Proximity effects in superconductor-ferromagnet heterostructures

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The proximity effect at superconductor-ferromagnet interfaces produces damped oscillatory behavior of the Cooper pair wave function within the ferromagnetic medium. This is analogous to the inhomogeneous superconductivity, predicted long ago by Fulde and Ferrell (P. Fulde and R. A. Ferrell, 1964, "Superconductivity in a strong spin-exchange field," Phys. Rev. **135**, A550–A563), and by Larkin and Ovchinnikov (A. I. Larkin and Y. N. Ovchinnikov, 1964, "Inhomogeneous state of superconductors," Zh. Eksp. Teor. Fiz. **47**, 1136–1146 [Sov. Phys. JETP **20**, 762–769 (1965)]), and sought by condensed-matter experimentalists ever since. This article offers a qualitative analysis of the proximity effect in the presence of an exchange field and then provides a description of the properties of superconductor-ferromagnet heterostructures. Special attention is paid to the striking nonmonotonic dependence of the critical temperature of multilayers and bilayers on the ferromagnetic layer thickness as well as to the conditions under which " π " Josephson junctions are realized. Recent progress in the preparation of high-quality hybrid systems has permitted the observation of many interesting experimental effects, which are also discussed. Finally, the author analyzes the phenomenon of domain-wall superconductivity and the influence of superconductivity on the magnetic structure in superconductor-ferromagnet bilayers.

CONTENTS

I.	Introduction	935	
II.	Paramagnetic Limit and Qualitative Explanation of the		
	Nonuniform Phase Formation	937	
	A. The (H, T) phase diagram	937	
	B. Exchange field in the ferromagnet	937	
	C. Why does the Fulde-Ferrell-Larkin-Ovchinnikov		
	state appear?	938	
	D. Generalized Ginzburg-Landau functional	938	
III.	Proximity Effect in Ferromagnets	939	
	A. Some generalities about superconducting proximit	y	
	effect	939	
	B. Damped oscillatory dependence of the Cooper pa	ir	
	wave function in ferromagnets	940	
	C. Density-of-states oscillations	942	
	D. Andreev reflection at the S/F interface	943	
IV.	Oscillatory Superconducting Transition Temperature	in	
	S/F Multilayers and Bilayers	944	
	A. First experimental evidence of the anomalous		
	proximity effect in S/F systems	944	
	B. Theoretical description of the S/F multilayers	945	
	C. 0 and π phases	947	
	D. Oscillating critical temperature	947	
V.	Superconductor-Ferromagnet-Superconductor π		
	Junction	950	
	A. General characteristics of the π junction	950	
	B. Theory of π junctions	950	
	C. Experiments with π junctions	954	
VI.	Complex S/F Structures	955	
	A. F/S/F spin-valve sandwiches	955	
	B. S-F-I-F'-S heterostructures and triplet proximity		
	effect	958	
/II.	Atomic Thickness S/F Multilayers	960	
	A. Layered ferromagnetic superconductors	960	

В.	Exactly solvable model of the π phase	960
VIII. Su	perconductivity Near the Domain Wall	962
IX. M	odification of Ferromagnetic Order by	
Sı	perconductivity	963
A.	Effective exchange field in thin S/F bilayers	963
В.	Domain structure	964
C.	Negative domain-wall energy	965
D.	Ferromagnetic film on a superconducting substrate	966
X. C	onclusions	966
Acknowledgments		967
Appendix		967
1.	Bogoliubov-de Gennes equations	967
2.	Eilenberger and Usadel equations for ferromagnets	967
References		

I. INTRODUCTION

Due to their incompatible nature, singlet superconductivity and ferromagnetic order do not coexist in bulk materials. Ginzburg (1956) first formulated the problem of the coexistence of magnetism and superconductivity considering an orbital mechanism by which superconductivity is suppressed (the interaction of the superconducting order parameter with a vector potential A of the magnetic field). After the advent of the BCS theory by Bardeen, Cooper, and Schrieffer (1957), it became clear that superconductivity in the singlet state could also be destroyed by an exchange mechanism. The exchange field, in a magnetically ordered state, tends to align spins of Cooper pairs in the same direction, thus preventing a pairing effect. This is the so-called *paramagnetic effect* (Saint-James et al., 1969). Anderson and Suhl (1959) demonstrated that ferromagnetic ordering is unlikely to appear in the superconducting phase. The main reason for this is the suppression of the zero-wave-vector component of the electronic paramagnetic susceptibility in

١

the presence of superconductivity. In such a situation the energy for ferromagnetic ordering decreases and, instead of ferromagnetic order, nonuniform magnetic ordering should appear. Anderson and Suhl (1959) called this state *cryptoferromagnetic*.

The 1977 discovery of ternary rare-earth (RE) compounds (RE)Rh₄B₄ and (RE)Mo₆X₈ (X=S, Se; for a review, see, for example, Maple and Fisher, 1982) provided the first experimental evidence of magnetism and superconductivity coexisting in stoichiometrical compounds. It turned out that in many of these systems superconductivity coexists with antiferromagnetic order, and the Néel temperature $T_N < T_c$, the critical temperature.

The more recent discovery of superconductivity in the quaternary intermetallic compounds [(RE)Ni₂B₂C, for a review, see, for example, Müller and Narozhnyi, 2001] offers another example of antiferromagnetism and superconductivity coexisting.

Indeed, superconductivity and antiferromagnetism can coexist quite peacefully because, on average, the exchange and orbital fields are zero at distances of the order of the Cooper pair size or superconducting coherence length. Even more interesting, reentrant superconductivity was observed in ErRh₄B₄ and HoMo₆S₈ (Maple and Fisher, 1982). For example, ErRh₄B₄ becomes a superconductor below T_c =8.7 K. When it is cooled to the Curie temperature $\Theta \approx 0.8$ K, an inhomogeneous magnetic order appears in the superconducting state. With further cooling the superconductivity is destroyed by the onset of a first-order ferromagnetic transition at the second critical temperature $T_{c2} \approx 0.7$ K. Another example of reentrant superconductivity is HoMo₆S₈ with T_c =1.8 K, $\Theta \approx 0.74$ K, and $T_{c2} \approx 0.7$ K.

As predicted by Anderson and Suhl (1959), a nonuniform magnetic order appears at the Curie temperature in these compounds. Its presence has been confirmed by neutron-scattering experiments. The period of this magnetic structure is smaller than the superconducting coherence length but larger than the interatomic distance. In some sense this structure is the realization of a compromise between superconductivity and ferromagnetism: for the superconductivity it is seen as antiferromagnetism, but for the magnetism it looks like ferromagnetism. Theoretical analysis, taking into account orbital and exchange mechanisms as well as magnetic anisotropy (for a review, see Bulaevskii et al., 1985), revealed that the coexistence phase is a domainlike structure with very small period. The region in which magnetism and superconductivity coexist in ErRh₄B₄ and HoMo₆S₈ is narrow, but in HoMo₆Se₈ the domain of coexistence survives until T=0 K.

The first truly ferromagnetic superconductors, UGe₂ (Saxena *et al.*, 2000) and URhGe (Aoki *et al.*, 2001), were discovered only recently. Apparently these systems have a triplet pairing character which permits the coexistence of superconducting with ferromagnetism. Indeed, superconductivity in URhGe (Aoki *et al.*, 2001) appears below 0.3 K in the ferromagnetic phase, while the Curie temperature is Θ =9.5 K; this makes the sin-

glet scenario of superconductivity rather improbable.

Though the coexistence of singlet superconductivity with ferromagnetism is very unlikely in bulk compounds, it may be easily achieved in artificially fabricated layered ferromagnet/superconductor (F/S) systems. Due to the *proximity effect* described later, the Cooper pairs can penetrate into the F layer and induce superconductivity there. In such a case we have a unique opportunity to study the properties of superconducting electrons under the influence of a large exchange field. In addition, it is possible to study the interplay between superconductivity and magnetism in a controlled manner, since by varying the layer thicknesses we change the relative strengths of two competing orderings. The behavior of the superconducting condensate under these conditions is quite peculiar.

A long time ago Larkin and Ovchinnikov (1964) and Fulde and Ferrell (1964) demonstrated that, in a pure ferromagnetic superconductor at low temperature, superconductivity may be nonuniform. Due to the incompatibility of ferromagnetism and superconductivity it is not easy to verify this prediction experimentally. Superconducting/ferromagnet systems are in some ways analogous to the nonuniform superconducting state. The Cooper pair wave function extends from superconductor to ferromagnetic with damped oscillatory behavior. This results in many new effects, which we discuss in this review: spatial oscillations of the electron density of states, a nonmonotonic dependence of the critical temperature of S/F multilayers and bilayers on the ferromagnet layer thickness, and the realization of " π " Josephson junctions in S/F/S systems. Spin-valve behavior in complex S/F structures gives another example of the interesting interplay between magnetism and superconductivity, an effect that is promising for potential applications. We also discuss localized domain-wall superconductivity in S/F bilayers and the inverse influence of superconductivity on ferromagnetism, which favors nonuniform magnetic structures. An interesting example of atomicthickness S/F multilayers is provided by layered superconductors like $Sm_{1.85}Ce_{0.15}CuO_4$ and RuSr₂GdCu₂O₈. For such systems the exchange field in the F layer also favors the π -phase behavior, with an alternating order parameter in adjacent superconducting layers.

Note that practically all interesting effects related to the interplay between superconductivity and magnetism in S/F structures occur at the nanoscale range of layer thicknesses. The observation of these effects became possible only recently due to the great progress in the preparation of high-quality hybrid F/S systems. The experimental progress and the promise of potential applications in turn stimulated a revival of interest in the interplay of superconductivity and ferromagnetism in heterostructures. It seems to be timely to review the research in this domain and consider the outlook for future work.

0.8

II. PARAMAGNETIC LIMIT AND QUALITATIVE EXPLANATION OF THE NONUNIFORM PHASE FORMATION

A. The (H, T) phase diagram

For a pure paramagnetic effect, the critical field of a superconductor H_p at T=0 may be found from a comparison of the energy gain ΔE_n due to the electron-spin polarization in the normal state and the superconducting condensation energy ΔE_s . In the normal state, the polarization of the electron gas changes its energy in the magnetic field by

$$\Delta E_n = -\chi_n \frac{H^2}{2},\tag{1}$$

where $\chi_n = 2\mu_B^2 N(0)$ is the spin susceptibility of the normal metal, μ_B is the Bohr magneton, 2N(0) is the density of electron states at Fermi level (per two spin projections), and the electron g factor is equal to 2.

On the other hand, in a superconductor the polarization is absent, but the BCS pairing energy decreases by

$$\Delta E_s = -N(0)\frac{\Delta_0^2}{2},\tag{2}$$

where $\Delta_0 = 1.76T_c$ is the superconducting gap at T=0. From the condition $\Delta E_n = \Delta E_s$, we find the Chandrasekhar (1962)-Clogston (1962) limit, or the paramagnetic limit at T=0,

$$H_p(0) = \frac{\Delta_0}{\sqrt{2}\mu_B}.$$
(3)

Note that this field represents the first-order phase transition from a normal to a superconducting state. The complete analysis (Saint-James et al., 1969) demonstrates that at T=0 this critical field is higher than the second-order phase transition $H_p^{\text{II}}(0) = \Delta_0/2\mu_B$, and the transition from a normal to a uniform superconducting state is of second order at $T^* < T < T_c$, where T^* =0.56 T_c , $H^* = H(T^*) = 0.61\Delta_0/\mu_B = 1.05T_c/\mu_B$. However, Fulde and Ferrell (1964) and Larkin and Ovchinnikov (FFLO) (1964) predicted in the framework of the model of pure paramagnetic effect the appearance of the nonuniform superconducting state with a sinusoidal modulation of the superconducting order parameter at the scale of the superconducting coherence length ξ_s . In this FFLO state, the Cooper pairs have a finite momentum, compared with zero momentum in conventional superconductors. Recently, Casalbuoni and Nardulli (2004) reviewed the theory of the inhomogeneous superconductivity applied to the condensed matter and quantum chromodynamics at high density and low temperature.

The critical field of the second-order transition into the FFLO state appears above the first-order transition line into a uniform superconducting state (Saint-James *et al.*, 1969). At T=0, it is $H^{\text{FFLO}}(0)=0.755\Delta_0/\mu_B$ whereas $H_p=0.7\Delta_0/\mu_B$. This FFLO state only appears in the temperature interval $0 < T < T^*$, and is sensitive to impurities (Aslamazov, 1968). In the dirty limit it is suppressed,



At temperatures below $T^* = 0.56T_c$ the second-order transition occurs from the normal to the nonuniform superconducting FFLO phase. The bold line corresponds to the first-order transition into the uniform superconducting state, and the dotted line represents the second-order transition into the nonuniform superconducting state.

and the first-order transition into the uniform superconducting state occurs instead. The phase diagram for three-dimensional (3D) superconductors in the pure paramagnetic effect model is presented in Fig. 1 (Saint-James *et al.*, 1969). Up to now, there were no unambiguous experimental proofs of this state. Note, however, that recently the magnetic-field-induced superconductivity has been observed in the quasi-two-dimensional organic conductor (BETS)₂FeCl₄ (Uji *et al.*, 2001) which is an excellent candidate for the FFLO state formation (Balicas *et al.*, 2001; Houzet *et al.*, 2002).

B. Exchange field in the ferromagnet

In a ferromagnet an exchange interaction between the electrons and magnetic moments may be considered as an effective Zeeman field. In the case of magnetic moments with spin S_i , localized in the sites r_i , their interaction with electron spins is described by the exchange Hamiltonian

$$H_{\rm int} = \int d^3 r \Psi^+(\mathbf{r}) \left\{ \sum_i J(\mathbf{r} - \mathbf{r}_i) \mathbf{S}_i \sigma \right\} \Psi(\mathbf{r}), \qquad (4)$$

where $\Psi(\mathbf{r})$ is the electron's spinor operator, $\sigma = \{\sigma_x, \sigma_y, \sigma_z\}$ are the Pauli matrices, and $J(\mathbf{r})$ is the exchange integral. Below the Curie temperature Θ , the average value of the localized spins $\langle \mathbf{S}_i \rangle$ is nonzero, and the exchange interaction may be considered as an effective Zeeman field $H^{\text{eff}} = (\langle S_i^z \rangle n / \mu_B) \int J(\mathbf{r}) d^3 r$, where *n* is the concentration of localized moments, and the spin quantization *z* axis is chosen along the ferromagnetic moment. It is convenient to introduce the exchange field *h* as

938

$$h = \mu_B H^{\text{eff}} = \langle S_i^z \rangle n \int J(\mathbf{r}) d^3 r = s(T) h_0, \qquad (5)$$

where $s(T) = \langle S_i^z \rangle / \langle S_i^z \rangle_{T=0}$ is the dimensionless magnetization and h_0 is the maximum value of an exchange field at T=0. The exchange field *h* describes the spin-dependent part of the electron's energy and the exchange Hamiltonian (4) is then

$$H_{\rm int} = \int d^3 r \Psi^+(\mathbf{r}) h \sigma_{\mathbf{z}} \Psi(\mathbf{r}).$$
(6)

If we also want to take into account the proper Zeeman field of magnetization M, then replace h in Eq. (6) by $h+4\pi M\mu_B$. The reader is warned that, in principle, if the exchange integral is negative, the exchange field will be in the direction opposite to the magnetic moments and the Jaccarino-Peter compensation effect (Jaccarino and Peter, 1962) is possible. However, in ferromagnetic metals, the contribution of the magnetic induction to the spin splitting is several orders of magnitude smaller than that of the exchange interaction and may be neglected. In the case of the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism of the ferromagnetic ordering, the Curie temperature $\Theta \sim h_0^2/E_F$ and in all real systems the exchange field $h_0 \ge \Theta, T_c$. This explains that for singlet superconductivity and ferromagnetism to coexist the conditions required are very stringent. Indeed, if Θ $> T_c$ the exchange field in a ferromagnet $h \ge T_c$, which strongly exceeds the paramagnetic limit. On the other hand, if $\Theta < T_c$ then, instead of the ferromagnetic transition inhomogeneous magnetic ordering appears (Maple and Fisher, 1982; Bulaevskii et al., 1985). The large value of the exchange field in a ferromagnet permits us to concentrate on the paramagnetic effect and neglect the orbital one. Note that well below the Curie temperature the magnetic induction $4\pi M$ in ferromagnets is of the order of several kOe.

C. Why does the Fulde-Ferrell-Larkin-Ovchinnikov state appear?

What is the physical origin of the superconducting order-parameter modulation in the FFLO state? The appearance of modulation in the superconducting order parameter is related to Zeeman's splitting of the electron's level under a magnetic field acting on electron spins. To demonstrate this, we consider the simplest case of the 1D superconductor.

In the absence of the field, a Cooper pair is formed by two electrons with opposite momenta $+k_F$ and $-k_F$ and opposite spins (\uparrow) and (\downarrow), respectively. The resulting momentum of the Cooper pair will be $k_F + (-k_F) = 0$. Under a magnetic field, because of Zeeman's splitting, the Fermi momentum of the electron with spin (\uparrow) will shift from k_F to $k_1 = k_F + \delta k_F$, where $\delta k_F = \mu_B H/v_F$ and v_F is the Fermi velocity. Similarly, the Fermi momentum of an electron with spin (\downarrow) will shift from $-k_F$ to $k_2 = -k_F$ $+ \delta k_F$ (see Fig. 2). Then, the resulting momentum of the Cooper pair will be $k_1 + k_2 = 2\delta k_F \neq 0$, which implies that



FIG. 2. Energy band of the 1D superconductor near the Fermi energy. Due to the Zeeman splitting the energy of the electrons with spin orientation along the magnetic field (\uparrow) decreases, dotted line, while the energy of the electrons with the opposite spin orientation (\downarrow) increases, dotted line. The splitting of the Fermi momenta is $\pm \delta k_F$, where $\delta k_F = \mu_B H/v_F$. The Cooper pair comprises one electron with spin (\uparrow) and momentum $k_F + \delta k_F$, and another electron with spin (\downarrow) and momentum $-k_F + \delta k_F$. The resulting momentum of the Cooper pair is nonzero: $k_F + \delta k_F + (-k_F + \delta k_F) = 2 \delta k_F \neq 0$.

the space modulation of the superconducting order parameter has a resulting wave vector $2\delta k_F$. Such an explanation presents how a nonuniform superconducting state forms in the presence of the field acting on electron spins, and, at the same time, demonstrates the absence of a paramagnetic limit (at $T \rightarrow 0$) for the 1D superconductor (Buzdin and Polonskii, 1987). For 3D (Larkin and Ovchinnikov, 1964; Fulde and Ferrell, 1964) or 2D (Bulaevskii, 1973) superconductors, it is not possible to choose the single wave vector δk_F which compensates the Zeeman splitting for all electrons on the Fermi surface, as δk_F depends on the direction of v_F , and the paramagnetic limit is preserved. However, the critical field for a nonuniform state at T=0 is always higher than a uniform one. When $T \ge \mu_B H$, at finite temperature, the smearing of the electron distribution function near the Fermi energy decreases the difference of energies between nonuniform and uniform states. From microscopical calculations, at $T > T^* = 0.56T_c$ the uniform superconducting phase is always favored (Saint-James et al., 1969).

D. Generalized Ginzburg-Landau functional

Qualitatively, the FFLO phase formation and the proximity effect in S/F systems may be described in the framework of the generalized Ginzburg-Landau expansion. Let us first recall the standard Ginzburg-Landau functional (see, for example, de Gennes, 1966a),

$$F = a|\psi|^{2} + \gamma |\vec{\nabla}\psi|^{2} + \frac{b}{2}|\psi|^{4}, \qquad (7)$$

where ψ is the superconducting order parameter and the coefficient *a* vanishes at the transition temperature T_c .

At $T < T_c$, the coefficient *a* is negative and the minimum of F in Eq. (7) occurs for a uniform superconducting state with $|\psi|^2 = -a/b$. If we also consider the paramagnetic effect of the magnetic field, all the coefficients in Eq. (7) will depend on the energy of the Zeeman splitting $\mu_B H$, i.e., an exchange field h in the ferromagnet. Note that we neglect the orbital effect, so there is no vector potential \mathbf{A} in Eq. (7). To take into account the orbital effect in the Ginzburg-Landau functional, we may substitute the gradient by its gauge-invariant form $\vec{\nabla} \rightarrow \vec{\nabla} - (2ie/c)\mathbf{A}$. The orbital effect is usually much more important for the superconductivity destruction than the paramagnetic one. This explains why in the standard Ginzburg-Landau theory there is no need to take into account the field and temperature dependence of the coefficients γ and b. However, when the paramagnetic effect becomes predominant, this approximation fails. What are the consequences? If it was simply a renormalization of the coefficients in the Ginzburg-Landau functional, the general superconducting properties of the system would basically be the same. However, qualitatively new physics emerges due to the fact that the coefficient γ changes its sign at the point (H^*, T^*) of the phase diagram; see Fig. 1. A negative sign of γ means that the minimum of the functional does not correspond to a uniform state, and a spatial variation of the order parameter decreases the energy of the system. To de-

scribe such a situation it is necessary to add a higherorder derivative term in the expansion (7), and the generalized Ginzburg-Landau expansion will be r(H,T)

$$F_{G} = a(H,T)|\psi|^{2} + \gamma(H,T)|\vec{\nabla}\psi|^{2} + \frac{\eta(H,T)}{2}|\vec{\nabla}^{2}\psi|^{2} + \frac{b(H,T)}{2}|\psi|^{4}.$$
(8)

The critical temperature of the second-order phase transition into a superconducting state can be found from solving the linear equation for the superconducting order parameter:

$$a\psi - \gamma\Delta\psi + \frac{\eta}{2}\Delta^2\psi = 0.$$
⁽⁹⁾

If we seek a nonuniform solution $\psi = \psi_0 \exp(i\mathbf{q} \cdot \mathbf{r})$, the corresponding critical temperature depends on the wave vector \mathbf{q} and is given by

$$a = -\gamma q^2 - \frac{\eta}{2} q^4. \tag{10}$$

Note that the coefficient *a* is given by $a = \alpha [T - T_{cu}(H)]$, where $T_{cu}(H)$ is the critical temperature of the transition into the uniform superconducting state. The gradient term in the Ginzburg-Landau functional is usually positive $\gamma > 0$, and the highest transition temperature occurs at $T_{cu}(H)$; this is realized for the uniform state with q=0. However, in the case $\gamma < 0$, 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 nonuniform FFLO state $T_{ci}(H)$ with the coefficient *a* given by

$$a = \alpha (T_{ci} - T_{cu}) = \frac{\gamma^2}{2\eta}.$$
(11)

This temperature is higher than the critical temperature T_{cu} of the uniform state. Therefore, the appearance of an FFLO state may simply be interpreted as a sign change of the gradient term in the Ginzburg-Landau functional. A more detailed analysis of the FFLO state using the generalized Ginzburg-Landau functional shows that it is not an exponential but a onedimensional sinusoidal modulation of the order parameter which gives the minimum energy (Buzdin and Kachkachi, 1997; Houzet et al., 1999). In fact, the generalized Ginzburg-Landau functional describes a new type of superconductor with very different properties, and the theory of superconductivity must be redone on the basis of this functional. The orbital effect in the generalized Ginzburg-Landau functional may be introduced with the usual gauge-invariant procedure $\vec{\nabla} \rightarrow \vec{\nabla}$ $-(2ie/c)\mathbf{A}$. The resulting expression for the superconducting current is different from the usual one and the critical field corresponds to higher Landau level solutions as well as new types of vortex lattices may exist (Houzet and Buzdin, 2000, 2001).

III. PROXIMITY EFFECT IN FERROMAGNETS

A. Some generalities about superconducting proximity effect

The contact of materials with different long-range ordering modifies their properties near the interface. In the case of a superconductor-normal-metal interface, the Cooper pairs can penetrate the normal metal at some distance. If the electron's motion is diffusive, this distance is proportional to the thermal diffusion length scale $L_T \sim \sqrt{D/T}$, where D is the diffusion constant. In the case of a pure normal metal the corresponding characteristic distance is $\xi_T \sim v_F/T$. Therefore, superconductinglike properties may be induced in the normal metal, and this phenomenon is called the proximity effect. Simultaneously the leakage of the Cooper pairs weakens the superconductivity near the interface with a normal metal. This effect is called the inverse proximity effect, and results in a decrease of the superconducting transition temperature in a thin superconducting layer in contact with a normal metal. If the thickness of a superconducting layer is smaller than a critical one, the proximity effect totally suppresses the superconducting transition. All these phenomena and earlier experimental and theoretical works on the proximity effect were reviewed by Deutscher and de Gennes (1969).

Note that the proximity effect is a rather general phenomenon not limited by the superconducting phase transition. For example, in the case of surface magnetism (White and Geballe, 1979) the critical temperature at the surface can be higher than the bulk one. As a result the magnetic transition at the surface induces the magnetization nearby. On the other hand, the volume significantly affects the surface transition characteristics.

However, a unique characteristic of the superconducting proximity effect is the Andreev reflection revealed at the microscopical level. Andreev (1964) demonstrated how single-electron states of the normal metal are converted into Cooper pairs and also explained the transformation at the interface of the dissipative electrical current into the dissipationless supercurrent. An electron with an energy below the superconducting gap is reflected at the interface as a hole. The corresponding charge 2e is transferred to the Cooper pair which appears on the superconducting side of the interface. The manifestation of this double charge transfer is that for perfect contact the subgap conductance appears to be twice the normal-state conductance. The classical work by Blonder, Tinkham, and Klapwijk (1982) gives the detailed theory of this phenomenon.

Andreev reflection plays a primary role for the understanding of quantum transport properties of superconductor-normal-metal systems. The interplay between Andreev reflection and the proximity effect was reviewed by Pannetier and Courtois (2000). The reader can find a detailed description of the Andreev reflection in normal-metal-superconductor junctions using scattering theory in the review by Beenakker (1997). A recent review by Deutscher (2005) is devoted to the Andreev reflection spectroscopy of the superconductors.

B. Damped oscillatory dependence of the Cooper pair wave function in ferromagnets

The physics of the oscillating Cooper pair wave function in a ferromagnet is similar to the physics of the superconducting order-parameter modulation in the FFLO state-see Sec. II.C. A qualitative picture of this effect has been provided by Demler, Arnold, and Beasley (1997). When a superconductor is in a contact with a normal metal, the Cooper pairs penetrate across the interface at some distance inside the metal. A Cooper pair in a superconductor consists of two electrons with opposite spins and momenta. In a ferromagnet the up-spin electron, defined the spin orientation along the exchange field, decreases its energy by h, while the downspin electron energy increases by the same value. To compensate this energy variation, the up-spin electron increases its kinetic energy, while the down-spin electron decreases its kinetic energy. As a result the Cooper pair acquires a center-of-mass momentum $2\delta k_F = 2h/v_F$, which implies the modulation of the order parameter with period $\pi v_F/h$. The direction of the modulation wave vector must be perpendicular to the interface, because only this orientation provides for a uniform order parameter in the superconductor.

To get an idea of the proximity effect in S/F structures, we may also start from a description based on the generalized Ginzburg-Landau functional (8). Such an approach is adequate for a small wave-vector modulation case, i.e., in the vicinity of the (H^*, T^*) point of the



FIG. 3. Schematic behavior of the superconducting order parameter near the (a) superconductor-normal metal and (b) superconductor-ferromagnet interfaces. The continuity of the order parameter at the interface implies the absence of the potential barrier. In the general case at the interface the jump of the superconducting order parameter occurs.

(H,T) phase diagram; otherwise, the microscopical theory must be used. This description corresponds to a very weak ferromagnet with an extremely small exchange field $h \approx \mu_B H^* = 1.05T_c$, which is nonrealistic since $h \gg T_c$. However, we discuss this case to get a preliminary understanding of the phenomenon. We address the question of the proximity effect for a weak ferromagnet described by the generalized Ginzburg-Landau functional (8). 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 (see Fig. 3).

The induced superconductivity is weak and, to deal with it, we use the linearized equation for the order parameter (9), which for our geometry is

$$a\psi - \gamma \frac{\partial^2 \psi}{\partial x^2} + \frac{\eta}{2} \frac{\partial^4 \psi}{\partial x^4} = 0.$$
 (12)

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

$$k_1^2 = \frac{|\gamma|}{2\eta} \left(\sqrt{1 + \frac{T - T_{ci}}{T_{ci} - T_{cu}}} - 1 \right), \tag{13}$$

$$k_2^2 = \frac{|\gamma|}{2\eta} \left(1 + \sqrt{1 + \frac{T - T_{ci}}{T_{ci} - T_{cu}}} \right).$$
(14)

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,

$$\psi(x) = \psi_i \exp(-k_1 x) \cos(k_2 x), \tag{15}$$

where the choice of the root for k is the condition $k_1 > 0$. The decay of the order parameter is then accompanied by its oscillation [Fig. 3(b)], which is a characteristic of the proximity effect for the system considered. Approaching the critical temperature T_{ci} the decaying wave vector vanishes $k_1 \rightarrow 0$, while the oscillating wave vector k_2 approaches the FFLO wave vector, $k_2 \rightarrow \sqrt{|\gamma|/\eta}$, and a FFLO phase emerges. Let us compare this behavior with the standard proximity effect (Deutscher and de Gennes, 1969) described by the linearized Ginzburg-Landau equation for the order parameter,

$$a\psi - \gamma \frac{\partial^2 \psi}{\partial x^2} = 0, \qquad (16)$$

with $\gamma > 0$. Here T_c simply coincides with T_{cu} , and the decaying solution is $\psi = \psi_0 \exp[-x/\xi(T)]$, where the coherence length $\xi(T) = \sqrt{\gamma/a}$ [Fig. 3(a)]. This simple analysis provides evidence for the appearance of order parameter oscillations in the presence of an exchange field. This is a fundamental difference between the proximity effect in S/F and S/N systems, and it is at the origin of peculiar characteristics of S/F heterostructures.

In real ferromagnets, the exchange field is large compared with the superconducting temperature and energy scales, and as such the gradients of the superconducting order-parameter variations are also large, and cannot be treated with the generalized Ginzburg-Landau functional. To describe the relevant experimental situation we need to use a microscopical approach. The most convenient scheme (see Appendix) the is the Bogoliubov-de Gennes equations or the Green's functions using the quasiclassical Eilenberger (1968) or Usadel (1970) equations.

In S/F systems if the electron-scattering mean free path *l* is small, the most natural approach is to use the Usadel equations for the Green's functions averaged over the Fermi surface (see the Appendix). Linearized over the pair potential $\Delta(x)$, the Usadel equation for the anomalous function $F(x, \omega)$ depending only on one coordinate *x* is

$$\left(|\omega| + ih \operatorname{sgn}(\omega) - \frac{D}{2} \frac{\partial^2}{\partial x^2}\right) F(x, \omega) = \Delta(x), \quad (17)$$

where $\omega = (2n+1)\pi T$ are the Matsubara frequencies, and $D = \frac{1}{3}v_F l$ is the diffusion coefficient. In the F region, we may neglect the Matsubara frequencies compared to the large exchange field $h \gg T_c$ (the pairing potential Δ is absent and we assume that the BCS coupling constant λ is zero there). This results in a very simple form of the Usadel equation for the anomalous function F_f in the ferromagnet

TABLE I. Characteristic length scales of S/F proximity effect.

Thermal diffusion length
$$L_T$$

Superconducting coherence length
$$\xi_s$$

Superconducting correlation decay length ξ_{1f} in a ferromagnet

Superconducting correlation oscillating length ξ_{2f} in a ferromagnet

$$\frac{v_{Fs}}{2\pi T}$$
 in pure limit

 $\sqrt{\frac{D}{2}}$

$$\sqrt{\frac{D_s}{2\pi T_c}}$$
 in dirty limit

$$\frac{v_{Ff}}{2\pi T}$$
 in pure limit
 $\xi_f = \sqrt{\frac{D_f}{h}}$ in dirty limit

$$\frac{v_{Ff}}{2h} \text{ in pure limit}$$
$$\xi_f = \sqrt{\frac{D_f}{h}} \text{ in dirty limit}$$

$$ih \operatorname{sgn}(\omega)F_f - \frac{D_f}{2}\frac{\partial^2 F_f}{\partial x^2} = 0,$$
 (18)

where D_f is the diffusion coefficient in the ferromagnet. For the geometry in Fig. 3 and $\omega > 0$, the decaying solution for F_f is

$$F_f(x,\omega > 0) = A \, \exp\left(-\frac{i+1}{\xi_f}x\right),\tag{19}$$

where $\xi_f = \sqrt{D_f}/h$ is the characteristic length of the superconducting correlation decay (with oscillations) in the F layer (see Table I). As a result of the condition $h \gg T_c$, this length is much smaller than the superconducting coherence length $\xi_s = \sqrt{D_s/2\pi T_c}$, i.e., $\xi_f \ll \xi_s$. The constant A is determined by the boundary conditions at the S/F interface. For example, in the case of a low resistivity of a ferromagnet, at first approximation the anomalous function in a superconductor F_s is independent of the coordinate and similar to the absence of the ferromagnet, i.e., $F_s = \Delta / \sqrt{\Delta^2 + \omega^2}$. If, in addition, the interface is transparent then the continuity of the function F at the F/S boundary gives $A = \Delta / \sqrt{\Delta^2 + \omega^2}$. For $\omega < 0$, we have $F_f(x, \omega < 0) = F_f^*(x, \omega > 0)$. In a ferromagnet, the role of the Cooper pair wave function is played by Ψ then decays as

$$\Psi \sim \sum_{\omega} F(x,\omega) \sim \Delta \exp\left(-\frac{x}{\xi_f}\right) \cos\left(\frac{x}{\xi_f}\right).$$
 (20)

We retrieve the damping oscillatory behavior of the order parameter (15); see Fig. 3(b). The conclusion we ob-

941

tain from the microscopic approach is that in the dirty limit the scale for the oscillation and decay of the Cooper pair wave function in a ferromagnet is the same.

In the case of a clean ferromagnet the damped oscillatory behavior of the Cooper pair wave function remains, although at zero temperature the damping is nonexponential and much weaker $(\sim 1/x)$. Indeed, the decaying solution of the Eilenberger equation in the clean limit (see the Appendix, Sec. 2) is

$$f(x,\theta,\omega) \sim \exp\left(-\frac{2(\omega+ih)x}{v_{Ff}\cos\theta}\right),$$
 (21)

where θ is the angle between the *x* axis and the Fermi velocity in a ferromagnet, and v_{Ff} is its modulus. After averaging over the angle θ and summing over the Matsubara frequencies ω we obtain

$$\Psi \sim \sum_{\omega} \int_{0}^{\pi} f(x,\theta,\omega) \sin \theta \, d\theta \sim \frac{1}{x} \exp\left(-\frac{x}{\xi_{1f}}\right) \sin\left(\frac{x}{\xi_{2f}}\right).$$
(22)

Here the decay length is $\xi_{1f} = v_{Ff}/2\pi T$ and the oscillating length is $\xi_{2f} = v_{Ff}/2h$ (see Table I). At low temperature $\xi_{1f} \rightarrow 0$ and the Cooper pair wave function decays slowly $\sim (1/x)\sin(x/\xi_{2f})$. An important difference with the proximity effect in the normal metal is the presence of shortranged oscillations of the order parameter with the temperature-independent period $2\pi\xi_{2f}$. In contrast with the dirty limit in a clean ferromagnet the characteristic lengths of the superconducting correlations' decay and oscillations are not the same. Halterman and Valls (2001)performed studies of the ferromagnetsuperconductor interfaces using the self-consistent numerical solution of microscopical Bogoliubov-de Gennes equations. They clearly observed the damped oscillatory behavior of the Cooper pair wave function of the type $\Psi \sim (1/x) \sin(x/\xi_{2f})$.

We conclude that at low temperatures the proximity effect in clean ferromagnet metals is long ranged. On the other hand, in the dirty limit the use of the Usadel equations gives an exponential decay of Ψ . This is due to the fact that the Usadel equations are obtained by averaging over the impurities configurations. Zyuzin et al. (2003) pointed out that at distances $x \ge \xi_f$ the anomalous Green's function F as well as the Cooper pair wave function has a random sample-specific sign, while the modulus does not decay exponentially. This leads to the survival of the proximity effect in the dirty ferromagnet limit at distances $x \ge \xi_f$. The use of the Usadel equations at such distances may be misleading. However, from a practical point of view the range of interest is $x < 5\xi_{f}$, because at larger distances it is difficult to observe the oscillating phenomena in experiment. In this range the use of the Usadel equation is adequate.

The characteristic length of the induced superconductivity variation in a ferromagnet is small compared with the superconducting length, and this implies the use of the microscopic theory of the superconductivity to describe the proximity effect in S/F structures. In this context, the calculations of the free energy of S/F structures in the framework of the standard Ginzburg-Landau functional (Ryazanov, Oboznov, Rusanov, *et al.*, 2001; Ryazanov, Oboznov, Veretennikov, Rusanov, *et al.*, 2001) cannot be justified. Indeed, neglecting higher gradient terms in the Ginzburg-Landau functional implies that the order parameter must be larger than the correlation length. In the ferromagnet the correlation length is ξ_f = $\sqrt{D_f/h}$ in the dirty limit and $\xi_f^0 = v_{Ff}/h$ in the clean limit. We see that the characteristic lengths of the orderparameter variation in a ferromagnet are similar. Therefore, higher gradient terms in the Ginzburg-Landau functional will be of the same order of magnitude as terms with the first derivative.

C. Density-of-states oscillations

Superconductivity creates a gap in the electronic density of states (DOS) near the Fermi energy E_F , i.e., the DOS is zero for energy E in the interval $E_F - \Delta < E$ $< E_F + \Delta$. It is natural that the induced superconductivity in S/N structures decreases DOS at E_F near the interface. Detailed experimental studies of this phenomenon have been performed by Moussy *et al.* (2001). The damped oscillatory dependence of the Cooper pair wave function in a ferromagnet suggests that similar behavior may be expected for the DOS variation due to the proximity effect. Indeed, the DOS $N(\varepsilon)$, where $\varepsilon = E - E_F$ is the energy calculated from the Fermi energy, is directly related to the normal Green's function in the ferromagnet $G_f(x, \omega)$ (Abrikosov *et al.*, 1975),

$$N_{f}(\varepsilon) = N(0) \operatorname{Re} G_{f}(x, \omega \to i\varepsilon),$$
 (23)

where N(0) is the DOS of the ferromagnetic metal. In the dirty limit taking the relationship between the normal and anomalous Green's functions $G_f^2 + F_f^2 = 1$ (Usadel, 1970) into account, and using F_f $=[(\Delta/\sqrt{\Delta^2 + \omega^2})]\exp\{-[(i+1)/\xi_f]x\}$, we obtain directly the DOS at the Fermi energy ($\varepsilon = 0$) in a ferromagnet (Buzdin, 2000) at the distance $x \ge \xi_f$,

$$N_f(\varepsilon = 0) \approx N(0) \left[1 - \frac{1}{2} \exp\left(-\frac{2x}{\xi_f}\right) \cos\left(\frac{2x}{\xi_f}\right) \right].$$
(24)

This simple calculation implies $\Delta \ll T_c$. At certain distances the DOS at the Fermi energy may be higher than in the absence of superconductor. This is in contradiction with the proximity effect in S/N systems. Such behavior has been observed experimentally by Kontos *et al.* (2001) in DOS measurements with planar-tunneling spectroscopy in Al/Al₂O₃/PdNi/Nb junctions; see Fig. 4.

For a PdNi layer thickness of 50 Å the $\cos(2x/\xi_f)$ term in Eq. (24) is positive and the DOS decrease inside the gap is due to the proximity effect. However, for a PdNi layer thickness of 75 Å the $\cos(2x/\xi_f)$ term changes sign and the DOS becomes larger than its normal effect value. Such inversion of the DOS permits us to estimate ξ_f for the PdNi alloy used by Kontos *et al.* (2001) as 60 Å.



FIG. 4. Measurements of the differential conductance by Kontos *et al.* (2001) for two Al/Al₂O₃/PdNi/Nb junctions with two different thicknesses (50 and 75 Å) of the ferromagnetic PdNi layer. A 1500-Å-thick aluminum layer was evaporated on SiO and then quickly oxidized to produce a Al₂O₃ tunnel barrier. Tunnel junction areas were defined by evaporating 500 Å of SiO through masks. A PdNi thin layer was deposited and then backed by a Nb layer.

Currently, there exists only one experimental work on the DOS in S/F systems, while several theoretical papers treat this subject in more detail. In a series of papers Halterman and Valls (2001, 2002, 2003) performed extensive theoretical studies of the local DOS behavior in S/F systems in a clean limit using the self-consistent Bogoliubov–de Gennes approach. They calculated the DOS spectra in both S and F regions and took into account the Fermi wave-vector mismatch, interfacial barrier, and sample size.

Fazio and Lucheroni (1999) performed numerical selfconsistent calculations of the local DOS in an S/F system using the Usadel equation. Impurity scattering on DOS oscillations has been studied by Baladié and Buzdin (2001) and Bergeret et al. (2002). One concludes that oscillations disappear in the clean limit. Here the calculations of the DOS oscillations made in the ballistic regime for the ferromagnetic film on the top of the superconductor (Zareyan et al., 2001, 2002) depend essentially on the boundary conditions at the ferromagnet-vacuum interface. Sun et al. (2002) used the quasiclassical version of the Bogoliubov-de Gennes equations for numerically calculating the DOS in the S/F system with a semiinfinite ferromagnet. They obtained in the clean limit oscillations of the DOS and presented a quantitative fit of the experimental data of Kontos et al. (2001). Astonishingly, in another quasiclassical approach using Eilenberger equations oscillations of the DOS are absent for an infinite electron mean free path (Baladié and Buzdin, 2001; Bergeret et al., 2002).

DOS oscillations in ferromagnets suggest similar oscillatory behavior of the local magnetic moment of the electrons. The corresponding magnetic moment induced by the proximity effect may be written as

$$\delta M = i\mu_B N(0)\pi T \sum_{\omega} \left[G_f(x,\omega,h) - G_f(x,\omega,-h) \right].$$
(25)

Assuming the low resistivity of a ferromagnet in the dirty limit at temperatures near T_c , the magnetic moment is

$$\delta M = -\mu_B N(0) \pi \frac{\Delta^2}{2T_c} \exp\left(-\frac{2x}{\xi_f}\right) \sin\left(\frac{2x}{\xi_f}\right).$$
(26)

Note that the total electron's magnetic moment in a ferromagnet is

$$M = \delta M + \mu_B N(0)h. \tag{27}$$

The local magnetic moment oscillates as does the DOS, and in some regions M may be higher than in the absence of superconductivity. The proximity effect also induces the local magnetic moment in a superconductor near the S/F interface at a distance of the order of superconducting coherence length ξ_s .

Proximity induced magnetism was studied using the Usadel equations by Krivoruchko and Koshina (2002) and Bergeret *et al.* (2004a, 2004b). Numerical calculations of Krivoruchko and Koshina (2002) revealed the damped oscillatory behavior of the local magnetic moment in a superconductor at the ξ_s scale with positive magnetization at the interface. On the other hand, Bergeret *et al.* (2004a) argued that the induced magnetic moment in a superconductor must be negative. This is related to the Cooper pairs located in space that one electron of the pair is in the superconductor, while the other is in the ferromagnet. The direction along the magnetic moment is favored for the electron in the ferromagnet which makes the spin of the other electron in the superconductor antiparallel.

Microscopic calculations of the local magnetic moment in the pure limit using the Bogoliubov-de Gennes equations (Halterman and Valls, 2004a) also revealed the damped oscillatory behavior of the local magnetic moment but at the atomic length scale. In the quasiclassical approach oscillations of the local magnetic moments disappear in the clean limit, similar to the case of DOS oscillations. The magnitude of the proximity induced magnetic moment is very small, and at the present time there is no evidence in experiments.

D. Andreev reflection at the S/F interface

Spin effects play an important role in the Andreev reflection at the S/F interface. An incident spin-up electron in a ferromagnet is reflected by the interface as a spin-down hole, and as a result a Cooper pair of electrons with opposite spins appears in a superconductor. Therefore both the spin-up and spin-down bands of electrons in a ferromagnet are involved in this process. de Jong and Beenakker (1995) were the first to demonstrate the major influence of spin polarization in a ferromagnet on the subgap conductance of the S/F interface. In fully spin-polarized metals all carriers have the same spin and Andreev reflection is totally suppressed. In general, with an increase of the spin polarization the subgap conductance drops from twice the normal-state conductance value to a small value for highly polarized metals. Following de Jong and Beenakker (1995), consider a simple intuitive picture of the conductance through a ballistic S/F point contact. Using scattering channels which are subbands which cross the Fermi level, the conductance at T=0 of a ferromagnet–normal-metal contact is given by the Landauer formula

$$G_{FN} = \frac{e^2}{h}N.$$
(28)

The total number of scattering channels N is the sum of the spin-up N_{\uparrow} and spin-down N_{\downarrow} channels $N=N_{\uparrow}$ $+N_{\downarrow}$, and spin polarization gives $N_{\uparrow}>N_{\downarrow}$. For the case of the superconductor in contact with the nonpolarized metal all electrons are reflected as holes, which doubles the number of scattering channels and the conductance itself. For the spin-polarized metal where $N_{\uparrow}>N_{\downarrow}$, all spin-down electrons are reflected as spin-up holes. However, only $N_{\downarrow}/N_{\uparrow}<1$ of spin-up electrons can be Andreev reflected. The subgap conductance of the S/F contact is then

$$G_{FS} = \frac{e^2}{h} \left(2N_{\downarrow} + 2N_{\uparrow} \frac{N_{\downarrow}}{N_{\uparrow}} \right) = 4 \frac{e^2}{h} N_{\downarrow}.$$
⁽²⁹⁾

Comparing this expression with Eq. (28) we see that $G_{FS}/G_{FN}=4N_{\uparrow}/(N_{\downarrow}+N_{\uparrow})<2$ and $G_{FS}=0$ for the full-polarized ferromagnet with $N_{\downarrow}=0$. If spin polarization is defined as $P=(N_{\uparrow}-N_{\downarrow})/(N_{\downarrow}+N_{\uparrow})$, then the suppression of the normalized zero-bias conductance gives the value of *P*:

$$\frac{G_{FS}}{G_{FN}} = 2(1-P).$$
 (30)

Subsequent experimental measurements of spin polarization with Andreev reflection (Soulen *et al.*, 1998; Upadhyay *et al.*, 1998) fully confirmed the efficiency of this method to probe ferromagnets. Andreev point-contact spectroscopy permits one to measure the spin polarization in a much wider range of materials (Zutic, Fabian, and Das Sarma, 2004) compared with spin-polarized electron tunneling (Meservey and Tedrow, 1994).

However, the interpretation of the Andreev reflection data on the conductance of the S/F interfaces and comparison of the spin polarization with tunneling data may be complicated by band-structure effects (Mazin, 1999). Zutic and Das Sarma (1999) and Zutic and Valls (1999, 2000) generalized the theoretical analysis of Blonder, Tinkham, and Klapwijk (1982) to the case of the S/F interface. An interesting result is that, in the absence of the potential barrier at the S/F interface, the spin polarization increases the subgap conductance. The condition of perfect transparency of the interface is $v_{F\uparrow}v_{F\downarrow}=v_s^2$, where $v_{F\uparrow}$ and $v_{F\downarrow}$ are the Fermi velocities for two spin polarizations in a ferromagnet, and v_s is the Fermi velocity in a superconductor. Vodopyanov and Tagirov (2003a) proposed a quasiclassical theory of Andreev reflection in F/S nanocontacts and analyzed the spin polarization calculated from conductance and tunneling measurements.

Note that a high spin polarization was measured in CrO_2 films P=90% and in $La_{0.7}Sr_{0.3}MnO_3$ films P=78% (Soulen *et al.*, 1998). The spin-polarized tunneling data for these systems are lacking.

Another interesting effect related to crossed Andreev reflection was predicted by Deutsher and Feinberg (2000) [see also Yamashita, Takahashi, and Maekawa (2003), Yamashita *et al.* (2003), and Deutsher (2004)]. The electric current between two ferromagnetic leads attached to the superconductor strongly depends on the relative orientation of the magnetization in these leads. If we assume that the leads are fully polarized, then the electron coming from one lead cannot experience Andreev reflection in the same lead. However, this reflection is possible in the second lead, provided its polarization is opposite, and the distance between the leads is smaller than the superconducting coherence length. The magnetic resistance between leads will be high for parallel orientation and low for antiparallel orientation.

IV. OSCILLATORY SUPERCONDUCTING TRANSITION TEMPERATURE IN S/F MULTILAYERS AND BILAYERS

A. First experimental evidence of the anomalous proximity effect in S/F systems

The damped oscillatory behavior of the superconducting order parameter in ferromagnets may produce commensurability effects between the period of the orderparameter oscillation ($\sim \xi_f$) and the thickness of a F layer. This results in a nonmonotonic superconducting transition temperature dependence on the F layer thickness in S/F multilayers. Indeed, for a F layer thickness smaller than ξ_f , the pair wave function in the F layer changes 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 absent and we call this state the "0" phase. On the other hand, if the F layer thickness becomes $\sim \xi_f$, the pair wave function may cross zero at the center of the F layer with an opposite sign or π shift of the phase of the superconducting order parameter in the adjacent S layers, which we call the " π " phase. An increase of the F layer thickness may provoke subsequent transitions from 0 to π phases, which superpose on the commensurability effect and result in a nonmonotonic dependence of the critical temperature on the F layer thickness. For the S/F bilayers, the transitions between 0 and π phases are impossible; the commensurability effect between ξ_f and F layer thickness nevertheless leads to a nonmonotonic dependence of T_c on the F layer thickness.

The predicted oscillatory-type dependence of the critical temperature (Buzdin and Kuprianov, 1990; Radovic *et al.*, 1991) was subsequently observed experimentally in Nb/Gd (Jiang *et al.*, 1995), Nb/CuMn (Mercaldo *et al.*, 1996), and Nb/Co and V/Co (Obi *et al.*, 1999)

multilayers, as well as in bilayers Nb/Ni (Sidorenko et al., 2003), trilayers Fe/V/Fe (Garifullin et al., 2002), Fe/Nb/Fe (Mühge et al., 1996), Nb/[Fe/Cu] layers (Vélez et al., 1999), and Fe/Pb/Fe (Lazar et al., 2000).

The strong pair-breaking influence of the ferromagnet and the nanoscopic range of oscillation period complicate the observation of this effect. Advances in thin-film processing techniques were crucial for the study of this phenomenon. The first indications on the nonmonotonic variation of T_c versus the F layer thickness was obtained by Wong et al. (1986) for V/Fe superlattices. However, in subsequent experiments of Koorevaar et al. (1994), no oscillatory behavior of T_c was found, while recent studies by Garifullin et al. (2002) of the superconducting properties of Fe/V/Fe trilayers revealed the reentrant T_c behavior as a function of the F layer thickness. Bourgeois and Dynes (2002) studied amorphous Pb/Ni bilayer quench-condensed films and observed only a monotonic depairing effect with an increase of the Ni layer thickness. In the work of Sidorenko et al. (2003), a comparative analysis of sample preparations was made and concluded that the molecular beam epitaxy grown samples do not reveal T_c oscillations, whereas magnetron sputtered samples do. This difference is attributed to the appearance of a magnetically "dead" interdiffused layer at the S/F interface which plays an important role for the molecular beam epitaxy grown samples. The transition-metal ferromagnets, such as Fe, have a strongly itinerant character of the magnetic moment which is very sensitive to the local coordination. In thin Fe layers, the magnetism may strongly decrease and even vanish. The best choice is to use the rare-earth ferromagnetic metal with localized magnetic moments. This has been done by Jiang *et al.* (1995) who prepared magnetron sputtered Nb/Gd multilayers, which clearly revealed the T_c oscillations; see Fig. 5.

In Fig. 5 the curves show a pronounced nonmonotonic dependence of T_c on the Gd layer thickness. An increase of T_c implies the transition from the 0 phase to the π phase. Note that previous experiments on the molecular beam epitaxy grown Nb/Gd samples (Strunk et al., 1994) revealed a steplike decrease of T_c with increasing Gd layer thickness. A comprehensive analysis of different sample's quality was made by Chien and Reich (1999). Aarts et al. (1997) studied in detail the proximity effect in a system consisting of the superconducting V and ferromagnetic $V_{1-x}Fe_x$ alloys and used interface transparency to understand the pair-breaking mechanism.

B. Theoretical description of the S/F multilayers

To provide a theoretical description of a nonmonotonic dependence of T_c , we consider the S/F multilayered system with a thickness of the F layer $2d_f$ and the S layer $2d_s$; see Fig. 6.

The x axis is chosen perpendicular to the layers with x=0 at the center of the S layer. The 0 phase corresponds to the same superconducting order-parameter



tion of the critical temperature of Nb/Gd multilayers vs thickness of Gd layer d_G for two different thicknesses of Nb layers: (a) $d_{\text{Nb}} = 600$ Å and (b) $d_{\text{Nb}} = 500$ Å. Dashed line in (a) is a fit by the theory of Radovic et al. (1991).



FIG. 6. S/F multilayer. The x axis is chosen perpendicular to the planes of the S and F layers with the thicknesses $2d_s$ and $2d_f$, respectively. (a) The curve $\Psi(x)$ represents schematically the behavior of the Cooper pair wave function in the 0 phase. Due to symmetry the derivative of Ψ (and F) is zero at the centers of the S and F layers. The case of the 0 phase is equivalent to the S/F bilayer with the S and F layer thicknesses d_s and $d_{\rm f}$, respectively. (b) The Cooper pair wave function in the π phase vanishes at the center of the F layers and $\Psi(x)$ is antisymmetric toward the center of the F layer.

sign in all S layers [Fig. 6(a)] while in the π phase the sign of the superconducting order parameter in adjacent S layers is opposite [Fig. 6(b)]. In the case of a S/F bilayer, the anomalous Green's function F(x) has zero derivative at the boundary with vacuum; see Eq. (32) below. Here the function F(x) is in the 0 phase at the centers of the S and F layers. As a result, the superconducting characteristics of a S/F bilayer with thickness d_s and d_f of the S and F layers, respectively, are equivalent to that of the S/F multilayer with double layer thickness ($2d_s$ and $2d_f$).

The approach based on the quasiclassical Eilenberger (1968) or Usadel (1970) equations is very convenient to deal with S/F systems (see the Appendix, Sec. 2). In fact, it is much simpler than the complete microscopical theory, it does not need detailed knowledge of all the characteristics of the S and F metals, and is applicable for scales larger than the atomic one. As such, it must work for thicknesses of the layers in the range 20-200 Å, which is of primary interest for S/F systems.

In the dirty limit, if the electron elastic scattering time $\tau = l/v_f$ is small, more precisely $T_c \tau \ll 1$ and $h\tau \ll 1$, the use of the Usadel equations is justified. The second condition, however, is much more restrictive due to a large value of the exchange field $(h \gg T_c)$. The Usadel equations deal only with the Green's functions $G(x, \omega)$ and $F(x, \omega)$ averaged over the Fermi surface. Moreover, to calculate the critical temperature of the second-order superconducting transition in S/F systems, consider only the limit of the small superconducting order parameter $(\Delta \rightarrow 0)$ in the Usadel equations. This linearization permits us to set $G = \text{sgn}(\omega)$ and in the form linearized over Δ , the Usadel equation for the anomalous function F_s in the S region is written as

$$\left(|\omega| - \frac{D_s}{2}\frac{\partial^2}{\partial x^2}\right)F_s = \Delta(x), \tag{31}$$

where D_s is the diffusion coefficient in the S layer. In the F region, the exchange field is present while the pairing potential Δ is absent, and the corresponding Usadel equation for the anomalous function F_f is Eq. (18).

The equations for F_s and F_f must be supplemented by the boundary conditions. At the superconductor-vacuum interface, the boundary condition is simply a zero derivative of the anomalous Green's function, which implies the absence of the superconducting current through the interface. The general boundary conditions for the Usadel equations at the superconductor-normal metal interface have been derived by Kupriyanov and Lukichev (1988) and near the critical temperature they read

$$\left(\frac{\partial F_s}{\partial x}\right)_{x=0} = \frac{\sigma_f}{\sigma_s} \left(\frac{\partial F_f}{\partial x}\right)_{x=0}$$

$$F_s(0) = F_f(0) - \xi_n \gamma_B \left(\frac{\partial F_f}{\partial x}\right)_{x=0},\tag{32}$$

where $\sigma_t(\sigma_s)$ is the conductivity of the F layer (S layer above T_c). The parameter γ_B characterizes the interface transparency $T=1/(1+\gamma_B)$ and is related to the S/F boundary resistance per unit area R_b via $\gamma_B = R_b \sigma_f / \xi_n$ (Kupriyanov and Lukichev, 1988). In analogy with the superconducting coherence length $\xi_s = \sqrt{D_s/2\pi T_c}$, we introduce the normal-metal coherence length ξ_n $=\sqrt{D_f/2\pi T_c}$. The boundary conditions correspond to the S/F interface x=0 and the positive direction of the x axis chosen along the outer normal to the S surface, i.e., the x axis is directed from the S to the F metal. It is worth noting that the boundary conditions for the Usadel equations (Kupriyanov and Lukichev, 1988) were obtained for superconductor-normal-metal interfaces, and their applicability for S/F interfaces is justified, provided that the exchange field in the ferromagnet is much smaller than the Fermi energy, i.e., $h \ll E_F$. For a ferromagnet with localized moments, such as Gd, this condition is always fulfilled, while it becomes more stringent for transition metals and violated for half metals. Recently, Vodopyanov and Tagirov (2003b) obtained the boundary conditions for the Eilenberger equations in the case of a strong ferromagnet. They used them to study the critical temperature of a S/F bilayer when the ferromagnet is in the clean limit. Nevertheless, the important question about the boundary conditions for Usadel equations at the superconductor-strong ferromagnet interface is still open.

Provided the solutions of Usadel equations in the F and S layers are known, the critical temperature T_c^* may be found from the self-consistency equation for the pair potential $\Delta(x)$ in a superconducting layer,

$$\Delta(x) = \pi T_c^* \lambda \sum_{\omega} F_s(x, \omega), \qquad (33)$$

where λ is BCS coupling constant in the S layer, while in the F layer it is supposed to be equal to zero. This equation is more conveniently written in the following form:

$$\Delta(x)\ln\frac{T_c^*}{T_c} + \pi T_c^* \sum_{\omega} \left(\frac{\Delta(x)}{|\omega|} - F_s(x,\omega)\right) = 0, \qquad (34)$$

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

The Usadel equations provide a good basis for the complete numerical solution of the problem of the transition temperature of S/F superlattices. First, such a solution has been obtained for a S/F system with no interface barrier by Radovic *et al.* (1988, 1991), using the Fourier-transform method, and this case was treated analytically by Buzdin and Kuprianov (1990) and Buzdin *et al.* (1992). The role of the S/F interface transparency has been elucidated by Proshin and Khusainov (1997) [for more references, see also the review by Tagirov (1998) and Izyumov *et al.* (2002)]. Recently, Fominov *et al.* (2002) performed a detailed analysis of the nonmono-

tonic critical temperature dependence of S/F bilayers for arbitrary interface transparency and compared the results of different approximations with exact numerical calculations.

Below we illustrate the appearance of a nonmonotonic superconducting transition temperature dependence for the case of a thin S layer, which has a simple analytical solution. More precisely, we consider the case $d_s \ll \xi_s$, which implies that variations of the superconducting order parameter and anomalous Green's function in the S layer are small. We may write the following expansion up to the x^2 -order term for F_s in the S layer centered at x=0:

$$F_s(x,\omega) = F_0 \left(1 - \frac{\beta_\omega}{2} x^2 \right), \tag{35}$$

where F_0 is the value of the anomalous Green's functions at the center of the S layer, and the linear over x term is absent due to the symmetry of the problem in both 0 and π phases (see Fig. 4). Using form (35) of F_s in the Usadel equation (31), we readily find

$$F_0 = \frac{\Delta}{\omega + \tau_s^{-1}},\tag{36}$$

where we have introduced the complex pair-breaking parameter $\tau_s^{-1} = (D_s/2)\beta_\omega$ and, in the first approximation over $d_s/\xi_s \ll 1$, the pair potential Δ may be considered as spatially independent. The pair-breaking parameter τ_s^{-1} is directly related to the logarithmic derivative of F_s at $x=d_s$,

$$\frac{F'_s(d_s)}{F_s(d_s)} \simeq -d_s \beta_\omega = -\frac{2d_s \tau_s^{-1}}{D_s}.$$
(37)

The boundary conditions (32) permit us to calculate the parameter τ_s^{-1} , provided the anomalous Green's function in the F layer is known:

$$\tau_s^{-1} = -\frac{D_s}{2d_s} \frac{\sigma_f}{\sigma_s} \frac{F_f'(d_s)/F_f(d_s)}{1 - \xi_f \gamma_B F_f'(d_s)/F_f(d_s)}.$$
(38)

C. 0 and π phases

The solution of the Usadel equation (18) in the F layer is straightforward but different for the 0 and π phases. Let us start first with a 0 phase. In such a case [see Fig. 6(a)], we take as a solution for $F_f(x)$ at $\omega > 0$ in the interval $d_s < x < d_s + 2d_f$ the function symmetrical relative to the plane $x=d_s+d_f$, i.e.,

$$F_f(x,\omega>0) = A \cosh\left[\frac{i+1}{\xi_f}(x-d_s-d_f)\right].$$
(39)

Therefore the pair-breaking parameter τ_{s0}^{-1} for the 0 phase at $\omega > 0$ is

$$\tau_{s,0}^{-1}(\omega > 0) = \frac{D_s}{2d_s} \frac{\sigma_f i + 1}{\sigma_s} \frac{\tanh\left(\frac{i+1}{\xi_f}d_f\right)}{1 + \frac{i+1}{\xi_f}\xi_n \gamma_B \tanh\left(\frac{i+1}{\xi_f}d_f\right)},$$
(40)

and does not depend on the Matsubara frequencies ω . For a negative ω we have $\tau_{s,0}^{-1}(\omega < 0) = [\tau_{s,0}^{-1}(\omega > 0)]^*$.

Now, let us address the case of the π phase. The only difference is that we must choose the asymmetrical solution for $F_t(x)$,

$$F_f(x,\omega>0) = B \sinh\left[\frac{i+1}{\xi_f}(x-d_s-d_f)\right],\tag{41}$$

and the corresponding pair-breaking parameter $\tau_{s,\pi}^{-1}$ is given by

$$\tau_{s,\pi}^{-1}(\omega > 0) = \left[\tau_{s,\pi}^{-1}(\omega < 0)\right]^*$$
$$= \frac{D_s}{2d_s} \frac{\sigma_f i + 1}{\sigma_s} \frac{\coth\left(\frac{i+1}{\xi_f}d_f\right)}{1 + \frac{i+1}{\xi_f}\xi_n \gamma_B \coth\left(\frac{i+1}{\xi_f}d_f\right)}.$$
(42)

We see that in all cases the pair-breaking parameter τ_s^{-1} is complex and depends only on the sign of the Matsubara frequency and not its value. As a result, using the self-consistency equation (34), we obtain the following expression for the critical temperature T_c^* of the S/F multilayer:

$$\ln \frac{T_c^*}{T_c} = \Psi\left(\frac{1}{2}\right) - \operatorname{Re} \Psi\left\{\frac{1}{2} + \frac{1}{2\pi T_c^* \tau_s}\right\},\tag{43}$$

where Ψ is the digamma function, and the pair-breaking parameter τ_s^{-1} is given by Eqs. (40) and (42) for the 0 and π phases, respectively. Expression (43) for T_c^* reminds one of the corresponding formula for the critical temperature of a superconductor with magnetic impurities (Abrikosov and Gor'kov, 1960), though the magnetic scattering time τ_s is complex in our system. If the critical temperature variation is small $[(T_c - T_c^*)/T_c \ll 1]$, the critical temperature shift (43) may be simplified,

$$\frac{T_c - T_c^{*}}{T_c} = \frac{\pi}{4T_c} \operatorname{Re}(\tau_s^{-1}).$$
(44)

D. Oscillating critical temperature

To illustrate the oscillatory behavior of the critical temperature, we consider the case of a transparent S/F interface $\gamma_B=0$. The critical temperatures T_c^{*0} and $T_c^{*\pi}$ for the 0 and π phases, respectively, are

$$\frac{T_c - T_c^{*0}}{T_c} = \frac{\pi}{4T_c\tau_0} \left(\frac{\sinh(2y) - \sin(2y)}{\cosh(2y) + \cos(2y)}\right),\tag{45}$$



FIG. 7. The dependence of the critical temperature on the thickness of the F layer for the 0 phase (solid line) and the π phase (dotted line) for the transparent S/F interface. Note that the highest transition temperature T_c^* corresponds to the lowest point. The dimensionless thickness of the F layer $2y = 2d_f/\xi_f$ and the first transition from the 0 to the π phase occurs at $2d_f = 2.36\xi_f$. The parameter is $\tau_0 = (2d_s\xi_f/D_s)\sigma_s/\sigma_f$.

$$\frac{T_c - T_c^{*\pi}}{T_c} = \frac{\pi}{4T_c\tau_0} \left(\frac{\sinh(2y) + \sin(2y)}{\cosh(2y) - \cos(2y)}\right),\tag{46}$$

where $\tau_0^{-1} = (D_s/2d_s\xi_f)\sigma_f/\sigma_s$ and $2y = 2d_f/\xi_f$ is the dimensionless F layer thickness. The critical temperature variation versus the F layer thickness is presented in Fig. 7.

We see that for small F layer thickness, the 0 phase has a higher transition temperature. The first crossing of the curves $T_c^{*0}(y)$ and $T_c^{*\pi}(y)$ occurs at $2y_c \approx 2.36$ and in the thickness range $2.36\xi_f < 2d_f < 5.5\xi_f$, the π phase has a higher critical temperature. The oscillations of the critical temperature rapidly decay with an increase of y, and it is not realistic to observe in experiment more than two periods of oscillations.

In general, the F-layer thickness dependence of the critical temperature (43) may be written for the 0 phase in the following form convenient for numerical calculations:

$$\ln \frac{T_c^{*0}}{T_c} = \Psi\left(\frac{1}{2}\right) - \operatorname{Re} \Psi\left\{\frac{1}{2} + \frac{2T_c}{T_c^{*0}\tilde{\tau}_0}\right\}$$
$$\times \frac{1}{\tilde{\gamma} + \frac{1-i}{2}\operatorname{coth}[(1+i)y]}, \qquad (47)$$

where the dimensionless parameter $\tilde{\tau}_0^{-1} = 1/4\pi T_c \tau_0$ and $\tilde{\gamma} = \gamma_B(\xi_n/\xi_f)$. The corresponding formula for the critical temperature for the π phase is simply obtained from Eq. (47) by the substitution coth \rightarrow tanh.

In Fig. 8, we present examples of the thickness dependence on the critical temperature for S/F multilayers at different interface transparencies.



FIG. 8. The critical temperature of the 0 phase (solid line) and the π phase (dashed line) as a function of the dimensionless thickness of the F layer $2y=2d_f/\xi_f$ for different S/F interface barriers $\tilde{\gamma}=\gamma_B(\xi_n/\xi_f)$. (a) The dimensionless pair-breaking parameter $\tilde{\tau}_0=4\pi T_c(2d_s\xi_f/D_s)\sigma_s/\sigma_f=21$. (b) The dimensionless pair-breaking parameter $\tilde{\tau}_0=20.05$.

The oscillations of the critical temperature are most pronounced for the transparent interface $\tilde{\gamma}=0$, and they rapidly decrease with an increase of the boundary barrier; at $\tilde{\gamma} \gtrsim 2$ the oscillations are hardly observable. Note that, for certain values of the parameters $\tilde{\tau}_0$ and $\tilde{\gamma}$, the $T_c^{*0}(d_f)$ dependence may show the infinite derivative, which indicates a superconducting transition from the second-order to the first-order one. This question was studied in detail by Tollis (2004). The increase of the boundary barrier not only decreases the amplitude of the critical temperature oscillations, but it also decreases the critical thickness of F layer y_c , corresponding to the 0- π phase transition. The limit $\tilde{\gamma} = \gamma_B(\xi_n/\xi_f) \gg 1$ is a rather special one. In such a case the S/F interface barrier becomes a tunnel barrier, and the critical thickness y_c may be much smaller than 1. Indeed, if the critical temperature variation is small (more precisely, if $\tilde{\gamma}\tau_0$ ≥ 1), the condition $\operatorname{Re}(\tau_{s,0}^{-1}) = \operatorname{Re}(\tau_{s,\pi}^{-1})$ is realized at



FIG. 9. Variation of the critical temperature of the Nb/Cu_{0.43}Ni_{0.57} bilayer with the F layer thickness (Ryazanov *et al.*, 2003). The theoretical fit (Fominov *et al.*, 2002) gives an exchange field value $h \sim 130$ K and an interface transparency parameter $\gamma_B \sim 0.3$.

$$d_f^c = \frac{\xi_f}{2} \left(\frac{3}{\tilde{\gamma}}\right)^{1/3},\tag{48}$$

and the $0-\pi$ phase transition is now related to tunneling through the F layer. This result is very different from low interface transparency, when the transition occurs due to the spatial oscillations of the anomalous Green's function. It is difficult to observe the low transparency regime of the $0-\pi$ transition with critical temperature measurements due to the fact that at $\tilde{\gamma} \ge 1$ the $T_c^*(d_f)$ oscillations become very small. On the other hand, measurements of the critical current in S/F/S junctions may be adequate to reveal the $0-\pi$ transition in this regime (see the next section).

It is interesting to note that at small F layer thickness $(d_f < \xi_f)$ the critical temperature decreases with an increase of the interface barrier [provided the condition $\tilde{\gamma}(d_f/\xi_f) < 1$ is fulfilled]; see Fig. 8. Such a counterintiutive behavior may be explained in the following way. The low penetration of the barrier prevents the quick return of the Cooper pair from the thin F layer. Therefore, the Cooper pair stays for a relatively long time in the F layer before going back to the S layer. As a result, the pair-breaking role of the exchange field in the F layer appears to be strongly enhanced.

The cases of S/F bilayers or F/S/F trilayers with parallel magnetization are equivalent to the 0-phase case for multilayers (with double F layers thickness) and the corresponding $T_{c,0}^*(d_f)$ dependence reveals a rather weak nonmonotonic behavior for finite transparency of the S/F interface (see Fig. 8). Comparison of the experimental data of Ryazanov *et al.* (2003) for the critical temperature of the bilayer Nb/Cu_{0.43}Ni_{0.57} vs the thickness of the ferromagnetic layer with the theoretical fit (Fominov *et al.*, 2002) is presented in Fig. 9.

Now let us address the following question: Is it possible to have a transition into a state with the phase difference other than 0 and π ? For example, a state with

phase difference $0 < \varphi_0 < \pi$ is expected at F layer thickness near d_f^c . The numerical calculations of Radovic *et al.* (1991) revealed the presence of an intermediate phase. However, the relative width of its existence near d_f^c was very small-around several percent only. On the other hand, analytical calculations show that for the thin S layer states without current (corresponding to the highest T_c^*) are only possible for the phase difference 0 or π . Also, in S/F/S junctions transitions between 0 and π states are discontinuous-see the discussion in the next section. The narrow region of the φ_0 phase obtained with numerical calculations (Radovic et al., 1991) is simply related with its accuracy $\sim 1\%$, and the width of this region may decrease with an increase of the accuracy. Nevertheless, there is another mechanism of the realization of the φ_0 phase due to the fluctuations of the thickness of the F layer. In such a case near the critical F layer thickness d_f^c regions of the 0 and π phases would coexist. If the characteristic dimensions of these regions are smaller than the Josephson length in the S/F structure, then the average phase difference would be different from 0 and π (Buzdin and Koshelev, 2003).

The quasiclassical Eilenberger and Usadel equations are not adequate for treating strong ferromagnets with $h \sim E_F$ because the period of Green's-function oscillations becomes comparable with the interatomic distance. On the other hand, the approach based on the Bogoliubov-de Gennes equations in the clean limit is universal. Halterman and Valls (2003, 2004a) applied it to study the properties of clean S/F multilayers, at low temperature. They obtained the excitation spectrum through numerical solution of the self-consistent Bogoliubov-de Gennes equations and discussed the influence of the interface barrier and Fermi energy mismatch on the local density of states. Comparing the energy of the 0 and π phases Halterman and Valls confirmed the existence of the phase transitions with an increase of the F layer thickness. It is of interest that the local density of states is quite different in the 0 and π phases, and its measurements could permit us to trace the 0- π transition. In more recent work, Halterman and Valls (2004b) showed that different order-parameter configurations may correspond to the local energy minima in S/F heterostructures.

Calculations of the energy spectrum in the S/F/S system in the 0 and π phases using the Eilenberger equations were performed by Dobrosavljevic-Grujic, Zikic, and Radovic (2000) for *s*-wave and *d*-wave superconductivity (Zikic *et al.*, 1999). The large peaks in the density of states were attributed to the spin-split bound states appearing due to Andreev reflection at the ferromagnetic barrier.

In a previous analysis the spin-orbit and magnetic scattering were ignored. Demler, Arnold, and Beasley (1997) theoretically studied the influence of the spinorbit scattering on the properties of S/F systems and demonstrated that it is quite harmful for the observation of oscillatory effects. A similar effect is produced by magnetic scattering which to some extent is always present in S/F systems due to the nonstoichiometry of the F layers (and it may be rather large when the magnetic alloy is used as the F layer). Calculations of the critical temperature of the S/F multilayers in the presence of magnetic scattering were first performed by Tagirov (1998). In the formalism presented in this section it is very easy to take into account the magnetic diffusion with the spin-flip scattering time τ_m —substitute the exchange field h in the linearized Usadel equation (17) by $h-i \operatorname{sgn}(\omega) \tau_m^{-1}$. This renormalization leads to a decrease of the damping length and an increase of the oscillation period, which makes the $T_c^*(d_f)$ oscillations less pronounced (Tagirov, 1998).

V. SUPERCONDUCTOR-FERROMAGNET-SUPERCONDUCTOR π JUNCTION

A. General characteristics of the π junction

A Josephson junction at equilibrium usually has a zero phase difference φ between two superconductors. The energy *E* of the Josephson junction may be written as (see, for example, de Gennes, 1966a)

$$E = \frac{\Phi_0 I_c}{2\pi c} (1 - \cos\varphi), \tag{49}$$

where I_c is the Josephson critical current, and the current-phase relation is $I_s(\varphi) = (2e/\hbar)\partial E/\partial \varphi = I_c \sin \varphi$. For the standard situation, the constant $I_c > 0$, and the minimum energy of a Josephson junction is achieved at $\varphi=0$. However, as demonstrated in the previous section in S/F multilayers transitions to the π phase may occur. This means that for the Josephson S/F/S junction (with the same thickness of the F layer which corresponds to the π phase in the multilayered system) the equilibrium phase difference would be π , and it is natural to call such a junction the π junction. For the π junction, the constant I_c in Eq. (49) is negative, and the transition from the 0 to the π state results in a sign change of the critical current, though the experimentally measured critical current is always positive and equal to $|I_c|$. The S/F/S junctions reveal the nonmonotonic behavior of the critical current as a function of the F layer thickness. The vanishing of critical current signals the transition from the 0 to the π state.

Negative Josephson coupling was first noted by Kulik (1965), who discussed the spin-flip tunneling through an insulator with magnetic impurities. Bulaevskii *et al.* (1977) presented arguments that under certain conditions such spin-flip tunneling could dominate direct tunneling and lead to a π -junction. Up to now there is no experimental evidence of the π coupling in the Josephson junctions with magnetic impurities. On the other hand, Buzdin *et al.* (1982) showed that in the ballistic S/F/S weak link I_c displays damped oscillations as a function of the F layer thickness and its exchange field. Later, Buzdin and Kuprianov (1991) demonstrated that these oscillations remain in the diffusive regime and the π coupling is an inherent property of the S/F/S junctions.



FIG. 10. Geometry of the S/F/S junction. The thickness of the ferromagnetic layers is $2d_f$ and both S/F interfaces have the same transparencies, given by the coefficient γ_B .

The characteristic thickness of the F layer corresponding to the transition from the 0 to the π phase is $\xi_f = \sqrt{D_f/h}$, and it is rather small (10–50 Å) in typical ferromagnets because of the large value of the exchange field ($h \ge 1000$ K). Experimental verification of the π coupling in S/F/S junction was not easy, due to the control of the F layer thickness. Finally, the first experimental evidence for a π junction was obtained by Ryazanov, Obozhov, Rusanov, *et al.* (2001a) for the Josephson junction with a weakly ferromagnetic interlayer of a Cu_xNi_{1-x} alloy. Such a choice of the F layer permitted us to have a ferromagnet with a relatively weak exchange field ($h \sim 100-500$ K) and therefore a relatively large ξ_f length.

B. Theory of π junctions

The complete quantitative analysis of the S/F/S junctions is rather complicated, because the ferromagnetic layer can modify superconductivity near the S/F interface. In addition, the boundary transparency and electron mean free path, as well as magnetic and spin-orbit scattering, are important parameters affecting the critical current.

To introduce the physics of π coupling, we concentrate on the approach based on the Usadel equation and consider the S/F/S junction with a F layer of thickness $2d_f$ (see Fig. 10) and identical S/F interfaces. In the case of small conductivity of the F layer or small interface transparency $\sigma_f \xi_s / \sigma_s \xi_f \ll \max(1, \gamma_B)$ we use rigid boundary conditions (Golubov *et al.*, 2004) with $F_s(-d_f) = \Delta e^{-i\varphi/2} / \sqrt{\omega^2 + \Delta^2}$ and $F_s(d_f) = \Delta e^{i\varphi/2} / \sqrt{\omega^2 + \Delta^2}$.

The solution of Eq. (18) in a ferromagnet satisfying the corresponding boundary conditions is

$$F(x) = \frac{\Delta}{\sqrt{\omega^2 + \Delta^2}} \left\{ \frac{\cos(\varphi/2)\cosh(kx)}{[\cosh(kd_f) + k\gamma_B\xi_n \sinh(kd_f)]} + \frac{i\sin(\varphi/2)\sinh(kx)}{[\sinh(kd_f) + k\gamma_B\xi_n \cosh(kd_f)]} \right\},$$
(50)

where the complex wave vector $k = \sqrt{2[|\omega| + i \operatorname{sgn}(\omega)h]/D_f}$. This solution describes the F(x) behavior near the critical temperature. Note that, in principle, at arbitrary temperature the boundary conditions are different from Eq. (32); see, for example, Gol-

ubov *et al.* (2004). However, in the limit of low S/F interface transparency ($\gamma_B \ge 1$), when the amplitude of the *F* function in the F layer is small, we use the linearized Usadel equation (18) at all temperatures. The only modification in the boundary conditions (32) is that F_s must be substituted by $F_s/|G_s|$ and γ_B by $\gamma_B/|G_s|$, where the normal Green's function in superconducting electrode is $G_s = \omega/\sqrt{\omega^2 + \Delta^2}$. Taking this renormalization into account in the explicit form of Eq. (50), we use the following formula for the supercurrent:

$$I_{s}(\varphi) = ieN(0)D_{f}\pi TS\sum_{-\infty}^{\infty} \left(F\frac{d}{dx}\tilde{F} - \tilde{F}\frac{d}{dx}F\right),$$
(51)

where $\overline{F}(x,h) = F^*(x,-h)$, *S* is the area of the cross section of the junction, and N(0) is the electron density of state for a one-spin projection. Expression (51) gives the usual sinusoidal current-phase dependence $I_s(\varphi) = I_c \sin(\varphi)$ with the critical current (Buzdin, 2003):

$$I_c = eSN(0)D_f \pi T \sum_{-\infty}^{\infty} \frac{\Delta^2}{\omega^2} \frac{2k/\cosh(2kd_f)}{\tanh(2kd_f)(1 + \Gamma_{\omega}^2 k^2) + 2k\Gamma_{\omega}},$$
(52)

where $\Gamma_{\omega} = \gamma_B \xi_n / |G_s|$. Expression (52) may be generalized to take into account the different interface transparencies $\gamma_{B1}, \gamma_{B2} \ge 1$, one can substitute $\Gamma_{\omega}^2 \rightarrow \gamma_{B1} \gamma_{B2} (\xi_n / |G_s|)^2$ and $2\Gamma_{\omega} \rightarrow (\gamma_{B1} + \gamma_{B2}) \xi_n / |G_s|$. Near T_c and in the case of transparent interface $\gamma_B \rightarrow 0$ (Buzdin and Kuprianov, 1991),

$$I_{c} = eSN(0)D_{f}\frac{\pi\Delta^{2}}{2T_{c}}\left|\operatorname{Re}\left[\frac{k}{\sinh(2kd_{f})}\right]\right|$$
$$= \frac{V_{0}}{R_{n}}4y\left|\frac{\cos(2y)\sinh(2y) + \sin(2y)\cosh(2y)}{\cosh(4y) - \cos(4y)}\right|, \quad (53)$$

where $2y=2d_f/\xi_f$ is the dimensionless F layer thickness, $R_n=2d_f/\sigma_f S$ is the resistance of the junction $[\sigma_f = 2e^2N(0)D_f$ is the conductivity of the F layer], and $V_0 = \pi\Delta^2/4eT_c$.

The dependence $I_c R_n/V_0$ vs 2y is presented in Fig. 11. The first vanishing of the critical current signals the transition from the 0 to the π state. It occurs at $2y_c \approx 2.36$ which is exactly the critical value of the F layer thickness in the S/F multilayer system corresponding to the 0- π -state transition, i.e., to the condition $T_c^{*0} = T_c^{*\pi}$ in Eqs. (45). The theoretical description of the S/F/S junctions with arbitrary interface transparencies near the critical temperature was proposed by Buzdin and Baladié (2003).

At low temperature or low S/F barrier the amplitude of the anomalous Green's function $F_f(x)$ is not small and we need to use the complete (nonlinearized) Usadel equation. In the large F layer thickness limit $d_f \ge \xi_f$ and $\gamma_B = 0$, an analytical solution was obtained by Buzdin and Kuprianov (1991), and the critical current is



FIG. 11. Critical current of the S/F/S Josephson junction near T_c as a function of the dimensionless thickness of the F layer $2y=2d_f/\xi_f$. There are no barriers at the S/F interfaces ($\gamma_B=0$), R_n is the resistance of the junction, and $V_0=\pi\Delta^2/2eT_c$.

$$I_c R_n = 64\sqrt{2} \frac{|\Delta|}{e} \mathcal{F}\left(\frac{|\Delta|}{T}\right) 2y \exp(-2y) \left| \sin\left(2y + \frac{\pi}{4}\right) \right|,$$
(54)

with

$$\mathcal{F}\left(\frac{|\Delta|}{T}\right) = \pi T \sum_{\omega>0}^{\infty} \frac{|\Delta|}{(\Omega+\omega)[\sqrt{2\Omega}+\sqrt{\Omega+\omega}]^2},$$
(55)

where $\Omega = \sqrt{\omega^2 + |\Delta|^2}$, and $\mathcal{F}(|\Delta|/T) \approx (\pi/128) |\Delta|/T_c$ at $T \approx T_c$ while at low temperature $T \ll T_c$, $\mathcal{F}(|\Delta|/T) \approx 0.071$.

Note that in the clean limit $(\tau h \ge 1)$ the thickness dependence of the critical current is very different (Buzdin *et al.*, 1982) and near T_c it is

$$I_c R_n = \frac{\pi \Delta^2}{4e} \frac{|\sin(4hd_f / v_F)|}{4hd_f / v_F},$$
(56)

i.e., the critical current decreases $\sim 1/d_f$ and not exponentially as in the dirty limit case. In general, in the clean limit the S/F proximity effect is not exponential, but a power-law one.

Expression (56) was obtained using the Eilenberger equations. In the case of a strong ferromagnet $h \leq E_F$, the oscillation periods of the Green's functions are on the order of the interatomic distance, and this approach no longer works. Using the Bogoliubov–de Gennes equations, Cayssol and Montambaux (2004) demonstrated that the quasiclassical result (56), where the only relevant parameter for the critical current oscillations is hd_f/v_F , is not applicable for the strong ferromagnets. This is related to an increase in suppression of the Andreev reflection channels with an increase of the exchange energy.

Using the Bogoliubov–de Gennes equations Radovic *et al.* (2003) studied the general case of the ballistic S/F/S junction for a strong exchange field, arbitrary interfacial transparency, and Fermi wave-vector mismatch. The

characteristic feature of such a ballistic junction is the short-period geometrical oscillations of the supercurrent as a function of d_f due to the quasiparticle transmission resonances. In the case of the strong ferromagnet, the period of $0-\pi$ oscillations becomes comparable with the period of geometrical oscillations, and their interplay provides very special $I_c(d_f)$ dependences. Radovic *et al.* (2003) also demonstrated that the current-phase relationship may strongly deviate from the simple sinusoidal one, and studied how it depends on the junction parameters. While the temperature variation of I_c is usually a monotonic decay with increasing temperature, near the critical thickness d_f corresponding to the 0- π transition, a nonmonotonic dependence I_c on temperature was obtained. Radovic et al. (2001) showed that at low temperature the characteristic multimode anharmonicity of the current-phase relation in clean S/F/S junctions implies the coexistence of stable and metastable 0 and π states. As a consequence, the coexistence of integer and halfinteger flixoid configuration of a superconducting quantum interference device (SQUID) was predicted. Note that for strong ferromagnets the details of the electron's energy bands become important for the description of the properties of the S/F/S junction.

The weak link between *d*-wave superconductors may also produce the π shift effect (for a review, see, for example, Van Harlingen, 1995). The situation of the Josephson coupling in a ferromagnetic weak link between *d*-wave superconductors was studied in the clean limit theoretically by Radovic *et al.* (1999).

It is interesting to note that in the limit $kd \ll 1$ (i.e., $d_f \ll \xi_f$) the oscillations of the anomalous function in the F layer are absent, but as it has been noted previously, for low transparency of the barrier $\gamma_B \gg 1$ the critical current can nevertheless change its sign. Indeed, in this limit, the expression for the critical current (52) reads

$$I_{c} = eN(0)D_{f}\pi TS \sum_{-\infty}^{\infty} \frac{2|\Delta|^{2}}{\omega^{2} + |\Delta|^{2}} \frac{1}{\gamma_{B}^{2}\xi_{n}^{2}2d_{f}} \left(\frac{1}{k^{2}} - \frac{2d_{f}^{2}}{3} - \frac{1}{\gamma_{B}\xi_{n}d_{f}k^{4}} \frac{|\omega|}{\sqrt{\omega^{2} + |\Delta|^{2}}}\right).$$
(57)

Usually from experiment, the Curie temperature Θ of the ferromagnet is higher than the superconducting critical temperature T_c . For the RKKY mechanism of ferromagnetic transition $\Theta \sim h^2/E_F$ and as a result the exchange field h appears to be much larger than the superconducting critical temperature T_c . For itinerant ferromagnetism, the exchange field is usually several times higher than the Curie temperature and the limit $h \gg T_c$ also holds. Taking this into account and performing the summation over Matsubara frequencies of the first two terms in the brackets of Eq. (57), we finally obtain (Buzdin, 2003)



FIG. 12. Temperature dependences of the critical thickness $2d_f^c$ of the F layer, corresponding to the crossover from the 0 to the π phase in the limit of very small boundary transparency for different values of the exchange field.

$$I_{c} = \frac{eN(0)SD_{f}\Delta\xi_{f}^{2}}{4\gamma_{B}^{2}d_{f}\xi_{n}^{2}} \Biggl\{ \frac{\Delta}{h} \Biggl[\Psi\Biggl(\frac{1}{2} + i\frac{h}{2\pi T}\Biggr) \\ - \Psi\Biggl(\frac{1}{2} + i\frac{\Delta}{2\pi T}\Biggr) + \text{c.c.} \Biggr] + \frac{2\pi T\Delta\xi_{f}^{2}}{\gamma_{B}\xi_{n}d_{f}} \\ \times \sum_{\omega > 0} \frac{\omega}{(\omega^{2} + \Delta^{2})^{3/2}} - \frac{4\pi}{3} \Biggl(\frac{d_{f}^{2}}{\xi_{f}^{2}}\Biggr) \tanh\Biggl(\frac{\Delta}{2T}\Biggr) \Biggr\}.$$
(58)

We start with an analysis of I_c over the d_f dependence in the limit of very large γ_B (more precisely, when $\gamma_B \gg h/T_c$). In such a case we neglect the term proportional to $1/\gamma_B$ in the brackets of Eq. (58), and then obtain that at $T \rightarrow 0$ the transition into the π phase occurs (I_c changes its sign) at

$$d_f^c \approx \xi_f \sqrt{\frac{2\Delta(0)}{h} \ln\left(\frac{h}{\Delta(0)}\right)}.$$
(59)

The condition $d_f \ll \xi_f$ is satisfied. For low boundary transparencies, the formula obtained by Buzdin and Baladié (2003) near the critical temperature in the limit $T_c/h \rightarrow 0$ also reveals the crossover between the 0 and π phase. On the other hand, no transition into the π phase was obtained in the analysis of the S/F/S system by Golubov *et al.* (2002b), which is related to the approximation for the anomalous function in ferromagnets when only the first term in the gradient expansion has been retained.

It is interesting to note that the critical F layer thickness d_f^c , when the transition from the 0 to π phase occurs, depends on the temperature. The corresponding temperature dependences are presented in Fig. 12 for different values of T_c/h ratios. We see that $d_f^c(T)$ decreases when the temperature decreases. This is a very



FIG. 13. Nonmonotonic temperature dependences of the normalized critical current for the low transparency limit. Curve 1, $h/T_c=10$ and $2d_f/\xi_f=0.84$; curve 2, $h/T_c=40$ and $2d_f/\xi_f=0.5$; curve 3, $h/T_c=100$ and $2d_f/\xi_f=0.43$.

general feature and it is true also for the subsequent 0- π transitions occurring at a higher F layer thickness. For some range of F layer thicknesses the transition from the 0 to π phase is possible when the temperature is lowered.

For the case when $1 \ll \gamma_B \ll h/T_c$, the Ψ function terms in Eq. (58) can be neglected, and at $T = T_c$ the critical thickness d_f^c is

$$d_f^c(T=T_c) = \frac{\xi_f}{2} \left(\frac{3\xi_f}{\gamma_B \xi_n}\right)^{1/3},\tag{60}$$

while at $T \rightarrow 0$ the critical thickness is smaller $d_f^c(T=0)$ $=(\xi_f/2)(6\xi_f/\pi\gamma_B\xi_n)^{1/3}$. The critical F layer thickness, given by Eq. (60), naturally coincides with the corresponding expression (48) obtained for S/F multilayers in the limit $h \gg T_c$. Examples of different nonmonotonic $I_c(T)$ dependences for low barrier transparency limit $\gamma_B \gg h/T_c$ are presented in Fig. 13. In fact, in the limit of low barrier transparency and thin F layer, superconducting electrons tunnel through ferromagnetically ordered atoms. The situation is reminiscent of the tunneling through magnetic impurities, considered by Kulik (1965) and Bulaevskii et al. (1977). More relevant is the analogy with the mechanism of the π -phase realization due to the tunneling through a ferromagnetic layer in the atomic S/F multilayer structure, which is considered in Sec. VII.

Fogelström (2000) considered the ferromagnetic layer as a partially transparent barrier with transmission depending on spin projections. This work may be considered as a further development of the Bulaevskii *et al.* (1977) approach. The Andreev bound states appearing near the spin-active interface within the superconducting gap are tunable with the magnetic properties of the interface. This can result in the switch of the junction from the 0 to π state by changing the transmission characteristics of the interface. This approach was also applied by Andersson, Cuevas, and Fogelström (2002) to study the coupling of two superconductors through a fer-



FIG. 14. Critical current I_c as a function of temperature for $Cu_{0.48}Ni_{0.52}$ junctions with different F layer thicknesses $2d_F$. At the thickness of the F layer of 27 nm the temperature mediated transition between the 0 and π phase occurs. Adapted from Ryazanov, Oboznov, Rusanov, *et al.*, 2001.

romagnetic dot. They demonstrated that the π junction is possible in this case as well. Using the Bogoliubov–de Gennes approach, Tanaka and Kashiwaya (1997) analyzed two superconductors separated by a δ -functional barrier with the spin-orientation dependent height.

Similar to the case of S/F multilayers we discuss the existence of the S/F/S junction with arbitrary equilibrium phase difference φ_0 . Naturally, the form of Eq. (49) for the energy of the junction gives the minima at $\varphi=0$ and $\varphi=\pi$ only. A more general expression for the Josephson-junction energy takes into account higher-order terms over the critical current which leads to the appearance of higher harmonics over φ in the current-phase relationship. Up to the second harmonic, the energy is

$$E = \frac{\Phi_0 I_c}{2\pi c} (1 - \cos\varphi) - \frac{\Phi_0}{2\pi c} \frac{I_2}{2} \cos 2\varphi, \tag{61}$$

and the current is

$$j(\varphi) = I_c \sin \varphi + I_2 \sin 2\varphi.$$
(62)

If the sign of the second harmonic term is negative $I_2 < 0$, then the transition from the 0 to π phase will be continuous, and the φ_0 junction becomes possible. In general, the φ_0 junction may exist if $j(\varphi_0)=0$ and $(\partial_j/\partial_{\varphi})_{\varphi_0}>0$. Calculations of the current-phase relationships for different types of S/F/S junctions (Radovic *et al.*, 2003; Golubov *et al.*, 2004; Cayssol and Montambaux, 2005) show that $\partial_j/\partial_{\varphi} < 0$, and therefore the transition between the 0 and π states appears discontinuous.

The presence of higher harmonics in the $j(\varphi)$ relationship prevents the vanishing of the critical current at the transition from the 0 to π state. This is always the case when the transition occurs at low temperature. Theoret-



FIG. 15. Critical current I_c at T=4.2 K 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. The theoretical fit corresponds to Eq. (A9) in the Appendix, Sec. 2, taking into account the presence of magnetic scattering with $\alpha = 1/\tau_s h = 1.33$ and $\xi_f = 2.4$ nm. The inset shows the temperature mediated 0- π transition for an F layer thickness of 11 nm.

ical studies of clean S/F/S junctions at $T < T_c$ (Buzdin *et al.*, 1982; Chtchelkatchev *et al.*, 2001; Radovic *et al.*, 2003) confirm this conclusion.

Zyuzin and Spivak (2000) argued that the mesoscopic fluctuations of the critical current may produce the $\pi/2$ superconducting Josephson junction. Such a situation is possible when the the F layer thickness is close to $2d_f^c$. Spatial variations of the F layer lead to a second harmonic term in Eq. (62) with $I_2 < 0$ (Buzdin and Koshelev, 2003), and thus the φ_0 junction becomes possible at $2d_f \approx 2d_f^c$.

C. Experiments with π junctions

The temperature dependence of the critical thickness d_f^c is at the origin of the temperature dependence of the critical current $I_c(T)$ observed by Ryazanov, Oboznov, Rusanov, *et al.* (2001) (see Fig. 14). With decreasing temperature for specific thicknesses of the F layer (around 27 nm), a maximum of I_c is followed by a steady decrease down to zero, after which I_c further increases.

This was the first unambiguous experimental confirmation of the 0- π transition via critical current measurements. Ryazanov, Oboznov, Rusanov, *et al.* (2001) explained their results with a small exchange field $h \sim T_c$. The Cu_xNi_{1-x} alloy used in their experiments has a Curie temperature $\Theta \sim 20-30$ K and the exchange field must be higher than 100 K. In consequence, the F layer thickness was in the range $d_f^c(0) < d_f < d_f^c(T_c)$, which provides the strong nonmonotonic temperature dependence of I_c . Also, the experimental estimate of $\xi_f \sim 10$ nm is too large for the expected value of the exchange field.

Recent systematic studies of the critical current in junctions with the $Cu_x Ni_{1-x}$ alloy as a F layer (Ryazanov *et al.*, 2004) have revealed very strong variation of I_c



FIG. 16. Experimental points correspond to critical current measurements, by Kontos *et al.* (2002), vs the PdNi layer thickness. The theoretical curve is the fit of Buzdin and Baladié (2003). The fitting parameters are $\xi_f \sim 30$ Å and $\pi \Delta^2 / eT_c \sim 110 \ \mu$ V.

with the F layer thickness. Indeed, the five orders of magnitude change of the critical current was observed in the thickness interval 12-26 nm. A natural explanation for such a strong thickness dependence is the magnetic scattering effect which is inherent to ferromagnetic alloys. The presence of magnetic scattering in Cu_rNi_{1-r} alloy S/F/S junctions was also noted by Sellier et al. (2003). Magnetic scattering strengthens a decrease of the critical current with an increase of the F layer thickness, and at the same time it increases the period of $I_c(2d_f)$ oscillations. A general expression for the $I_c(2d_f)$ dependence, taking into account magnetic scattering, is given in the Appendix, Sec. 2, Eq. (A9). Attempts to describe the experimental data of Ryazanov et al. (2004) on the $I_c(2d_f)$ dependence with this expression provided hints on the existence of another minimum $I_c(2d_f)$ at a smaller F layer thickness—around 10 nm. Very recent experiments with junctions with F layer thickness up to 7 nm have confirmed this prediction (Ryazanov *et al.*, 2005); see Fig. 15. The existence of the first $0-\pi$ transition at $2d_f \approx 11$ nm means that previously reported transitions in $Cu_x Ni_{1-x}$ junctions were actually the transitions from the π to 0 phase (and not, as was assumed, from the 0 to π phase). This means that now it is also possible to fabricate the π junctions with a 10⁴ times higher critical current. Note that the first measurements (Frolov et al., 2004) of the current-phase relation in the S/F/S junction with the Cu_{0.47}Ni_{0.53} F layer provided no evidence of the second harmonic in the $j(\varphi)$ relationship at the 0- π transition. These measurements were performed using the junction with a F layer thickness around 22 nm, i.e., near the second minimum on the $I_c(2d_f)$ dependence. The much higher critical current near the first minimum (at $2d_f \approx 11$ nm) may occur to be very helpful for a search of the second harmonic.

The results of Ryazanov, Oboznov, Rusanov, *et al.* (2001) on the temperature induced crossover between 0

1.8

1.6

and π states were recently confirmed in the experiments of Sellier *et al.* (2003). Kontos *et al.* (2002) observed damped oscillations of the critical current as a function of the F layer thickness in Nb/Al/Al₂O₃/PdNi/Nb junctions. The measured critical currents with the theoretical fit (Buzdin and Baladié, 2003) are presented in Fig. 16. Blum *et al.* (2002) reported the strong oscillations of the critical current with the F layer thickness in Nb/Cu/Ni/Cu/Nb junctions.

Bulaevskii et al. (1977) pointed out that the π junction incorporated into a superconducting ring would generate a spontaneous current and a corresponding magnetic flux would be half a flux quantum Φ_0 . The appearance of the spontaneous current is related to the fact that the ground state of the π junction corresponds to the phase difference π and this phase difference will generate a supercurrent in the ring which short-circuits the junction. Naturally the spontaneous current is generated if there are any odd number of π junctions in the ring. This has been exploited in an elegant way by Ryazanov, Oboznov, Veretennikov, and Rusanov (2001) to provide unambiguous proof of the π -phase transition. Ryazanov, Oboznov, Veretennikov, and Rusanov (2001) observed the half-period shift of the external magnetic-field dependence of the transport critical current in triangular S/F/S arrays. The F layer thickness of the S/F/S junctions was chosen such that at high temperature the junctions were the usual 0 junctions, and transformed into the π junctions with a decrease in temperature (Ryazanov, Oboznov, Rusanov, et al., 2001).

Guichard et al. (2003) performed similar phasesensitive experiments using dc SQUID with a π junction. The total current I flowing through the SQUID is the sum of the currents I_a and I_b flowing through the two junctions, $I = I_a + I_b$. If the junctions have the same critical currents I_c and both are 0 junctions, then I_a $=I_c \sin \varphi_a$ and $I_b=I_c \sin \varphi_b$, where φ_a and φ_b are the phase differences across the junctions. Neglecting the inductance of the loop of the SQUID, the phase differences satisfy the usual relation (Barone and Paterno, 1982) $\varphi_a - \varphi_b = 2\pi \Phi / \Phi_0$, where Φ is the flux of the external magnetic field through the loop of the SQUID. The maximum critical current of the SQUID is I_{max} $=2I_c \cos(\pi \Phi/\Phi_0)$. When one of the junctions (let us say b) is the π junction with the same critical current, the current flowing through it is $I_b = -I_c \sin \varphi_b = I_c \sin(\varphi_b)$ $+\pi$). Therefore, the maximum critical current of the SQUID in this case is $I_{\text{max}}^{\pi} = 2I_c \cos(\pi \Phi/\Phi_0 + \pi/2)$, and the diffraction pattern is shifted by half a quantum flux. If both junctions are the π junctions, the diffraction pattern is identical to the diffraction pattern of the SQUID with two 0 junctions. Namely, this was observed in experiment by Guichard et al. (2003) with SQUID containing junctions with PdNi ferromagnetic layers; see Fig. 17.

Recently, Bauer *et al.* (2004) measured with micro-Hall sensor the magnetization of a mesoscopic superconducting loop containing a PdNi ferromagnetic π junction. These measurements also provided a direct



SQUID 0-0

T=5.54 K

FIG. 17. Experiments of Guichard *et al.* (2003) on the diffraction pattern of SQUID with 0 and π junctions. There is no shift of the pattern between 0-0 and π - π SQUIDs. The $\Phi_0/2$ shift is observed between 0- π and 0-0 or π - π SQUIDs. The 0 and π junctions were obtained by varying the PdNi layer thickness.

evidence of the spontaneous current induced by the π junction.

VI. COMPLEX S/F STRUCTURES

A. F/S/F spin-valve sandwiches

The strong proximity effect in superconductormetallic ferromagnet structures could lead to the phenomenon of spin-orientation-dependent superconductivity in F/S/F spin-valve sandwiches. Such behavior was predicted by Buzdin *et al.* (1999) and Tagirov (1999) and recently observed in experiment by Gu *et al.* (2002). Note that de Gennes (1966b) considered theoretically a

2.5

SQUID π-π

T=5.37 K



FIG. 18. Earlier observation by Deutscher and Meunier (1969) of the spin-valve effect on In film between oxidized FeNi and Ni layers. The resistive measurements of the critical temperature are presented in zero field: dashed line, after application of the 1-T field parallel to the ferromagnetic layers; solid line, after application of the -1-T field and subsequently +0.03-T field to return the magnetization of the FeNi layer.

system consisting of a thin S layer in between two ferromagnetic insulators. He argued that the parallel orientation of the magnetic moments is more harmful for superconductivity because of the presence of the nonzero averaged exchange field acting on the surface of the superconductor. This prediction has been confirmed in experiment by Hauser (1969) on In film sandwiched between two Fe_3O_4 films, and by Deutscher and Meunier (1969) on a In film between oxidized FeNi and Ni layers; see Fig. 18. Curiously, the experiments of Deutscher and Meunier (1969) correspond more closely to the case of metallic F/S/F sandwiches as these authors report rather low interface resistance.

To consider the spin-orientation effect in metallic F/S/F sandwiches we use the notations analogous to that of Sec. IV. More precisely, to have a direct connection with the corresponding formula of Sec. IV, we assume that the F layer thickness is d_f and the S layer thickness is $2d_s$; see Fig. 19.



FIG. 19. Geometry of the F/S/F sandwich. The thickness of the S layer is $2d_s$ and two F layers have identical thicknesses d_f .

To provide a simple theoretical description we consider the case $d_s \ll \xi_s$ with only two orientations of the ferromagnetic moments: parallel and antiparallel. For arbitrary orientations the ferromagnetic moment needs the introduction of triplet components of the anomalous Green's functions. The first attempt of such an analysis was made by Baladié *et al.* (2001), but using an incomplete form of the Usadel equation. The full correct calculations for this case were performed by Bergeret *et al.* (2003), Fominov, Golubov, and Kupriyanov (2003a), and Volkov *et al.* (2003).

In fact, we only need to analyze the case of the antiparallel orientation of ferromagnetic moments because the parallel orientation case is equivalent to the 0 phase in a S/F multilayered structure (Sec. IV) with the F layers two times thinner than in a F/S/F sandwich. In other words, our choice of notation allows the parallel orientation case to correspond to the critical temperature for the 0 phase from Sec. IV. To analyze the antiparallel orientation case, we follow the approach used in Sec. IV, but we need to retain the linear over x term in the expansion of the anomalous Green's function in the S layer in Eq. (35),

$$F_s(x,\omega) = F_0 \left(1 + \alpha_\omega x - \frac{\beta_\omega}{2} x^2 \right).$$
(63)

With the help of the Usadel equation (31), we find that F_0 has the form (36) with the pair-breaking parameter τ_s^{-1} determined by

$$\frac{4d_s\tau_s^{-1}}{D_s} = 2d_s\beta_\omega \simeq \frac{F_s'(-d_s)}{F_s(-d_s)} - \frac{F_s'(d_s)}{F_s(d_s)} - \frac{d_s}{2} \left[\frac{F_s'(d_s)}{F_s(d_s)} + \frac{F_s'(-d_s)}{F_s(-d_s)} \right]^2.$$
(64)

Let us suppose that the exchange field is positive (+h) in the right F layer and then for $d_s+d_f>x>d_s$ we have

$$F_f(x,\omega>0) = A \cosh\left[\frac{i+1}{\xi_f}(x-d_s-d_f)\right],\tag{65}$$

while for the left F layer, the exchange field is negative and for $-d_s-d_f \le x \le -d_s$ we have

$$F_{f}(x,\omega > 0) = B \cosh\left[\frac{1-i}{\xi_{f}}(x-d_{s}-d_{f})\right].$$
 (66)

Taking into account the explicit form of $F_f(x)$ and the boundary conditions (32), we find for the antiparallel alignment case $F'_s(d_s)/F_s(d_s) = -[F'_s(-d_s)/F_s(-d_s)]^*$ and the pair-breaking parameter for this case $\tau_s^{-1} = \tau_{s,AP}^{-1}$ may be written as

$$\tau_{s,AP}^{-1} \simeq -\frac{D_s}{2d_s} \operatorname{Re}\left(\frac{F_s'(d_s)}{F_s(d_s)}\right) + \frac{D_s}{2} \left[\operatorname{Im}\left(\frac{F_s'(d_s)}{F_s(d_s)}\right)\right]^2.$$
(67)

The second term on the right-hand side of Eq. (67) is important in the limit of small d_f and we will omit it further. The boundary conditions (32) permit us to calculate the parameter τ_s^{-1} , provided the anomalous Green's function in the F layer is known. For the parallel



FIG. 20. Influence of the S/F interface transparency [parameter $\tilde{\gamma} = \gamma_B(\xi_n/\xi_f)$] on the T_c^* vs d_f dependence (Baladié and Buzdin, 2003). The thickness of the F layer is normalized by ξ_f . The dimensionless pair-breaking parameter $\tilde{\tau}_0 = 4\pi T_c (2d_s\xi_f/D_s)\sigma_s/\sigma_f$ is chosen constant and equal to 4. The solid line corresponds to the antiparallel case, and the dashed line to the parallel case. One can distinguish four characteristic types of behavior: (a) weakly nonmonotonic decay to a finite value T_c^* , (b) reentrant behavior for the parallel orientation, and (c) and (d) monotonic decay to $T_c^* = 0$ with (d) or without (c) switching to a first-order transition in the parallel case. In (d), the dotted line represents schematically the first-order transition line.

alignment of the ferromagnetic moments it is $\tau_{s,P}^{-1} = \tau_{s,0}^{-1}$, where $\tau_{s,0}^{-1}$ is given by Eq. (40), while for the antiparallel alignment it is

$$\tau_{s,AP}^{-1} = \operatorname{Re}(\tau_{s,0}^{-1}) = \operatorname{Re}(\tau_{sP}^{-1}).$$
(68)

As a result, we obtain the following simple formula for the critical temperature T_c^P with parallel orientation and T_c^{AP} with antiparallel orientation:

$$\ln \frac{T_{c}^{P}}{T_{c}} = \Psi\left(\frac{1}{2}\right) - \operatorname{Re}\Psi\left\{\frac{1}{2} + \frac{1}{2\pi T_{c}^{P}\tau_{s,0}}\right\},\tag{69}$$

$$\ln \frac{T_c^{AP}}{T_c} = \Psi\left(\frac{1}{2}\right) - \Psi\left\{\frac{1}{2} + \operatorname{Re}\left(\frac{1}{2\pi T_c^{AP}\tau_{s,0}}\right)\right\}.$$
 (70)

The different kinds of $T_c(d_f)$ curves are presented in Fig. 20.

We see that the interface transparency is an important factor, controlling the spin-valve effect in F/S/F structures. It is interesting to note that an optimum condition for the observation of this effect in the case of the non-negligible interface transparency is the choice $d_f \sim (0.1-0.4)\xi_f$.



FIG. 21. The calculated dependence of the superconducting transition temperature vs inverse reduced half thickness d^*/d_s of the superconducting layer for parallel and antiparallel alignments for the transparent interface ($\gamma_B=0$) and thick ferromagnetic layer ($d_f \gg \xi_f$). The effective length is $d^* = (\sigma_f/\sigma_s)(D_s/4\pi T_c)(h/D_f)^{1/2}$.

In the case when the F layer thickness exceeds ξ_f , the critical temperature negligibly depends on d_f . The case for the transparent S/F interface ($\gamma_B=0$) was considered by Buzdin *et al.* (1999), and critical temperatures for parallel and antiparallel alignments are presented in Fig. 21. The finite interface transparency decreases the spinvalve effect, and for $\tilde{\gamma}_B > 5$ the dependence of critical temperatures on the mutual orientation of ferromagnetic moments is hardly observed.

The thermodynamic characteristics of F/S/F systems were studied theoretically by Baladié and Buzdin (2003) and Tollis (2004) using the Usadel formalism and it was noted that the superconductivity always remains gapless.

Bagrets *et al.* (2003) developed a microscopic theory with F/S/F systems based on the direct solution of the Gor'kov equations for the normal and anomalous Green's functions. The main mechanism of the electron scattering in F layers was assumed to be of the *s*-*d* type. The results of this microscopical analysis were in accordance with the quasiclassical approach and provided a reasonable quantitative description of the experimental data of Obi *et al.* (1999) on the $T_c(d_f)$ dependence in Nb/Co multilayers.

Krunavakarn *et al.* (2004) generalized the approach of Fominov *et al.* (2002) to perform exact numerical calculations of the nonmonotonic critical temperature in F/S/F sandwiches. They demonstrated also that the Takahashi-Tachiki (1986) theory of the proximity effect is equivalent to the approach using the Usadel equations.

Bozovic and Radovic (2002) studied theoretically the coherent transport current through F/S/F double-barrier junctions. The exchange field and the interface barrier reduce Andreev reflection due to the enhancement of the normal reflection. Interestingly, the conductance is always higher for parallel alignment of the ferromagnetic moments. A similar conclusion was obtained in work of Yamashita, Takahashi, and Maekawa (2003). Such behavior is related with the larger transmission for the normal tunneling current in this orientation. The calculations also revealed the periodic vanishing of Andreev reflection at the energies above the superconducting gap.

The case of insulating F layers (de Gennes, 1966b) corresponds to when the superconducting electrons feel the exchange field on the surface of the S layer. We describe this case taking the limit $d_f \rightarrow 0$ with τ_{s0}^{-1} $=ih(\tilde{a}/d_s)$, where \tilde{a} is the interatomic distance, which describes the region near the S/F interface where the exchange interaction (described by the exchange field h) with electron spins takes place. In fact it simply means that, for the parallel orientation case, the superconductor is under the influence of an averaged exchange field $h = h(\tilde{a}/d_s)$, while for the antiparallel orientation this field is absent. Careful theoretical analysis of a system consisting of the superconducting film sandwiched between two ferromagnetic semiconducting insulators with different oriented magnetization was performed by Kulić and Endres (2000) for both singlet and triplet superconductivity cases. In the case of a triplet superconductivity, the critical temperature depends not only on the relative orientation of the magnetization but also on its absolute orientation.

B. S-F-I-F'-S heterostructures and triplet proximity effect

Many theoretical works were devoted to the analysis of more complex S/F systems. Proshin *et al.* (2001) (see also Izyumov *et al.*, 2002) studied the critical temperature of S/F multilayers with alternating magnetization of adjacent F layers. Izyumov *et al.* (2000, 2002) also proposed the 3D LOFF state in F/S contacts. However, this conclusion was based on controversial boundary conditions, corresponding to different in-plane 2D wave vectors on both sides of the contact—see the Comment by Fominov, Kupriyanov, and Feigelman (2003) and the Reply of Khusainov and Proshin (2003).

Koshina and Krivoruchko (2001) (see also Golubov *et al.*, 2002a) studied the Josephson current of two proximity S/F bilayers separated by an insulating (I) barrier and demonstrated that in such S/F-I-F/S contacts the π phase may appear even at very small F layer thickness (smaller than ξ_f). The π -phase transition in this case is related to a rotation of $\pi/2$ with the anomalous Green's function F on the S/F boundary in addition to a jump of its modulus. To demonstrate this we consider a thin F layer of thickness $d_f \ll \xi_s$ in contact with a superconductor. If x = 0 corresponds to the S/F interface, and $x=d_f$ is the outer surface of the F layer, then the solution of the linearized Usadel equation in the ferromagnet is

$$F_f(x,\omega>0) = A \cosh\left[\frac{i+1}{\xi_f}(x-d_f)\right].$$
(71)

Using the boundary condition (32) we obtain

$$F_f(x,\omega > 0) \simeq F_f(0,\omega > 0) = \frac{F_s(0,\omega > 0)}{1 + 2i\gamma_B \xi_n d_f / \xi_f^2}.$$
 (72)

In the case of low interface transparency $\gamma_B \xi_n d_f / \xi_f^2 \ge 1$, the jump of the phase of the *F* function at the interface is approximately equal to $-\pi/2$:

$$F_f(0,\omega>0) \approx F_s(0,\omega>0) \exp\left(-i\frac{\pi}{2}\right) \frac{\xi_f^2}{\gamma_B \xi_n d_f}.$$
 (73)

Koshina and Krivoruchko (2001) and Golubov *et al.* (2002a) argued that at each S/F interface in the S/F-I-F/S contact a phase jump $-\pi/2$ occurs, and the total phase jump in an equilibrium state would be π .

Kulić and Kulić (2001) calculated the Josephson current between two superconductors with a helicoidal magnetic structure. They found that the critical current depends on the relative orientation θ of the magnetic moments on the banks of contact

$$I_c = I_{c0}(1 - R_{\pm} \cos \theta),$$
(74)

where $R_{-}(R_{+})$ corresponds to the same (opposite) helicity of the magnetization in the banks. Depending on the parameters of the helicoidal ordering, the value of R_{\pm} may be either smaller or larger than 1. If $R_{\pm} > 1$, then I_{c} may be negative for some misorientation angles θ , which provides evidence of the π phase. Interestingly, by tuning the magnetic phase θ , it is possible to provoke a switch between the 0 and π phase. As seen from Eq. (74), the critical current of the Josephson junction is maximal for the antiparallel orientation ($\theta = \pi$) of the magnetizations in the banks.

Bergeret et al. (2001a) studied the Josephson current between two S/F bilayers and pointed out the enhancement of the critical current for ferromagnetic moments aligned antiparallel. They demonstrated that at low temperatures the critical current in a S/F-I-F/S junction becomes larger than in the absence of the exchange field (i.e., if the ferromagnetic layers are replaced by normalmetal layers with h=0). In more detail (taking into account different transparency of S/F interfaces and different orientations of the magnetization in the banks), studied these junctions were theoretically by Krivoruchko and Koshina (2001), Chtchelkatchev et al. (2002), Golubov et al. (2002a), and Li et al. (2002). Blanter and Hekking (2004) used the Eilenberger and Usadel equations to calculate the current-phase relation of the Josephson junction with the composite F layer, consisting of two ferromagnets with opposite magnetizations.

Bergeret *et al.* (2001b) and Kadigrobov *et al.* (2001) analyzed using the Usadel equations the proximity effect in S/F structures with local inhomogeneity of the magnetization. They obtained the conclusion that varying the space magnetization generates the triplet component of the anomalous Green's function $(\sim \langle \Psi_{\uparrow} \Psi_{\uparrow} \rangle)$ which penetrates in the ferromagnet at distances much larger than ξ_{f} . It is not, however, the triplet superconductivity itself because the corresponding triplet order parameter would be equal to zero, unlike the superfluidity in ³He, for example. In general, the triplet components of the anomalous Green's function always appear in the description of the singlet superconductivity in the rotation of space the exchange field. For example, they were introduced by Bulaevskii et al. (1980) in the theory of coexistence of superconductivity with helicoidal magnetic order. An important finding of Bergeret et al. (2001b) and Kadigrobov et al. (2001) was the demonstration that the triplet component is insensitive to the pair breaking by the exchange field. Therefore its characteristic decay length is the same as in the normal metal, i.e., $\xi_{T,d} = \sqrt{D_f/2\pi T}$ long-range proximity effect could explain the experiments on S/F mesoscopic structures (Giroud et al., 1998; Petrashov et al., 1999), where a considerable increase of the conductance below the superconducting critical temperature was observed at distances much larger than ξ_{f} .

In their subsequent works Bergeret et al. (2003) and Volkov et al. (2003) studied the manifestation of this triplet component in S/F multilayered structures. The most striking effect is the dependence of the critical current in multilayered S/F structures on the relative orientation of the ferromagnetic moments. For the collinear orientation, the triplet component is absent, and provided the thickness of the ferromagnetic layer $d_f \gg \xi_f$, the critical current is exponentially small. On the other hand, if the orientation of the magnetic moments is noncollinear then the triplet component of the superconducting condensate appears. Its decay length $\xi_{T,d}$ is much larger than ξ_f , and this triplet component provides the coupling between the adjacent superconducting layers. When the F layer thicknesses are in the interval $\xi_{T,d} \gg d_f \gg \xi_f$, this coupling then occurs to be strong. As a result, the critical current is maximal for the perpendicular orientation of the adjacent ferromagnetic moments, and it may significantly exceed the critical current for parallel orientation. Due to the mesoscopic fluctuations (Zyuzin *et al.*, 2003), the decay of the critical current for the magnetic moments oriented collinearly is not exponential. Nevertheless, for this orientation it would be very small, and this does not change the main conclusion of the long-range triplet proximity effect. A lot of interesting physics emerges in the case of S/F systems with genuine triplet superconductors. For example, the proximity effect depends on the mutual orientation of the magnetic moments of the Cooper pairs and ferromagnets.

The long-range triplet proximity effect was predicted to exist in the dirty limit. An interesting problem is how it evolves in the clean limit. In this regime there is no characteristic decay length for the anomalous Green's function in a ferromagnet [see Eqs. (21) and (22)], and the angular behavior of the critical current in S/F multilayers may be quite different. If, for example, we apply the Eilenberger equations for the description of a clean S/F/F'/S structure with antiparallel ferromagnetic layers with equal thicknesses, the exchange field is completely eliminated (Blanter and Hekking, 2004). Therefore, the critical current will be the same as for the nonmagnetic interlayers. Here it is difficult to believe that for the perpendicular orientation of the magnetic moments the critical current could be even higher. The microscopical calculations using the Bogoliubov–de Gennes equations of the properties of S/F multilayers with noncollinear orientation of the magnetic moments would be of interest.

Barash *et al.* (2002) studied the Josephson current in S-FIF-S junctions in the clean limit within the quasiclassical theory of superconductivity, based on the so-called Ricatti parametrization (Schopol and Maki, 1995). They obtained the nonmonotonic dependences of the critical current on the misorientation angle of the ferromagnetic moments. However, even for a rather high transparency of the I barrier (D=0.8), the maximum critical current onccurred for the magnetic moments oriented antiparallel.

VII. ATOMIC THICKNESS S/F MULTILAYERS

A. Layered ferromagnetic superconductors

In this section, we consider an atomic-scale multilayer F/S system, where the superconducting (S) and ferromagnetic (F) layers alternate. When the electron transfer integral between the S and F layers is small, superconductivity can coexist with ferromagnetism in adjacent layers. Andreev *et al.* (1991) demonstrated that the exchange field in F layers favors the π -phase behavior of superconductivity, when the superconducting order parameter alternates its sign on the adjacent S layers.

Nowadays several types of layered compounds, where superconducting and magnetic layers alternate, are known. For example, in Sm_{1.85}Ce_{0.15}CuO₄ (Sumarlin et al., 1992), which reveals superconductivity at T_c =23.5 K, the superconducting layers are separated by two ferromagnetic layers with magnetic moments oriented oppositely and the Néel temperature is T_N =5.9 K. Several years ago, a new class of magnetic superconductors based on the layered perovskite ruthenocuprate compound RuSr₂GdCu₂O₈ comprising CuO₂ bilayers and RuO_2 monolayers were synthesized (see, for example, McLaughlin et al., 1999, and references cited there). In RuSr₂GdCu₂O₈, the magnetic transition occurs at $T_M \sim 130-140$ K and superconductivity appears at $T_c \sim 30-50$ K. Recent measurements of the interlayer current in small-sized RuSr₂GdCu₈ single crystals showed the intrinsic Josephson effect (Nachtrab et al., 2004). Apparently, it is a weak ferromagnetic order which occurs in this compound. Although magnetization measurements provide evidence of a small ferromagnetic component, the neutron-diffraction data on RuSr₂GdCu₂O₈ (Lynn et al., 2000) revealed the dominant antiferromagnetic ordering in all three directions. Later, the presence of a ferromagnetic in-plane component of about $(0.1-0.3)\mu_B$ has been confirmed by neuscattering on isostructural RuSr₂YCu₂O₈ tron

(Tokunaga *et al.*, 2001). In addition, in an external magnetic field the ferromagnetic component grows rapidly at the expense of the antiferromagnetic one.

Due to the progress of multilayer preparation methods, the fabrication of artificial atomic-scale S/F superlattices has become possible. An important example is the YBa₂Cu₃O₇/La_{2/3}Ca_{1/3}MnO₃ superlattice (Sefrioui et al., 2003; Holden et al., 2004). The manganite half metallic compound La_{2/3}Ca_{1/3}MnO₃ (LCMO) exhibits colossal magnetoresistance and its Curie temperature is $\Theta = 240$ K. The cuprate high- T_c superconductor $YBa_2Cu_3O_7$ (YBaCuO) with $T_c=92$ K has a similar lattice constant as LCMO allows us to prepare the highquality YBaCuO/LCMO superlattices with different F and S layer thickness ratios. The proximity effect in these superlattices occurs to be long ranged. For a fixed superconducting layer thickness, the critical temperature is dependent on the LCMO layer thickness in the 100nm range (Sefrioui et al., 2003; Peña et al., 2004). This is unusual because the YBaCuO and LCMO are strongly anisotropic layered systems with small coherence length in a direction perpendicular to the layers (0.1-0.3 nm). A somewhat similar giant proximity effect has been recently reported in nonmagnetic trilayer junctions $La_{1.85}Sr_{0.15}CuO_4/La_2CuO_{4+d}/La_{1.85}Sr_{0.15}CuO_4$ (Bozovic et al., 2004) and in superconductor-antiferromagnet YBa₂Cu₃O₇/La_{0.45}Ca_{0.55}MnO₃ superlattices (Pang *et al.*, 2004). The observed giant proximity effect defies the conventional explanations. Bozovic et al. (2004) suggested that it may be related to resonant tunneling, but at the moment the question about the nature of this effect is open.

B. Exactly solvable model of the π phase

Let us consider the exactly solvable model (Andreev et al., 1991) of alternating superconducting and ferromagnetic atomic metallic layers. For simplicity, we assume that the electron's motion inside the F and S layers is described by the same energy spectrum $\xi(\mathbf{p})$. Three basic parameters characterize the system: t is the transfer energy between the F and S layers, λ is the Cooper pairing constant which is assumed to be nonzero in S layers only, and h is the constant exchange field in the F layers. The Hamiltonian of the system can be written as

$$H = \sum_{\vec{p},n,i,\sigma} \xi(\mathbf{p}) a_{ni\sigma}^{\dagger}(\mathbf{p}) a_{ni\sigma}(\mathbf{p}) + H_{\text{int1}} + H_{\text{int2}} + t[a_{ni\sigma}^{\dagger}(\mathbf{p})a_{n,-i,\sigma}(\mathbf{p}) + a_{n+1,-i,\sigma}^{\dagger}(\mathbf{p})a_{ni\sigma}(\mathbf{p}) + \text{H.c.}],$$

$$H_{\text{int1}} = \frac{g}{2} \sum_{\vec{p}_1,\vec{p}_2,n,\sigma} a_{n1\sigma}^{\dagger}(\mathbf{p}_1) a_{n,1,-\sigma}^{\dagger}(-\mathbf{p}_1) \times a_{n,1,-\sigma}(-\mathbf{p}_2) a_{n1\sigma}(\mathbf{p}_2),$$

$$H_{\text{int2}} = -h \sum \sigma a_{n,-1,\sigma}^{\dagger}(\mathbf{p}) a_{n,-1,\sigma}(\mathbf{p}), \qquad (75)$$

where $a_{ni\sigma}^{\dagger}$ is the creation operator of an electron with spin σ in the *n*th elementary cell and momentum **p** in

 \vec{p}, n, σ

layer *i*, where *i*=1 for the S layer and *i*=-1 for the F layer, and *g* is the pairing constant. An important advantage of this model is that the quasiparticle Green's functions can be calculated exactly and a complete analysis of the superconducting characteristic is possible. Assuming that the order parameter changes from cell to cell in the form $\Delta_n = |\Delta| e^{ikn}$, the self-consistency equation for the order parameter $|\Delta|$ reads

$$1 = -\lambda T_c^* \lambda \sum_{\omega} \int_{-\infty}^{\infty} d\xi \int_0^{2\pi} \frac{dq}{2\pi} \times \frac{\tilde{\omega}_+ \tilde{\omega}_-}{\tilde{\omega}_+ \tilde{\omega}_- |\Delta|^2 - (\omega_- \tilde{\omega}_- - |T_{q+k}|^2)(\omega_+ \tilde{\omega}_+ - |T_q|^2)},$$
(76)

where $\lambda = gN(0)$ and $\omega_{\pm} = i\omega \pm \xi(p)$, $\tilde{\omega}_{\pm} = \omega_{\pm} + h$. The quasimomentum q is perpendicular to the direction of the layers, and $T_q = 2t \cos(q/2)e^{iq/2}$. In the limit of a small transfer integral $t \ll T_c$, where T_c is the bare mean-field critical temperature of the S layer in the absence of coupling (t=0), we arrive at the following equation for the critical temperature T_c^* :

$$\ln \frac{T_c^*}{T_c} = -\pi T_c^* t^2 \sum_{\omega} \frac{4}{|\omega| (4\omega^2 + h^2)} + \pi T_c t^4 \cos k \sum_{\omega} \frac{12\omega^4 - 7\omega^2 h^2 - h^4}{|\omega|^3 (\omega^2 + h^2) (4\omega^2 + h^2)^2}.$$
 (77)

The critical temperature T_c^* is close to the bare critical temperature T_c and from Eq. (77), for h=0, the maximal T_c^* corresponds to k=0, i.e., the superconducting order parameter is the same at all layers. It is worth noting that as the exchange field on the F layers grows, tunneling becomes energetically more costly, so the leading second-order term in t falls as $1/h^2$ for large h and the critical temperature increases. This is related to the fact that, due to the decrease of the coupling, the effective exchange field induced on the S layers decreases with an increase of h. For $h \ge T_c$, the coefficient of the $\cos k$ term has a negative sign and the maximal T_c^* corresponds to $k = \pi$, and the transition occurs with the π phase with an alternating order parameter $\Delta_n = |\Delta|(-1)^n$. Numerical calculations (Andreev et al., 1991) give for the critical value of the exchange field (at which kchanges from 0 to π) $h_c=3.77T_c$, and the complete (h,T)phase diagram is presented in Fig. 22.

At T=0 the transition to the π phase occurs at $h_{c0} = 0.87T_c$. The analysis of Prokić *et al.* (1999) and Houzet *et al.* (2001) shows that the perpendicular critical current vanishes at the transition from the 0 to the π phase and the Josephson coupled superconducting planes are decoupled. Strictly speaking, the critical current vanishes only in the $\sim t^4$ approximation; see Eq. (77). The term $\sim t^8$ gives the contribution $\sim t^8 \cos 2k$, and the critical current at the transition to the π phase will decrease to a small value $\sim I_c(t/T_c)^8$. Note that the sign of the second harmonic in the $j(\varphi)$ relation generated by this $\sim t^8$ term is positive, and therefore the transition from the 0 to the π phase is discontinuous.



FIG. 22. The (T,h) phase diagram of the atomic S/F multilayer in the limit of the small transfer integral $t \ll T_c$.

As a result, if the exchange field is in the interval $h_{c0} \le h \le 3.77 T_c$, the 0- π transition may be easily observed with decreasing temperature due to the nonmonotonic behavior of the Josephson plasma frequency and the parallel London penetration (Houzet et al., 2001). However, a typical value of the exchange field is rather high and more probable is $h \ge T_c$, and the system will be in the π phase at any temperature. This is consistent with the recent experiments of Nachtrab et al. (2004) on $RuSr_2GdCu_2O_8$ presenting no evidence of superconducting planes decoupling with temperature. In $RuSr_2GdCu_2O_8$, the superconducting pairing is probably of the *d*-wave type. This case was analyzed theoretically by Prokić and Dobrosavljević-Grujić (1999), and the scenario of the π -phase appearance is similar to the s-wave superconductivity. Calculations of the electronic density of states by Prokić and Dobrosavljević-Grujić (1999) and Prokić et al. (1999) revealed some changes inherent to the 0- π transition, but the experimental identification of the π phase in atomic-scale S/F superlattices is an extremely difficult task. In principle, if the superlattice consists of an even number of superconducting layers, then the phase of the order parameter will differ by π , and the entire system will function as a Josephson π junction. The spontaneous current in a superconducting loop containing such a π junction was observed with an experiment analogous to the one made by Bauer *et al.* (2004).

The model (75) permits us to analyze the transition from the quasi-2D to 3D system with an increase of the transfer intergral t. At $t \leq T_c$, instead of the π phase, the LOFF state with modulation along the superconducting layers appears and the system becomes analogous to the 3D superconductor in a uniform exchange field (Houzet and Buzdin, 2002).

Buzdin and Daumens (2003) considered the spin-valve effect in the F/S/F structure consisting of three atomic layers and described by the model (75). Analogous to the F/S/F spin-valve sandwiches (see Sec. VI), the critical temperature is maximal for the antiparallel orientation of the ferromagnetic moments. However, at low temperature, the situation is inversed. Namely, the superconducting gap occurs to be larger for the parallel orientation of the ferromagnetic moments. This counterintuitive result of the inversion of the proximity effect may be understood by the example of the ferromagnetic half metal. Indeed at T=0, the disappearance of the Cooper pair in a S layer means that two electrons with opposite spin must exit. If the neighboring F layers of half metals are parallel, then, for one spin orientation, they are both insulators and the electron with this spin orientation cannot enter it. This results in the impossibility of the pair destruction. On the other hand, for the antiparallel orientation of the F layers, in the electronspin orientation there is an adjacent normal layer and a Cooper pair can leave the S layer. Such behavior contrasts with the diffusive model prediction (Baladié and Buzdin, 2003; Tollis, 2004) but is in accordance with the T=0 results obtained with the multiterminal model for S/F hybrid structures (Apinyan and Mélin, 2002). Apparently, it is a special property of the clean limit of the atomic-layer S/F model, and it disappears in the case of several consecutive S layers per unit cell (Mélin and Feinberg, 2004).

VIII. SUPERCONDUCTIVITY NEAR THE DOMAIN WALL

In the previous discussion of the properties of S/F heterostructures, we have implicitly assumed that the ferromagnet has uniform magnetization, i.e., there are no domains. In practice the domains appear in ferromagnets quite easily and special conditions are usually needed to obtain the single domain ferromagnet. In the standard situation, the size of the domains is much larger than the superconducting coherence length, $\xi_f \ll \xi_s$, and therefore the Cooper pair will sample the uniform exchange field. However, for the S/F proximity effect near the domain wall a special situation occurs, where the magnetic moments and the exchange field rotate. The Cooper pairs feel the exchange field averaged over the superconducting coherence length. Naturally, such an averaged field will be smaller near the domain wall, which leads to a local decrease of the pair-breaking parameter. As a result, we may expect that superconductivity would be more robust near the domain wall. In particular, the critical temperature T_{cw} for the superconductivity localized near the domain wall would be higher than that of the uniform S/F bilayer T_c^* . For bulk ferromagnetic superconductors, the critical temperature of the superconductivity localized near the domain wall was calculated by Buzdin et al. (1984). The experimental results of the domain-wall superconductivity in Ni_{0.80}Fe_{0.20}/Nb bilayers (with Nb thickness around 20 nm) were observed by Rusanov et al. (2004). The Néel-type domain walls in Permalloy $(Ni_{0.80}Fe_{0.20})$ are responsible for the local increase of the critical temperature around 10 mK. The width of the domain walls w in Permalloy films used by Rusanov et al. (2004) is rather large, $w \sim 0.5 \ \mu m$, i.e., much larger than the superconducting coherence length of niobium. The rotation angle α of the exchange field at the distance ξ_s is estimated as $\alpha \sim \xi_s / w$, and the averaged exchange field h^{av} is smaller than the field h distant from the domain wall: $(h-h^{av})/h \sim (\xi_s/w)^2$. Therefore, a relative decrease of the pair-breaking parameter τ_s^{-1} in Eq.

$$\frac{T_{cw} - T_c^*}{T_c^*} \sim (\xi_s/w)^2,$$
(78)

which is of the same order of magnitude as the effect observed on the Ni_{0.80}Fe_{0.20}/Nb bilayers. Keeping in mind the temperature dependence of the superconducting coherence length $\xi(T) \sim \xi_s \sqrt{T_c^*/|T - T_c^*|}$, we see that the condition for domain-wall superconductivity is $\xi(T_{cw}) \sim w$.

In the case of a very thin domain wall, the variation of the exchange field is steplike and the local suppression of the pair-breaking parameter occurs at a small distance of order $\xi_f \ll \xi_s$ near the domain wall. The situation resembles the enhancement of the superconducting pairing near the twin planes (Khlyustikov and Buzdin, 1987). The variation of the pair breaking occurring over a distance ξ_f induces a superconducting order parameter over a distance $\xi(T_{cw})$ near the domain wall and the effective relative decrease of the pair-breaking parameter will be of the order of $\xi_f/\xi(T_{cw})$. Therefore, if the shift of the critical temperature of the S/F bilayer is comparable with T_c , i.e., $(T_c - T_c^*)/T_c \sim 1$, the critical temperature T_{cw} of the superconductivity, localized near the domain wall, may be estimated from the condition $(T_{cw} - T_c^*)/T_c^*$ $\sim \xi_f / \xi(T_{cw})$. As a result we have

$$\frac{T_{cw} - T_c^*}{T_c^*} \sim (\xi_f / \xi_s)^2, \tag{79}$$

which is around 1–5 % for typical values of ξ_f and ξ_s . A small width of the domain walls is expected in the experiments of Kinsey, Burnell, and Blamire (2001) on the critical current measurements of Nb/Co bilayers. The domain walls occurred to be responsible for the critical current enhancement below T_c^* =5.24±0.05 K. In the presence of domains walls the nonzero critical current has been observed at 5.4±0.05 K, slightly above T_c^* .

It is worth noting that the effect of increasing the critical temperature in the vicinity of a domain wall is weak for very large and very thin domain walls. The optimum thickness, when the effect may be relatively strong, is $w \sim \xi_s$.

In the case of a perpendicular easy axis the branching of the domains occurs near the surface of magnetic film. If the scale of this branching is smaller than the superconducting coherence length, the effective exchange field is averaged, and the pair-breaking parameter will be strongly decreased. This mechanism has been proposed by Buzdin (1985) to explain superconductivity at low temperature in reentrant ferromagnetic superconductors. A similar effect takes place in S/F bilayers and in such a case the superconductivity would be extremely sensitive to the domain structure. A rather weak magnetic field would suffice to modify the branching of domains and suppress superconductivity.

Up to now we have concentrated on the interplay between superconductivity and ferromagnetism causing the passing of electrons across the S/F interface known as the proximity effect. However, if the magnetic field created by the ferromagnet penetrates into a superconductor, it switches on the orbital mechanism of superconductivity and the magnetic interaction. The situation when superconductivity and the magnetic interaction occurs is only when the ferromagnet is an insulator, or the buffer oxide layer separates the superconductor and the ferromagnet. Hybrid S/F systems were studied in connection with controlled flux pinning. Enhancement of the critical current was observed experimentally for superconducting films with arrays of submicron magnetic dots and antidotes see, for example, Van Bael, Raedts, et al. (2002) and Van Bael, Van Look, et al. (2002) and references cited therein], and for S/F bilayers with a domain structure in ferromagnetic films (García-Santiago et al., 2000). A theory of vortex structures and pinning in S/F systems at rather low magnetic field has been elaborated on by Lyuksyutov and Pokrovsky (1998), Bulaevskii et al. (2000), Erdin et al. (2002), and Milosevic et al. (2002a). This subject is discussed in detail in the recent review by Lyuksyutov and Pokrovsky (2005).

Nucleation of superconductivity with the domain structure was theoretically studied by Aladyshkin *et al.* (2003) and Buzdin and Melnikov (2003) in the case of magnetic film with perpendicular anisotropy. The conditions for a superconductivity are more favorable near the domain walls. Recently domain-wall superconductivity was revealed in an experiment by Yang *et al.* (2004). They deposited on the single-crystal ferromagnetic BaFe₁₂O₁₉ substrate a 10-nm Si buffer layer and then a 50-nm Nb film. The strong magnetic anisotropy of BaFe₁₂O₁₉ assures that its magnetization is perpendicular to the Nb film. The very characteristic R(T) dependences and pronounced hysteresis effects were found in resistance measurements in an applied field.

A different situation occurs if the magnetization of the F layer lies in the plane (parallel magnetic anisotropy). Then any type of domain walls will be a source of the magnetic field for the adjacent S layer, and the domain wall locally weakens superconductivity. This idea was proposed by Sonin (1988) in a S layer to create a superconducting weak link (Josephson junction) attached to the domain wall.

Lange *et al.* (2003) used a nanoengineered lattice of magnetic dots on superconducting films for field-induced superconductivity. An applied external magnetic field provided the compensation of the magnetic field of the dots and increased the critical temperature. The idea of such a compensation effect was proposed a long time ago by Ginzburg (1956) for ferromagnetic superconductors.

The analysis of superconducting states appearing near the magnetic dots (when the upper critical field depends on the angular momentum of the superconducting nucleus wave function) was done by Cheng and Fertig (1999) and Milosevic *et al.* (2002b).



FIG. 23. S/F bilayer with domain structure in the ferromagnetic layer. The period *D* of the domain structure $(D=2\pi/Q)$ is smaller than the superconducting coherence length ξ_s .

IX. MODIFICATION OF FERROMAGNETIC ORDER BY SUPERCONDUCTIVITY

A. Effective exchange field in thin S/F bilayers

The influence of ferromagnetism on superconductivity is strong, and it leads to many experimentally observed consequences. Is the inverse also true? In other words, can superconductivity affect or even destroy ferromagnetism? To address this equation, we start by comparing the characteristic energy scales for superconducting and magnetic transitions. The energy gain per atom at the magnetic transition is of the order of the Curie temperature Θ . On the other hand, the condensation energy per electron at the superconducting transition [Eq. (2)] is much smaller than T_c , and it is only about $\sim T_c(T_c/E_F)$ $\ll T_c$. Usually the Curie temperature is higher than T_c and ferromagnetism appears to be much more robust compared with superconductivity. Therefore superconductivity can hardly destroy the ferromagnetism, but it may, nevertheless, modify it, if such modification does not cost too much energy. As an example consider bulk ferromagnetic superconductors ErRh₄B₄, HoMo₆S₈, and $HoMo_6Se_8$, where, in the superconducting phase, ferromagnetism is transformed into a domain phase with the domain size smaller than the superconducting coherence length ξ_s (Maple and Fisher, 1982; Bulaevskii *et al.*, 1985). A similar effect was predicted by Buzdin and Bulaevskii (1988) for a thin ferromagnetic film on a superconductor. To illustrate this effect, we consider the S/F bilayer with the S layer thickness d_s smaller than the superconducting coherence length ξ_s and the F layer thickness $d_f \ll \xi_f \ll d_s$; see Fig. 23.

In the case of a transparent S/F interface, the pairbreaking parameter is given by Eq. (40), and

$$\tau_{s,0}^{-1}(\omega > 0) = i\hbar \frac{D_s}{D_f} \frac{d_f}{d_s} \frac{\sigma_f}{\sigma_s},\tag{80}$$

which means that the effective exchange field in the superconductor is $\tilde{h} \approx h(d_f/d_s)[(D_s/D_f)\sigma_f/\sigma_s]$. The condition of a transparent interface implies that the Fermi momenta are equal in both materials and this permits us to write the effective field as

$$\tilde{h} = h(d_f/d_s)v_{Fs}/v_{Ff},\tag{81}$$

where v_{Fs} and v_{Ff} are the Fermi velocities in S and F layers, respectively. Note, however, that for strong ferromagnets the condition for perfect transparency of the interface is different, $v_{F\uparrow}v_{F\downarrow}=v_s^2$, where $v_{F\uparrow}$ and $v_{F\downarrow}$ are the Fermi velocities for two spin polarizations in a ferromagnet (Zutic and Valls, 1999; Zutic *et al.*, 2004).

In fact, in the case considered of thin F and S layers the situation is analogous to magnetic superconductors with an effective exchange field \tilde{h} , which may also depend on the coordinates (y, z) in the plane of bilayer. Let us demonstrate this important point. Keeping in mind the domain structure (see Fig. 23), where the exchange field depends only on the z coordinate, we may write the Usadel equations in the F and S layers,

$$-\frac{D_f}{2} \left[G \left(F + \frac{\partial^2}{\partial z^2} F \right) - F \left(\frac{\partial^2}{\partial x^2} G + \frac{\partial^2}{\partial z^2} G \right) \right] + \left[\omega + ih(z) \right] F = 0, \qquad (82)$$

$$-\frac{D_s}{2} \left[G\left(\frac{\partial^2}{\partial x^2}F + \frac{\partial^2}{\partial z^2}F\right) - F\left(\frac{\partial^2}{\partial x^2}G + \frac{\partial^2}{\partial z^2}G\right) \right] + \omega F$$
$$= \Delta G. \tag{83}$$

Now let us perform the averaging procedure by integrating these equations over x. Due to the small thickness of the F and S layers, the Green's functions G and F vary little with x and may be considered as constants. The integration of the terms with the second derivatives on xwill generate $\partial F/\partial x$ and $\partial G/\partial x$ terms taken at the interfaces. At the interfaces with vacuum these derivatives vanish and the boundary conditions (32) permit us to rely on the derivatives of F function on both sides of the S/F interface [the same relation is true for the G function, due to the normalization condition Eq. (A6)]. Excluding the derivatives $(\partial F/\partial x)_{d_s}$ and $(\partial G/\partial x)_{d_s}$, we obtain the standard Usadel equation but for the averaged (over the S layer thickness) Green's functions \bar{F} and \bar{G} ,

$$\left[\omega + i\tilde{h}(z)\right]\bar{F} - \frac{D_s}{2}\left[\bar{G}\frac{\partial^2}{\partial z^2}\bar{F} - \bar{F}\frac{\partial^2}{\partial z^2}\bar{G}\right] = \Delta\bar{G},\qquad(84)$$

where the effective field is $\tilde{h}(z) = h(z)(d_f/d_s)(D_s/D_f)\sigma_f/\sigma_s = h(d_f/d_s)v_{Fs}/v_{Ff}$ and the condition $d_f/d_s \ll 1$ is used to neglect the small renormalization of D_s and ω . Introducing an effective field $\tilde{h}(z)$ in the case of a thin bilayer is quite natural and rather general. The same effective field may be introduced in the framework of the Eilenberger equations.

B. Domain structure

In the case of the uniform ferromagnetic ordering in the F layer, superconductivity can exist only if \tilde{h} does not exceed the paramagnetic limit: $\tilde{h} < 1.24T_c$. This means that the thickness of the F layer must be extremely small $d_f < (T_c/h)d_s$; even for $d_s \sim \xi_s$, taking $T_c \sim 10$ K and $h \sim 5000$ K, the maximum thickness of the F layer is only around 1 nm. However, ferromagnetic superconductors (Maple and Fisher, 1982; Bulaevskii *et al.*, 1985) provide an example of domain coexistence phases with an exchange field larger than the paramagnetic limit.

We apply the theory of magnetic superconductors (Bulaevskii et al., 1985) to the description of the domain structure with wave vector $Q \ge \xi_s^{-1}$ in the S/F bilayer; see Fig. 23. The pair-breaking parameter associated with the domain structure is $\tau_s^{-1} \sim \tilde{h}^2 / vQ$ (Bulaevskii *et al.*, 1985), where $v = v_{Fs}$ is the Fermi velocity in the S layer. Let us write the domain-wall energy per unit area as $\sigma/\pi a^2$, where a is the interatomic distance. The domain-wall energy in the F film per unit length of the wall will be $d_f(\sigma/\pi a^2)$. Note that we consider the case of relatively small domain-wall thickness $w \ll Q^{-1} \ll \xi_s$ and constant σ , where the domain-wall energy is of the order of Curie temperature Θ for an atomic thickness domain wall but may be smaller for thicker domain walls. The change of the superconducting condensation energy density due to the pair-breaking effect of the domain structure is of the order of $N(0)\Delta^2/\Delta\tau_s$. Therefore, the density (per unit area) of the energy $E_{\rm DS}$ related to the domain structure reads

$$E_{\rm DS} \sim N(0) d_s \Delta \frac{\tilde{h}^2}{vQ} + d_f \frac{\sigma Q}{a^2}.$$
(85)

Its minimum is reached at

$$Q^2 = \frac{d_s}{d_f} \frac{N(0)\Delta a^2 \tilde{h}^2}{\sigma v} \sim \frac{1}{a\xi_0} \frac{d_s}{d_f} \frac{\tilde{h}^2}{\sigma E_F},$$
(86)

where $\xi_0 = \hbar v / \pi \Delta$. The factor which favors the existence of the domain structure is the superconducting condensation energy $E_s \sim -N(0)d_s\Delta^2$ per unit area. The domain structure decreases the total energy of the system if $E_{DS}+E_s < 0$, and we obtain the following condition:

$$T_c \gtrsim (\tilde{h}^2 \sigma d_f / d_s)^{1/3} = \tilde{h} (\sigma / h)^{1/3}.$$
 (87)

Due to the small factor $\sigma/h \ll 1$ this condition is less restrictive than the paramagnetic limit $(T_c > 0.66\tilde{h})$. Nevertheless, the conditions for the formation of the domain structure remain rather stringent. To minimize the d_f/d_s ratio (and the effective exchange field) it is better to choose the largest possible d_s thickness. However, the maximum thickness of the region, where superconductivity will be affected by the presence of the F layer, is of the order of ξ_s . Then, even in the case of the bulk superconductor $d_s^{\max} \sim \xi_s$, the condition of the domain-phase formation reads

$$T_c \gtrsim h \frac{d_f}{\xi_s} (\sigma/h)^{1/3}.$$
(88)

We conclude that for the domain-phase observation it is better to choose a superconductor with a large coherence length ξ_s and a ferromagnet with low Curie temperature and small energy of the domain walls.

The transition into the domain state is a first-order one, and as all transitions related to the domain walls, it would be highly hysteretic. This circumstance may complicate its experimental observation. To overcome this difficulty, it may be helpful to fabricate the S/F bilayer with a ferromagnet with a low Curie temperature Θ $< T_c$. In such a case, initially we expect the appearance of nonuniform magnetic structure below Θ . This system would be analogous to ferromagnetic superconductors ErRh₄B₄, HoMo₆S₈, and HoMo₆Se₈.

Bergeret et al. (2000) argued that the appearance of a nonhomogeneous magnetic order in a F film deposited on the bulk superconductor occurs via a second-order transition and the period of the structure goes to infinity at the critical point. They considered the helicoidal magnetic structure with a wave vector Q and the magnetic moment lying in the plane of the film. The increase of the magnetic energy due to the rotation of the moments was taken to be proportional to Q^2 . However, the magnetic structure considered is known to generate a magnetic field at a distance $\sim Q^{-1}$ from the film. The contribution coming from this field produces a magnetic energy proportional to Q and not to Q^2 in a small wavevector regime. This circumstance qualitatively changes the conclusions of Bergeret et al. (2000) and makes the transition into a nonhomogeneous magnetic state a firstorder one.

The experiments of Mühge *et al.* (1998) on ferromagnetic resonance measurements in Fe/Nb bilayers revealed a decrease of the effective magnetization below T_c for bilayers with $d_f < 1$ nm. This thickness is compatible with the estimate (88), but the analysis of the experimental data by Garifullin (2002) reveals the possibility of the formation of islands at a small thickness of the Fe layer, which may complicate the interpretation of the experimental results.

C. Negative domain-wall energy

In the previous analysis, the energy of the domain walls was considered to be constant independent of the presence of the superconducting layer. It is a good approximation for a thin domain wall $w \ll \xi_s$. However, superconductivity localized near the domain walls occurs for the local enhancement of the superconducting condensation energy, which may give a negative contribution to the domain-wall energy. We estimate this effect for a thick $w \ge \xi_s$ domain wall. The effect is maximum for the S/F bilayer with the relative variation of the criti-

cal temperature $(T_c - T_c^*)T_c \sim 1$ at $d_s \sim \xi_s$. We will assume these conditions to be satisfied. Following the same reasoning as in domain-wall superconductivity, we estimate the relative local decrease of the pair-breaking parameter as $\delta(\tau_s^{-1})/\tau_s^{-1} \sim (\xi_s/w)^2$. Therefore, the local negative contribution to the domain-wall energy (per unit length) resulting from the superconductivity reads as

$$\delta E_s \sim -N(0)\Delta^2(\xi_s/w)^2 w d_s. \tag{89}$$

The proper magnetic energy of the domain wall is $E_{\rm DW} \sim d_f(\sigma/\pi a^2)$, and for a large domain wall $\sigma \sim \Theta(a/w)$. The condition of the vanishing of the total energy of the domain wall $\delta E_s + E_{\rm DW} = 0$ gives

$$\frac{T_c^2}{E_F}\frac{\xi_s^3}{wa} \sim d_f \sigma \sim \Theta \frac{a}{w} d_f, \tag{90}$$

where the estimate $d_s \sim \xi_s$ is used. Finally, we conclude that the energy of the domain wall may be negative for the system with

$$T_c \gtrsim \Theta \frac{a}{l} \frac{d_f}{\xi_s},\tag{91}$$

where *l* is the electron mean free path. We have taken into account that $\xi_s \sim \sqrt{\xi_0 l}$ and $a/\xi_0 \sim T_c/E_F$. If the condition (91) is fulfilled, the following scenario emerges. The temperature decrease below T_c^* will decrease the energy of the domain walls, which are usually present in a ferromagnet. The concentration of the domain walls will increase and finally, when the domain-wall energy changes sign, a dense domain structure appears. The average distance between domains walls in such a structure would be of the order of the domain-wall thickness itself. Note that for the case of small domain wall thickness the superconducting contribution to its energy is negligible and instead of Eq. (91) we obtain the nonrealistic condition $T_c \ge \Theta(d_f / \xi_f) \xi_s / l$. We have taken into account only the exchange mechanism of the interaction between magnetism and superconductivity. The orbital effect gives an opposite contribution to the domain-wall energy, as a result of the out-of-plane magnetic field near the domain wall, which generates screening currents in the superconducting layer.

At present, there are no clear experimental evidences for the domain structure formation in S/F bilayers. The experiments of Mühge *et al.* (1998) on ferromagnetic resonance measurements in Fe/Nb bilayers presented a decrease of the effective magnetization below T_c^* for bilayers with $d_f < 1$ nm. This thickness is compatible with the estimate (88), but the magnetic moment decreases continuously below T_c^* . In addition, the analysis of the experimental data by Garifullin (2002) reveals the possibility of the formation of islands at a small thickness of the iron layer thus reducing its magnetic stiffness. The condition (91) is apparently fulfilled in the experiments of Mühge *et al.* (1998). Therefore, the decrease of the domain-wall energy may be at the origin of the observed effect.



FIG. 24. The ferromagnetic film with perpendicular anisotropy on a superconducting substrate.

D. Ferromagnetic film on a superconducting substrate

Bulaevskii and Chudnovsky (2000) and Bulaevskii *et al.* (2002) demonstrated that the pure orbital effect could decrease the equilibrium domain width in a ferromagnetic film on the superconducting substrate. The ferromagnet with a perpendicular magnetic anisotropy is either an insulator or separated from the superconductor by a thin insulating (e.g., oxide) layer; see Fig. 24.

In such a case the ferromagnetic film and the superconductor are coupled only by the magnetic field. It is well known (Landau and Lifshitz, 1982) that the positive energy of the magnetic field favors small domains, so that the stray field does not spread at large distances. On the other hand, the positive domain-wall energy favors a large domain size. The balance of these two contributions gives the equilibrium domain width $l_N \sim \sqrt{wd_f}$. In the presence of a superconductor, the screening currents modify the distribution of the magnetic field near the S/F interface and give an additional positive contribution to the energy of the magnetic field. This results in a shrinkage of the domain width. The energy E_D of the domain structure on the superconducting substrate reads (Bulaevskii and Chudnovsky, 2000; Bulaevskii et al., 2002)

$$E_D \sim 3\bar{l} + \frac{2\bar{l}_N^2}{\bar{l}} - \frac{16\bar{l}}{7\zeta(3)} \times \sum_{k\ge 0} \frac{1}{(2k+1)^2 [2k+1+\sqrt{(2k+1)^2+16\bar{l}^2}]}.$$
(92)

Here $\bar{l}=l/4\pi\lambda$ and $\bar{l}_N=l_N/4\pi\lambda$ are the reduced widths of domains on a superconducting and normal substrate, respectively, and λ is the London penetration depth. The minimization of E_D over \bar{l} gives the equilibrium width of domains. In the limit $\lambda \to \infty$ the influence of superconductivity vanishes and $l=l_N$. The limit $\lambda \to 0$, when the magnetic field does not penetrate inside the superconductor, was considered by Sonin (2002). In this limit the shrinkage of the domain widths is maximum and l Helseth *et al.* (2002) studied the change of the Bloch domain-wall structure in a ferromagnetic thin film on a superconducting substrate with the in-plane magnetization of the domains. It occurs that the wall experiences a small shrinkage, which corresponds to an increase of the energy of the domain wall.

Recently, Dubonos et al. (2002) demonstrated experimentally the influence of the superconducting transition on the distribution of the magnetic domains in mesoscopic ferromagnet-superconductor structures. This finding makes the observation of the effect predicted by Bulaevskii and Chudnovsky (2000) and Bulaevskii et al. (2002) plausible. Rearrangement of the domains normally results in a resistance change in metallic ferromagnets. In this context Dubonos et al. (2002) noted that domain walls' displacement due to the superconducting transition could be long-range resistive proximity effects previously observed in mesoscopic Ni/Al structures (Petrashov et al., 1999) and Co/Al structures (Giroud et al., 1998). Note also that Aumentado and Chandrasekhar (2001) studied the electron transport in a submicron ferromagnet (Ni) in contact with a mesoscopic superconductor (Al) and demonstrated that the interface resistance is very sensitive to the magnetic state of the ferromagnetic particle.

X. CONCLUSIONS

The most striking peculiarity of the proximity effect between a superconductor and ferromagnet produces damped oscillatory behavior of the Cooper pair wave function in the ferromagnet. This results in a nonmonotonic dependence of the critical temperature of S/F multilayers on the F layer thickness, as well as in the formation of π junctions in S/F/S interfaces. The minimum energy of the π junction is realized for the phase difference $\pm \pi$, and a spontaneous supercurrent may appear in a circuit containing the π junction. Two possible directions of the supercurrent reflect the doubly degenerate ground state. In contrast to the usual junction such a state is achieved without an external applied field. The qubit (or quantum bit) is the analog of a bit for quantum computation, described by a state in a two-level quantum system (Nielsen and Chuang, 2000). Superconductor/ferromagnet systems present a way to create an environmentally decoupled (so-called "quiet") qubit (Ioffe et al., 1999) using a S/F/S junction.

The π junctions permit a realization of complementary logic. In the metal-oxide semiconductor logic family the combination of the semiconducting *n*-*p*-*n* junctions with the complementary *p*-*n*-*p* junctions allows a significant simplification of the circuitry. The same is possible for Josephson-junctions' devices and circuits when π junctions are used (Terzioglu and Beasley, 1998). The logic cells with π junctions play the role of complementary devices to the usual Josephson logic cells.

Recently, Ustinov and Kaplunenko (2003) proposed using the π junction as a phase shifter in rapid singleflux quantum circuits. The relatively large geometrical inductance, which is required by the single-flux quantum storage, may be replaced by the much smaller π junction. The advantage of using π junctions is the ability to scale the dimension of superconducting logic circuits down to the submicron size. In addition, the use of the π junction as a phase shifter substantially increases the parameter margins of the circuits.

As discussed in Sec. III.D, the exchange interaction strongly affects Andreev reflection at the F/S interface providing a powerful tool to probe ferromagnets and measure their spin polarization.

The structures consisting of 0 and π Josephson junctions exhibit quite unusual properties. Bulaevskii *et al.* (1978) demonstrated that a spontaneous vortex carrying flux $\Phi_0/2$ appears at the boundary between 0 and π junctions [see also Xu *et al.* (1995) and Goldobin *et al.* (2002)]. A periodic structure consisting of small alternating 0 and π Josephson junctions will have an equilibrium averaged phase difference φ_0 value in the interval $-\pi$ $<\varphi_0 < \pi$, depending on the ratio of the 0 and π junction lengths (Mints, 1998; Buzdin and Koshelev, 2003). Superconductor/ferromagnet heterostructures provide the possibility of the realization of such φ junction with a very special two-maxima current-phase relation and Josephson vortices carrying partial fluxes $\Phi_0(\varphi_0/\pi)$ and $\Phi_0(1-\varphi_0/\pi)$.

The possibility to combine in a controlled manner paramagnetic and orbital interaction mechanisms between superconductivity and magnetism makes the physics of S/F heterostructures quite rich and promising for potential applications. Also the recent observation of strong vortex pinning in S/F hybrid structures, the spinvalve effect in F/S/F systems, and domain-wall superconductivity provide a good perspective to the creation of new electronics devices. Recent progress in controlling the fabrication of high-quality heterostructures and interfaces was crucial in this domain. Further development on microfabrication technology will permit one to expect other interesting findings in the near future.

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APPENDIX

1. Bogoliubov-de Gennes equations

Since the characteristic length of the induced superconductivity variation in a ferromagnet is small compared with a superconducting length, this implies using the microscopic theory of superconductivity to describe the proximity effect in S/F structures. A microscopical approach to study superconducting properties in the ballistic regime (the clean limit) in spatially varying fields is the use of the Bogoliubov-de Gennes equations (de Gennes, 1966a). The equations for electron and hole wave functions $u_1(\mathbf{r})$ and $v_1(\mathbf{r})$ are

$$[H_0 - h(\mathbf{r})]u_{\uparrow}(\mathbf{r}) + \Delta(\mathbf{r})v_{\downarrow}(\mathbf{r}) = E_{\uparrow}u_{\uparrow}(\mathbf{r}),$$

$$\Delta^*(\mathbf{r})u_{\uparrow}(\mathbf{r}) - [H_0 + h(\mathbf{r})]v_{\downarrow}(\mathbf{r}) = E_{\uparrow}v_{\downarrow}(\mathbf{r}), \qquad (A1)$$

where E_{\uparrow} is the quasiparticle excitation energy, $H_0 = -\hbar^2 (\nabla^2 / 2m) - E_F$ is the single-particle Hamiltonian, $h(\mathbf{r})$ is the exchange field in the ferromagnet, and the spin quantization axis is chosen along its direction. Equations for the wave functions with opposite spin orientation $u_{\downarrow}(\mathbf{r})$ and $v_{\uparrow}(\mathbf{r})$ and the excitation energy E_{\downarrow} are obtained from Eq. (A1) with the substitution $h \rightarrow -h$. Note that the solution $(u_{\downarrow}, v_{\uparrow})$ with energy E_{\downarrow} may be obtained from the solution of Eq. (A1), if we choose $u_{\downarrow} = v_{\uparrow}, v_{\uparrow} = -u_{\downarrow}$, and $E_{\downarrow} = -E_{\uparrow}$. The pair potential in the superconductor is determined by the self-consistent equation

$$\Delta(\mathbf{r}) = \lambda \sum_{E_{\uparrow}>0} u_{\uparrow}(\mathbf{r}) v_{\downarrow}^{*}(\mathbf{r}) [1 - 2f(E_{\uparrow})], \qquad (A2)$$

where f(E) is the Fermi distribution function $f(E) = 1/[1 + \exp(E/T)]$, and λ is the BCS coupling constant.

Assuming that the Cooper pairing is absent in the ferromagnet, we have $\Delta(\mathbf{r})=0$. This occurs when analytical solutions of the Bogoliubov-de Gennes equations are obtained with spatially varying pair potentials which are very rare. However, these equations provide a good basis for the numerical calculations to treat different aspects of S/N and S/F proximity effects.

2. Eilenberger and Usadel equations for ferromagnets

Another microscopical approach to the theory of superconductivity uses the electronic Green's functions. The Green's function technique for superconductors has been proposed by Gor'kov who introduced in addition to the normal Green's function $G(\mathbf{r}_1, \mathbf{r}_2)$ the anomalous (Gor'kov) function $F(\mathbf{r}_1, \mathbf{r}_2)$ (see, for example, Abrikosov *et al.*, 1975). This technique is a very powerful tool, but the corresponding Green's functions in a general case

appear to be rather complicated and oscillate as a function of the relative coordinate $\mathbf{r}_1 - \mathbf{r}_2$ on interatomic distances. On the other hand, the characteristic length scales for superconductivity in S/F systems are of the order of the layers thicknesses or damping decay length for the induced superconductivity and, then, they are much greater than the atomic length. This smooth variation is described by the center-of-mass coordinate $\mathbf{r} = (\mathbf{r}_1 + \mathbf{r}_2)/2$ in the Green's functions. The quasiclassical equations for the Green's functions averaged over rapid oscillations on the relative coordinate have been proposed by Eilenberger (1968) [and also by Larkin and Ovchinnikov (1968)].

The Eilenberger equations are transportlike equations for the energy-integrated Green's functions $f(\mathbf{r}, \omega, \mathbf{n})$ and $g(\mathbf{r}, \omega, \mathbf{n})$, depending on the center-of-mass coordinate \mathbf{r} , Matsubara frequencies $\omega = \pi T(2n+1)$, and the direction of the unit vector \mathbf{n} normal to the Fermi surface. For the case of S/F multilayers we restrict ourselves to situations when all quantities only depend on one coordinate x, chosen perpendicular to the layers. Introducing the angle θ between the \mathbf{x} axis and the direction of the vector \mathbf{n} (the direction of the Fermi velocity), we write the Eilenberger equations in the presence of an exchange field h(x) in the form [see, for example, Bulaevskii *et al.* (1985) and a recent review on the physics of Josephson junctions by Golubov *et al.* (2004)]

$$\begin{pmatrix} \omega + ih(x) + \frac{1}{2\tau}G(x,\omega) \end{pmatrix} f(x,\theta,\omega) + \frac{1}{2}v_F \cos \theta \frac{\partial f(x,\theta,\omega)}{\partial x}$$
$$= \left(\Delta(x) + \frac{1}{2\tau}F(x,\omega) \right) g(x,\theta,\omega),$$

$$G(x,\omega) = \int \frac{d\Omega}{4\pi} g(x,\theta,\omega), \quad F(x,\omega) = \int \frac{d\Omega}{4\pi} f(x,\theta,\omega),$$
$$f(x,\theta,\omega)f^{\dagger}(x,\theta,\omega) + g^{2}(x,\theta,\omega) = 1,$$
(A3)

where the function $f^+(x, \mathbf{n}, \omega)$ satisfies the same equation as $f(x, -\mathbf{n}, \omega)$ with $\Delta \rightarrow \Delta^*$ and the presence of impurities is described by the elastic scattering time $\tau = l/v_f$. The functions $G(x, \omega)$ and $F(x, \omega)$ are the Green's functions averaged over the Fermi surface. The Eilenberger equations are completed by the self-consistency equation for the pair potential $\Delta(x)$ in a superconducting layer:

$$\Delta(x) = \pi T \lambda \sum_{\omega} F(x, \omega). \tag{A4}$$

The BCS coupling constant λ is spatially independent in a superconducting layer, while in a ferromagnetic layer it is equal to zero. In a superconducting layer, the selfconsistency equation may also be written in the following convenient form:

$$\Delta(x) \ln \frac{T}{T_c} + \pi T \sum_{\omega} \left(\frac{\Delta(x)}{|\omega|} - F(x, \omega) \right) = 0,$$
 (A5)

where T_{c0} is the bare transition temperature of the superconducting layer in the absence of proximity effect.

Note that the Eilenberger equations as presented provide a natural choice for the spin quantization axis along the direction of the exchange field, and the only difference with the standard form of these equations is the substitution of the Matsubara frequency ω by $\omega + ih(x)$.

Usually, the electron-scattering mean free path in S/F/S systems is rather small. As such in the dirty limit, the angular dependence of the Green's functions is weak, and the Eilenberger equations can be replaced by the much simpler Usadel (1970) equations. In fact, the conditions required for using the Usadel equations are $T_c\tau \ll 1$ and $h\tau \ll 1$. The second condition is much more restrictive due to a large value of the exchange field ($h \gg T_c$). The Usadel equations only apply to Green's functions $G(x, \omega)$ and $F(x, \omega)$ averaged over the Fermi surface:

$$-\frac{D}{2}\left[G(x,\omega,h)\frac{\partial^2}{\partial x^2}F(x,\omega,h)\right]$$
$$-F(x,\omega,h)\frac{\partial^2}{\partial x^2}G(x,\omega,h)\left] + \left[\omega + ih(x)\right]F(x,\omega,h)$$
$$= \Delta(x)G(x,\omega,h),$$

$$G^{2}(x,\omega,h) + F(x,\omega,h)F^{*}(x,-h,\omega) = 1,$$
 (A6)

 $D = \frac{1}{3}v_F l$ is the diffusion coefficient which is different in the S and F regions and the equation for the function $F^+(x,h,\omega)$ is the same as for $F(x,\omega,h)$ with the substitution $\Delta \rightarrow \Delta^*$. Here the only difference with the standard form of the Usadel equations is the substitution ω by $\omega + ih(x)$.

The equations for the Green's functions in the F and S regions must be completed by the corresponding boundary conditions at the interfaces. For the Eilenberger equations they were derived by Zaitsev (1984) and for the Usadel equations by Kupriyanov and Lukichev (1988). These boundary conditions take into account the finite transparency (resistance) of the interfaces; see Eq. (32).

The most important pair-breaking mechanism in the ferromagnet is the exchange field h. However, disorder in the lattice of magnetic atoms creates the centers of magnetic scattering. In ferromagnetic alloys, used as the F layer in S/F/S Josephson junctions, the role of magnetic scattering may be quite important. Note that even in the case of a perfect ordering of the magnetic atoms, the spin waves will generate magnetic scattering. The natural choice of the spin-quantization axis used implicitly above is along the direction of the exchange field. The magnetic scattering and spin-orbit scattering mix up the up and down spin states. Therefore to describe this situation it is needed to introduce two normal Green's functions $G_1 \sim \langle \psi_{\uparrow} \psi_{\uparrow}^{+} \rangle$, $G_2 \sim \langle \psi_{\downarrow} \psi_{\downarrow}^{+} \rangle$ and two anomalous one $F_1 \sim \langle \psi_{\uparrow} \psi_{\downarrow} \rangle$, $F_2 \sim \langle \psi_{\uparrow} \psi_{\uparrow} \rangle$. The microscopical Green's function theory of superconductors with magnetic impurities and spin-orbit scattering was proposed by Abrikosov and Gorkov (1960, 1962). The generalization of the Usadel equations (A6) to this case gives

$$-\frac{D}{2}\left[G_1\frac{\partial^2}{\partial x^2}F_1 - F_1\frac{\partial^2}{\partial x^2}G_1\right] + \left[\omega + ih\right]$$
$$+ \left(\frac{1}{\tau_z} + \frac{2}{\tau_x}\right)G_1\left]F_1 + G_1(F_2 - F_1)\left(\frac{1}{\tau_x} - \frac{1}{\tau_{so}}\right)$$
$$+ F_1(G_2 - G_1)\left(\frac{1}{\tau_x} + \frac{1}{\tau_{so}}\right) = \Delta(x)G_1,$$

$$G_1^2(x,\,\omega\,,h) + F_1(x,\,\omega\,,h)F_1^+(x,h,\omega) = 1\,, \tag{A7}$$

and the similar equation for F_2 with the indices substitution $1 \leftrightarrow 2$. Here 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} describes the intensity of the magnetic scattering via exchange interaction and we follow the notation of the paper Fulde and Maki (1966). In the spatially uniform case the equations (A7) are equivalent to those of the Abrikosov-Gorkov theory (1960, 1962) (see also Fulde and Maki, 1966). Demler *et al.* (1997) analyzed the influence of the spin-orbit scattering on the critical temperature of the S/F multilayers and Krivoruchko and Petryuk (2002) on the critical current of SFIFS tunnel structures.

The ferromagnets used as F layers in S/F heterostructures reveal strong uniaxial anisotropy. Then the magnetic scattering in the plane (xy) perpendicular to the anisotropy axis is negligible. Moreover due to the relatively small atomic numbers of the F layer atoms the spin-orbit scattering is expected to be weak. In such a case there is no spin mixing scattering anymore and the Usadel equations retrieve the initial form (A6) with the substitution of the Matsubara frequencies by $\omega \rightarrow \omega$ $+G/\tau_s$, where $\tau_s^{-1} = \tau_z^{-1} \approx \tau_2^{-1} \langle S_z^2 \rangle / S^2$ may be considered as a phenomenological parameter describing the intensity of the magnetic scattering (Buzdin, 1985). The linearized Usadel equation in the ferromagnet reads

$$\left(|\omega| + ih \operatorname{sgn}(\omega) + \frac{1}{\tau_s}\right) F_f - \frac{D_f \partial^2 F_f}{2 \partial x^2} = 0.$$
 (A8)

If $\tau_s T_c \ll 1$, we may neglect $|\omega|$ in Eq. (A7) and the exponentially decaying solution has the form

$$F_f(x, \omega > 0) = A \exp[-x(k_1 + ik_2)],$$
(A9)

with $k_1 = (1/\xi_f)\sqrt{\sqrt{1+\alpha^2}+\alpha}$ and $k_2 = (1/\xi_f)\sqrt{\sqrt{1+\alpha^2}-\alpha}$, where $\alpha = 1/\tau_s h$. In the absence of magnetic scattering, the decaying and oscillating wave vectors are the same, $k_1=k_2$. Magnetic scattering decreases the characteristic decay length and increases the period of oscillations. In practice, this means that a decrease of the critical current of the S/F/S junction with an increase of d_f will be stronger. Note that the spin-orbit scattering (in contrast to magnetic scattering) decreases the pair-breaking effect of the exchange field (Demler *et al.*, 1997) and both scattering mechansims decrease the amplitude of the oscillations of the Cooper pair wave function. In some sense the spin-orbit scattering is more harmful for these oscillations because they completely disappear at τ_{so}^{-1} behavior of T'_c in S/F multilayers is an indirect proof of the weakness of the spin-orbit scattering.

The expression for the $I_c(2d_f)$ dependence (54) may be generalized to take into account magnetic scattering,

$$I_c R_n = 64 \frac{\pi T}{3} \operatorname{Re} \left[\sum_{\omega>0}^{\infty} \frac{2q_{\omega} y \exp(-2q_{\omega} y) \Phi_{\omega}}{\left[\sqrt{(1-\eta_{\omega}^2) \Phi_{\omega} + 1} + 1 \right]^2} \right],$$
(A10)

where

$$\Phi_{\omega} = \frac{\Delta^2}{(\Omega + \omega)^2}, \quad q_{\omega} = \sqrt{2i + 2\alpha + 2\omega/h}, \quad (A11)$$

$$\eta_{\omega}^2 = \frac{\alpha}{\alpha + i + \omega/h}.$$

Near T_c and in the limit $h \ge T_c$ and $2d_f k_2 \ge 1$ the following analytical expression can be obtained for the critical current:

$$I_{c} = \frac{\pi S \sigma_{f} \Delta^{2} k_{1}}{2 e T_{c}} \left[\cos(2d_{f}k_{2}) + \frac{k_{2}}{k_{1}} \sin(2d_{f}k_{2}) \right] \\ \times \exp(-2d_{f}k_{1}).$$
(A12)

We see that due to magnetic scattering the decay length of the critical current $\xi_{f1}=1/k_1$ may be substantially smaller than the oscillating length $\xi_{f2}=1/k_2$.

As noted above, the ability to use the Usadel equations, $h\tau \ll 1$, is rather restrictive in ferromagnets due to the large value of the exchange field. Therefore, it is of interest to retain in the Usadel equations the first correction in the parameter $h\tau$. The first attempts to calculate this correction were made by Proshin and Khusainov (1998) and Tagirov (1998) and resulted in the renormalization of the diffusion constant of the F layer $D_f \rightarrow D_f [1 - 2ih\tau \operatorname{sgn}(\omega)]$. Later on, a similar renormalization has been proposed by Baladié and Buzdin (2001) and Bergeret et al. (2001c). A critical analysis of this renormalization by Fominov et al. (2002) (see also Fominov, Kupriyanov, and Feigelman, 2003, and Khusainov and Proshin, 2003) revealed the inaccuracy of this renormalization, but did not provide the answer. The careful derivation of the Usadel equation for an F layer retaining the linear correction over $h\tau$ was made by Buzdin and Baladié (2003) and resulted in a somewhat different renormalization of the diffusion constant $D_f \rightarrow D_f [1]$ $-0.4ih\tau$ sgn(ω)]. The coefficient in $h\tau$ appears to be rather small which provides more confidence in the description of F layers using the Usadel equations. Note that this renormalization of the diffusion constant increases the decay characteristic length and decreases the period of oscillations, which is opposite to the influence of magnetic scattering.

The Usadel equations give a description of Green's functions only on average. Zyuzin *et al.* (2003) pointed out that, due to the mesoscopic fluctuations, the decay of the anomalous Green's function F_f at distances much

larger than ξ_f is not exponential. As a result, the Josephson effect in S/F/S systems may be observed even with a thick ferromagnetic layer.

The Eilenberger and Usadel equations adequately describe weak ferromagnets, where $h \ll E_F$ and the spin-up $v_{F\uparrow}$ and spin-down $v_{F\downarrow}$ Fermi velocities are the same. When the parameters of the electron spectra of the spin-up and spin-down bands are very different, the quasiclassical approach fails. However, if the characteristics of the spin bands are similar, the Eilenberger and Usadel equations are still applicable. Performing the derivation of the Eilenberger equation in such a case, it may be demonstrated that the Fermi velocity v_F in Eq. (A3) must be substituted by $(v_{F\uparrow} + v_{F\downarrow})/2$ and the scattering rate $1/\tau$ by $(1/\tau_{\uparrow}+1/\tau_{\downarrow})/2$. In consequence, the diffusion coefficient D_f in the Usadel equation becomes $(1/6)(v_{F\uparrow}+v_{F\downarrow})^2/(1/\tau_{\uparrow}+1/\tau_{\downarrow})$. Let us stress that such renormalization is justified only for close values of $v_{F\uparrow}$ and $v_{F|}$ (as well as τ_{\uparrow} and τ_{\downarrow}). Otherwise, the Bogoliubov-de Gennes equations must be used for the description of the proximity effect in strong ferromagnets.

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