

## Josephson Junction through a Thin Ferromagnetic Layer: Negative Coupling

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We investigate Josephson coupling through a ferromagnetic thin film using superconductor-insulator-ferromagnet-superconductor planar junctions. Damped oscillations of the critical current are observed as a function of the ferromagnetic layer thickness. We show that they result from the exchange energy gained or lost by a quasiparticle Andreev-reflected at the ferromagnet-superconductor interface. The critical current cancels out at the transition from positive (“0”) to negative (“ $\pi$ ”) coupling, in agreement with theoretical calculations.

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Current can flow without dissipation through a thin insulating layer or a weak link separating two superconductors. Because of the quantum character of the superflow a phase difference,  $\Delta\theta$ , between the superconductors appears. The current-phase relationship of the link is a periodic function of  $\Delta\theta$ . For a tunnel barrier, as first found by Josephson [1], it is given by  $I = I_c \sin(\Delta\theta)$ , where  $I_c$  is the critical current. In this Letter, we show that  $I_c$  can change sign when the superconductors are coupled via a thin ferromagnetic layer. Referring to the Josephson current-phase relationship, this change corresponds to a  $\pi$ -phase shift of  $\Delta\theta$ . Thus Josephson junctions presenting a negative coupling are usually called  $\pi$  junctions. We specifically observe the transition from a positive to a negative coupling or equivalently from “0” to “ $\pi$ ” junctions by increasing the ferromagnetic layer thickness.

The possibility of a negative Josephson coupling was first raised by Kulik [2]. It was attributed to the spin flip of the electrons forming a Cooper pair when tunneling through an insulator containing uncorrelated magnetic impurities. This channel, which coexists with direct tunneling, was suggested to reduce the critical current of the junction. Whether or not it can overtake direct tunneling, leading to a negative critical current, remains uncertain.

More recently Buzdin *et al.* [3] showed that in ballistic superconductor/ferromagnet/superconductor weak links (S/F/S),  $I_c$  displays damped oscillations around zero as a function of  $\theta_F = 2E_{ex}d_F/\hbar v_F$  leading to  $\pi$  coupling when  $I_c$  is negative.  $E_{ex}$  is the exchange energy,  $d_F$  the thickness of  $F$ , and  $v_F$  the Fermi velocity. Physically, this is a consequence of the phase change of the pair function induced in  $F$  by the proximity effect [4]. In fact, energy conservation requires that a Cooper pair entering into the ferromagnet receives a finite momentum,  $\Delta p = \hbar v_F/2E_{ex}$ , from the spin splitting of the up and down bands [5]. By quantum mechanics,  $\Delta p$  modifies the phase,  $\theta_x = \Delta p \cdot x$ , of the pair wave function that increases linearly with the distance,  $x$ , from the S/F interface. As the Josephson critical current is proportional to the pair amplitude in  $F$ ,  $I_c$  follows the sign of the order parameter in  $F$ . It is either positive or negative depending on the phase  $\theta_F$  accumulate in  $F$ .

Microscopically the transfer of Cooper pairs through the ferromagnet occurs via Andreev reflections [6]. An electronlike excitation in  $F$  with energy lower than the superconducting energy gap,  $\Delta$ , cannot enter into the superconductor. It is reflected at the  $F/S$  interface as a hole and it is then reflected back as an electron at the opposite  $S/F$  interface as illustrated in the inset of Fig. 1 [8]. The constructive interference of the electronlike and holelike excitations gives rise to Andreev bound states (ABS) in  $F$ , which carry the supercurrent. The same mechanism accounts for the Josephson coupling through a normal metal [8], but in a ferromagnet the spectrum of the ABS is affected by the spin splitting of the spin bands as the Andreev reflections reverse the spin of the quasiparticles [9]. Therefore the spectral current density,  $Q(\epsilon)$  that defines the current density per bound state in  $F$  is a function of the exchange energy,  $E_{ex}$ . In the dirty limit,  $Q(\epsilon)$  can be obtained by solving [7] the Usadel equations

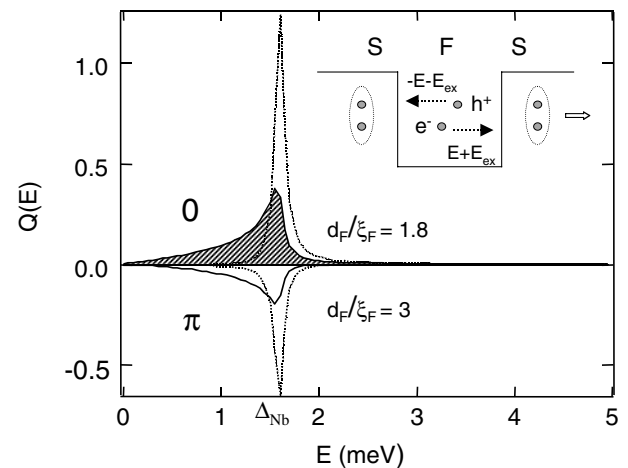


FIG. 1. Spectral current density,  $Q(\epsilon)$ , as a function of energy for SFS (full line) and SIFS (dashed line) Josephson junctions.  $Q(\epsilon)$  is calculated from the Usadel equation [7] for two different  $d_F/\xi_F$  ratios. The “0” ( $d_F/\xi_F = 1.8$ ) and “ $\pi$ ” ( $d_F/\xi_F = 3$ ) couplings result from positive (dashed area) and negative integrals, respectively. The inset illustrates the transfer mechanism for Cooper pairs through  $F$  by Andreev reflections.

[10]. Because of the strong dephasing due to the exchange energy, the order parameter in  $F$  decays on a short length scale given by  $\xi_F = \sqrt{\hbar D/E_{ex}}$ , the coherence length in  $F$ , where  $D$  is the diffusion constant. When the thickness of  $F$  is on the order of  $\xi_F$  one can use the “long junction” approximation [11] to evaluate  $Q(\epsilon)$ . If the energy gap is smaller than  $E_{ex}$  and the Thouless energy  $E_{Th} = \hbar D/d_F^2$ , as in the experiment described below, the energy dependence of  $Q(\epsilon)$  is basically fixed by the BCS superconducting density of states [12]. Therefore, the spectral current density is peaked at the superconducting energy gap as shown in Fig. 1. It is either positive or negative as a function of  $d_F/\xi_F$  as well as the critical current  $I_c$  [13] at zero temperature which is obtained by integrating  $Q(\epsilon)$ .

The existence of “ $\pi$ ” coupling was first suggested to explain the nonmonotonic dependence of the critical temperature observed in  $F/S$  multilayers as a function of  $d_F$  [14]. But it is only very recently that density of states [4] and Josephson [15] measurements have confirmed this hypothesis. The critical current of  $SFS$  junctions has been shown to increase, decrease, possibly cancel, and increase again lowering temperature close to zero coupling (i.e.,  $I_c R_n \leq 1$  nV for  $d_F \approx 2\xi_F$ ). On the other hand, planar junctions where a thin insulating barrier is inserted between the superconducting counterelectrode and the ferromagnet allow measurement of the Josephson coupling as a function of  $d_F/\xi_F$  or equivalently  $\sqrt{E_{ex}/E_{Th}}$ . As the tunnel barrier raises the junction resistance,  $R_n$ ,  $I_c$  is lowered to measurable values (typically 10–100  $\mu$ A) for any  $d_F/\xi_F$  ratio. In a superconductor/insulator/ferromagnet/superconductor (SIFS) junction the energy spectrum of the Andreev bound states in  $F$  is modified by the insulating layer [16]. However, we have calculated [17] the current spectral density as reported in Fig. 1 and found that it shows the same general behavior as observed in  $SFS$  junctions, including the change of sign as a function of  $d_F$ .

Planar Nb/Al/Al<sub>2</sub>O<sub>3</sub>/PdNi/Nb junctions were fabricated by  $e$ -gun thin film evaporation in a typical base pressure of  $10^{-9}$  Torr, rising to  $10^{-8}$  Torr during deposition. The film thickness was monitored during growth to better than 1 Å by a quartz balance. The junction had the standard four terminal cross geometry [4], defined during evaporation by shadow masks. A Si wafer was first covered by 500 Å of SiO. Then, a 1500 Å thick Nb strip was evaporated and backed by 500 Å of Al. The critical temperature of the Nb strip was 9.2 K and its residual-resistance-ratio about 4. The Al surface oxidation was performed in a glow discharge during 1 min, and completed in a 10 Torr oxygen atmosphere during 2 min. A square window of 1 mm  $\times$  1 mm left in a 500 Å thick SiO layer deposited on the Al defined the junction area. Eight junctions were aligned on the same Nb/Al strip. A PdNi strip (thickness 40–150 Å) perpendicular to the Nb/Al strip was then evaporated on each junction and covered by a 500 Å thick Nb counterelectrode ( $T_c = 8.7$  K). The junction resistance varied from 50 to 400 m $\Omega$ . The entire

process was carried out without breaking the vacuum in the deposition chamber. The Ni concentration was about 12% as checked by Rutherford backscattering spectrometry on samples evaporated at the same time. At this concentration, the PdNi is an itinerant ferromagnet [18], whose Curie temperature is about 100 K (see inset Fig. 4). The junction was polarized by an ac current, the  $I$ - $V$  characteristics being monitored on a digital commercial oscilloscope set in the X-Y mode and directly recorded from the oscilloscope. Measurements were performed down to 1.5 K; the minimum detectable value of  $I_c$  was about 1  $\mu$ A. A  $\mu$ -metal shield reduced the residual magnetic field on the sample to  $10^{-2}$  G.

The Nb/Al/Al<sub>2</sub>O<sub>3</sub>/Nb junctions without a ferromagnetic layer showed high quality tunneling. At low temperature the Nb/Al bilayer is a homogeneous superconductor. The  $I$ - $V$  characteristic is hysteretic as expected for SIS junctions [20]. The subgap conductance at 1.5 K is lower  $10^{-2}$  of the high energy conductance corresponding to negligible current leakage as shown in the inset of Fig. 2. The  $I_c R_n$  product is 1.25 mV with junction to junction fluctuations of at most 15%. The inset of Fig. 2 also shows the  $I$ - $V$  characteristics of a Nb/Al/Al<sub>2</sub>O<sub>3</sub>/PdNi/Nb junction with a thin layer of PdNi ( $d_F = 50$  Å). The critical current is strongly reduced while the subgap conductance is enhanced. Both effects result from the suppression of the order parameter in  $F$  by the exchange field. The superconducting correlations are so efficiently reduced at the

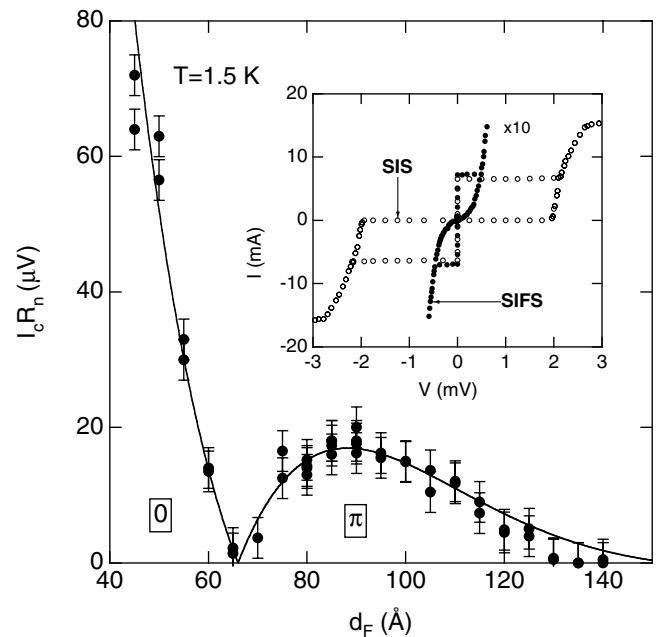


FIG. 2. Josephson coupling as a function of thickness of the PdNi layer (full circles). The critical current cancels out at  $d_F \approx 65$  Å indicating the transition from “0” to “ $\pi$ ” coupling. The full line is the best fit obtained from the theory [Eq. (1)] as described in the text. Inset shows typical  $I$ - $V$  characteristics of two junctions with (full circles), and without (empty circles) PdNi layer.

Al/Al<sub>2</sub>O<sub>3</sub>/PdNi interface that the dissipative branch of the  $I$ - $V$  curve reflects the energy dependence of the density of states in the Nb/Al bilayer.

The Josephson coupling,  $I_c R_n$ , as a function of the thickness of the ferromagnetic layer is presented in the main body of Fig. 2. We chose this representation to eliminate fluctuations in the critical current that are simply due to fluctuations in the junction resistance. For each value of  $d_F$ , the  $I_c R_n$  product of two different junctions is reported, with typical sample to sample variations of less than 10%, illustrating the reproducibility of the junction fabrication technique. The Josephson coupling is of the order of 10–20  $\mu$ V, i.e., 100 times lower than that measured on junctions without PdNi. The sign of  $I_c$  cannot be determined from the  $I$ - $V$  characteristics and the transition from "0" to " $\pi$ " coupling is revealed as a zero of the  $I_c R_n$  product. The latter occurs at  $d_F \approx 65$  Å, consistent with our recent measurements of the density of states in  $F$  [4]. Figure 2 also shows the best fit to the theory in which we have calculated the  $I_c R_n$  product by integrating the spectral current density found solving the Usadel equations for a SIFS junction as shown in Fig. 1. For a small transparency PdNi/Nb interface, the Usadel equations can be linearized. We obtain for  $\gamma_b \gg 1$  [17] an analytical expression for  $I_c R_n$ :

$$I_c R_n = \frac{\pi \Delta}{2e} \frac{1}{\gamma_b} \frac{\cos \frac{d_F}{\xi_F} \cosh \frac{d_F}{\xi_F} + \sin \frac{d_F}{\xi_F} \sinh \frac{d_F}{\xi_F}}{\cos^2 \frac{d_F}{\xi_F} \cosh^2 \frac{d_F}{\xi_F} + \sin^2 \frac{d_F}{\xi_F} \sinh^2 \frac{d_F}{\xi_F}}, \quad (1)$$

where  $\gamma_b = \rho_B \sigma_F / \xi_F$ ,  $\rho_B$  is the PdNi/Nb interface resistance and  $\sigma_B$  the conductance of PdNi layer. The Josephson coupling decreases linearly with  $\gamma_b$  as do the number of Andreev reflections at the  $S/F$  interface [21]. Although Eq. (1) displays exponentially damped oscillations, its first maximum occurs for a lower thickness than that predicted by earlier calculations in SFS junctions [13]. This is due to the effect of the insulating layer on the Andreev amplitude at the  $F/S$  interface. The small scattering of the experimental data around the fit indicates small sample to sample variations of PdNi/Nb interface resistance. The fitting parameters are  $\gamma_b = 5.3$  and  $\xi_F = 28$  Å. The value of  $\gamma_b$  is the same as that deduced from the thickness dependence of the zero energy density of states [4] while the coherence length in PdNi is smaller possibly because of the few percent increase in the Ni concentration. The corresponding exchange energy is  $E_{ex} = 35$  meV, taking  $v_F = 2 \times 10^7$  cm/s [22] and the mean free path in the PdNi equal to  $d_F$  as found by resistivity measurements on bare PdNi thin films.

In Fig. 3 the critical current,  $I_c$  as a function of the magnetic flux is presented for (i) a junction without PdNi and (ii) three junctions with different thickness of PdNi but roughly the same  $I_c R_n$  product. One of the latter gives "0" coupling (60 Å) and two " $\pi$ " coupling (90 and 110 Å). The diffraction patterns are those expected for a square junction with homogeneous current density [20] provided a

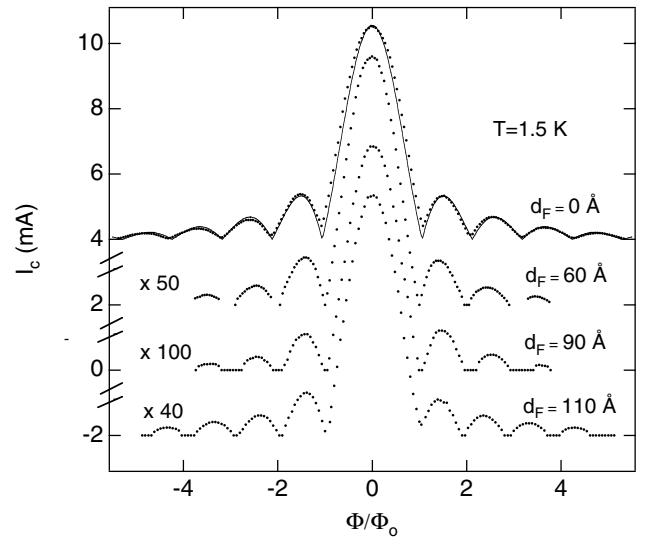


FIG. 3. Critical current as a function of magnetic flux for four different junctions, three with and one without PdNi. The data are shifted vertically for clarity. The full line is the Fraunhofer pattern expected for a square junction with homogeneous current density and a misalignment of  $7^\circ$  between the field and the junction edge. The junctions with PdNi have about the same coupling. The diffraction patterns are not affected by the PdNi magnetization.

small misorientation of  $7^\circ$  between the field direction and one edge of the junction is included (see fit in Fig. 3). The critical current cancels out when an integer of flux quanta settles into the junction. As the field penetrates in a volume whose cross section is given by  $w(2\lambda + d_F)$ , where  $\lambda$  is the penetration depth and  $w = 1$  mm the junction width, an estimate of  $\lambda$  for  $H = 0.2$  G corresponding to the first quantum flux, is  $\lambda = 500$  Å, consistent with the value expected for Nb thin films [20]. Importantly, the maximum critical current occurs at zero applied field independently of the ferromagnetic layer thickness. This shows that the Josephson coupling presented in Fig. 2 is indeed the maximum coupling as assumed. It also indicates that the field produced by the thin PdNi film falls off rapidly in the superconductor when one moves away from the  $S/F$  interface. Therefore no orbital dephasing is generated in  $S$  by the ferromagnetic layer which acts only through the spin degree of freedom as expected in the Pauli limit [23].

The temperature dependence of the critical current is found by integrating the spectral current density,  $Q(\epsilon)$  multiplied by the distribution function [7]. Far from  $d_F \approx 3\pi/4\xi_F$ , where the critical current cancels out,  $I_c$  is mainly determined by the temperature dependence of the density of states on each side of the junction. As the energy dependence of the density of states in the Al and PdNi is not exactly the BCS density of states, the temperature dependence of  $I_c$  shows a more linear behavior [19] than that originally found by Ambegaokar-Baratoff [24] for two bulk superconductors separated by a tunnel barrier. This linear behavior is shown in Fig. 4 where the temperature

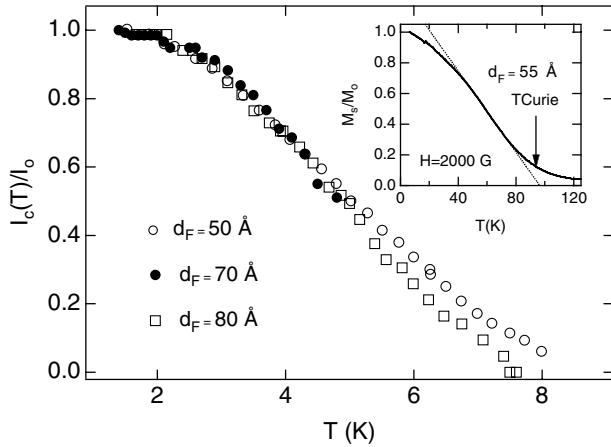


FIG. 4. Normalized temperature dependence of the critical current for three different PdNi thicknesses. Note linear dependence expected for superconductor-insulator-normal-superconductor tunnel junction [19]. Inset shows normalized saturation magnetization as a function of temperature, measured via anomalous Hall effect for a bare PdNi thin film.

dependence of the normalized critical current for three junctions with different PdNi thickness is reported ( $d_F = 50 \text{ \AA}$  for “0” coupling,  $d_F = 80 \text{ \AA}$  for “ $\pi$ ” coupling and  $d_F = 70 \text{ \AA}$  near the transition). The critical current of the junction with a  $70 \text{ \AA}$  thick PdNi layer is too small to be measured at  $T > 5 \text{ K}$ .

When  $d_F \approx 3\pi/4\xi_F$ , where the critical current cancels, the spectral current density  $Q(\epsilon)$ , changes sign as a function of energy. Thus, changing the occupation of the ABS by raising the temperature allows to shift the spectral weight from a positive to a negative current density or equivalently from forward to back-ward ABS. This results in a temperature induced “0-junction”–“ $\pi$ -junction” transition as recently observed in SFS junctions [15]. However, we estimate that such a transition requires being as close to the critical point as the  $I_c R_n$  product is lower than  $1 \mu\text{V}$ . Because of relatively high junction resistance the minimum  $I_c R_n$  product measurable in our SFIS junctions is as low as a few  $\mu\text{V}$  and hence too large to reveal a sign change as function of temperature. As a final check we measured the temperature dependence of the saturation magnetization of bare PdNi thin films ( $d_F = 40\text{--}150 \text{ \AA}$ ) via the anomalous Hall effect [25]. As shown in the inset of Fig. 4 for  $d_F = 55 \text{ \AA}$  the normalized saturation magnetization,  $M_s/M_0$ , which is proportional to the Hall signal at  $H = 2000 \text{ G}$ , shows very small variations at low temperature ( $T < 10 \text{ K}$ ) corresponding to a few percent change in  $M_s$ . Thus the long range magnetic order at low temperature is well defined and the small change in  $M_s$  has negligible effects on the critical current temperature dependence.

In summary, we have shown that the critical current of SIFS Josephson junction is an oscillating function of the thickness of the ferromagnetic layer. The zero of the critical current obtained for  $d_F \approx 3\pi/4\xi_F$  indicates a change

of sign of  $I_c$ , leading to “ $\pi$  coupling.” This is a consequence of the superconducting order parameter oscillations induced in the ferromagnet by the proximity effect as recently observed by tunneling spectroscopy [4]. Such ferromagnetic-based “ $\pi$ -junctions” may represent a new element in the architecture of novel quantum devices as recently suggested [26].

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