Phase Sensitive Experiments in Ferromagnetic-Based Josephson Junctions

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We have measured the ground state of ferromagnetic Josephson junctions using a single dc SQUID (superconducting quantum interference device).We show that the Josephson coupling is either positive (0 coupling) or negative (π coupling) depending on the ferromagnetic layer thickness. As expected, the sign change of the Josephson coupling is observed as a shift of half a quantum flux in the SQUID diffraction pattern when operating in the linear limit.

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Introduction.—A series of experiments have recently shown that the ground state of a Josephson junction separating two superconductors can be defined by a negative energy. Josephson junctions with such a negative coupling are commonly called π junctions as negative coupling is obtained introducing a π-phase shift in the current-phase relationship of the junction. π coupling was first observed in high temperature superconductors (HTCS) [1] and attributed to a sign change of the superconducting order parameter on the Fermi surface suggesting unconventional pairing. It has been also reported in ^{3}He and related to the p-wave symmetry of the superfluid condensate in the B phase [2]. More recently experiments with conventional superconductors have shown that π coupling does not necessarily require unconventional pairing. In superconductor/normal/superconductor (SNS) junctions, changing the quasiparticle distribution function in the normal layer can reverse the direction of the dissipationless current through the junction [3]. Similarly, a negative supercurrent can circulate when the normal layer becomes ferromagnetic (F).

In SFS junctions, π coupling was first suggested to explain the nonmonotonic dependence of the critical temperature [4] as a function of the ferromagnetic thickness in SF multilayers, in agreement with theoretical calculations [5]. Oscillations of the Josephson critical current as a function of the temperature [6] and the ferromagnetic layer thickness [7] were also observed and interpreted as a transition from 0 to π coupling as predicted by Buzdin *et al.* [8]. The manifestation of these oscillations in the superconducting density of states measured in F by planar tunneling spectroscopy provided a further microscopic signature of π coupling [9].

Here, we present a phase sensitive experiment that probes the superconducting phase directly by the interference of the quantum mechanical phase of two ferromagnetic Josephson junctions. π coupling originates from the microscopic transport mechanism at a S/F interface, i.e., Andreev reflection [10]: an incoming electron in F with energy lower than the superconducting gap is reflected as a hole with opposite spin while a Cooper pair is transferred into S. The electron and the hole accumulate a phase difference of $\delta\varphi = \Delta K x$ depending on the traveled distance x from the interface. In a normal metal, the difference between the hole and the electron momenta, $\Delta k = 2E/\hbar v_F$, depends on the energy, E, of the quasiparticles. As Andreev reflections reverse the quasiparticle spin, in a ferromagnet the electron and the hole accumulate an extra momentum $\Delta K = \Delta k + Q$, with $Q = 2E_{\mathrm{ex}}/\hbar v_F$ coming from the spin splitting of the conduction bands. In general the exchange energy is much larger than the superconducting energy gap; thus ΔK is practically independent of the quasiparticle energy and is equal to Q. As a consequence, the phase difference between electron and hole generates a sign reversal oscillating term in the real part of the superconducting order parameter as a function of the distance from the S/F interface. Therefore, when the ferromagnetic layer is coupled with another superconductor, the Josephson critical current through the junction also oscillates as a function of the ferromagnetic layer thickness. Negative critical current gives rise to a negative Josephson coupling. The oscillation length scale and damping in the clean limit is given by $\xi_{\mathrm{F}} = 1/Q = \hbar v_F/2E_{\mathrm{ex}}$, ranging from some angstroms for ferromagnetic materials such as Fe, Co, or Ni to some nanometers for ferromagnetic alloys such as PdNi or CuNi [6,9] where E_{ex} is smaller. Here we have introduced the Andreev reflections in the case where the quasiparticle momenta is a good quantum number; the basic physics is unchanged in the dirty limit [8,11].

The 0 to π transition can be detected using a dc SQUID. In a dc SQUID with junction critical currents I_{ca} and I_{cb}, the total current flowing through the device is $I = I_{ca}\sin\varphi_a + I_{cb}\sin\varphi_b$, where φ_a and φ_b are the gauge invariant phase differences across the junctions. The effective magnetic flux in the loop is given by $\phi = \phi_{\mathrm{ext}} + LI_S$, where ϕ_{ext} is the external flux and I_S is the shielding circulating current. The phase around the loop is subject to the constraint $\varphi_a - \varphi_b = 2\pi\frac{\phi}{\phi_0} + 2\pi n + \delta_{ab}$, where δ_{ab} accounts for the sum of the intrinsic phase differences along the two junctions and, in unconventional superconductors, the intrinsic phase difference of the condensate. For a dc SQUID with negligible loop inductance ($LI_c \ll \phi_0$, where ϕ_0 is the quantum flux) and equal junction critical currents, i.e., $I_{ca} = I_{cb} = I_0$, the critical current modulates with applied flux from a maximum of $2I_0$ to zero current according to $I_c(\phi_{\rm ext}) = 2I_0|\cos[\pi\frac{\phi_{\rm ext}}{\phi_0} + \delta_{ab}/2]|$ [12]. Thus, for conventional superconductors, if one of the two junctions is a π junction the diffraction pattern is shifted of half a quantum flux as $\delta_{ab} = \pi$. If both junctions are π junctions, $\delta_{ab} = 2\pi$, the diffraction pattern is shifted by a flux quantum and it is identical to the diffraction pattern of a SQUID with two 0 junctions. Recently a superconducting array of identical S/F/S π junctions has been realized in which the transition to the π state is induced by the temperature [13]. Here we present a 0-π interferometer, which involves only one dc SQUID with a thickness dependent transition to the π state.

Sample.—The SQUIDs were obtained by lift-off after angle evaporation through resin masks. The mask was fabricated from a trilayer, PES(0.8 μm)/Ge(45 nm)/PMMA(110 nm), where the poly PhenyleneEtherSulfone (PES) is a thermostable polymer [14]. After patterning the PMMA (polymethylmethacrylate) by electron beam lithography and removing the unprotected Ge layer by reactive ion etching the bottom layer PES was etched by a combination of wet process and oxygen plasma. More details on the fabrication process will be given elsewhere [15]. The samples were deposited by e-gun evaporation in a typical base pressure of 10^{-9} Torr, rising to 10^{-8} Torr during deposition. They were prepared in five steps with the arrows in Fig. 1 indicating the direction of the different evaporations: (1) evaporation of the first Nb layer (25 nm), (2) direct oxidation of the Nb just after deposition, (3) evaporation of the first PdNi layer ($d_{\rm F1}$), (4) evaporation of the second PdNi layer ($d_{\rm F2}$), and (5) evaporation of the Nb counter electrode (25 nm). The outer dimensions of the superconducting rectangle are 8×12 μm^2; the junction size is 0.5 μm $\times$ 0.7 μm. The geometrical inductance of the loop is about $L_G \approx 40$ pH [16,17] so that the SQUID is in the linear limit for critical currents of some microamperes ($LI_c = 0.1\phi_0 < \phi_0/2$). On the same substrate, we prepared four types of masks corresponding to 0-0, 0-π, and π-π SQUIDs with ferromagnetic layers in each junction and to a 0-0 SQUID without a ferromagnetic layer. For 0-0 and π-π SQUIDs, both junctions are obtained from the same PdNi evaporation.

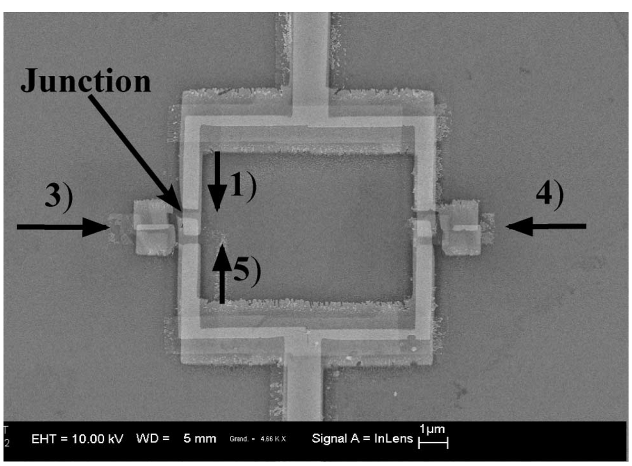

FIG. 1. Picture of a 0-π SQUID. The arrows correspond to the different evaporation directions as described in the text.

The ferromagnetic layer thickness, $d_{\rm F1}$ and $d_{\rm F2}$ corresponding to 0 and π coupling with similar critical currents, were chosen following the recently reported [7] dependence of the Josephson coupling as a function of the PdNi thickness. As the Ni concentration measured by Rutherford backscattering in that case was smaller (12% [7]) than the one (18%) used here, the ferromagnetic layer thickness was renormalized to account for the increased exchange energy. Assuming that the increase in $E_{\rm ex}$ is linear with Ni concentration as confirmed by a Curie temperature measurement on reference samples [18], the transition from 0 to π junction is expected at about 4.6 nm. The evaporated ferromagnetic layer thickness for a 0 junction was 4.2 nm, and for a π junction it was 8 nm.

Results and discussions.—The $I(V)$ curves were monitored on a digital oscilloscope with the junction current supplied by a sinusoidal oscillator ranging from 100 to 200 Hz. The critical current was measured with a sinusoidal ac current of 0.04 μA amplitude superposed on a dc current fixed by a dichotomy method around the $dV/dI(I)$ transition of the SQUID. The magnetic field was supplied by a superconducting coil without earth field shielding. In order to account for this field offset we always measured the relative shift between two SQUIDs at the same time. Measurements of critical current modulations of identical SQUIDs separated by a distance of 1.5 mm showed that the field inhomogeneity was below 1%. We made also sure that after cooling, heating above the Nb critical temperature did not effect the relative zero of the magnetic field. Figure 2 shows the temperature dependence of the resistance for a Nb/NbO$_x$/PdNi/Nb SQUID and for a Nb/NbO$_x$/Nb SQUID. The cross section of the SQUID junctions is sketched in the inset of Fig. 2. The critical temperature, T_c, of the SQUID without PdNi is 7 K. Note that the NbO$_x$ layer decouples the Nb upper layer (25 nm) from the lower one (25 nm). The T_c of the SQUID with PdNi is lower due to the reduced critical temperature of the PdNi/Nb bilayer. The T_c of π-π and 0-0 SQUIDs with PdNi is about $T_c = 5.3$ K and 5.6 K, respectively. The T_c of 0-0 SQUIDs is slightly larger due to the thinner PdNi thickness. As the Nb layer is relatively thin the proximity effect with PdNi produces a strong depairing. When the Nb thickness is risen up to 50 nm and no oxidation of the bottom Nb layer is performed, the SQUIDs T_c with and without PdNi are nearly the same. The $I(V)$ curve (Fig. 2 inset) for a 0-π-Nb/NbO$_x$/PdNi/Nb SQUID with a typical critical current of

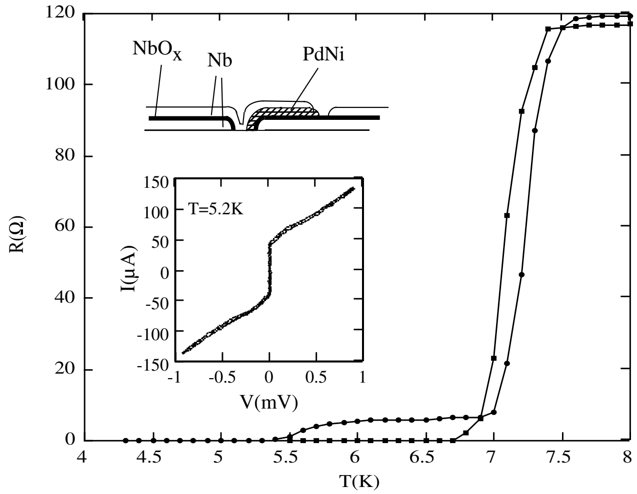

FIG. 2. $R(T)$ curve for a Nb/NbO$_x$/PdNi/Nb SQUID and for a Nb/NbO$_x$/Nb SQUID. Inset: a drawing of the cross section of a single SQUID junction and $I(V)$ characteristic for a 0-π Nb/NbO$_x$/PdNi/Nb SQUID.

40 μA at 5.2 K is nonhysteretic as expected near T_c in the resistive shunted junction model [12]. Two main transitions can be separated: first, the junction with a resistance of 8 Ω, second, the Nb/PdNi bilayer with a critical current of 70 μA and a resistance of 2 Ω. We observed that SQUIDs without NbO$_x$ show a much lower junction resistance and a much higher critical current (typically 200 μA at $T = 7$ K for a Nb layer thickness of 50 nm). They can be hardly used to operate in the linear limit as required. Even when the bottom Nb layer is oxidized, the SQUID is in the linear limit only close to T_c.

The main result of this Letter is shown in Fig. 3: the modulation curves $I_c(B)$ for a 0-0, a 0-π and a π-π SQUID show no shift between a 0-0 SQUID and a π-π SQUID, whereas a shift of $\phi_0/2$ is observed between a 0-π SQUID and a 0-0 SQUID or π-π SQUID. We have reproduced these results on five samples per SQUID type. For each of them we have also checked that there are no changes when warming up above the T_c of Nb and cooling down several times. This rules out aging effects due to a vortex distribution. We always observed the expected $\phi_0/2$ shift for small critical currents (a few μA). Note that the flux quantum of 20 μT is the same for SQUIDs containing ferromagnetic junctions or not and corresponds to the outer dimensions of the SQUID. This is probably due to the phase gradient produced by the finite supercurrents in the loop. An evaluation of the effective penetration length in the dirty limit,

$$\lambda_{\rm eff}(l,T) = \frac{\lambda_L}{\sqrt{1-(T/T_c)^2}}\sqrt{\left(\frac{\xi_{\rm BCS}^{T_c^{\rm Nb}=7\,{\rm K}}}{l}+1\right)},$$

where $l = 9$ nm [18], $\xi_{\rm BCS} = 0.180\hbar v_F/k_B T_c = 51.4$ nm with $v_F = 2.77 \times 10^7$ cm/s [19] gives $\lambda_{\rm eff} = 0.15$ μm at $T = 5.3$ K, comparable to the SQUID arm width

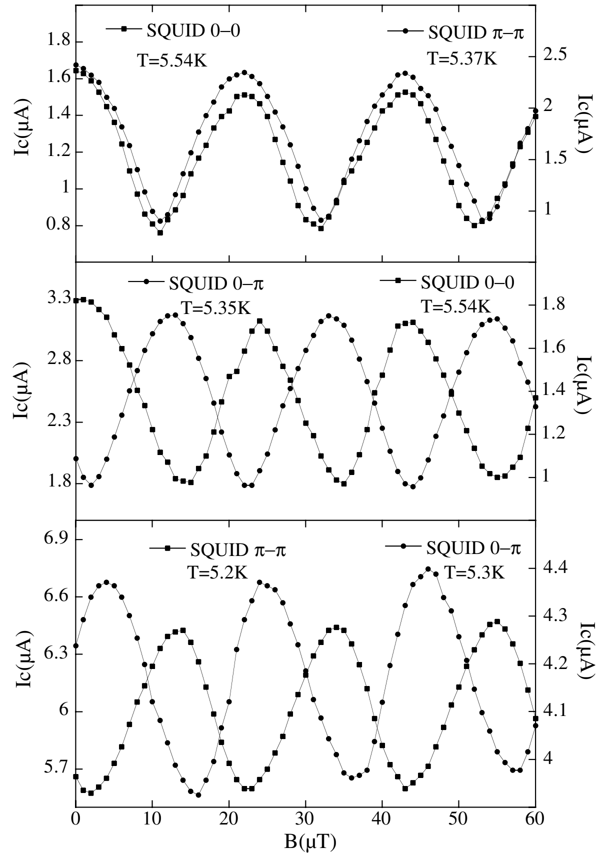

FIG. 3. Critical current modulations showing the expected no-shift between a 0-0 SQUID and a π-π SQUID and of $\phi_0/2$ between a 0-π SQUID and a 0-0 SQUID or π-π SQUID. Each couple of the SQUIDs has been measured at the same time. As the T_c of 0-0, 0-π, π-π SQUIDs are different (as explained in the text), the $I_c(B)$ curves are observed at different temperatures in order to measure in the same range of critical currents for each couple.

(0.5 μm). Therefore the phase quantization in the SQUID loop cannot neglect the shielding supercurrents as usually assumed. The amplitude of the $I_c(B)$ modulations is not $\Delta I_c/I_c = 100\%$ as expected for an ideal symmetric dc SQUID in the linear limit. A reduced modulation depth is usually found in damped SQUIDs with a critical current imbalance between the SQUID arms or large geometric inductance. First, assuming SQUIDs with identical junction critical currents, we estimate the decrease in the modulation depth taking into account the finite screening. From the geometrical inductance $L_G = 40$ pH we find a screening factor $\beta_L/2\pi = L_G I_c/\phi_0$ of about 0.1. Similarly, the increase in the extra phase gradient due to finite supercurrents can be simulated by a kinetic inductance $L_K = \mu_0 \lambda_{\rm eff}^2 t/\sigma$ and hence a screening factor $\beta_L/2\pi = L_K I_c/\phi_0$ [20], where t is the circumference and σ is the SQUID arm cross section. We estimate $L_K = 33$ pH, comparable to the geometric inductance. Although the amplitude of the critical current modulations decreases lowering the temperature

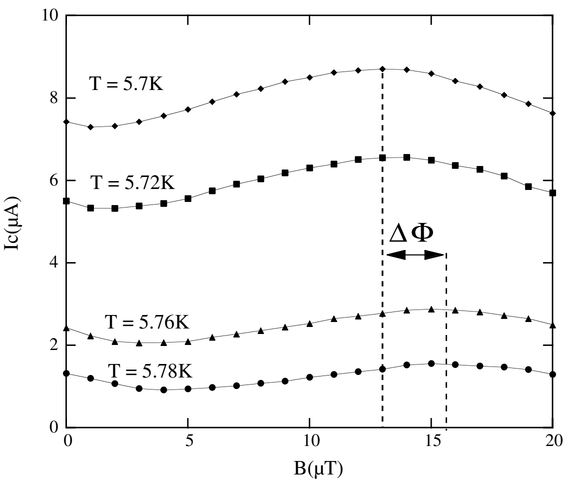

FIG. 4. $I_c(B)$ curves for a 0-0 SQUID with decreasing temperature. The curve is shifted by $\Delta\phi = 0.13\phi_0$ due to critical current imbalance between the SQUID junctions.

as expected because of the increase in the screening factor, the measured modulation depth is usually smaller than that expected theoretically ($\approx 70\%$) obtained adding the geometrical and kinetic inductances. In order to analyze the impact of the junction critical current difference on the critical current modulations we cut one SQUID arm by a focused ion beam and measured the critical current of the junction of the uncut arm. The critical current imbalance at the temperatures indicated in Fig. 3 is still too small to explain the reduction in the critical current oscillations for all the SQUIDs that we have measured [21]. On the other hand, we note a larger critical current imbalance for some 0-0 and 0-π SQUIDs. This may rise from the fact that a small variation in the ferromagnetic layer thickness produces a bigger change of the Josephson coupling for 0 junctions than π junctions [7].

SQUIDs with a significant critical current imbalance show a shift of the diffraction pattern lowering temperature as reported in Fig. 4. This shift results from the self-flux generated by the current imbalance in the SQUID arms. Lowering the temperature, the contribution of the self-flux to the total flux in the SQUID loop increases as does the critical current. The same effect has been observed recently in HTCS SQUIDs [22]. A critical current difference ΔI should result in a $I_c(B)$ shift of $\Delta B = L\Delta I_c/2$ [23]. For the SQUID shown in Fig. 4 the observed shift of $0.13\phi_0$ is consistent to the corresponding measured critical current difference of 6 μA at 5.7 K and the total inductance $L = L_G + L_K \approx 75$ pH.

In summary, we have produced niobium dc SQUIDs based on ferromagnetic Josephson junctions. The phase difference of the junctions within the SQUID is fixed by the ferromagnetic layer thickness. SQUIDs with π junction show a half quantum flux shift in the diffraction pattern. Ferromagnetic π junctions could be easily implemented as a π shifter in superconducting networks and devices. In particular, they may be used in macroscopic quantum mechanics experiments and applications in superconducting based Q-bits.

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