

Superconducting proximity effect in a diffusive ferromagnet with spin-active interfaces

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We reconsider the problem of the superconducting proximity effect in a diffusive ferromagnet bounded by tunneling interfaces, using spin-dependent boundary conditions. This introduces for each interface a phase-shifting conductance G_ϕ which results from the spin dependence of the phase shifts acquired by electrons upon scattering on the interface. We show that G_ϕ strongly affects the density of states and supercurrents predicted for superconducting/ferromagnetic hybrid circuits. We show the relevance of this effect by identifying clear signatures of G_ϕ in the data of T. Kontos *et al.* [Phys. Rev. Lett. **86**, 304 (2001), **89**, 137007 (2002)].

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Superconducting/ferromagnetic (S/F) hybrid structures raise the fundamental question of what happens when two phases with different broken symmetries interact. When a F metal with uniform magnetization is connected to a BCS superconductor, the singlet electronic correlations characteristic of the S phase propagate into F via Andreev reflections which couple electrons and holes with opposite spins and excitation energies. In the diffusive case, this propagation occurs on a scale limited by the ferromagnetic exchange field. The decay of the correlations in F is accompanied by oscillations of the superconducting order parameter because the exchange field induces an energy shift between the correlated electrons and holes.^{1,2} This has been observed experimentally through oscillations of the density of states (DOS) in F ,³ or of the critical current I_0 through $S/F/S$ structures,⁴⁻⁷ with the thickness of F or the temperature. Remarkably, the oscillations of I_0 have allowed one to obtain π junctions, i.e., Josephson junctions with $I_0 < 0$,⁸ which could find applications in the field of superconducting circuits.⁹

The interface between a ferromagnet and a nonmagnetic material can scatter electrons with spin that is parallel or antiparallel to the magnetization of the ferromagnet with different phase shifts. The *spin dependence* of the interfacial phase shifts (SDIPS) is a general concept in the field of spin-dependent transport. The SDIPS implies that spins noncollinear to the magnetization precess during the scattering by the interface. This so-called spin mixing is expected to drastically affect the behavior of F /normal metal systems¹⁰ when several F electrodes with noncollinear magnetization are used. The same phenomenon is predicted to occur in F /coulomb blockade island,¹¹ and F /Luttinger liquid¹² hybrid circuits. In S/F hybrid systems,¹³⁻¹⁵ the SDIPS is even predicted to affect the system in collinear configurations, due to the coupling of electrons and holes with opposite spins by the Andreev reflections. However, few experimental signatures of the SDIPS have been identified up to now (e.g., Ref. 13 proposes for the data of Ref. 16 an interpretation based on the SDIPS).

In this Rapid Communication, we reconsider the problem of the superconducting proximity effect in a diffusive F . Up to now the tunnel S/F contacts used to produce this effect were described (see, e.g., Ref. 2) with spin-independent boundary conditions (BC) derived in Ref. 17 for the spin-

degenerate case. Instead of that, we use spin-dependent BC based on Ref. 15. These BC introduce a phase-shifting conductance G_ϕ which takes into account the SDIPS. We show that G_ϕ strongly affects the phase and the amplitude of the oscillations of the DOS or I_0 with the thickness of F . Our approach thus provides a framework for future work on S/F diffusive circuits with tunneling interfaces. We show its relevance by a comparison with the data of Refs. 3 and 5 which shows that strong experimental manifestations of the SDIPS have already been observed through the superconducting proximity effect.

We consider a S/F hybrid circuit with a single F electrode homogeneously magnetized in direction \vec{z} . In the diffusive limit, the electrons in a superconducting or ferromagnetic electrode α can be described with quasiclassical and diffusive Green's functions \check{G}_α in the Keldysh \otimes Nambu \otimes spin space (we use the notations of Ref. 15). The BC at a S/F interface can be calculated by assuming that the interface potential locally dominates the Hamiltonian, i.e., at a short distance it causes only ordinary scattering (with no particle-hole mixing). We characterize this scattering with transmission and reflection amplitudes $t_{n,\sigma}^{S(F)}$ and $r_{n,\sigma}^{S(F)}$ for electrons coming from the $S(F)$ side in channel n with a spin σ parallel ($\sigma = \uparrow$) or antiparallel ($\sigma = \downarrow$) to \vec{z} . In practice, the planar S/F contacts used to induce the superconducting proximity effect in a diffusive ferromagnet are likely to be in the tunnel limit,^{18,19} due, e.g., to a mismatch of band structure between S and F , thus we assume $T_n = \sum_\sigma |t_{n,\sigma}^S|^2 \ll 1$. We also consider that the system is weakly polarized. Following Ref. 15 and 20, the BC at the right-hand side F of a S/F interface is

$$2g_F \check{G}_F \frac{\partial \check{G}_F}{\partial x} = \left[G_t \check{G}_S + iG_\phi \sigma_z \check{\tau}_3 + \frac{G_{MR}}{2} \check{D}_+ \check{G}_F \right] + [iG_\chi \check{G}_S \check{D}_- + iG_\xi \check{D}_- \check{G}_F, \check{G}_F] \quad (1)$$

with $\check{D}_\pm = \sigma_z \check{\tau}_3 \check{G}_S \pm \check{G}_S \sigma_z \check{\tau}_3$. Here, σ_z and $\check{\tau}_3$ are Pauli matrices in spin and Nambu space, respectively. The conductivity of F times the area of the junction, noted g_F , is assumed to be spin independent. The coefficient $G_t = G_Q \sum_n T_n$ is the tunneling conductance, $G_{MR} = G_Q \sum_n (|t_{n,\uparrow}^S|^2 - |t_{n,\downarrow}^S|^2)$ is the magnetoresistance term which leads to a spin polarization of the current, and $G_\phi = 2G_Q \sum_n (\rho_n^f - 4[\tau_n^S/T_n])$ is the phase-shifting conduc-

tance, with $\tau_n^S = \text{Im}[r_{n,\uparrow}^S r_{n,\downarrow}^{S*}]$, $\rho_n^F = \text{Im}[r_{n,\uparrow}^F r_{n,\downarrow}^{F*}]$ and $G_Q = e^2/h$. These three terms already appeared in Ref. 15 for studying normal electrodes in contact with S and F reservoirs (with no proximity effect in F). The extra terms in $G_\xi = -G_Q \sum_n \tau_n^S$ and $G_\chi = G_Q \sum_n T_n (\rho_n^F + \tau_n^S)/4$ occur because there are superconducting correlations at both sides of the interface. Note that G_ϕ , G_χ , and G_ξ can be finite only if the phase shifts acquired by the electrons upon reflection or transmission at the interface are spin dependent. The exact values of these conductance coefficients depend on the microscopic structure of the interface. However, we can estimate their relative orders of magnitude in a rectangular potential barrier model by describing the ferromagnetism of F with an exchange field E_{ex} that is much smaller than the spin-averaged Fermi energy E_F of F . This gives expressions of G_{MR} , G_ϕ , G_χ , and G_ξ linear with E_{ex}/E_F . The tunnel limit can be reached by considering a strong mismatch between the Fermi wave vectors in S and F (case 1) or a high enough barrier (case 2). In both limits we find $|G_{MR}|, |G_\chi|, |G_\xi| \ll G_t$, which allows us to neglect these terms in the following. In case 1, we find $|G_\phi| \ll G_t$ whereas in case 2, $|G_\phi|$ can be larger than G_t . Thus we will study the consequences of the spin-dependent BC for an arbitrary value of $|G_\phi|/G_t$. In addition, in case 1 we find $G_\phi < 0$ but in case 2, the sign of G_ϕ depends on the details of the barrier, thus we will consider both signs for G_ϕ .

In equilibrium, we can use normal and anomalous quasi-classical Matsubara Green's functions parametrized, respectively, as $\cos(\Lambda_\sigma)$ and $\sin(\Lambda_\sigma) \exp(i\varphi_\sigma)$ to describe the normal excitations and the condensate of pairs (see, e.g., Ref. 21). The spatial variations of the superconducting correlations in F are described by the Usadel equations $\partial Q_\sigma / \partial x = 0$ and

$$\partial^2 \Lambda_\sigma / \partial x^2 = k_\sigma^2 \text{sgn}(\omega_n) \sin(\Lambda_\sigma) / \xi_F^2 + Q_\sigma^2 \cos(\Lambda_\sigma) / \sin^3(\Lambda_\sigma),$$

with $\xi_F = (\hbar D / E_{ex})^{1/2}$, $\omega_n = (2n+1)\pi k_B T$. Here, $Q_\sigma = \sin^2(\Lambda_\sigma) \partial \varphi_\sigma / \partial x$ is the spectral current (constant with x) and D the diffusion coefficient. We introduced $k_\sigma = \{2[i\sigma \text{sgn}(\omega_n) + |\omega_n|/E_{ex}]\}^{1/2}$ for later use.² Neglecting G_{MR} , G_χ , and G_ξ in (1) yields

$$g_F \frac{\partial \Lambda_\sigma}{\partial x} = iG_\phi \sigma \sin(\Lambda_\sigma) + G_t [\cos(\Lambda_S) \sin(\Lambda_\sigma) - \sin(\Lambda_S) \cos(\Lambda_\sigma) \cos(\varphi_\sigma - \varphi_S)], \quad (2)$$

$$g_F \frac{\partial \varphi_\sigma}{\partial x} \sin(\Lambda_\sigma) = G_t \sin(\Lambda_S) \sin(\varphi_\sigma - \varphi_S). \quad (3)$$

In Eqs. (2) and (3), we used rigid BC for S , i.e., $\Lambda_\sigma = \Lambda_S = \arctan[\Delta / \omega_n]$, with Δ the gap of S .

In the following, we consider the limit of a weak proximity effect in F , i.e., $\Lambda_\sigma = \theta_\sigma$ for $\omega_n > 0$ and $\Lambda_\sigma = \pi - \theta_\sigma$ for $\omega_n < 0$ with $|\theta_\sigma(x)| \ll 1$. We first study geometries with $Q_\sigma = 0$, i.e., no supercurrent flows through the device. In this case, the proximity effect in F can be probed through measurements of the density of states $N(\varepsilon) = N_0 \{1 - \sum_\sigma \text{Re}[\theta_\sigma^2(x)]/4\}$ [with $\omega_n = -i\varepsilon + 0^+$ and $\text{sgn}(\omega_n) = 1$]. The simplest case of a single S/F interface with F at $x > 0$ yields

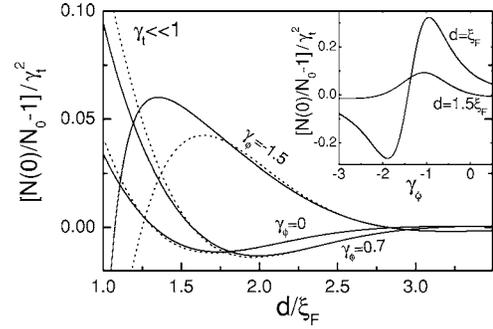


FIG. 1. Zero energy density of states at $x=d$ in a $S/F/I$ structure, in terms of $\{[N(0)/N_0]-1\}/\gamma_t^2$ as a function of d/ξ_F , for $\gamma_t \ll 1$ and different values of γ_ϕ (full lines). The dotted lines show $4\{[N(0)/N_0]-1\}/\gamma_t^2$ at $x=d$ in a semi-infinite S/F structure with the same values of γ_t and γ_ϕ . The inset shows the DOS at $x=d$ as a function of γ_ϕ for the $S/F/I$ structure.

$$\theta_\sigma^{SF}(x) = \frac{\gamma_t \sin(\Lambda_S)}{\gamma_t |\cos(\Lambda_S)| + i\gamma_\phi \sigma \text{sgn}(\omega_n) + k_\sigma} e^{-k_\sigma x / \xi_F} \quad (4)$$

with $\gamma_{t(\phi)} = G_{t(\phi)} \xi_F / g_F$. In the limit $\Delta \ll E_{ex}$ where $k_\sigma = 1 + i\sigma \text{sgn}(\omega_n)$, the weak proximity effect hypothesis leading to (4) is valid for any values of γ_ϕ and ε if $\gamma_t \ll 1$. Since k_σ has finite real and imaginary parts, $\theta_\sigma^{SF}(x)$ shows the well-known exponentially damped sinusoidal oscillations with d . The remarkable point in (4) is that γ_ϕ shifts these oscillations and modifies their amplitude [see Fig. 1 which shows the DOS following from (4)]. We also study the $S/F/I$ geometry, with F at $x \in [0, d]$ and the insulating layer I at $x > d$, for later comparison with the experimental data of Ref. 3. Using (2) for the S/F interface and $\partial \theta_\sigma / \partial x = 0$ for F/I yields

$$\theta_\sigma^{SFI}(x) = \theta_\sigma^d \cosh\left((x-d) \frac{k_\sigma}{\xi_F}\right) \left[\cosh\left(k_\sigma \frac{d}{\xi_F}\right) \right]^{-1} \quad (5)$$

with

$$\theta_\sigma^d = \frac{\gamma_t \sin(\Lambda_S)}{[\gamma_t |\cos(\Lambda_S)| + i\gamma_\phi \sigma \text{sgn}(\omega_n) + k_\sigma \tanh(k_\sigma d / \xi_F)]}$$

In the limit $\Delta \ll E_{ex}$ and $d \geq \xi_F$, the θ linearization leading to (5) is again valid for any γ_ϕ and ε if $\gamma_t \ll 1$. From Fig. 1, γ_ϕ has qualitatively the same effect on $\theta_\sigma^{SFI}(x)$ as on $\theta_\sigma^{SF}(x)$. More quantitatively, for $d \geq \xi_F$ one has $\theta_\sigma^{SFI}(x=d) / \theta_\sigma^{SF}(x=d) = 2$ (Ref. 22) and for lower values of d , this ratio depends on d .

Another way to probe the superconducting proximity effect in F is to measure the supercurrent through a $S/F/S$ Josephson junction. We consider a junction with F at $x \in [0, d]$ and a right (left) superconducting reservoir, called $R(L)$ at a constant phase $(-)\varphi_S/2$. A supercurrent $I_S = \pi g_F k_B T \sum_{n \in \mathbb{Z}, \sigma = \pm 1} Q_\sigma(\omega_n) / 2e$ flows through this device.² We focus on the asymmetric limit $\gamma_t^R \ll \gamma_t^L$, which corresponds to the experiment of Ref. 5, and assume $\gamma_\phi^R = 0$.²³ We allow L and R to have different superconducting gaps $\Delta^{R(L)}$, so that $\Lambda_\sigma = \Lambda_S^{R(L)}$ in $R(L)$. Solving this problem perturbatively with respect to the $S/F/I$ case yields

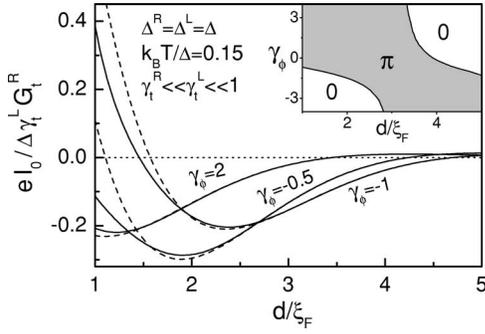


FIG. 2. Critical current I_0 of an asymmetric $S/F/S$ junction as a function of d/ξ_F , calculated from Eq. (6) for $\gamma_t^R \ll \gamma_t^L \ll 1$, $\Delta^{L(R)} = \Delta \ll E_{ex}$ and $k_B T/\Delta = 0.15$ (full lines). The dashed lines show the large d/ξ_F approximation of Eq. (7). The inset is a phase diagram indicating the equilibrium state of the junction (0 or π) depending on γ_ϕ and d/ξ_F .

$$Q_\sigma(\omega_n) = \theta_\sigma^d \gamma_t^R \sin(\Lambda_S^R) \sin(\varphi_S) \left[\xi_F \cosh\left(k_\sigma \frac{d}{\xi_F}\right) \right]^{-1}, \quad (6)$$

where θ_σ^d corresponds to the expression given above with $\Lambda_S = \Lambda_S^L$ and $\gamma_{t(\phi)} = \gamma_{t(\phi)}^L$. The supercurrent has the form $I_S = I_0 \sin(\varphi_S)$ because most of the phase drop occurs at R . In the limit $\Delta^L = \Delta^R = \Delta \ll E_{ex}$, $\gamma_t^L \ll 1$ and $d/\xi_F \gg 1$, (6) yields

$$\frac{eI_0}{\gamma_t^L G_t^R \Delta} = \pi \tanh\left(\frac{\Delta}{2k_B T}\right) \left[\frac{\sin\left(\frac{d}{\xi_F} + \lambda(\gamma_\phi^L)\right)}{[1 + (1 + \gamma_\phi^L)^2]^{1/2}} e^{-d/\xi_F} \right] \quad (7)$$

with $\lambda(\gamma_\phi^L) = \arg[-(1 + \gamma_\phi^L)]$. It is already known that the state of the junction depends on d . Equation (7) shows that γ_ϕ^L shifts the oscillations of the $I_0(d)$ curve. Thus, for a given value of d , the state of the junction can be 0 as well as π , depending on γ_ϕ^L . Figure 2 shows that this effect still occurs when one goes beyond the large d/ξ_F approximation. Note that in the limit $\Delta \ll E_{ex}$ and $\gamma_t^L \ll 1$ used to obtain (7), it is not possible to find a temperature crossover for the sign of I_0 as observed in Refs. 4 and 6. However, we expect to find such a temperature crossover with a $0/\pi$ or $\pi/0$ transition, depending on the value of γ_ϕ^L , if the energy dependence of k_σ is taken into account.²⁵

To show the relevance of our approach, we compare our predictions with the measurements of Refs. 3 and 5. We first consider the $|I_0|$ measured in an asymmetric $S/F/S$ junction, i.e., Nb/Pd_{1-x}Ni_x/Al_{ox}/Al/Nb with $x \sim 0.1$ and $\gamma_t^L/\gamma_t^R \sim 10^5$. We assume that the contacts have $T_n \ll 1$, which allows to use Eqs. (2) and (3). We will use the experimentally determined values $\Delta^{Al/Nb} = 0.6$ meV and $\Delta^{Nb} = 1.35$ meV $\ll E_{ex}$, which implies $k_\sigma \sim 1 + i\sigma$, and $T = 1.5$ K. Samples with different thicknesses d of PdNi were measured (see Fig. 3). Interpreting these data requires a careful analysis of the influence of d on the different parameters. We have $g_F = 2e^2 N_0 D A$ and $\xi_F = \sqrt{\hbar D/E_{ex}}$, with $D = v_F l/3$ and A the conductors cross section. Curie temperature measurements show that the exchange field E_{ex} increases linearly with d .²⁶ In addition, we first assume that the mean free path l is constant with d , as confirmed by resistivity measurements for $d > d_0$

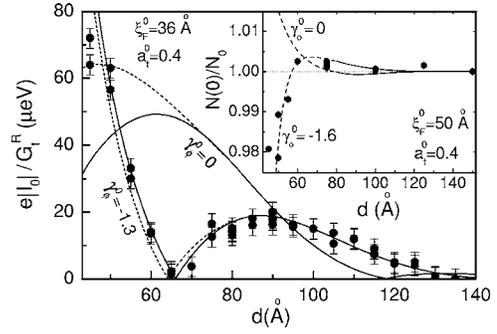


FIG. 3. Critical current measured as in Ref. 5 through Nb/Pd_{1-x}Ni_x/Al_{ox}/Al/Nb junctions as a function of the thickness d of Pd_{1-x}Ni_x (symbols). The lines are theoretical curves calculated from Eq. (6) for $d > d_0$ (full lines) and $d < d_0$ (dashed lines) with the fitting parameters $a_t^0 = 0.4$, $\xi_F^0 = 36$ Å and the experimentally determined parameters $\Delta^{Nb} = 1.35$ meV, $\Delta^{Al/Nb} = 0.6$ meV, $d_0 = 80$ Å and $T = 1.5$ K. The data are well fitted with $\gamma_\phi^0 = -1.3$. We also show the theory for $\gamma_\phi^0 = 0$. Inset: DOS measured by Ref. 3 in Nb/Pd_{1-x}Ni_x/Al_{ox}/Al, as a function of d . The full and dotted lines show the DOS at $x=d$ calculated from the second-order generalization of (5) (see text), for $d > d_0$ and $d < d_0$, respectively. We used $\xi_F^0 = 50$ Å and $\gamma_\phi^0 = -1.6$ or $\gamma_\phi^0 = 0$, all the other parameters being unchanged.

$= 80$ Å. This allows us to parametrize the problem with $\gamma_t^L = a_t^0 \xi_F^0 / \sqrt{d_0 d}$ and $\xi_F = \xi_F^0 \sqrt{d_0/d}$ where ξ_F^0 and $a_t^0 = G_t^L d_0 / g_F$ are constant with d . We also assume that G_ϕ^L is proportional to E_{ex} as found above in the rectangular barrier model for $E_{ex} \ll E_F$, so that we take $\gamma_\phi^L = \gamma_\phi^0 \sqrt{d/d_0}$ with γ_ϕ^0 constant with d . We neglect γ_ϕ^R due to the existence of a strong insulating barrier at R .²⁴ The absolute amplitude of E_{ex} was not determined exactly, so that ξ_F^0 can be considered as a fitting parameter as well as a_t^0 and γ_ϕ^0 . This makes in total three fitting parameters but we expect to find for a_t^0 a value close to the value 0.2 found from minigap measurements in Nb/Pd.²⁵ We have calculated $|I_0|$ by summing (6) on energy and spin. It is not possible to account for the data with $\gamma_\phi^0 = 0$. On the contrary, a good agreement with the experiment is obtained by using $a_t^0 = 0.4$, $\xi_F^0 = 36$ Å, and $\gamma_\phi^0 = -1.3$ (full lines in Fig. 3).^{26,27} We have checked that this choice of parameters fulfills the hypothesis $|\theta_\sigma(x)| \ll 1$ made in our calculations. Remarkably, for $d \sim d_0$ in Fig. 3, the theory for $\gamma_\phi^0 = -1.3$ gives $I_0 < 0$ in agreement with subsequent experiments,^{27,28} whereas it gives $I_0 > 0$ for $\gamma_\phi^0 = 0$ if one keeps the same orders of magnitude for a_t^0 and ξ_F^0 . For $d < d_0$, l is linear with d , which we have taken into account by using $\xi_F = \xi_F^0$, $\gamma_\phi^L = \gamma_\phi^0$, and $\gamma_t^L = a_t^0 \xi_F^0 / d$, with the same values of a_t^0 , γ_ϕ^0 , and ξ_F^0 as previously given (dashed lines in Fig. 3). This approach gives a surprising agreement with the data, which seems to indicate that the Usadel description still works for $d < d_0$ although l is linear with d .²⁹ Kontos *et al.* have also performed DOS measurements in Nb/Pd_{1-x}Ni_x/Al_{ox}/Al,³ prior to the I_0 measurements. We have assumed again that E_{ex} was linear with d in these measurements, to try to interpret the $N(0) = f(d)$ curve with the same fitting procedure as for I_0 . We have generalized Eq. (5) to second order in θ_σ because the values of d/ξ_F are slightly lower than for the I_0 measure-

ments. Again it is impossible to interpret the data with $\gamma_\phi^0 = 0$. We obtain a satisfactory fit by choosing $\xi_F^0 = 50 \text{ \AA}$ and $\gamma_\phi^0 = -1.6$, all the other parameters used being the same as in the previous case. Finding a ξ_F^0 higher than for the I_0 data is in agreement with the fact that the samples used for measuring the DOS were realized with a lower concentration x of Ni.

In summary, we have studied the effect of spin-dependent boundary conditions on the superconducting proximity effect in a diffusive ferromagnet bounded by tunneling interfaces. We have shown that the phase-shifting conductances G_ϕ , describing the spin activity of the interfaces in this context,

strongly affect the behavior of the system and allow a consistent microscopic explanation of the DOS and supercurrent data of Refs. 3 and 5. This suggests that such effects will have to be considered in any future work on S/F hybrid circuits. In the context of spintronics, this approach might also provide a way to characterize spin-active interfaces.

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