

Spin screening and antiscreening in a ferromagnet/superconductor heterojunctionF. S. Bergeret^{1,2} and N. García^{1,2}¹*Laboratorio de Física de Sistemas Pequeños y Nanotecnología, CSIC, Serrano144, E-28006 Madrid, Spain*²*Laboratório de Filmes e Superfícies, UFSC, 88040-900 Florianópolis, Brazil*

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We present a theoretical study of spin-screening effects in a ferromagnet-superconductor (FS) heterojunction. It is shown that the magnetic moment of the ferromagnet is screened or antiscreened, depending on the polarization of the electrons at the Fermi level. If the polarization is determined by the electrons of the majority (minority) spin band then the magnetic moment of the ferromagnet is screened (antiscreened) by the electrons in the superconductor. We propose experiments that may confirm our theory: for ferromagnetic alloys with certain concentration of Fe or Ni ions there will be screening or antiscreening, respectively. Different configurations for the density of states are also discussed.

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The coexistence and mutual influence of ferromagnetism and conventional superconductivity in heterostructures has been studied intensively in the past years due to the great progress in preparing high-quality multilayered systems (for a review see Ref. 1). These two long-range phenomena are antagonistic: while in the superconducting state, electrons form Cooper pairs with opposite spin, in a ferromagnet the exchange field tries to align the spin of the electrons. Their coexistence in a bulk material is hardly possible and only takes place for exchange fields smaller than the characteristic superconducting energy.^{2,3} The situation changes if the superconducting and ferromagnetic regions are spatially separated (e.g., in heterostructures). In this case the coexistence is possible even if the exchange field exceeds the value of the superconducting order parameter Δ , and their mutual influence is due to the so-called proximity effect: when a superconductor (S) is brought in electrical contact with a normal metal (N) the superconducting condensate may penetrate into N over a distance of the order of $\sqrt{D_N/T}$, where D_N is the diffusive coefficient. If the normal metal is a ferromagnet (F) the penetration length is drastically reduced due to the destructive action of the exchange field h on the Cooper pairs. Each electron of a pair is in a different spin band. These bands are shifted by an energy h and therefore, if h is very large the Cooper pair breaks up. In that case the condensate penetrates into the F region over a distance of the order of $\sqrt{D_F/h}$ and undergoes some characteristic oscillations.⁴ In order to have a stronger proximity effect, i.e., weaker exchange fields, experimentalists are using dilute magnetic alloys. For example, in the experiments of Ref. 5 Cu-Ni alloys have been used in order to observe the change of sign of the Josephson critical current in a superconductor-ferromagnet-superconductor (SFS) structure. This effect was predicted many years ago.⁶

Another interesting effect (the inverse proximity effect) was studied recently in Ref. 7. The authors proposed a physical picture according to which some Cooper pairs share the electrons between the superconductor and the ferromagnet. It was discussed that while the spin of the electron in F prefers to be parallel to the magnetic moment of F , the spin of the electron in S is automatically antiparallel to the magnetization. In S a (screening) magnetic moment is induced which

penetrates over the characteristic superconducting length ξ_s . Although this intuitive idea might be true in some cases, it cannot be the whole story because the magnetization, as we will see, is not always the relevant parameter. For example for a nonitinerant ferromagnet the effect will be zero or negligible. The reason is that according to the physical picture the electrons involved in this effect are only those of the condensate which, as it is well known, are around the Fermi level (FL). Therefore the screening in the superconductor cannot be determined by the magnetization of the ferromagnet, which involves the integral over *all* the electrons, but rather by the polarization of the electrons at the FL as we will show below. In Ref. 8 the magnetization of a ballistic SF system was studied. However, the authors have not discussed the inverse proximity effect and instead they found a magnetization leakage from F to S over distances of the order of the Fermi wavelength. In the present paper we are not interested in such small scales. The magnetic leakage found in Ref. 8 can be included by taking a renormalized thickness of F . Also in Ref. 9, leakage of the magnetic moment into S was reported.

It is clear from the physics involved in FS junctions that the inverse proximity effect is related to the properties of the conducting electrons. This implies that the main role is played by the densities of states (DoS) for electrons with spin up and spin down at the FL [$\nu_{\pm}(0)$], which in general are different. The polarization at the FL does not necessarily have the sign of the magnetization. In particular the result in Ref. 7 was obtained for the case that the polarization at the FL is due to the majority electrons, and therefore has the same sign as the magnetization (see Fig. 1). However, it is well known from band-structure calculations that ferromagnetic metals show a very complicated band structure and in some cases like Ni, Co, and many other materials, the polarization at the FL is due to minority electrons.^{10,11} In this case at the FL $\nu_{-}(0) > \nu_{+}(0)$ (see Fig. 2), and therefore according to the physical picture given above, the magnetization induced in the superconductor has the same sign as in F (antiscreening).

The aim of this paper is to perform a general theory which explains this physical picture. We show using the method of the Green's function (GFs) that the change of the magnetization of the system is proportional to the difference of DoS

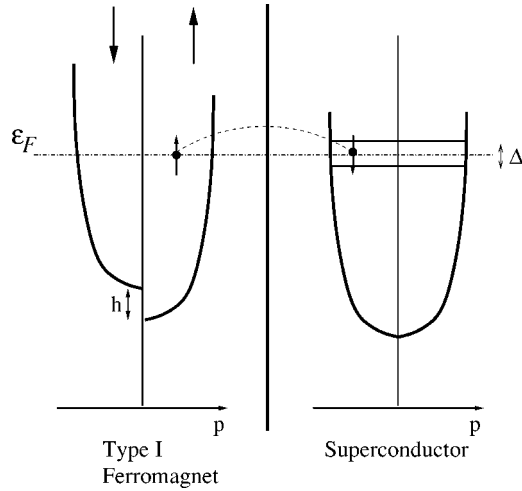


FIG. 1. SF system. The ferromagnet shows a type I BS. The DoS at the Fermi level for majority band (spin up) is larger than the DoS of the minority band. The two electrons connected by a dashed line represent a Cooper pair which contributes to the inverse proximity effect.

at the FL. The magnetization of the system is reduced if the polarization at the Fermi level is controlled by spin majority and enhanced if it is dominated by spin minority. We propose different experiments and applications that may confirm our predictions. We distinguish between two types of ferromagnetic metals: (a) with a conduction band structure (BS) of type I (Fig. 1) and (b) materials with BS of type II (Fig. 2). The density of states (DoS) at the Fermi level of the majority spin band is larger (smaller) than the DoS of the minority band in the case of materials with BS of type I (II). In order to model both types of materials we chose a simple model for the ferromagnet, which catches the main physics of the system. We assume that both spin bands have the same shape and are shifted by the exchange energy h . The Hamiltonian describing the ferromagnet is given by

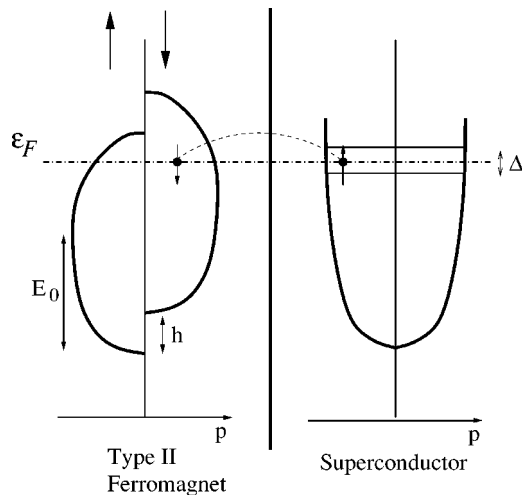


FIG. 2. SF system. The ferromagnet shows a type II BS. The DoS at the Fermi level for minority band is larger than the DoS of the majority band.

$$H_F = \sum_{\{p,s,i\}} \{a_{sp}^+ [\xi_p \delta_{pp'} + U_i(p,p') - h(\hat{\sigma}_3)_{ss'}] a_{s'p'}\}. \quad (1)$$

Here ξ_p is the energy of the quasiparticles (counted from the Fermi energy ϵ_F) and $U_i(p,p')$ is the scattering potential from the i impurity. The last term describes the ferromagnetic interaction which is written in the mean-field approximation and leads to the shift of the spin bands. In the free-electron model and defining E_0 as the midband energy, we assume that the momentum is

$$p = \sqrt{2mE} \quad \text{for } E < E_0, \quad (2)$$

$$p = \sqrt{2m(2E_0 - E)} \quad \text{for } E > E_0 \quad (3)$$

Of course one can choose another shape for the curves $E(p)$. However, the main results of this paper do not depend on this choice. Notice also that there may be another type of materials for which the Fermi energy lies for one spin band above E_0 and for the other spin band below E_0 . The generalization of our results for this case is straightforward.

The Green's functions G_{\pm} for the spin-up and spin-down electrons corresponding to the Hamiltonian (1) are

$$G_{\pm}(\omega_n, \mathbf{p}) = [i\omega_n - \xi_p \mp h/2 + (\text{sgn } \omega/2\tau)]^{-1}, \quad (4)$$

where $\omega_n = \pi T(2n+1)$ is the Matsubara frequency and τ is the momentum relaxation time caused by the impurities. The DoS for spin-up and -down electrons are

$$\nu_{\pm}(\omega) = \frac{m}{2\pi^2} \sqrt{2m(\epsilon_F \pm h + \omega)} \quad (5)$$

for energies above E_0 , and

$$\nu_{\pm}(\omega) = \frac{m}{2\pi^2} \sqrt{2m(2E_0 - \epsilon_F \mp h - \omega)} \quad (6)$$

for energies below E_0 . The *total* magnetization M_F is obtained by integration over all ω 's and therefore is positive in both cases. However, we emphasize that the spin polarization at the FL for materials with BS of type I is parallel to M_F , while for materials of type II is in the opposite direction.

The superconductor is described by the usual BCS Hamiltonian in the mean field approximation

$$\hat{H}_{BCS} = H_0 - \sum_{\{p,s\}} \{\Delta a_{sp}^+ a_{sp}^+ + \text{c.c.}\}, \quad (7)$$

where H_0 is the free-electron part which contains also scattering by impurities. Δ is the superconducting order parameter. The index s denotes spin and p momentum. The Cooper pairs forming the condensate have total momentum equals zero and are in singlet state (\bar{s} and \bar{p} stay for $-s$ and $-p$, respectively). We are interested in the inverse proximity effect, in particular how the magnetization M of the system changes due to the presence of the superconducting correlations. The total Hamiltonian of the system is $H = H_F + H_{BCS}$. Finding the GFs for the SF structure is a quite difficult task and some simplifications have to be made. We use here the well-known quasiclassical approach (see, e.g., Ref. 12). The quasiclassical GFs are obtained by integrating the microscopic GFs over ξ_p and only contain information about elec-

trons close to the Fermi surface. This restriction does not limit our analysis since only these electrons participate in the proximity effect. In order to obtain the equations for the quasiclassical GF, one assumes that all energies involved in the problem are small in comparison to ϵ_F , in particular $h \ll \epsilon_F$. The quasiclassical equations are derived in many papers and therefore we skip here the derivation (see, for example, Ref. 12). Tagirov generalized these equations for the case that the momenta at the Fermi level $p_{F\pm}$ of both spin bands are different.¹³ According to Eqs. (5) and (6) $p_{F\pm} = 2\pi^2 v_{\pm}(0)/m$. For a diffusive system one obtains the general Usadel equation^{13,14}

$$D \nabla (\check{g} \nabla \check{g}) - \omega_n [\hat{\tau}_3 \hat{\sigma}_0, \check{g}] + i v_F \delta p_F [\hat{\tau}_3 \hat{\sigma}_3, \check{g}] = -i [\check{\Delta}, \check{g}]. \quad (8)$$

The GF $\check{g} = \hat{g} \hat{\tau}_3 + \hat{f} i \hat{\tau}_2$ is a 4×4 matrix in the spin ($\hat{\sigma}$) and particle-hole space ($\hat{\tau}$) and $\omega_n = \pi T(2n+1)$ are the Matsubara frequencies. In the S region $D = D_S$, $\delta p_F \equiv p_{F+} - p_{F-} = 0$, and $\check{\Delta} = \Delta i \hat{\tau}_2 \hat{\sigma}_3$ (the phase of Δ is chosen to be zero). While in the F layer, $D = D_F$, $\Delta = 0$, $\delta p_F \neq 0$, and v_F is the Fermi velocity for vanishing exchange field. The term proportional to $v_F \delta p_F$ is related to the effective exchange field acting on the electrons at the Fermi level. In the limit under consideration ($h \ll \epsilon_F$), $h = v_F |\delta p_F|$.¹³ Note that the sign of this term depends on whether F has a BS of type I or II. Equation (8) is complemented by proper boundary conditions.^{13,15}

In order to avoid cumbersome calculations we make a further simplification which does not change the qualitative validity of our results. We assume that the F and S layers are thinner than the characteristic length of variation of the GFs. In that case one can average Eq. (8) over the thicknesses and define an effective exchange field $h_{\text{eff}} \equiv v_F \delta p_F (v_F^{I,II} d_F) / (v_F^{I,II} d_F + v_S d_S)$, where $v_F^{I,II}$ are the corresponding DoS at zero value of the exchange field for case I and II. We also define $\Delta_{\text{eff}} \equiv \Delta (v_S d_S) / (v_S d_S + v_F^{I,II} d_F)$. Within this approximation and the assumption that the SF interface is perfect, Eq. (8) can be transformed into an algebraic equation for \check{g} complemented by the normalization condition $\check{g}^2 = 1$. The solution of this set of equations can be found easily (see, e.g., Refs. 16 and 17).

Our aim is to calculate the magnetization per unit area induced in the superconductor⁷

$$M_S = -i \mu_B \pi v_S d_S T \sum_{\omega_n} \text{Tr} \hat{\sigma}_3 \hat{g}, \quad (9)$$

where μ_B is the Bohr magneton. If the F layer is very thin, the expression for the component of \hat{g} proportional to $\hat{\sigma}_3$ is

$$g_3 = -i \frac{h_{\text{eff}} \Delta_S^2}{(\omega_n^2 + \Delta_S^2)^{3/2}}. \quad (10)$$

Inserting this expression in Eq. (9) we obtain for the magnetization (per unit area) induced in S at $T=0$

$$M_S^{I,II} = \mathcal{N} \mu_B (v_F \delta p_F) \frac{v_F^{I,II} d_F}{v_F^{I,II} d_F + v_S d_S} v_S d_S, \quad (11)$$

where \mathcal{N} is a positive numerical factor of the order of unity. For finite temperatures and according to Eq. (10), the induced magnetization is a monotonically decaying function of the temperature which vanishes when $T=T_C$ as expected. It was shown in Ref. 7 that the component g_3 of the GF induced in the superconductor penetrates over the length ξ_S . Thus, if the thickness of the superconductor is larger than the coherence length ξ_S , Eq. (11) can be used for estimates if one substitutes d_S by ξ_S .

Equation (11) confirms our intuitive picture given in the introduction. Depending on the sign of δp_F which is proportional to $v_+(0) - v_-(0)$, the magnetization induced in S is antiparallel (case I, $\delta p_F > 0$) or parallel (case II, $\delta p_F < 0$) to the magnetization in F . From Eq. (11) one can see that the maximum induced magnetic moment in S is related to the density of electrons at the FL v_S . This quantity approximately equal to Δv_S , i.e., corresponds to $10^{-3} - 10^{-4}$ Bohr magneton per atom. This is a very small quantity and therefore it will be difficult to observe this effect with usual magnetic material as Fe or Ni. In order to check these effects one should try with dilute materials, ferromagnetic semiconductors,¹⁸ or in materials with very low magnetization as, for example, seems to be the case of graphite and polymerized fullerenes.¹⁹

By deriving Eq. (11) we have assumed that the SF transparency is high enough. However, it is known that in many experiments the SF interfaces are not perfect and the transparency may be very low. In this case the proximity effect is weak and one can linearize Eq. (8). This limit was considered by the authors of Ref. 7 for a F layer with a BS of type I. In that case the induced magnetization decreases as R_b^{-2} by increasing the interface resistance R_b . This result is also valid in the case of type II BS. The main difference is that in the latter case, and according to our theory, the induced magnetization will be parallel to the magnetization of F and hence the total magnetic moment will increase. Thus, high values of R_b will suppress the inverse proximity effect in both cases. An increase of the interface resistance can be due to a formation of an oxide layer between the metals or band mismatch.

We propose possible experiments that will illuminate the correctness or incorrectness of our theory. For ferromagnetic alloys with, for example, certain iron concentration, as the systems VFe/V or PdFe/V used in Ref. 20 and Ref. 21, respectively, there will be a screening effect because in these alloys the majority electrons at the FL aligned with the magnetization. However, for the case of ferromagnetic alloys with Ni ions (e.g., the junction NiCu/Nb used in Ref. 5), antiscreening will take place due to the fact that the electrons of Ni at the FL are dominated by minority electrons.

If the widths of the conduction band are very different it is clear from the physical picture that there is no possibility to have Cooper pairs sharing their electrons between the ferromagnet and the superconductor because the momenta matching is very bad. In that case the proximity effect, i.e., the penetration of Cooper pairs into the F region, is negligibly

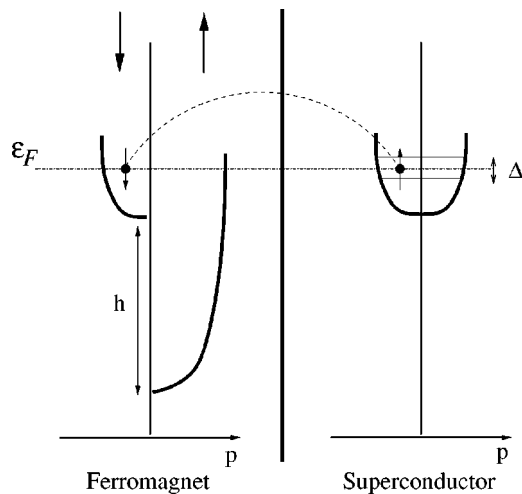


FIG. 3. SF structure consisting of a ferromagnet with a large exchange splitting h . The band width of the minority spin-band is approximately equal to the band width of the superconductor.

small. However, one can imagine the situation depicted in Fig. 3, where the exchange field in F is so strong that the Fermi momenta for electrons with spin up and down are very different (this is similar to the situation of a half metal).²² For example, if the width of the minority band is similar to the width of the band of the superconductor, then according to our theory the inverse proximity effect will lead to an enhancement of the total magnetic moment, since only the electrons of the minority band can be paired with electrons of S . It can also occur that the majority spin-band width corresponds to the S band width. In that case we predict a decrease of the total magnetic moment when T is lowered below T_C . Thus, the effect considered in this paper can be used in order to study the electronic properties of ferromagnetic materials at the Fermi level. One can perform an experiment by measuring the magnetization for temperatures above and below the superconducting temperature. If by lowering the temperature the magnetization is enhanced, then it is clear that at the Fermi level the minority spin band dominates, and vice versa. The situation depicted in Fig. 3

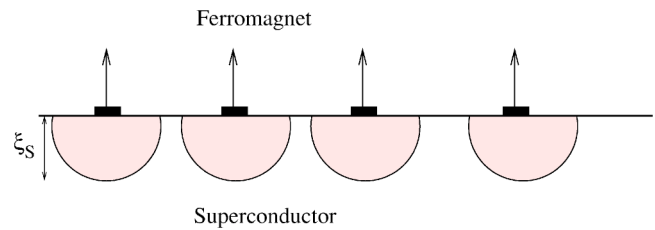


FIG. 4. SF systems. The magnetization in F is due to the magnetic moment of certain regions. The screening or antiscreening in S is only possible in regions of size ξ_s (circles)

may correspond to the case of some high T_C superconductors which in general have very low Fermi energies.

One can also use this effect for examining if a ferromagnetic material has either its magnetic moments concentrated in small regions or distributed homogeneously (see Fig. 4). The number of magnetic moment which can be screened (or antiscreened) is proportional to $\Delta v_S(\xi_s S)$, where S is the cross section of the magnetized region. It is clear from our analysis that if the magnetization of F is due to highly magnetized small regions, the relative change of magnetization is negligible, while if the magnetic moments are homogeneously distributed, the effect of screening or antiscreening might be more pronounced.

In conclusion we have studied the inverse proximity effect in a SF system. Superconducting correlations leads to the formation of Cooper pairs which share their electrons between the superconductor and the ferromagnet. Depending on the polarization of the electrons at the Fermi level we predict a screening or an antiscreening of the magnetic moment. If the DoS at the Fermi level of the majority band is larger than the DoS of the minority one, then the magnetization of the system is reduced by lowering the temperature below the superconducting temperature. In the opposite case we predict an enhancement of the magnetization. Such effects may be useful to examine the electronic properties at the Fermi level and the distribution of magnetic moments of ferromagnetic metals.

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