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Josephson interferometry and Shapiro step measurements of superconductor-ferromagnet-superconductor 0-π junctions

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We report the observation of the 0-π junction state in superconductor-ferromagnet-superconductor Josephson junctions arising from variations in the effective barrier thickness d/ξ_F. By varying the temperature, the junctions can be tuned from the 0- to the π-state through an intermediate regime in which parts of the junction are in each state. A signature of the 0-π regime is that the usual peak in the critical current diffraction pattern at zero magnetic field becomes a dip, evidence for sign changes in the critical current density. In some junctions, the zero-field critical current does not vanish at any temperature and half-integer Shapiro steps are observed, indicating the presence of spontaneous supercurrents circulating within the junctions around the 0-π interfaces.

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In the past few years considerable attention has been directed toward the understanding and realization of π Josephson junctions.[1] Transitions between 0 and π junction states were demonstrated in transport measurements on superconductor-ferromagnet-superconductor (SFS) junctions as a function of temperature[2] and barrier thickness,[3] and in mesoscopic superconductor–normal metal–superconductor junctions as a function of the barrier thermalization voltage.[4,5] Further experiments provided phase-sensitive evidence of the π junction state, including dc superconducting quantum interference device (SQUID) interferometry,[6,7] direct measurements of the current-phase relation,[8] and probes of spontaneous currents in dc (Refs. 6 and 9) and rf π SQUIDs.[10] These findings demonstrate the feasibility of basic π junction circuits that have been proposed for both quantum and ultrafast classical computing implementations.[11–13]

In this Rapid Communication, we report a study of SFS junctions in which the critical current density varies spatially within the junction due to small inhomogeneities in the thickness and/or strength of the exchange field in the ferromagnetic barriers. As a result, regions of the junctions transition into the π junction state at different temperatures, resulting in a 0-π junction with regions in each state.[14] We are able to manipulate the 0 and π critical current densities by changing the temperature, and monitor the spatial variation of the critical current via magnetic field modulation measurements. We find that when the net critical currents of the 0 and π regions become comparable in magnitude, the energy of the junction is minimized by generation of spontaneous circulating currents flowing around the interfaces between them. These currents prevent the overall junction critical current from vanishing at any temperature, the usual signature of the 0-π transition. It also couples to applied microwave fields, producing the half-integer Shapiro steps that we observe in a narrow temperature range around the minimum critical current. A similar effect has been observed in nearly symmetric dc SQUIDs with an applied magnetic flux of $\frac{1}{2}\Phi_0$ ($\Phi_0=h/2e$).[15]

A π Josephson junction is characterized by a negative critical current. The mechanism of the π state in SFS Josephson junctions is the spatial oscillation of the proximity-induced order parameter inside the ferromagnetic barrier which arises from its exchange field.[16] The critical current density J_c is predicted to oscillate and decay with increasing barrier thickness d according to[17]

$$J_c(d) \sim \left[\cos\left(\frac{d}{\xi_{F2}}\right) + \frac{\xi_{F1}}{\xi_{F2}} \sin\left(\frac{d}{\xi_{F2}}\right)\right]\exp\left(-\frac{d}{\xi_{F1}}\right), \quad (1)$$

where ξ_{F1} is the decay length and $2\pi\xi_{F2}$ is the oscillation period of the order parameter. This expression is valid for $d \gg \xi_{F1}$. The lengths ξ_{F1} and ξ_{F2} can be extracted by fitting the measured $J_c(d)$ to Eq. (1). In our junctions at $T=4.2$ K, $\xi_{F1} \approx 1.3$ nm and $\xi_{F2} \approx 3.7$ nm, and the first two nodes of J_c occur for $d \approx 11$ and 22 nm.[18] The lengths ξ_{F1} and ξ_{F2} vary with temperature according to

$$\frac{\xi_{F1,F2}(T)}{\xi_{F1,F2}(0)} = \left(\frac{E_{ex}}{[(\pi k_B T)^2 + E_{ex}^2]^{1/2} \pm \pi k_B T}\right)^{1/2}, \quad (2)$$

where E_{ex} is the ferromagnet exchange energy and $\xi_{F1,F2}(0)$ are the values at zero temperature. By using a weakly ferromagnetic alloy $Cu_{0.47}Ni_{0.53}$ with $T_{Curie} \sim 60$ K as a barrier material, temperature changes in the 1–4 K range have a significant effect on the suppression and modulation of the induced pair correlations in the ferromagnetic interlayer, allowing us to tune through the 0-π transition by changing the temperature.

The SFS junctions were patterned by optical lithography. Base and counterelectrode superconducting layers were dc-sputtered Nb with thicknesses 100 and 240 nm, respectively, separated by an 11 nm barrier layer of rf-sputtered $Cu_{0.47}Ni_{0.53}$ and a 20–30 nm layer of dc-sputtered Cu. The ferromagnetic layer thickness is chosen near the first 0-π transition thickness, while the Cu layer protects the barrier during processing. Junction sizes were 4×4 μm^2 or 10×10 μm^2, defined by a window in an insulating SiO layer

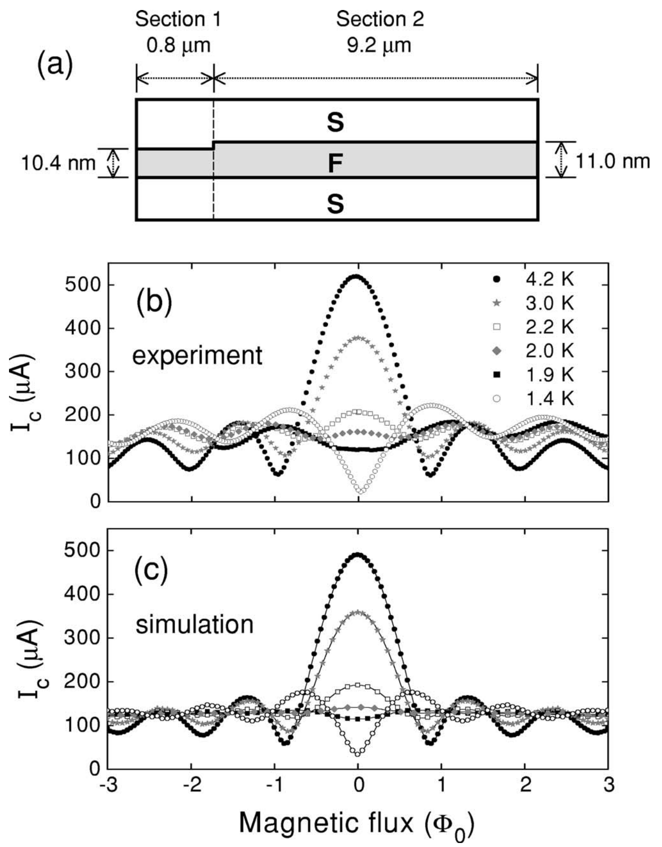

FIG. 1. (a) Stepped ferromagnetic barrier deduced from critical current vs applied magnetic flux measurements. (b) Diffraction patterns at a series of temperatures showing deviations from Fraunhofer behavior at low temperatures. (c) Simulated diffraction patterns using the deduced ferromagnetic barrier profile.

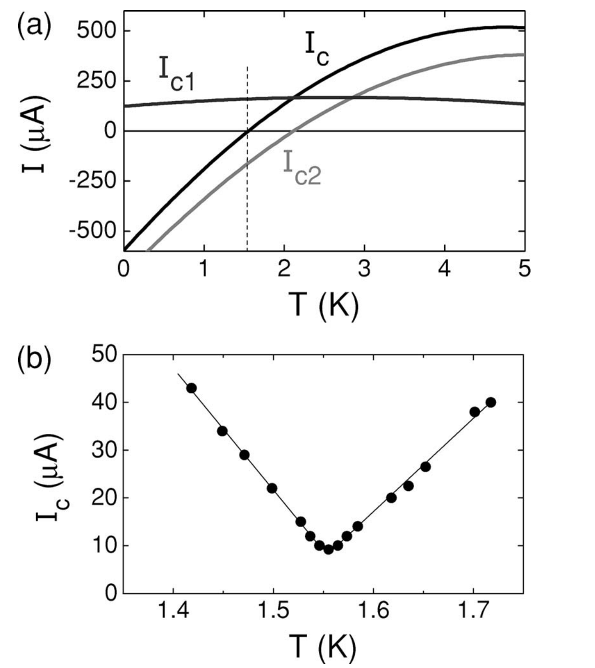

FIG. 2. (a) Calculated temperature variation of the critical current for junction sections 1 and 2 and for the entire junction. The thin region remains in the 0 state at all temperatures while the thick region crosses from the 0 state into the π state. (b) Measured critical current vs temperature at zero applied magnetic field dips but remains finite.

deposited on top of the CuNi/Cu barrier. Because SFS junctions have small I_cR_N products $\sim 1-100$ nV, a commercial dc SQUID with a standard resistor $R_{st} \approx 10$ mΩ is used as a potentiometer with sensitivity 1 pV to perform transport measurements. This translates into a critical current measurement resolution of 100 nA. A uniform magnetic field up to 100 G can be applied through the junction barrier from a solenoid coil to measure the magnetic field dependence of the critical current, and a rf current component at frequencies f_{rf}=0.3–1.3 MHz is superimposed on the dc bias current to observe ac-induced Shapiro steps in the current-voltage characteristics.

Measurements of the critical current I_c vs the applied magnetic flux Φ threading the junction barrier reveal that the critical current distribution is often not uniform across the junction. Figure 1(b) shows a series of $I_c(\Phi)$ curves in the temperature range 1.4–4.2 K for a 10×10 μm^2 junction. At T=4.2 K, $I_c(\Phi)$ has a Fraunhofer-like shape but with nonvanishing supercurrents at the side minima. This can occur in a junction with a localized region of high critical current density. In the temperature interval 1.4–1.9 K a minimum in the critical current is observed at zero field, indicating that regions of opposite-polarity critical current density exist in the junction.

The temperature evolution of the $I_c(\Phi)$ patterns indicates that some fraction of the junction width makes a transition from the 0 state to the π state as the temperature is lowered, while the remaining part stays in the 0 state. The critical current nonuniformity likely arises from spatial variations in the barrier thickness across the junction but could also be caused by inhomogeneities in the ferromagnet exchange energy or by variations in the S-F interface transparency. Figure 1(a) shows the step barrier geometry deduced by fitting the measured diffraction patterns in Fig. 1(b) in the short-junction approximation in which magnetic fields from the tunneling current are neglected. We obtain good agreement as demonstrated in Fig. 1(c). It is not surprising that the small 6 Å step has such a dramatic effect on the diffraction patterns since close to the 0-π transition the critical current density in our junctions changes by 1000 A/cm^2 per 1 nm change in the barrier thickness. For the barrier profile in Fig. 1(a) and the experimental data for $J_c(d)$, we use Eqs. (1) and (2) to calculate the temperature dependences of the zero-field critical currents I_{c1} of the thin narrow region, I_{c2} of the wide thick region, and I_c, the total junction critical current. These are plotted in Fig. 2(a). We see that I_{c1} is relatively constant while I_{c2} decreases and changes sign at $T \approx 2.1$ K, causing the I_c to vanish at $T_{\pi 0} \approx 1.55$ K. However, measurements plotted in Fig. 2(b) show that $I_c(\Phi=0)$ does not go fully to zero at $T_{\pi 0}$, instead reaching a minimum value of ≈ 10 μA, suggesting that the short-junction approximation is not fully valid.

Shapiro steps induced in zero applied magnetic field at drive frequency f_{rf} are also anomalous for this junction, exhibiting not only the usual steps at integer multiples of the

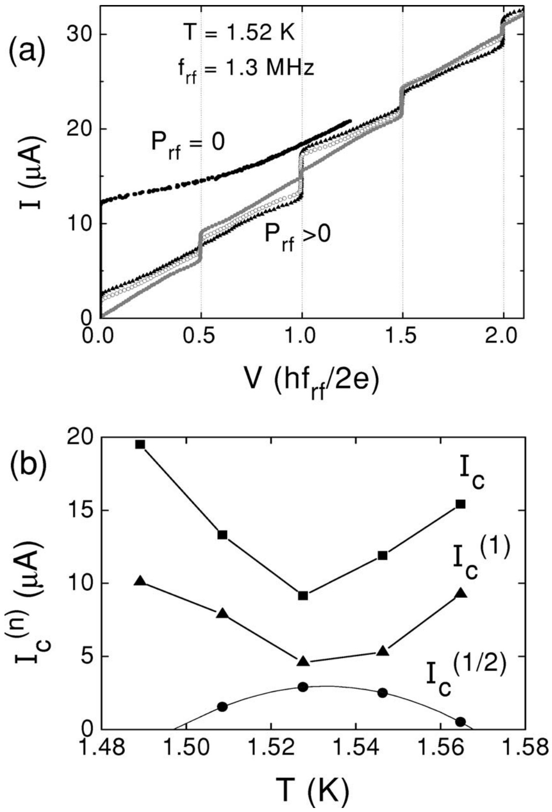

FIG. 3. (a) Current-voltage characteristics showing rf-induced Shapiro steps both at the usual voltages $nhf_{rf}/2e$ and at half-integer values $nhf_{rf}/4e$. (b) Temperature dependence of the maximum (power-optimized) critical current steps, showing that the integer steps scale with the junction critical current whereas the half-integer steps occur only at temperatures near the minimum in the critical current.

Josephson voltage $V_n = n(hf_{rf}/2e)$, but also steps at half-integer Josephson voltages such as $V_{1/2} = hf_{rf}/4e$ and $V_{3/2} = 3hf_{rf}/4e$ when the temperature is close to $T_{\pi 0}$. An example is given in Fig. 3(a) for $f_{rf} = 1.3$ MHz. Figure 3(b) shows the maximum value of the ($V=0$) Josephson supercurrent, and the maximum amplitudes of the $n=\frac{1}{2}$ and the $n=1$ Shapiro steps obtained by adjusting the rf amplitude. At temperatures far from $T_{\pi 0}$, only integer Shapiro steps were observed. Half-integer steps appear near the minimum in the critical current in a temperature range of width ~60 mK. Note that the minimum in the critical current measured in Fig. 3(b) is about 25 mK lower than that in Fig. 2(b) as a result of room-temperature annealing of the ferromagnetic barrier during the 3 days between measurements.[2] In other junctions in which the critical current does vanish at $T_{\pi 0}$, half-integer Shapiro steps were not observed at any temperatures.

These phenomena can be explained by self-field effects that must be taken into account in finite-width 0-π junctions. At temperatures close to $T_{\pi 0}$, spontaneous currents circulate around interfaces between 0 and π regions to lower the total energy of the system.[14,19,20] These circulating currents generate magnetic flux through the junction that prevents the total critical current from vanishing at any applied magnetic field. In addition, they can resonate with a rf bias current applied at twice the Josephson frequency, resulting in half-integer Shapiro steps. A detailed calculation of the spatial distribution of spontaneous currents and the net critical current in this regime requires a self-consistent solution of the sine-Gordon equations in the junction. Computation of the amplitudes of the half-integer Shapiro steps further requires numerical simulations of the junction phase dynamics. However, we can understand the onset and effects of the spontaneous currents in a 0-π junction in analogy to a dc SQUID with finite geometric inductance.

We consider a dc SQUID with a 0 junction of critical current $I_{c0} > 0$ and a π junction of critical current $I_{c\pi} < 0$ in a loop of inductance L. In such a SQUID, phase coherence around the SQUID loop precludes both junctions being in their low-energy states and the junction phases ϕ_0 and ϕ_π depend on the inductance parameter $\beta_L = 2\pi L I_c/\Phi_0$, where $I_c = (|I_{c0}| + |I_{c\pi}|)/2$, and the critical current asymmetry $\alpha = (|I_{c0}| - |I_{c\pi}|)/2I_c$. For $\beta_L < 2\alpha/(1-\alpha^2)$, it is energetically favorable to switch the phase of the junction with the smaller critical current magnitude into its high-energy state, e.g., if $|I_{c0}| > |I_{c\pi}|$, then $\phi_0 = \phi_\pi = 0$. Under the influence of a harmonic ac drive, ϕ_0 and ϕ_π wind in phase and only integer Shapiro steps can be observed. When $\beta_L > 2\alpha/(1-\alpha^2)$, the SQUID energy is lowered by generation of a spontaneous circulating current and $\phi_0 \neq \phi_\pi$. In this regime, the phase differences ϕ_0 and ϕ_π are no longer synchronized. Hence, the spontaneous circulating current $J = (I_c/\beta_L)(\phi_0 - \phi_\pi)$ can phase lock to the driving frequency in such a way that it switches direction an even or odd number of times during each period of the drive signal, corresponding to integer or half-integer Shapiro steps. This phenomenon has previously been studied in an equivalent system, an ordinary dc SQUID with an applied flux of $\frac{1}{2}\Phi_0$. Measurements and simulations showed half-integer Shapiro steps with amplitudes that increase as the SQUID inductance is increased and the critical current asymmetry is decreased.[15]

In a 0-π junction, the existence of spontaneous circulating currents, and hence half-integer Shapiro steps, depends on the ratio of the widths of the 0 and π regions of the junction w_0 and w_π to their corresponding Josephson penetration depths λ_{J0} and $\lambda_{J\pi}$.[14] Here, $\lambda_J = (\hbar/2e\mu_0 d_m J_c)^{1/2}$, where the magnetic barrier thickness $d_m = 2\lambda + d$ depends on the penetration depth of the superconductor λ and the barrier thickness d. The phase diagram denoting the regimes of uniform phase and spontaneous current flow is shown in Fig. 4. Regimes with uniform phase differences $\phi=0$ or $\phi=\pi$ are separated by a narrow region in which the phase differences vary across the junction due to spontaneous circulating currents. The circulating currents onset along lines defined by $\lambda_{J\pi}\tanh(w_0/\lambda_{J0}) = \lambda_{J0}\tan(w_\pi/\lambda_{J\pi})$ and $\lambda_{J0}\tanh(w_\pi/\lambda_{J\pi}) = \lambda_{J\pi}\tan(w_0/\lambda_{J0})$.[14] Also shown is the path in the phase diagram followed by the nonuniform junction described in Fig. 2(a) as the temperature is varied from 1 to 2.1 K, the temperature at which the wide segment of the junction crosses into the 0 state. For this junction, $d_m \approx 220$ nm, determined from the period of the critical current diffraction patterns. We see that spontaneous currents are expected only in the narrow temperature range 1.53–1.57 K. This is comparable to the

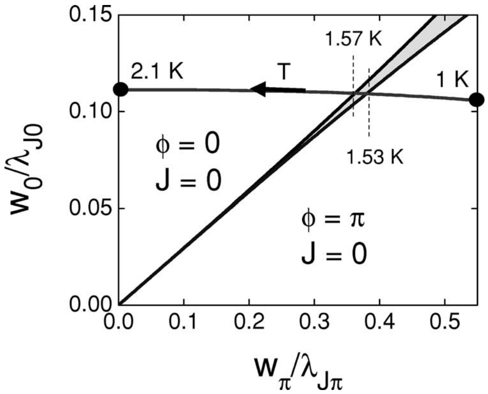

FIG. 4. Phase diagram mapping the region of w_0/λ_{J0} and $w_\pi/\lambda_{J\pi}$ values for which a spontaneous circulating current flows around the 0-π step edge. In the shaded region, the phase drop across the junction ϕ and current density J vary spatially along the junction width. Outside this region, the phase is uniform with value 0 or π and no spontaneous currents flow in the junction.

temperature range 1.50–1.56 K in which we observe half-integer Shapiro steps, suggesting that they have the same origin. The heights of the half-integer steps and the minimum value of the critical current, as estimated from a simple treatment of the junction as a 0-π dc SQUID as described above, are also in good agreement with the measurements.

Other nonuniform junctions that we have measured had either smaller areas so that w_0 and w_π were smaller, or smaller barrier thickness steps so that λ_{J0} and $\lambda_{J\pi}$ were larger. In either case, the temperature range of spontaneous currents was predicted to be immeasurably small (<1 mK), explaining why we were not able to observe half-integer Shapiro steps or a finite minimum critical current in these junctions.

Both half-integer Shapiro steps and nonvanishing critical currents at zero applied magnetic flux have previously been observed in SFS junctions and could arise from a secondary Josephson component proportional to $\sin 2\phi$ in the current-phase relation (CPR).[21] However, the amplitude of the second-order component in the CPR was predicted to remain finite in the wide range of temperatures close to a 0-π transition barrier thickness,[22–24] while the amplitudes of half-integer Shapiro steps in our junctions are only seen within 50 mK around $T_{\pi 0}$. Moreover, half-integer Shapiro steps were not observed in more uniform 0-π junctions in which spontaneous currents were calculated to be negligible. We therefore conclude that no $\sin 2\phi$ component was observed in our junctions. These phenomena are of great importance for quantum and digital computing applications that rely on the current-phase relation of π Josephson junctions since such junctions will most likely exhibit critical current variations of the scale reported here.

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