \) with the \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \) crystal \( b \)-axis is varied in units of 5 degrees, so that tunneling can be probed in \( 360^\circ/5^\circ = 72 \) different directions in the \( ab \) plane (see Fig. 1 in [20]). The growth of untwinned \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \) films [21], as well as detailed order parameter studies [20], and ZES-assisted quasiparticle tunneling [22] in these particular junctions are reported elsewhere. All 72 junctions are 4 \( \mu \)m wide.

We first measured the quasiparticle conductance spectra \( G(V) \) of all 72 junctions for a wide range of temperatures \( T \) (4.2-77 K) and magnetic fields \( B \) (0-7 T). A quantitative comparison of these measurements with calculations made on the basis of an \( S_d\)S\( _s \) tunnel junction model (with the \( S_d \) superconductor being Nb and the \( S_s \) superconductor being \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \)) using quasiclassical techniques was recently published [22]. It was found that all observed features are consistent with a convolution of density of states with broadened ZES formed at the \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} /\text{Au/Nb junction interfaces [22]} \). Here we only summarize some of the most important findings from a qualitative point of view. We observed the same qualitative picture independent of the tunneling direction. At 4.2 K and a small \( B \) of 0.01 T, which is large enough to completely suppress the \( dc \) Josephson current, well-defined Nb coherence peaks and a dip at the center of a broadened zero-bias conductance peak (ZBCP) are observed (see Fig.1). As superconductivity is suppressed in Nb, by increasing \( T \) from 4.2 K up to slightly below the critical temperature of Nb (\( T_{c,Nb} \approx 9.1 \) K) or \( B \) from 0.1 T up to slightly below the second critical field of Nb (\( B_{c2,Nb} \approx 1.15 \) T) the Nb coherence peaks become suppressed and the ZBCP-presence gradually manifest. Close to the critical temperature \( T_{c,Nb} \) (see Fig. 1a) or to 0.4 T (see Fig. 1b) no trace is left of the Nb coherence peaks, while the ZBCP is fully developed. That provides clear evidence for the formation of ZES. Increasing \( T \) or \( B \) even further (from \( T_{c,Nb} \) up to 77
K, or \( B \) from 0.4 T to \( B_{c2,\text{Nb}} \) and further to 7 T), however, a significant difference appears between the \( T \) and \( B \) dependence of \( G(V) \). The ZBCP (its amplitude and width) is essentially not affected by an increase of \( B \), while by increasing \( T \) the ZBCP becomes strongly suppressed and widens. In particular, we could not observe any trace of a ZBCP at 77 K. The remarkable insensitivity of \( G(V) \) to the tunneling direction strongly suggest the existence of ZES in all tunneling orientations in the \( ab \) plane, including the [100] and [010] directions. We believe this is a signature of diffusive reflection or scattering, possibly due to microscopic interface roughness.

3. Shapiro steps measurements

To identify \( J_2 \) we searched for half-integer Shapiro steps. It is well known that if the CPR is pure sinusoidal \((Jc_2 = 0 \text{ in Eq. } (1))\) microwave radiation of frequency \( f \) will induce Shapiro steps at integer \( n \) multiples of the voltage \( V_0 \), satisfying the Josephson voltage-frequency relation \( f/V_0 = 0.486 \text{ GHz}/\mu\text{V} \). If \( Jc_2 \) is finite also half-integer Shapiro steps should appear at multiples of \( V_0/2 \) [23]. If half-integer Shapiro steps are not observed then the presence of a significant \( J_2 \) in the CPR can be ruled out. We performed a very detailed search in the entire frequency range where integer Shapiro steps could be observed, carefully examining every 10 MHz frequency interval within the 1-20 GHz region. We repeated this approach for all junctions investigated. Typical sets of current-voltage characteristics are shown in Figs.2a-2c for three junctions: [100], [110] and [110]-5°. Well-defined integer Shapiro steps in accordance with the theoretical expectations are clearly visible. We detected pronounced integer Shapiro steps up to \( n=21 \) (as in Fig.2(a)) or even higher in some cases. We also measured the

Figure 1: Representative conductance spectra of (a) 4 junctions with different tunneling directions at 4.2 K and just below \( T_{c,Nb} \), and (b) a [110]-oriented junction for ten different magnetic field values from 0 T (in black) up to 7 T (in black). The inset in (b) shows details of the low voltage spectra.

![Figure 1](image-url)
amplitude of the integer Shapiro steps as a function of the microwave current amplitude. Some typical examples are shown in Figs.2(d)-2(f) for three junctions: [110], [110]+5°. We found no trace of half-integer Shapiro steps in any of the junctions, although we paid particular attention to those microwave amplitudes where the integer Shapiro steps or the $I_c$ vanishes and consequently the half-integer Shapiro steps are expected to be most pronounced. In particular, as can be inferred from Figs.2d, increasing the microwave power first fully suppresses $I_c$ and thereafter the first integer Shapiro step. However, no signature of the first half-integer Shapiro step is observed. Moreover, the fact that $I_c$ is fully suppressed by microwaves (see Figs.2(d)-2(f)) is a further confirmation that $J_2$ is insignificantly small as non-zero minima are expected for $I_c$ in case $J_2$ has considerable amplitude [23]. Taking into account our finite resolution in detecting the Shapiro steps an upper bound on $J_2$ of about 1% from $J_c$ is found, both being measured for the same crystal orientation.

The ramp-edge junctions investigated here might not be ideally described in the frame of an $S_dIS_n$ model. However, as our previously report showed [22], as far as quasiparticle tunneling is concerned there is a good quantitative agreement between the measured conductance spectra and calculations made on a basis of an $S_dIS_n$ tunnel junction model using quasiclassical techniques. Looking into the Josephson tunneling, in the frame of Green’s function formalism $J_2$ is calculated by integrating over all transverse wave vectors [9]:

$$J_2 = \frac{2e}{\pi \hbar} \int_{-\alpha}^{\alpha} dE f(E) \int_{-\pi/2}^{\pi/2} \frac{d\alpha}{2} \cos \alpha J(\alpha, E)$$

Figure 2: (a-c) Integer Shapiro steps (indicated by vertical arrows) at 4.2 K of [010], [110], and [110]+5°-oriented junctions at different microwave amplitudes. For clarity, the current-voltage characteristics in (b) are shifted in diagonal direction shown by the gray line. (d-f) Amplitude of the first three integer Shapiro steps and of the critical current versus the normalized microwave-current amplitude for a [110], [110]+5°, and [110]+5° junction.
where \( J(\alpha, E) = 2\pi |M|^{2} E^{2} g_{\text{YBCO}}^{\text{eh}}(\alpha) g_{\text{Nb}}^{\text{eh}}(\alpha, E) \) are the pair-correlation functions in the two superconductors, \( M(\alpha) \) is the matrix element between Nb and YBCO, \( \delta'(E) \) is the derivative of the Dirac delta function, and \( \alpha \) is the angle between a reflected wave and the normal to the junction interface. From Eq.(2) it follows that junction roughness has a dramatic influence on \( J_{2} \). For a smooth junction the tunneling process does not affect the transverse momentum of the quasiparticle and \( J_{2} \) has to be observed in experiments. We believe our junctions are rough on the scale of a Fermi wavelength. In this case a quasiparticle in one transverse direction in Nb can get scattered to any transverse direction \( \alpha \) in YBCO. This results in an averaging of the pair-correlation functions over different directions \( \alpha \). Since \( g_{\text{YBCO},\text{Nb}}^{\text{eh}}(\alpha, E) \) are antisymmetric functions of \( \alpha \) this averaging process makes \( J_{2} \) disappear completely. Our assumption of rough junctions is also consistent with ZES formation in all tunneling orientations in the \( ab \) plane including the [100] and [010] directions, in high contrast to the case of smooth junctions.

4. Conclusions

In summary, we provided evidence in support of ZES formation in untwinned, \( d \)-wave YBa\(_{2}\)Cu\(_{3}\)O\(_{7-\delta}\)/Nb junctions. However, in contrast to the theoretical predictions [7, 8, 10, 11, 14-17], Shapiro step measurements reveal no trace of a ZES-induced Josephson current \( J_{2} \). We believe it is scattering due to junction roughness on the scale of a Fermi wavelength that completely suppresses \( J_{2} \). Our results suggest that the nature of \( J_{2} \) in various types of \( d \)-wave junctions, not only in the ramp-edge junctions investigated here, is more subtle than previously anticipated due to its strong sensitivity to scattering. Therefore, the observation of a ZES-induced Josephson current may prove a very difficult task in experiments. They also suggest that YBa\(_{2}\)Cu\(_{3}\)O\(_{7-\delta}\)/Nb \( d \)-wave junctions have a purely sinusoidal CPR which is essential to take into consideration for their implementation as qubits [1, 12] or \( \pi \)-junctions in digital circuits [2].

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