SUPERCONDUCTOR-FERROMAGNET HETEROSTRUCTURES

A. I. Buzdin¹, M. Fauré¹, and M. Houzet²

¹Université Bordeaux I, CPMOH, 33400 Talence Cedex, France ² Commissariat à l'Énergie Atomique, DSM, Département de Recherche Fondamentale sur la Matière Condensée, SPSMS, F-38054 Grenoble, France

Abstract: We provide a general description of superconductor/ferromagnet structures and study the evolution of the critical temperature, critical current and magnetization variation with the thickness of the ferromagnetic layer. Special attention is given to the influence of the magnetic scattering on the properties of such hybrid systems.

Key words: magnetism, superconductivity, magnetic impurities

1. INTRODUCTION

Ferromagnetism and singlet superconductivity are two antagonistic orderings. Indeed, a magnetic field can destroy conventional superconductivity via the orbital effect and via the paramagnetic effect and therefore, they usually try to avoid each other. The competition between these two orderings has always been a subject of great interest for both theoretical and experimental physics. The question of their coexistence was first addressed by Ginzburg as soon as 1956 [1]. In fact, it was demonstrated later that а modulated magnetic structure (cryptoferromagnetic state [2]) appears instead of ferromagnetism in a singlet superconductor. As a review of the problem of singlet superconductivity and magnetism coexistence, see [3]. Moreover, Larkin and Ovchinnikov, and Fulde and Ferrell demonstrated in 1964 that the superconductivity of a pure ferromagnetic superconductor may be non uniform at low temperature ([4] and [5]). It is unfortunately not easy to verify this prediction on experiment because of the incompatibility of ferromagnetism and superconductivity in bulk materials.

However, their interplay may be studied when the two orderings are spatially separated, which is obtained in artificially made superconductor/ferromagnet (S/F) structures. These hybrid systems give us the unique possibility to study the properties of superconducting electrons under the influence of a huge exchange field acting on the electron spins. In such systems, Cooper pairs can penetrate into the F layer and induce superconductivity there, which is the so called proximity effect. In addition, it is possible to study the interplay between superconductivity and magnetism in a controlled manner, since varying the layer thicknesses changes the relative strength of the two competing phenomena.

Note that almost all the interesting effects related to superconductivity and magnetism interplay in S/F structures occur at a nanoscopic scale. The observation of these effects became possible only a few years ago thanks to recent progress in the preparation of high-quality hybrid layers.

The most striking features of S/F systems are the highly non monotonic behaviors of the critical temperature T_c and the critical current I_c with the thickness of the ferromagnetic layer. In S/F/S junctions and S/F multilayers, this is related to $0-\pi$ transitions, which are studied in the present work. Another interesting manifestation of the proximity effect is the variation of the magnetization in both types of layers.

A general review of S/F structures was reported in [6]; see also [7]. Here, we would like to concentrate on the influence of magnetic scattering on the properties of S/F systems. Although the oscillatory behavior of T_c and I_c is well known, a noticeable difference between theoretical calculations and experimental results still exists. It could be understood by the introduction of an additional scattering mechanism in theoretical descriptions. Indeed, magnetic impurities, spin-wave or non stoechiometric lattices... can play an important role as the spin-flip process has dramatic consequences on superconductivity (on the contrary of non magnetic impurities which have very little impact). More precisely, the pair-breaking effect induced by magnetic impurities leads to the decrease of the decay length of T_c and I_c and to the increase of the oscillations period. Note that the question of the spin flip scattering was firstly addressed by Tagirov [8], while Demler *et al.* studied the spin-orbit scattering role [9].

In the present work, we study the critical temperature and current and magnetization in S/F bilayers and multilayers in the framework of the Usadel equations and report on the spin-flip scattering influence.

2. PROXIMITY EFFECT IN S/F SYSTEMS

2.1 Generalized Ginzburg-Landau functional

The physics of the proximity effect can be qualitatively described by a standard Ginzburg-Landau functional in Superconductor/Normal (S/N) metal structures (see, for example [10]):

$$F = a |\psi|^2 + \gamma |\overline{\nabla} \psi|^2 + \frac{\sigma}{2} |\psi|^4, \qquad (1)$$

where \forall is the superconducting order parameter, and the coefficient $a \propto (T - T_c)$ vanishes at the transition temperature T_c .

It should be noted that expression (1) is valid only if the effect of the exchange field h may be neglected. In the F layer, the functional has to be modified. The coefficients a, b, and γ are dependent on h. In particular, the gradient term coefficient γ becomes negative for a relatively large value of h/T > 1. In that case, it is then necessary to add a higher order derivative term. Finally, the generalized Ginzburg-Landau expansion may be written as following:

$$F_{G} = a(h,T)|\psi|^{2} + \gamma(h,T)|\nabla \psi|^{2} + \frac{\eta(n,T)}{2}|\nabla \psi|^{2} + \frac{\sigma(n,T)}{2}|\psi|^{4}.$$
 (2)

The critical temperature of the second order phase transition into a superconducting state may be found from the solution of the linear equation for the superconducting order parameter

$$a\psi - \gamma\Delta\psi + \frac{\eta}{2}\Delta^2\psi = 0.$$
(3)

If we seek for a non-uniform solution $\psi = \psi_0 \exp(i\vec{q}\vec{r})$, the corresponding critical temperature depends on the wave-vector \vec{q} and is given by the expression $a = -\gamma q^2 - \eta q^4/2$. The coefficient *a* can be written as $a = \alpha (T - T_{cu}(h))$, where $T_{cu}(h)$ is the critical temperature of the transition into the uniform superconducting state.

In a standard situation, the gradient term in the Ginzburg-Landau functional is positive, $\gamma > 0$, and the highest transition temperature coincides with $T_{cu}(h)$; it is realized for the uniform state with q = 0. However, when $\gamma < 0$, the maximum critical temperature corresponds to the finite value of the modulation vector $q_0^2 = -\gamma / \eta$ and the corresponding transition temperature into the non-uniform state $T_{ci}(h)$ is given by $a = \alpha (T_{ci} - T_{cu}) = \gamma^2 / (2\eta)$. It is higher than the critical temperature T_{cu} of the uniform state. Therefore, we see that the non uniform state appearance, called FFLO state, may simply be interpreted as a change of the sign of the gradient term in the Ginzburg-Landau functional.

2.2 Damped oscillatory decay of the cooper pair wave function in ferromagnets

To get some idea about the peculiarity of the proximity effect in S/F structures, we may start with the description based on the generalized Ginzburg-Landau functional (2). Such an approach is adequate for a small wave-vector modulation case, otherwise a microscopic theory must be used. This situation corresponds to a very weak ferromagnet with an extremely small exchange field $h \approx T_c$. We address the question of the proximity effect for a weak ferromagnet described by the generalized Ginzburg-Landau functional (2). More precisely, we consider the decay of the order parameter in the normal phase, i. e. at $T > T_{ci}$ assuming that our system is in contact with another superconductor with a higher critical temperature, and the x axis is chosen perpendicular to the interface.

The induced superconductivity is weak and we may use the linearized equation for the order parameter (3), which is written for our geometry as

$$a\psi - \gamma \frac{\gamma}{2} + \frac{\gamma}{2} + \frac{\gamma}{2} \frac{\gamma}{2} = 0.$$
 (4)

The solutions of this equation in the normal phase are of the type $\psi = \psi_0 \exp(kx)$, with a complex wave-vector $k = k_1 + ik_2$, and

$$k_1^2 = \frac{|V|}{2\pi} \left| 1 + \frac{1}{T} \frac{1}{T} - 1 \right|, \tag{5}$$

$$k_2^2 = \frac{|\nu|}{2\pi} \left| \left| 1 + \frac{1}{T} - \frac{1}{C} + 1 \right| \right|.$$
(6)

If we choose the gauge with the real order parameter in the superconductor, then the solution for the decaying order parameter in the ferromagnet is also real

$$\psi \propto \exp[-k_1 x] \cos[k_2 x], \tag{7}$$

where the choice of the root for k corresponds to $k_1 > 0$. So the decay of the order parameter is accompanied by oscillations (Fig. 1b), which is the characteristic feature of the proximity effect in the considered system.

Let us compare this behavior with the standard proximity effect [11] described by the linearized Ginzburg-Landau equation for the order parameter

$$a\psi - \gamma \frac{\tau}{2} = 0, \qquad (8)$$

with $\gamma > 0$. In such a case T_c simply coincides with T_{cu} and the decaying solution is $\psi = \psi_0 \exp(-x/\xi_n)$ where the coherence length $\xi_n = \sqrt{\gamma/a}$ (Fig. 1a).

This simple analysis brings in evidence the appearance of the oscillations of the order parameter in the presence of an exchange field.

Thus, there is a fundamental difference between the proximity effect in S/F and S/N systems.



Figure 1. Schematic behaviour of the superconducting order parameter near the interface (a) superconductor/normal metal and (b) superconductor/ferromagnet.

The oscillations of the superconducting order parameter in S/F systems may also be understood when considering a Cooper pair picture. Indeed, a Cooper pair is usually formed by two electrons with opposite momenta k_F and $-k_F$ and opposite spins. The resulting momentum of the Cooper pair $k_F + (-k_F) = 0$. When a magnetic field is applied, because of the Zeeman's splitting, the Fermi momentum of the electron with the spin parallel to the field (up) will shift from k_F to $k_1 = k_F + \delta k$, where $\delta k = \mu_B h/v_F$, v_F being the Fermi velocity and h the exchange field in the F layer. Similarly, the Fermi momentum of the down spin electron will shift from $-k_F$ to $k_2 = -k_F + \delta k$ (see Fig. 2). Then, the resulting momentum of the Cooper pair is $k_1 + k_2 = 2\delta k \neq 0$, which implies the space modulation of the superconducting order parameter with a resulting wave-vector $2\delta k$.



Figure 2. Energy band of a 1D superconductor near the Fermi surface.

Therefore, we can see again that Ψ does not only decays into the F layer, but it is also space modulated, which gives the following behaviour of the pair wave function:

$$\psi \propto \exp\left[-x/\xi_{f_1}\right]\cos\left[x/\xi_{f_2}\right],\tag{9}$$

where $\xi_{f1} \equiv 1/k_1$ and $\xi_{f2} \equiv 1/k_2$ are the decaying and oscillations length of the superconducting correlations in the F layer (see Fig 1(b)) while it only decays in S/N systems.

2.3 Consequences of the superconducting order parameter oscillations in S/F systems

The damped oscillatory behavior of the superconducting order parameter in ferromagnets may produce commensurable effects between the period of the order parameter oscillation (given by ξ_f) and the thickness of a F layer. This results in the striking non monotonic superconducting transition temperature dependence on the F layer thickness in S/F multilayers and bilayers. Indeed, for a F layer thickness smaller than ξ_f , the pair wave function in the F layer changes a little and the superconducting order parameter in the adjacent S layers must be the same. The phase difference between the superconducting order parameters in the S layers is zero, which is the so-called 0-phase.

On the other hand, if the F layer thickness becomes of the order of ξ_{f} , the pair wave function may go trough zero at the center of the F layer providing the state with the opposite sign (or π shift of the phase) of the superconducting order parameter in the adjacent S layers, which is the π - phase. The increase of the thickness of the F layers may provoke subsequent transitions from 0- to π -phases, results in a very special dependence of the critical temperature on the F layer thickness.

For S/F bilayers, the transitions between 0 and π -phases are impossible. The commensurable effect between ξ_f and the F layer thickness nevertheless leads to the non-monotonous dependence of T_c on the F layer thickness due to the commensurability effect between the period of superconducting wave function oscillation and the thickness of the F layer.

The first experimental indications on the non-monotonous variation of T_c versus the thickness of the F layer was obtained by Wong *et al.* [12] for V/Fe superlattices. However, the strong pair-breaking influence of the ferromagnet and the nanoscopic range of the oscillations period complicated the observation of this effect. Advances in thin film processing techniques were therefore crucial for the study of this subtle phenomenon. The predicted oscillatory type dependence of the critical temperature was finally clearly observed in 1995 in Nb/Gd [13] and then in other systems (for more detail, see [6]).

Another consequence of the superconducting order parameter modulation is the damped oscillatory behavior of the critical current of a S/F/S junction.

A S/F/S sandwich realizes a Josephson junction in which the weak link between the two superconductors is ensured by the ferromagnetic layer. The supercurrent $I_s(\varphi)$ flowing across the structure can be expressed as $I_s(\varphi) = I_c \sin(\varphi)$, where I_c is the critical current and φ stands for the phase difference between the two superconducting layers. A standard junction has at equilibrium $I_c > 0$ and $\varphi = 0$, and therefore, no current exists. It may appear however that I_c becomes negative, which implies that the equilibrium phase difference is $\varphi = \pi$ and the ground state undergoes a π phase shift, namely the π junction.

The first unambiguous experimental evidence of the $0-\pi$ transition with the temperature variation via critical current measurements was observed by Ryazanov *et al.* in 2001 [14]. Sellier *et al.* recently obtained a similar result [15], while Kontos *et al.* [16] observed the damped oscillations of the critical current as a function of the F layer thickness in Nb/Al/Al₂O₃ /PdNi/Nb junctions.

3. THEORETICAL FRAMEWORK

3.1 Usadel equations

Real ferromagnets present rather large exchange fields and the Ginzburg-Landau functional is not an adequate approach for S/F systems description. A microscopic theory has to be used to theoretically describe the proximity effect in such structures. The most convenient schemes are the use of the Boboliubov-de Gennes equations [10] or the Green's functions in the framework of the quasiclassical Eilenberger [17] or Usadel equations [18]. If the electron scattering mean free path ℓ is small (which is usually the case in S/F systems), the most natural approach is to choose the Usadel equations for the Green's functions averaged over the Fermi surface.

The normal Green's function will be noted $G_{s(f)}$ in the S(F) layer, while the anomalous Green's function is $F_{s(f)}$.

In the general case, magnetic and spin-orbit scatterings mix up the up and down spins states. Choosing the spin quantization axis along the direction of the exchange field, and introducing the Green functions $G_1 \sim \langle \Psi \uparrow \Psi \uparrow \rangle$ and $F_1 \sim \langle \Psi \uparrow \Psi \downarrow \rangle$ (G_2 and F_2 for the opposite spin orientations), we may write the nonlinear Usadel equation in the following form

$$\begin{pmatrix} L & \langle z & x \rangle \end{pmatrix}$$
 (10)

where τ_{so}^{-1} is the spin-orbit scattering rate while the magnetic scattering rates are $\tau_z^{-1} = \tau_2^{-1} \langle S_z^2 \rangle / S^2$ and $\tau_x^{-1} = \tau_2^{-1} \langle S_x^2 \rangle / S^2$. The rate τ_2^{-1} is proportional to the square of the exchange interaction potential, and we follow the notations of work [19]. For the spatially uniform case, equation (10) naturally gives the same result as the microscopic approach developed in [20,19].

The ferromagnets that are usually used in S/F heterostructures contain elements with relatively small atomic numbers. Therefore, the spin-orbit scattering may be neglected, and henceforth, $\tau_{so}^{-1} = 0$. The influence of the spin-orbit scattering on the critical temperature of S/F bilayers was studied in [9].

In addition, the uniaxial anisotropy strongly suppresses the perpendicular fluctuations of the local exchange field, that is $\tau_x^{-1} \rightarrow 0$. In such a case, the Usadel equation is simplified and may be written in the F layer as

$$-\frac{D_f}{2}(G_f \nabla^2 F_f - F_f \nabla^2 G_f) + (\omega + ih + \tau_m^{-1} G_f) F_f = 0, \qquad (11)$$

where $\tau_m^{-1} = \tau_2^{-1} \langle S_z^2 \rangle / S^2$ may be considered as a phenomenological parameter. Here, there is no spin mixing scattering anymore. Therefore, there is no need to retain the spin indexes 1,2.

In that case, we may use the parametrization of the normal and anomalous Green's functions $G = \cos \theta(x)$ and $F = \sin \theta(x)$ when the pair potential can be chosen real. For $\omega > 0$, the Usadel equations are

$$\omega \sin \theta_s - \frac{D_s}{2} \frac{1}{x^2} = \Delta(x)$$
 in the S layer and (12)

$$\left| \omega + ih + \frac{\cos \varphi_{f}}{2} \right| \sin \varphi_{f} - \frac{\omega_{f}}{2} \frac{\cos \varphi_{f}}{2} = 0 \text{ in the F layer.}$$
(13)

Note that the Usadel equations are nonlinear but may be linearized over the pair potential $\Delta(x)$ near T_c or when the S/F interface has low transparency.

3.2 Oscillating Cooper pair wave function

For a semi-infinite bilayer without magnetic impurities, the decaying solution for F_f is

$$F_f(x,\omega > 0) = A \exp\left| -\frac{1+\varepsilon}{\varepsilon} x \right|, \qquad (14)$$

where $\xi_f = \sqrt{D_f/h}$ is the characteristic length of the superconducting correlations decay (with oscillations) in F- layer. In real ferromagnets, the exchange field is very large compared with the superconducting order parameter ($h >> T_c$). Consequently, ξ_f is much smaller than the superconducting coherence length $\xi_s = \sqrt{D_s/(2\pi T_c)}$.

The constant A is determined by the boundary conditions at the S/F interface.

In a ferromagnet, the role of the Cooper pair wave function role is played by Ψ :

$$\Psi \sim \sum F(x,\omega) \sim \Delta \exp\left| -\frac{x}{r} \left| \cos \left| \frac{x}{r} \right| \right|.$$
 (15)

Thus, the damping oscillatory behavior of the order parameter is retrieved.

It can be seen from this microscopic approach that the decay length ξ_{f1} and the oscillation period ξ_{f2} are quite the same for a ferromagnet in the dirty limit with no magnetic impurities.

In presence of magnetic scattering, the decaying solution has the form

$$F_f(x, \omega > 0) = A \exp(-(k_1 + ik_2)x), \text{ which implies}$$
(16)

$$\psi \sim \Delta \exp\left(-\frac{x}{\xi_{f1}}\right)\cos\left(\frac{x}{\xi_{f2}}\right),$$

Where in the limit $h >> T_c$

$$k_1 = \frac{1}{z} \sqrt{\sqrt{1 + \alpha^2 + \alpha}} = \frac{1}{z}, \qquad (17)$$

$$k_1 = \frac{1}{\xi_f} \sqrt{\sqrt{1 + \alpha^2 - \alpha}} = \frac{1}{\xi_{f2}},$$
(18)

with $\alpha = \frac{1}{l_{\alpha}}$

If the spin-flip scattering time becomes relatively small $\alpha >> 1$, i.e. the magnetic impurities concentration is not negligible, the decaying length can become substantially smaller than the oscillating length, see Fig.3. This results in the much stronger decrease of the critical temperature in multilayers and critical current in S/F/S junctions with the increase of the F layer thickness.



Figure 3. Schematic evolution of Ψ without magnetic scattering (solid line) and with magnetic scattering (dashed line). Note that the oscillations do not disappear in presence of magnetic scattering, but become very small.

4. OSCILLATORY SUPERCONDUCTING TRANSITION TEMPERATURE IN S/F SYSTEMS

4.1 Theoretical description of S/F multilayers

We consider a S/F multilayered system with a thickness $2d_F$ of the F layers and $2d_S$ of the S layers, see Fig. 4 (this case is equivalent to a S/F bilayer of thicknesses d_F and d_S respectively).



Figure 4. Geometry of the studied multilayered system.

The critical temperature is determined by the self consistent equation for the superconducting gap:

$$\Delta \ln \frac{T_c}{T} + \pi T_c^* \sum \left| \frac{\Delta}{1-1} - F_s(x,\omega) \right| = 0$$
⁽¹⁹⁾

where T_c is the bare transition temperature of the superconducting layer in the absence of the proximity effect.

Consequently, the anomalous Green's function in the S layer has to be determined to find the critical temperature. It can be deduced from the anomalous Green's function in the F layer, and the boundary conditions at the S/F interface [21]:

$$\left| \frac{\sigma r_s}{2} \right| = \frac{\sigma_n}{2} \left| \frac{\sigma^2 f}{2} \right|$$
(20)

$$F_{\mathcal{S}}(0) = F_{f}(0) - \xi_{n} \gamma_{B} \left| \frac{\gamma_{f}}{\gamma} \right|$$
(21)

with $\sigma_n(\sigma_s)$ the conductivity of the F(S) layer, $\xi_n = \sqrt{D_f/(2\pi T_c)}$ and $\gamma_B = R_B \sigma_n/\xi_n$ is related to the S/F resistance per unit area R_B .

The anomalous Green's function in the F layer F_f is the solution of the linearized Usadel equation. Therefore, taking into account the symmetry of the system, it can be written as

$$F_f(x,\omega) = A \cosh[k(x - d_s - d_f)] \text{ in the 0 phase and}$$
(22)

$$F_f(x, \omega) = A \sinh[k[x - d_s - d_f]]$$
 in the π phase where (23)

$$k = k_1 + ik_2 = \frac{1}{\xi} \sqrt{i + \alpha}$$
 and $\xi_f = \sqrt{\frac{D_f}{z}}$.

Finally, when considering that the superconducting layer is thin, i.e. $d_s << \xi_s$, F_s varies a little in the S layer and is

$$F_{S}(x,\omega) = F_{0} \left[1 - \frac{\gamma_{\omega}}{2} x^{2} \right], \qquad (24)$$

where F_0 is the value of the anomalous Green's function at the center of the S layer:

$$F_0 = \frac{1}{2} (25)$$

The parameter τ_s^{-1} a pair-breaking parameter, that plays the same role as the corresponding parameter in the Abrikosov-Gorkov theory of superconductivity with magnetic impurities [22]. Note however that in our case, it can be complex. It is written as

$$\tau_{s,0}^{-1}(\omega > 0) = \tau_0^{-1} \frac{q}{2(1+1)} \frac{q}{2(1+1)}, \text{ in the 0 phase}$$
(26)

$$\tau_{s,\pi}^{-1}(\omega > 0) = \tau_0^{-1} \frac{q^{-1} \cdots q^{-1} q^{-1}}{q^{-1} \cdots q^{-1}} \text{ in the } \pi \text{ phase.}$$
(27)

The parameter $\tau_0^{-1} = \frac{\omega_s}{2 J} \frac{\sigma_n}{\sigma} \frac{1}{\kappa}$ while $\tilde{\gamma} = \frac{5 n}{\kappa} \gamma_B$, $q = k \xi_f$ and

 $d_f = d_f / \xi_f$.

Next, the critical temperature T_c^* is directly determined from the following equation

$$\ln\left|\frac{1}{T}\right| = \Psi\left|\frac{1}{2}\right| - \operatorname{Re}\Psi\left\{\frac{1}{2} + \frac{1}{2T}\right\}$$
(28)

4.2 Critical temperature versus F layer thickness

In Fig. 5, the critical temperature T_c^* has been plotted for two values of the magnetic scattering time for transparent interfaces. It can be deduced that the magnetic scattering decreases the damping length and increases the oscillation period. The decrease of the decay length obviously makes the observation of the oscillations more difficult.





Figure 5. Influence of the spin-flip scattering on the evolution of the critical temperature. (a) $\tau_0 = \frac{11}{h}$ and $\alpha = 0$. (b) $\tau_0 = \frac{11}{h}$ and $\alpha = \frac{1}{2}$.

Moreover, the intriguing evolution of T_c^* in bilayers with the finite interface transparency must be underlined (see Fig. 6). First, if $\alpha = 0$, there is no magnetic scattering and it could intuitively be believed that, the higher the barrier, the less is the influence of the proximity effect and therefore, $T_c^*(\tilde{\gamma} >> 1) > T_c^*(\tilde{\gamma} \sim 1)$. However, it can be seen from Fig. 6 that the critical temperature is a decreasing function of the barrier $\tilde{\gamma}$ for a small thickness of the F layer. This counter-intuitive behavior can be qualitatively understood as following. The probability for a Cooper pair to leave the S layer is smaller for a low transparent interface ($\tilde{\gamma} >> 1$). Nevertheless, the probability for this pair to come back again in the S layer is much higher for a transparent interface. Indeed, when the F layer is thin, the reflection of the Cooper pair at the other interface of the F layer allows the pair to cross again the first interface, which is easier when $\tilde{\gamma}$ is small. Consequently, the staying time in the F layer increases with the barrier, and when this time becomes bigger than the coherence time of the Cooper pair, the pair is destroyed, leading to a weakened superconductivity and therefore, the critical temperature decreases with the barrier. On the other hand, if d_f is not so small, the Cooper pair is hardly reflected by the external interface of the F layer whatever the value of $\tilde{\gamma}$ is and the critical temperature is expected to increase with the barrier.



Figure 6. Influence of the interface transparency on the evolution of the critical temperature.

Let us now consider briefly the general case, with spin-orbit and perpendicular spin flip scattering. An additional parameter has to be introduced, namely

$$\mathcal{A}_{\perp} = \frac{1}{L} \left(\frac{1}{L} - \frac{1}{L} \right) ,$$

and the parameter α becomes $\alpha = \frac{1}{L} \left(\frac{1}{L} + \frac{1}{L} \right)$.

The Usadel equation (10) can be linearized and the complex pair breaking parameter may be determined. The results are as following

In the 0 phase, $q \tanh[q\tilde{d}_f]$ in expression (26) is replaced by

(29)
$$1 + \xi_n \gamma_B q^* \tanh(q^* d_f)$$

where
$$q^2 = 2\left(\frac{\omega}{r} + \alpha - \alpha_{\perp} + i\sqrt{1 - \alpha_{\perp}^2}\right)$$
, (30)

and
$$\beta = -\alpha_{\perp} \frac{\alpha_{\perp} - \alpha_{\perp}}{\sqrt{\alpha_{\perp}}}$$
. (31)

In the π phase, $tanh(qd_f)$ is replaced by $coth(qd_f)$ in expression (29).

Therefore, the influence of 'perpendicular' spin-flip scattering and spinorbit scattering is quite similar to the influence of 'parallel' spin-flip processes, in the sense that it also implies the decrease of the decaying length and the increase of the oscillations period. However, a special situation arises when $\alpha_{\perp} > 1$. Then, the oscillations of the Cooper pair wave function are completely destroyed. Similar conclusion for spin-orbit mechanism was obtained in [9]. In fact, the influence of the 'perpendicular' magnetic scattering is analogous to the spin-orbit scattering. Probably the role of 'perpendicular' spin-flip or spin-orbit scatterings is important for the understanding of experimental results where no oscillations of the critical temperature were detected. Besides, note that the critical temperature oscillations can not disappear when there is only 'parallel' spin flip.

5. BEHAVIOR OF THE CRITICAL CURRENT

The constant I_c in the relation $I_s = I_c \sin \varphi$ is negative in the π phase while it is positive in the 0 phase. Thus, the transition from the 0 to π state may be considered as a change of the sign of the critical current, though the experimentally measured critical current is always positive and is equal to $|I_c|$. The $0-\pi$ transition occurs at each minimum of $|I_c|$.

The so called ' π junction' was first predicted for S/F/S structures by Buzdin *et al.* in 1982 in the clean limit [23], and later in the more realistic case of the diffusive limit [24]. Although the critical current behavior was a subject of intensive theoretical study, the experimental observation of the π state was difficult to obtain because the characteristic thickness of the F layer corresponding to the crossover from 0 to π state ξ_f is rather small. The first experimental evidence was finally reported by Ryazanov *et al.* in 2001[14] as a function of the temperature and later by Kontos *et al.* as a function of the ferromagnetic layer. The experimental data that are now available [14-16] on S/F/S junctions can be qualitatively understood in the framework of the existing approach. However, further development of the theory is needed for a more complete description. Below, we consider in more detail the influence of the magnetic scattering on the properties of S/F/S junctions. Experimental hints on the presence of relatively strong spin-flip effects were obtained in [15,25].

The studied geometry is a S/F/S junction of a thickness $2d_f$ of the F layer and large superconducting electrodes (see Fig. 7).



Figure 7. Geometry of the studied S/F/S Josephson junction.

The dirty limit conditions are supposed to be fulfilled. Therefore, the Usadel equations may be used. The supercurrent is determined by the following expression

$$I_{s}(\varphi) = ieN(0)D_{f}\pi TS \sum \left| F_{f} \frac{\alpha}{L}F_{f} - F_{f} \frac{\alpha}{L}F_{f} \right|, \qquad (32)$$

where $\tilde{F}_f(x,h) = F_f^*(x,-h)$, S is the area of the cross section of the junction and N(0) is the electron density of states per one spin projection.

5.1.1 Linearized Usadel equations

In the limit $T \to T_c$, the amplitude of F_f is small and the linearized Usadel equations are valid. Using the rigid boundary conditions, which are valid if $\sigma_n \xi_s / \sigma_f \xi_f << \max(\gamma_B, 1)$, the critical current may be easily calculated

$$I_{c} = \frac{2q}{\xi} \operatorname{Re} \left[\sum_{\alpha} \frac{2}{2 + \lambda^{2}} \frac{2q}{\operatorname{sink}(2 - \widetilde{J})(1 + \widetilde{\sigma}^{2} - \widetilde{z}) + 2 - \widetilde{\sigma} \operatorname{sock}(2 - \widetilde{J})} \right]$$
(33)

where $\tilde{\gamma} = \gamma_B \xi_n / \xi_f$, and q = a + ib. Expression (34) takes into account the magnetic scattering and

 $a = \sqrt{\sqrt{1 + \alpha^2} + \alpha}$ and

$$b = \sqrt{1 + \alpha^2 - \alpha}$$
, with $\alpha = 1/(\tau_m h)$.

This formula can be generalized to the case when the interface barriers are different (noted γ_{B1} and $\gamma_{B1} \gamma_{+Bp}$). Then, $\tilde{\gamma}^2$ must be replaced by $\gamma_{B1}\gamma_{B2}(\xi_n/\xi_f)^2$ and $\tilde{\gamma}$ by $\frac{\gamma_{B1}\gamma_{B2}\xi_n}{2}$.

Besides, this expression is also valid at all temperatures if $\gamma_B >> 1$, with the substitution $\tilde{\gamma} \to \tilde{\gamma} / G_s$. G_s is the normal Green function in the superconducting electrodes $G_s = \omega / \sqrt{\omega^2 + \Delta^2}$.

The evolution of the critical current for different values of the barrier transparency is given in Fig. 8.



Figure 8. Evolution of the critical temperature with the thickness of the F layer for different interface transparencies.

Besides, if $\tilde{\gamma} >> 1$, the previous expression may be simplified and the critical current becomes

$$I_c = \frac{2}{\varepsilon} \frac{1}{\varepsilon} \operatorname{Re} \left| \frac{2}{\varepsilon^2 + 1/2} \right|.$$
(34)

When there is no magnetic scattering, the transition into the π phase occurs in the limit $T \rightarrow 0$ at

Superconductor-ferromagnet heterostructures

 $\tilde{d}_f^c = \sqrt{\frac{2}{n}} \ln \left| \frac{n}{n} \right|$, see [26] and [6] for further detail.

It can be seen from the previous expression that in the absence of magnetic scattering, the exchange field determines the critical thickness. Therefore, when $\alpha = 0$, the critical thickness may be much smaller than ξ_{f} .

In presence of magnetic scattering, if $T_c / h < \alpha < 1$, the first minimum is achieved at $\tilde{d}_f^c = 3\alpha$. Note that the experimental measuring of the critical thickness allows a direct determination of the magnetic scattering.

When the spin flip scattering controls the system, i. e. $\alpha > 1$, then the subsequent $0-\pi$ transitions occur at

$$d_f^c = \frac{\psi + n\pi}{2L}$$
, where ψ is defined by $\tan \psi = \frac{\pi}{L}$.

In that case, the critical thickness d_f^c is larger than ξ_f .

If one interface is completely transparent $\gamma_{B1} = 0$ and the other interface has a large barrier $\gamma_{B2} >> 1$, then we obtain the following expression for the critical current near T_c

$$I_c = \frac{2}{\varepsilon} \frac{1}{\varepsilon} \frac{1}{\varepsilon} \frac{1}{\varepsilon} \operatorname{Re} \left[\frac{2}{\varepsilon} \frac{1}{\varepsilon} \right]. \tag{35}$$

It vanishes for $d_f^c = \frac{n}{AT}$.

Thus, a vanishing barrier interface tends to increase the critical thickness, as can be seen from Fig. 8.

It can be seen that the critical thickness increases with the increase of the spin flip rate. At the same time, the magnetic scattering strongly increases the damping of I_c with the increase of the F layer thickness. Consequently, the magnetic scattering role is quite controversial for the experimental observation of the $0-\pi$ transitions. Indeed, even though the increase of the critical thickness leads to an easier observation of the transitions, the decrease of the decay length is on the contrary quite harmful for experiments.

5.1.2 Transparent interfaces

For a transparent interface, the linearized Usadel equation can not be used at low temperature and the complete nonlinear equation must be solved. Introducing the dimensionless parameters $\tilde{\omega} = \omega / h$, $\alpha = 1/(\tau_m h)$ and $y = x/d_f$, it becomes

$$-\frac{1}{2}\frac{\partial^2 f}{\partial a^2} + (\tilde{\omega} + i + \alpha \cos \theta_f)\sin \theta_f = 0 \text{ in the F layer.}$$
(36)

A S/F/S junction presents two interfaces. In the limit of relatively large thickness of the F layer $d_f > \xi_f$, the decay of the Cooper pairs wave function occurs independently near each interface. It can therefore be treated separately enough to consider the behavior of the anomalous Green's function near each S/F interface, assuming that the F layer thickness is infinite. one interface.

Although Usadel equation (36) is nonlinear, one may find its exact solution. Using the first integral of (36) with the following boundary conditions:

$$\Theta_f(y \to \infty) = \left| \frac{\sigma \sigma_f}{2\pi} \right| = 0$$

We obtain the following relation

$$g = \frac{v}{1 + 2 + 2 + 1 + 2}$$
(37)

where $q = \sqrt{2(\tilde{\omega} + i + \alpha)}$ and $p^2 = \alpha / (\tilde{\omega} + i + \alpha)$. The function $g = g_0 \exp(-2qy)$, where the constant g_0 is determined by the continuity of the Green's functions at the interface. In the case of the rigid boundary conditions, the inverse proximity effect may be neglected. As a result,

$$g_0 = \frac{1}{\sqrt{(1 + 1^2)^2}}$$
 and $U(n) = \frac{1}{\sqrt{(1 + 1^2)^2}}$,

where $\Omega = \sqrt{\omega^2 + \Delta^2}$.

The anomalous Green's function at the center of the F layer may be taken as the superposition of the two decaying F_f functions. As a result, the current-phase relation is sinusoidal and the critical current becomes

$$I_c = 64 \frac{1}{r} \operatorname{Re} \left[\sum_{r} \frac{1}{r} \frac{1}{r} \operatorname{Re} \right] \left[\sum_{r} \frac{1}{r} \frac{1}{r} \frac{1}{r} \right]$$
(38)

where $d_f = d_f / \xi_f$.

This expression generalizes the corresponding formula from Ref. [24] to the case of finite magnetic scattering.

The critical current is proportional to the small factor $\exp(-2qd_f)$. The terms neglected in our approach are much smaller and are of the order of $\exp(-4qd_f)$. Therefore, they give a tiny second harmonic term in the current-phase relation.

When $T \rightarrow T_c$, expression (36) may be simplified and we have

$$I_c = \frac{m (2 - y)^2}{T + y} \frac{\partial}{\partial t} \exp(-2ad_f) \sin(2yd_f + \psi), \qquad (39)$$

where $\tan \psi = \frac{\alpha}{r}$ and q = a + ib. Note that if the magnetic scattering is negligible $\alpha \to 0$, $\psi = \frac{\alpha}{r}$ while if $\alpha > 1$, $\frac{\alpha}{r} < \psi < \frac{\alpha}{2}$.

The two characteristic lengths, namely the decay length and the oscillations period, appear in expression (40). Accordingly, this formula should be very helpful to fit experimental data.

Recent systematic studies of the thickness dependence of the critical current in junctions with $Cu_x Ni_{1-x}$ alloy as a F layer [25] revealed a very strong variation of I_c with the F layer thickness. Indeed, a five orders of magnitude change of the critical current was observed in the thickness interval (12-26) nm. The magnetic scattering effect - inherent to all the ferromagnetic alloys- is probably at the origin of this behavior. Besides, the

presence of a rather strong magnetic scattering in $Cu_x Ni_{1-x}$ alloy S/F/S junctions was also noted in [15, 26, 27].

The theoretical fit based on Eq. (39) shows good agreement with the experimental data [28] if the spin-flip scattering is taken into account (see Fig. 9).



Figure 9. Critical current of Cu_{0.47} Ni_{0.53} junctions as a function of the F layer thickness (Ryazanov *et al.*, 2005). Two 0- π transitions are revealed. For this fit, the choice for the parameters was: $\alpha = 1.33$ and $\xi_f = 2.4 nm$. The insert shows the temperature mediated 0- π transition for a F layer thickness of 11 nm.

In presence of 'perpendicular' magnetic scatterings, up and down spins states are mixed and equation (10) has to be used. When the temperature is close to the critical temperature, this expression may be linearized. In that case, the critical current becomes

$$I_{c} = I_{0} \operatorname{Re} \left[\sum_{\alpha} \frac{\Delta q}{1 + (2 + \tilde{q} + 1))} \right] + \beta \frac{\operatorname{cost}(q + f)}{1 + (1 + \tilde{q} + 1))} \left\| q + q^{*} \beta \frac{\operatorname{stat}(q + f)}{1 + (1 + \tilde{q} + 1))} \right\|,$$

where $I_0 = \frac{\alpha + \zeta_0 + \omega_f}{\xi_f}$, $\Omega^2 = \omega^2 + \Delta^2$ and β is defined by expression (31).

Note that when $\alpha_{\perp} \rightarrow 0$, the previous expression gives the critical current when there is only spin flip in the direction of the exchange field. The oscillations of the critical current (and the $0-\pi$ transitions) completely disappear if $\alpha_{\perp} > 1$. Then, the experimental observation of these oscillations [14-16, 26] may be considered as an indication of the weakness of spin-orbit and 'perpendicular' spin-flip scatterings effects.

Besides, this expression may be written in the same form as expression (40) which is used to fit experimental data. However, the expressions for the corresponding parameters are rather lengthy and therefore are not presented here.

6. ELECTRONIC MAGNETIZATION VARIATION IN S/F SYSTEMS

The mutual influence of ferromagnetism and superconductivity has until now been considered through its consequences on the evolution of the superconducting critical temperature and critical current. Nevertheless, the proximity effect can also manifest itself by a modification of the electronic magnetization. Indeed, the presence of the ferromagnetic magnetization leads to a magnetization onset in the S layer. Similarly, the ferromagnetic layer may present a variation of its magnetization.

It should be underlined that this topic has already been investigated in the dirty limit by Krivoruchko *et al.* [29] and Bergeret *et al.* [30] and in clean multilayered S/F structures by Halterman and Valls [31].

6.1 Ferromagnet at the contact with a superconductor

We consider a S/F system, with a thickness d_s of the S layer and an infinite thickness of the F layer. The x axis is chosen to be perpendicular to the layer, with the origin at the vacuum-S layer interface.

The magnetization of the F layer is

$$M = M_p + M_s, \tag{40}$$

where M_p is the magnetization due to the Pauli paramagnetism while M_s stems from the superconductivity contribution. M_s may be expressed as

$$M_s = iN(0)\pi I \sum (G_{f\uparrow\uparrow} - G_{f\downarrow\downarrow}), \qquad (41)$$

where G is the normal Green function in the F layer and may be deduced from F_f thanks to the normalization condition $G_f^2 - F_f^2 = 1$.

For $T \sim T_c$, the anomalous Green's function is small, and therefore, $G_f(\omega) \sim 1$ - $F_f^2/2$. Also taking into account that $F_{f\downarrow\uparrow} = F_{f\uparrow\downarrow}^* = F_f^*$, the magnetization may be presented as

$$M_s = \iota N(0)\pi I \sum_{i} (F_f(\omega > 0) - F_f(\omega > 0))$$
(42)

Note that (42) gives in fact the part of the magnetization related to the presence of superconducting correlations. Since $T \rightarrow T_c$, the linearized Usadel equation may be solved to find the anomalous Green's function. If the interface is supposed to be transparent, i. e. $\gamma_B = 0$, calculations give the following final expression of the magnetization

$$M_{s} = -2N(0)\pi T\Delta^{2} \exp(-2x/\xi_{f}) [A(\omega)\sin(2x/\xi_{f}) + B(\omega)\cos(2x/\xi_{f})], \quad (43)$$

with $A(\omega) = \sum_{f} \frac{1}{((\omega + \pi^{-1})^{2} + (\pi^{-1})^{2})^{2}} \omega (\omega + 2\pi_{0}^{-1}),$

and
$$B(\omega) = \sum \frac{1}{((\omega + \tau^{-1})^2 + (\tau^{-1})^2)^2} 2\tau_0^{-1}(\omega + \tau_0^{-1}),$$

where $\tau_0^{-1} = \frac{\omega_s}{2 J} \frac{\sigma_n}{\sigma} \frac{1}{\varepsilon}$ is the pair breaking parameter.

In the case of very low S/F interface transparency, the magnetization becomes

$$M_{s} = -N(0)\pi T\Delta^{2} \exp(-2/\xi_{f}) \cos(2x/\xi_{f}) \sum_{f} \frac{1}{(m+\sigma^{-1}/\pi)^{2} w^{2}}.$$
 (44)

A qualitative evolution of the magnetization variation is given in Fig. 10.

Figure 10. Qualitative oscillating evolution of the magnetization variation in the F layer.

In all cases, the total magnetization of the F layer should present an oscillating behavior. Interestingly, the electronic magnetization at some distance may be larger than in the absence of superconductivity.

6.2 Magnetization variation in the S layer in a thin S/F bilayer

Let us now consider a thin S/F bilayer (see Fig. 11) with $d_f << \xi_f$ and $d_s << \xi_s$.



Figure 11. Geometry of the studied system.

The magnetization in the S layer may be easily determined in this case. For a transparent interface, it reads

Therefore, the inverse proximity effect leads to the appearance of a negative magnetization in the superconducting layer. Ref. [31] explains this fact quite simply. Namely, although the Cooper pairs inside the F layer do not contribute to a magnetization onset, a Cooper pair with one electron in

each layer does. Indeed, the electron in the F layer has its spin parallel to the exchange field (up), while the second electron of the pair has a spin down. Therefore, each of theses Cooper pairs gets a favorite orientation, with the up spin electron in the F layer and the down spin electron in the S layer. That is how a magnetization appears in the S layer, whose direction is the opposite of the one in the F layer.

7. CONCLUSION

We have investigated the particularities of the proximity effect in S/F multilayered systems in the dirty limit. We demonstrated that the spin-flip scattering may strongly influence the behavior of the critical temperature and critical current. Indeed, it decreases the decay length and increases the oscillations period. Moreover, perpendicular magnetic scatterings can even lead to the complete destruction of the oscillations. It should therefore be taken into account for theoretical fits of experimental data for systems with weak magnetic anisotropy.

Besides, another manifestation of the proximity effect in S/F bilayer is the appearance of a negative magnetization in the S layer, while the magnetization of the F layer is being oscillating.

ACKNOWLEDGMENTS

The authors are grateful for V. Ryazanov for useful discussions and J. Cayssol for critical reading of the manuscript. This work was supported in part by ESF PI Shift Program.

REFERENCES

- [1] V. L. Ginzburg, Zh. Teor. Fiz. 31, 202 (1956) [Sov. Phys. JETP 4, 153 (1957)].
- [2] P. W. Anderson, and H. Suhl, Phys. Rev. 116, 898 (1959).
- [3] L. N. Bulaevskii, A. I. Buzdin, M. L. Kulic, and S. V. Panjukov, Advances in Physics 34, 175 (1985).
- [4] A. I. Larkin, and Y. N. Ovchinnikov, Zh. Eksp. Teor, Fiz. 47, 1136 (1964) [Sov. Phys. JETP 20, 762 (1965)].
- [5] P. Fulde, and R. A. Ferrell, Phys. Rev. B 135, A550 (1964).
- [6] A. I. Buzdin, Rev. Mod. Phys. (in press).
- [7] A. A. Golubov, M. Yu. Kuprianov, and E. Il'ichev, Rev. Mod. Phys. 76, 411 (2004).

- [8] L. R. Tagirov, Physica C 307, 145 (1998).
- [9] E. A. Demler, G. B. Arnold, and M. R. Beasley, Phys. Rev. B 55, 15174 (1997).
- [10] P. G. De Gennes, Superconductivity of Metals and Alloys, Benjamin, New York, 1966.
- [11] G. Deutscher, and P. G. De Gennes, in *Superconductivity*, 1005-1034, edited by R. D. Parks, Marcel Dekker, New York, 1969.
- [12] H. K. Wong, B. Y. Jin, H. Q. Yang, J. B. Ketterson, and J. E. Hilliard, J. Low Temp. Phys. 63, 307 (1986).
- [13] J. S. Jiang, D. Davidovic, D. H. Reich, and C. L. Chien, Phys. Rev. Lett. 74, 314 (1995).
- [14] V. V. Ryazanov, V. A. Oboznov, A. V. Veretennikov, A. Yu. Rusanov, A. A. Golubov, and J. Aarts, Phys. Rev. Lett. 86,2427 (2001).
- [15] H. Sellier, C. Baraduc, F. Lefloch, and R. Calemczuk, Phys. Rev. B 68, 054531 (2003).
- [16] T. Kontos, M. Aprili, J. Lesuer, F. Genet, B. Stephanidis, and R. Boursier, Phys. Rev. Lett. 89, 137007 (2002).
- [17] G. Eilenberger, Z. Phys. 214, 195 (1968).
- [18] L. Usadel, Phys.Rev. Lett. 25, 507 (1970).
- [19] P. Fulde and K. Maki, Phys. Rev. 141, 275 (1966).
- [20] A. A. Abrikosov and L. P. Gor'kov, Zh. Eksp. Theor. Fiz. 39, 1781 (1960) [Sov. Phys. JETP 12, 1243 (1961)].
- [21] M.Y. Kuprianov, and V. F. Lukichev, Zh. Eksp. Teor, Fiz. 94, 147 (1982) [Sov. Phys. JETP 67, 1163 (1988)].
- [22] A. A. Abrikosov, L. P. Gorkov, *Methods of Quantum Field Theory in Statistical Physics*, Prentice-Hall, New Jersy, 1963 (English version).
- [23] A. I. Buzdin, L. N. Bulaevskii, and S. V. Panyyukov, Pis'ma Zh. Eksp. Teor, Phys. 35, 147 (1982) [JETP Lett 35, 178 (1982)].
- [24] A. I. Buzdin, and M.Y. Kuprianov, Pis'ma Zh. Eksp. Teor, Phys. 53, 308 (1991) [JETP Lett 53, 321 (1991)].
- [25] V. V. Ryazanov, V. A. Oboznov, A. S. Prokov'ev, V. V. Bolginov, and A. K. Feofanov, J. Low Temp. Phys. 136, 385 (2004).
- [26] A. I. Buzdin, Pis'ma Zh. Eksp. Teor. Fiz. 78, 1073 (2003) [JETP Lett.78, 583 (2003)].
- [27] L. Cretinon, A. K. Kupta, H. Sellier, F. Lefloch, M. Fauré, A. I. Buzdin, and H. Courtois cond-matt 0502050.
- [28] V. V. Ryazanov, V. A. Oboznov, V. V. Bolginov, A. K. Feofanov, and A. I. Buzdin, to be published (2005).
- [29] V. N. Krivoruchko and E. A. Koshina, Phys. Rev. B 66, 014521 (2002).
- [30] K. Halterman and O. T. Valls, Phys. Rev. B 69, 014517 (2004).
- [31] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Phys. Rev. B 69,174504 (2004).