Direct Observation of the Transition from the Conventional Superconducting State to the π State in a Controllable Josephson Junction

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We measure the full supercurrent-phase relation of a controllable π junction around the transition from the conventional 0 state to the π state. We show that around the transition the Josephson supercurrent-phase relation changes from $I_{sc} \simeq I_c \sin(\varphi)$ to $I_{sc} \simeq I_c \sin(2\varphi)$. This implies that, around the transition, two minima in the junction free energy exist, one at $\varphi = 0$ and one at $\varphi = \pi$, whereas only one minimum exists at the 0 state (at $\varphi = 0$) and at the π state (at $\varphi = \pi$). Theoretical calculations based on the quasiclassical theory are in good agreement with the observed behavior.

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The fundamental process which enables supercurrent transport in superconductor-normal-metal-superconductor (SNS) junctions is Andreev reflection. As a result, a spectrum of supercurrent carrying states is formed in the junction normal region which consists of both positive and negative contributions to the supercurrent. By populating the negative contributions and depopulating the positive ones, it is, in principle, possible to obtain a situation where the supercurrent flows in the negative direction with respect to the macroscopic phase difference φ between the superconducting electrodes. This corresponds to a π junction [1] in which the stable zero current state of the junction is located at a value $\varphi = \pi$ in contrast with $\varphi = 0$ for a conventional junction. This effect is predicted in ballistic systems [2,3] as well as in diffusive systems [4–7] and has recently been observed in controllable π junctions based on niobium-gold, niobium-silver, or aluminium-silver in the diffusive limit [8–10]. Other superconducting systems that exhibit a π shift in macroscopic phase difference are bicrystals [11] or "*s*-*d*" contacts in ceramic superconductors [12] and π junctions using a dilute ferromagnet f' as the "normal" region of a S-f'-S junction [13]. The state of the junction, i.e., a 0 state or a π state, depends in most of these systems on the sample design or on temperature. This is in contrast with controllable π junctions where the population of the supercurrent carrying states and, hence, the state of the junction is determined by the application of a control voltage V_c over additional contacts connected to the junction normal region. If $V_c < V_{c,\text{critical}}$, a geometry and temperature dependent critical value, the junction is in the 0 state, and, if $V_c > V_{c,\text{critical}}$, the junction is in the π state.

The question arises how the transition occurs from the 0 state to the π state, i.e., how the energy landscape evolves from having one minimum at $\varphi = 0$ to having one minimum at $\varphi = \pi$. Does the energy landscape become completely flat, indicating a total absence of Josephson coupling or does the minimum at $\varphi = \pi$ start to develop while the one at $\varphi = 0$ is still present? The latter implies that there is a region around the transition where the energy landscape has two minima, one at $\varphi = 0$ and one at $\varphi = \pi$. This is of considerable interest with respect to possible applications in quantum computing, which rely on systems with a double degenerate ground state [14,15]. The free energy of the junction, $W(\varphi)$, is given by [16]

$$W(\varphi) = \frac{\phi_0}{2\pi} \int_0^{\varphi} I_{sc}(\varphi')\, d\varphi. \tag{1}$$

Hence, a double minimum in the free energy implies a doubling in the periodicity of the supercurrent-phase relation: $I_{sc} = I_c \sin(\varphi) \Rightarrow I_{sc} = I_c \sin(2\varphi)$ (I_c is the critical current of the junction). In this Letter, we present an experiment in which we measure the full supercurrent-phase relation of a controllable π junction, in particular, around the 0 to π transition. We observe at the transition a doubling in the periodicity of the supercurrent-phase relation of the junction indicating the presence of two minima in $W(\varphi)$, one at $\varphi = 0$ and one at $\varphi = \pi$. We show that the measurements are in good agreement with predictions based on the well-established quasiclassical theory on diffusive SNS junctions [17].

The measurements are performed using a controllable π SQUID (superconducting quantum interference device) as shown in Fig. 1. A loop of Nb (50 nm thick) with a surface area of 70 μm^2 has two controllable π junctions as weak links. Each controllable π junction consists of a 50 nm thick Ag rectangle with length $L = 750$ nm and width $w = 600$ nm connected by means of a 150 nm wide silver wire to the center of another, V-shaped silver wire that we will call control channel. The silver rectangle overlaps the niobium over a distance of 200 nm forming a coplanar SNS junction with clean Nb-Ag interfaces.

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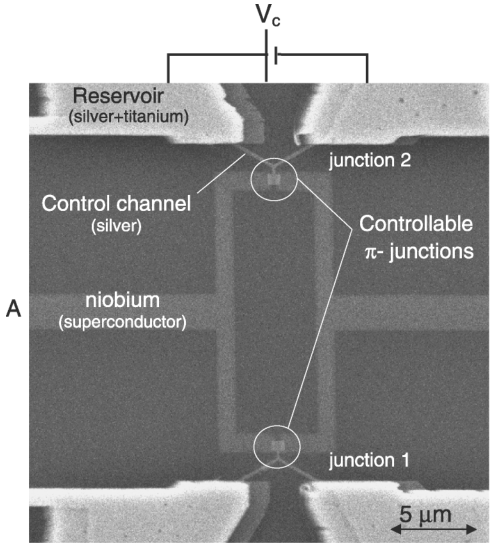

FIG. 1. Controllable π SQUID. Two controllable π junctions form the weak links of a dc SQUID. Each junction consists of a coplanar Nb-Ag-Nb SNS junction with clean interfaces of which the normal region is coupled to the control channel. The electron distribution in the normal region is modified using V_c causing the transition to a π state at sufficiently large values of V_c.

The diffusion constant of the silver is $D = 0.023\ \mathrm{m}^2/\mathrm{s}$ obtained from the measured square resistance $R_\square = 0.023\ \Omega$. This gives a Thouless energy $E_T = \hbar D/L^2 = 27\ \mu\mathrm{eV}$ for the SNS junction. The control channel is attached at each end to two very large thermal reservoirs. The sample fabrication is described in detail in Refs. [18,19]. By applying a voltage V_c, we create a nonthermal quasiparticle energy distribution function $f(E, V_c)$ with a (rounded) double step shape in the center of the silver control channel and, hence, in the normal region of the junction [8,19,20]. The consequence of this is a reduction in magnitude of the critical current and a subsequent transition to a π state as a function of V_c. The advantage of the SQUID geometry is that it enables one to measure the dependence of $I_{sc}(\varphi)$ over the entire range of the phase φ over one of the junctions in the SQUID loop: The critical current of the SQUID is given by $I_{c,\mathrm{SQUID}} = \max|I_{sc1}(\varphi_1) + I_{sc2}(\varphi_2)|$, where the phases over the two junctions are related to the flux ϕ in the SQUID according to $\varphi_2 = \varphi_1 + 2\pi\frac{\phi}{\phi_0}$ ($\phi_0 = 2.07 \times 10^{-15}$ Wb represents the quantum of magnetic flux). In the limit of negligible self-inductance $(2\pi L I_c')/\phi_0 < 1$ (with $I_c' = \min|I_{c1}, I_{c2}|$), ϕ is equal to the external flux ϕ_{ext}. If we furthermore assume that $I_{c1} \gg I_{c2}$, then only φ_2 is modified as a function of ϕ_{ext}. In this limit, it can be shown that the critical current of the SQUID is given by

$$I_{c,\mathrm{SQUID}}(\phi_{\mathrm{ext}}) = I_{c1} + I_{sc2}\left(\frac{\pi}{2} + 2\pi\frac{\phi_{\mathrm{ext}}}{\phi_0}\right), \qquad (2)$$

where we assume that I_{c1} is reached at $\varphi_1 = \frac{\pi}{2}$. Hence, $I_{c,\mathrm{SQUID}}(\phi_{\mathrm{ext}})$ represents the supercurrent-phase relation of junction 2 with $\varphi_2 = \frac{\pi}{2} + 2\pi\frac{\phi_{\mathrm{ext}}}{\phi_0}$.

In the experiment, performed at 1.4 K, we bias the SQUID (from contacts A to B in Fig. 1) with a *low frequency* bias current ($f \sim 80$ Hz) slightly larger than the critical current. Hence, the modifications of $I_{c,\mathrm{SQUID}}(\phi_{\mathrm{ext}})$ are transferred into a voltage signal (with a minus sign) which we measure using a standard lock-in technique as a function of the external magnetic field for different values of V_c. The advantage over a dc measurement is a large decrease in noise due to the limited bandwidth. At equilibrium ($V_c = 0$), the critical current of the SQUID is 22 μA, 11 μA for each junction [21]. The self-inductance of the SQUID is estimated to be 65 pH [19] which yields $(2\pi L I_c')/\phi_0 = 2.4$. At $V_c \approx V_{c,\mathrm{critical}}$, the critical current of the top junction, I_{c2}, is strongly suppressed and we are in the limit discussed above: $I_{c1} \gg I_{c2}$ and $(2\pi L I_c')/\phi_0 < 1$. The results of the measurements are shown in Fig. 2. The dashed grey line represents the situation where $V_c < V_{c,\mathrm{critical}}$. We observe an almost sinusoidal dependence of the voltage on ϕ_{ext} with a voltage minimum (supercurrent maximum) at $\phi_{\mathrm{ext}} = 0$ indicating that both junctions are in the 0 state. The solid grey line represents the situation at $V_c > V_{c,\mathrm{critical}}$ with a voltage minimum at $\phi_{\mathrm{ext}} = \phi_0/2$ corresponding to a π state of junction 2. The black lines represent the situation around the transition ($V_c \approx V_{c,\mathrm{critical}} = 0.602$ mV). The sinusoidal form of the grey curves starts to disappear until we observe a doubling in the periodicity at $V_c = 0.602$ mV shown by the middle curve. Hence, $I_{sc2}(\varphi_2) \approx I_{c2,\mathrm{transition}} \sin(2\varphi_2)$ with $I_{c2,\mathrm{transition}}$ estimated to be $I_{c2,\mathrm{transition}} \approx 70$ nA [22].

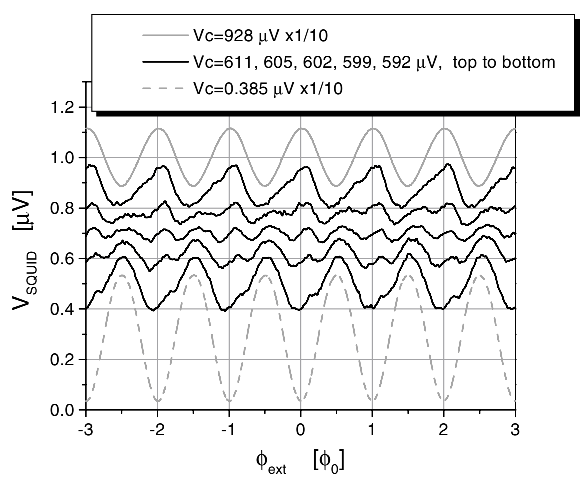

FIG. 2. Voltage over the SQUID (from contacts A to B in Fig. 1) at a bias current slightly larger than $I_{c,\mathrm{SQUID}}$ as a function of the ϕ_{ext} for different values of V_c. The amplitudes of the two grey curves are multiplied by 1/10. Around $V_{c,\mathrm{critical}} = 602\ \mu\mathrm{V}$, a doubling in periodicity of the voltage oscillations is observed.

To demonstrate how the energy landscape of the junction evolves around the transition, we can calculate $W(\varphi_2)$ using the data presented in Fig. 2 together with Eqs. (1) and (2). The result is shown in Fig. 3 for five values of V_c around $V_{c,\mathrm{critical}}$. As can be seen, the free energy of the junction evolves smoothly from the situation in which it has one minimum at $\varphi = 2\pi n$, via the situation where it has two local minima, to the situation where only the minimum at $\varphi = 2\pi(n + 1/2)$ is left. Identical behavior of higher harmonics in the Josephson current-phase relation has been observed in $YBa_2Cu_3O_{7-x}$ grain boundary junctions [23].

To compare these experimental results to the predictions from the quasiclassical theory, we solve the Usadel equation numerically to find the energy spectrum of the supercurrent carrying states in the normal region of the junction at different phases, $J(E, \varphi)$, shown for three values of φ in Fig. 4 (E is the energy with respect to the Fermi level). We consider a quasi-one-dimensional setup without the effect of the control probes on $J(E, \varphi)$. However, as long as the width of the wire connecting the normal region of the junction to the control channel is much smaller than the junction length, it does not affect the energy scales of $J(E, \varphi)$ above a few times E_T, only its overall magnitude [24]. The relation between the supercurrent through the junction I_{sc}, $J(E, \varphi)$, and $f(E, V_c)$ is given by [4–7]

$$I_{sc}(V_c, \varphi) = \frac{1}{2R_n}\int_{-\infty}^{\infty} dE[1 - 2f(E, V_c)]J(E, \varphi), \quad (3)$$

where R_n is the normal state resistance of the junction. Qualitatively, we can understand the presence of two minima in the free energy around the transition. It is a consequence of the phase dependence of $J(E, \varphi)$ shown in Fig. 4. At $\varphi = 0.1\pi$, shown by the dashed line, the positive contributions of $J(E, \varphi)$ shift to higher energies with respect to the solid curve at $\varphi = 0.5\pi$. At $\varphi = 0.9\pi$, a shift to lower energies is observed. The (rounded) double step shape of the energy distribution function implies that $f(E, V_c) \simeq 0.5$ in a window eV_c around the Fermi energy, a value which causes the integrand of Eq. (3) to vanish. The value of V_c that creates a form of $f(E, V_c)$ which causes the full integral Eq. (3) to vanish for $\varphi = 0.5$ due to an exact compensation of the (depopulated) positive states with the (almost completely populated) negative states would result in a positive supercurrent in Eq. (3) at $\varphi = 0.1\pi$ and a negative value of the supercurrent at $\varphi = 0.9\pi$ due to the phase dependence of the position of the positive contributions in $J(E, \varphi)$. Hence, the periodicity of the supercurrent-phase relation doubles, resulting in two minima in the junction free energy. This behavior is similar to the prediction for a ballistic SNS junction [2,3].

To be able to compare the observed behavior with the theoretical calculation of $J(E, \varphi)$, we calculate $f(E, V_c)$ as a function of inelastic electron-electron and electron-phonon interactions in the control channel. The effective strength of both interaction processes is obtained from a theoretical fit of $I_c(V_c)$ of a single controllable π junction with exactly identical control channel and reservoirs as junction 2 (see Ref. [19]). We then calculate $I_{sc2}(V_c, \varphi_2)$ by means of Eq. (3). We use E_T and the equilibrium critical current of junction 2 (at T_B and $V_c = 0$) $I_{c2,0}$ as fit parameters to obtain a quantitative agreement between the calculation and the data. We find $E_T = 31$ μeV and $I_{c2,0}R_n = 17.6$ μeV. The result is shown in Fig. 5. We observe that the calculated curves show the same behavior as the measurements. The amplitude of $I_{c2,\mathrm{transition}} =$ 105 nA (using $R_n = 1.6$ Ω), consistent with our previous estimate. The barrier height at the transition can be estimated using $I_{c2,\mathrm{transition}}$ and Eq. (1) to be 108 μeV. Model calculations indicate that an increase in the amplitude of the second harmonic, and, hence, an increase in

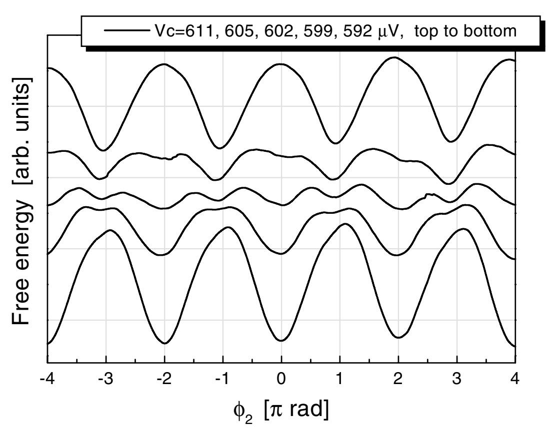

FIG. 3. Experimental free energy of junction 2 as a function of φ_2, $W(\varphi_2)$, obtained from the data presented in Fig. 2, Eq. (1), and Eq. (2).

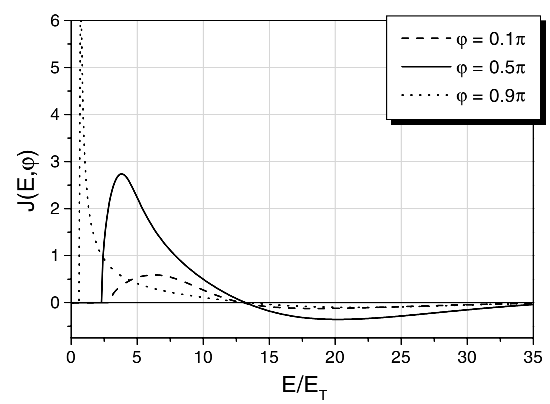

FIG. 4. Supercurrent carrying density of states $J(E, \varphi)$ for three different values of the macroscopic phase difference φ, using $\Delta/E_T = 0.52$ with Δ the superconducting gap of the S electrodes and E the energy with respect to the Fermi level. The positive contributions in $J(E, \varphi)$ shift to lower energies at higher values of φ.

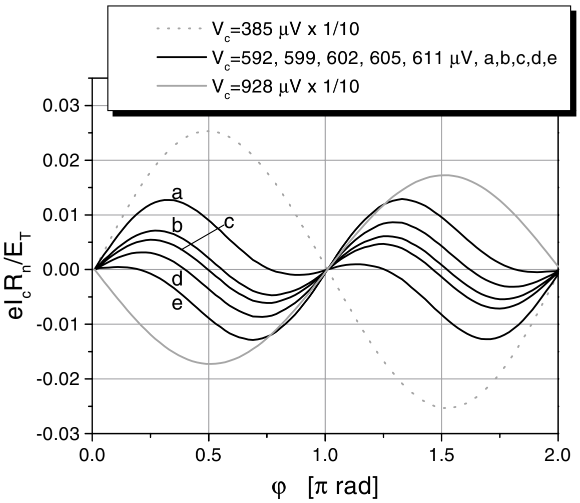

FIG. 5. Theoretical critical current as a function of φ for the top junction of the device shown in Fig. 1.

barrier height between the 0 and the π state, would be possible by measuring at a lower bath temperature.

In conclusion, we have measured the full supercurrent-phase relation of a controllable π junction around its transition and observed a doubling in the periodicity of the supercurrent-phase relation around the transition. This doubling of the periodicity as well as the relative amplitude of the double Josephson current are in good agreement with theoretical predictions. The implication is that the free energy of the junction around the transition evolves as follows. It starts with one minimum at $\phi = 0$ in the 0 state, then, close to the transition, develops a second minimum at $\phi = \pi$. This evolves from a local to a global minimum as the one at $\phi = 0$ slowly vanishes in the π state.

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[22] The estimation of $I_{c2,\mathrm{transition}}$ is done by a linear interpolation of I_{c2} and $\Delta V_{\mathrm{SQUID}}$: We measured that $I_{c2} = 11\ \mu\mathrm{A}$ results in $\Delta V_{\mathrm{SQUID}} = 7.8\ \mu\mathrm{V}$ (at $V_c = 0$), so $\Delta V_{\mathrm{SQUID}} = 0.05\ \mu\mathrm{V}$ at $V_c = 602\ \mu\mathrm{V}$ should give $I_{c2,\mathrm{transition}} = 70$ nA. This is correct only in first order, but a better estimate of I_{c2} from the data in Fig. 2 is impossible for several reasons: (i) $\max|I_{c,\mathrm{SQUID}}| - \min|I_{c,\mathrm{SQUID}}|$ depends strongly on the effect of the self-inductance $(2\pi L I_c')/\phi_0$. Moreover, we use a symmetrical bias current so the relation between $\Delta V_{\mathrm{SQUID}}$ and $\max|I_{c,\mathrm{SQUID}}| - \min|I_{c,\mathrm{SQUID}}|$ depends not only on the bias current but also on the fraction I_{c1}/I_{c2} if $(2\pi L I_c')/\phi_0 \neq 0$. (ii) In the situation around the transition, where $I_{sc1} \approx I_{c1}\sin(\varphi)$ and $I_{sc2} \approx I_{c2,\mathrm{transition}}\sin(2\varphi)$, the magnitude of the critical current of the SQUID depends on the current direction. The result is a reduction in the signal which is extremely sensitive to the exact bias current.
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