Thermodynamic properties of ferromagnet/superconductor/ferromagnet nanostructures

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The theoretical description of the thermodynamic properties of ferromagnet/superconductor/ferromagnet ($F/S/F$) systems of nanoscopic scale is proposed. Their superconducting characteristics strongly depend on the mutual orientation of the ferromagnetic layers. In addition, depending on the transparency of S/F interfaces, the superconducting critical temperature can exhibit four different types of dependences on the thickness of the F layer. The obtained results permit one to give some practical recommendations for the spin-valve effect experimental observation. In this spin-valve sandwich, we also expect a spontaneous transition from a parallel to antiparallel ferromagnetic moment orientation, due to the gain in the superconducting condensation energy.

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I. INTRODUCTION

The peculiar character of the proximity effect in superconducting/ferromagnet (S/F) systems is due to the strong exchange field acting on the electrons in the ferromagnet and provoking the oscillatorylike behavior of the superconducting order parameter. Several interesting phenomena inherent to the S/F hybrid structures have been predicted and subsequently observed in experiments: there is nonmonotonic dependence of the critical temperature in S/F structures on the thickness of the ferromagnetic layer;[1–7] also see the review[8] on the competition between superconductivity and magnetism in ferromagnet/superconductor heterostructures, the π-junction realization in $S/F/S$ systems,[9–12] and the local quasiparticle density of states oscillation in S/F structures.[13–15]

In recent years, great progress has been achieved in the preparation of high-quality hybrid S/F systems, especially high-quality interfaces, which could be quite interesting for possible applications. In particular, a very promising system is the $F/S/F$ spin-valve sandwich, where spin-orientation-dependent superconductivity was predicted in Refs. 16–18.

In this paper, we present the results of detailed theoretical studies of the properties of $F/S/F$ systems containing a thin superconducting layer (compared to the superconducting coherence length). We analyze the influence of the F-layer thickness and the S/F interface transparency on the spin-valve superconductivity effect. The last part of the paper is devoted to the thermodynamic properties of the spin valve: we calculate the superconducting order parameter and the superconducting condensation energy for parallel and antiparallel spin orientations of the F layers. We also discuss the possibility of a spontaneous phase transition, by decreasing the temperature, from parallel to antiparallel spin orientations.

II. GENERAL EQUATIONS

We will concentrate on studies of the properties of an $F/S/F$ trilayer system with F layers of thickness d_f and an S layer of thickness d_s; see Fig. 1. Assuming that dirty limit conditions hold in all layers, we may use the complete set of Usadel equations[19] in the superconducting layer and in the F layers. In the superconducting layer the Usadel Green functions F and G satisfy

$$-\frac{D_s}{2}\vec{\nabla}[G(x,\omega)\vec{\nabla}F(x,\omega)-F(x,\omega)\vec{\nabla}G(x,\omega)]+\omega F(x,\omega)=\Delta(x)G(x); \tag{1}$$

in F layers they verify[14]

$$(\omega+ih)F(x,\omega)-\frac{D_f}{2}\vec{\nabla}[G(x,\omega)\vec{\nabla}F(x,\omega)-F(x,\omega)\vec{\nabla}G(x,\omega)]=0; \tag{2}$$

and in both layers,

$$G^2(x,\omega)+F(x,\omega)F^*(x,-\omega)=1, \tag{3}$$

where D_s and D_f are the diffusion coefficients in the S and F layers, respectively. $\omega=2\pi T(n+1/2)$ are the Matsubara frequencies and $h(x)$ is the exchange field in the F layers. In the case of a parallel orientation of the magnetization of the F layers, the exchange field is $h(x)=h$ for $x<-d_s/2$ and $x>d_s/2$ whereas in the antiparallel case $h(x)=h$ for x

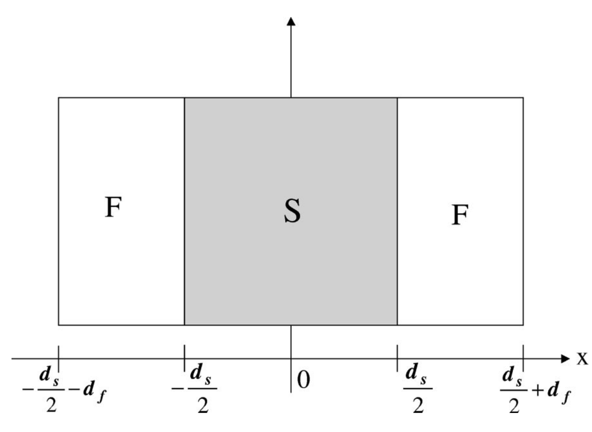

FIG. 1. Geometry of the $F/S/F$ sandwich. The thickness of the S layer is d_s, and d_f is the thickness of the F layers.

$>d_s/2$ and $h(x)=-h$ for $x<-d_s/2$. The Usadel equations are completed by the self-consistency equation in the form[20]

$$\Delta \ln\frac{T}{T_c}+\pi T\sum_{\omega}\left(\frac{\Delta}{|\omega|}-F_s\right)=0, \tag{4}$$

and by the boundary conditions at the S/F boundaries,[21]

$$\frac{\partial F_s}{\partial x}=\gamma\frac{\partial F_f}{\partial x},$$

$$F_s=F_f\pm\xi_f\gamma_B\frac{\partial F_f}{\partial x}, \tag{5}$$

where $\gamma=\sigma_s/\sigma_f$, σ_f (σ_s) is the conductivity of the F layer (the S layer above T_c), $\xi_f=\sqrt{D_f/2h}$, $\xi_s=\sqrt{D_s/2T_c}$ is the superconducting coherence length of the S layer, and the parameter $\gamma_B=R_b\sigma_f/\xi_f$, where R_b is the S/F boundary resistance per unit area. In the second boundary condition, the sign before the spatial derivative of F_f depends on the relative orientation of the x axis and the normal of the ferromagnet surface. If the normal is parallel to the x axis ($x=d_s/2$) a minus sign is required; in the other case ($x=-d_s/2$), a plus sign is required. The parameter γ_B is directly related to the transparency of the interface $T=1/(1+\gamma_B)$.[22] The limit $T=0$ ($\gamma_B=\infty$) corresponds to a vanishingly small boundary transparency, and the limit $T=1$ ($\gamma_B=0$) corresponds to a perfectly transparent interface. At the interface between the vacuum and the ferromagnet, the boundary condition is simply written as $\partial F_f/\partial x=0$.[22]

III. USADEL EQUATIONS FOR A THIN SUPERCONDUCTING INTERLAYER

The mutual influence of the superconductivity and ferromagnetism reveals interesting effects for an S-layer thickness smaller than or of the order of magnitude of the superconducting coherence length ξ_s; otherwise we have practically independent bulk superconductor and ferromagnetic systems. In addition, the case $d_s\ll\xi_s$ has an analytical solution; that is the reason why we will suppose this condition to be satisfied in the following analysis. In this limit, the small spatial variations of the Green functions in the S layer can be taken into account by a simple expansion to the order x^2,

$$F_s=F_0\left(1+\alpha x+\frac{\beta}{2}x^2\right), \tag{6}$$

$$G_s=G_0\left(1+ax+\frac{b}{2}x^2\right), \tag{7}$$

where F_0 and G_0 are values of the anomalous and normal Green functions at the center of the S layer. Using Eqs. (1) and (3), we finally obtain an effective Usadel equation for thin superconducting layers:

$$\left[\omega-\frac{D_s\beta}{2G_0}-\frac{D_s\alpha^2F_0^2}{4G_0^3}\right]F_0=\Delta G_0. \tag{8}$$

The coefficients α and β in expression (8) have to be found by using the boundary conditions at the F/S interfaces. As may be easily demonstrated from the boundary conditions, the term containing α is smaller by a factor $(d_s/\xi_s)\ll 1$ than the term with β; consequently this term can be neglected. Thus, in our approximation of a thin S layer, the Usadel equations take the simple form

$$\left[\omega-\frac{D_s\beta}{2G_0}\right]F_0=\Delta G_0, \tag{9}$$

$$F_0^2+G_0^2=1, \tag{10}$$

where the coefficient β plays the role of a pair-breaking parameter. The boundary conditions on the function F_s, following from Eq. (6), are

$$(F_s'/F_s)_{-d_s/2}=\alpha-d_s\beta/2,$$

$$(F_s'/F_s)_{d_s/2}=\alpha+d_s\beta/2. \tag{11}$$

By adding and subtracting the previous equations we can find the coefficients α and β from the boundary conditions on F_s. It is easy to demonstrate that the ratios $(F_s'/F_s)_{-d_s/2}$ and $(F_s'/F_s)_{d_s/2}$ are directly related to the corresponding ratios in the ferromagnet, using boundary conditions (5):

$$(F_s'/F_s)_{\pm d_s/2}=\frac{\gamma(F_f'/F_f)_{\pm d_s/2}}{1\mp\xi_f\gamma_B(F_f'/F_f)_{\pm d_s/2}}. \tag{12}$$

In Sec. IV, we will determine the critical temperature of the S-layer under general transparency conditions at the S/F interfaces. In a second part we will study the thermodynamics of the $F/S/F$ structure at an arbitrary temperature, in the limit of high and low transparencies.

IV. SPIN ORIENTATION DEPENDENCE OF THE CRITICAL TEMPERATURE

Close to the critical temperature and assuming that the exchange field in the ferromagnet is sufficiently strong ($h\gg T_c$), the Usadel equation in the ferromagnet can be simplified as

$$-\frac{\partial^2F_f(x,\omega)}{\partial x^2}+\frac{2ih\,\mathrm{sgn}(\omega)}{D_f}F_f(x,\omega)=0 \tag{13}$$

Using the boundary condition at a vacuum interface, we readily find solutions for the Usadel Green functions in the parallel case (the upper script P refers to the parallel case) for positive ω (the case $\omega<0$ is obtained by making the substitution $k_n\rightarrow k_n^*$),

$$F_f^P(x>d_s/2)=A\cosh k_n[x-(d_f+d_s/2)],$$

$$F_f^P(x<-d_s/2)=B\cosh k_n[x+(d_f+d_s/2)], \tag{14}$$

with $k_n=(1+i)\sqrt{h/D_f}$. Analogously for the antiparallel case (the upper script A refers to the antiparallel case),

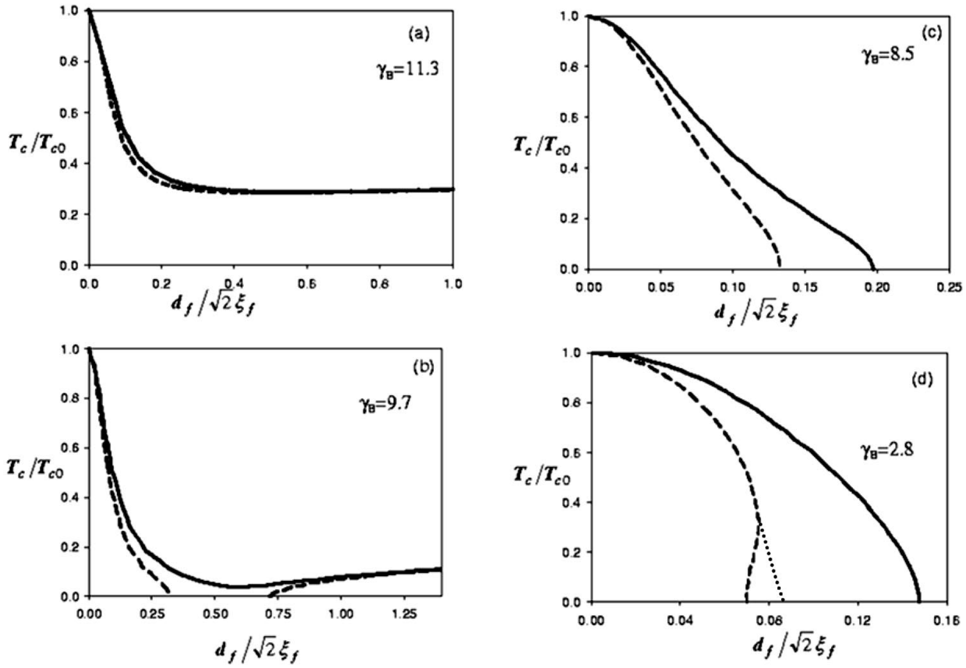

FIG. 2. Characteristic types of $T_c(d_f)$ behavior. The thickness of the F layer is normalized to the F-layer characteristic length ξ_f. The parameter $\pi\tau_0 T_{c0}$ is chosen to be constant and equal to 1. The full line corresponds to the antiparallel case, and the dashed line to the parallel case. One can distinguish four characteristic types of $T_c(d_f)$ behavior: (a) nonmonotonic decay to a finite value of 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 presents schematically the first order transition line.

$$F_f^A(x>d_s/2)=C\cosh k_n[x-(d_f+d_s/2)],$$

$$F_f^A(x<-d_s/2)=D\cosh k_n^*[x+(d_f+d_s/2)], \qquad (15)$$

These solutions immediately give the value of the ratios $(F_f'/F_f)_{\pm d_s/2}$ and, consequently [see Eq. (12)] the ratios $(F_s'/F_s)_{\pm d_s/2}$:

$$(F_s'/F_s)_{d_s/2}=-\frac{\gamma k_n \tanh(k_n d_f)}{1+\xi_f k_n \gamma_B \tanh(k_n d_f)},$$

$$(F_s'/F_s)^P_{-d_s/2}=-(F_s'/F_s)_{d_s/2},$$

$$(F_s'/F_s)^A_{-d_s/2}=\frac{\gamma k_n^* \tanh(k_n^* d_f)}{1+\xi_f k_n^* \gamma_B \tanh(k_n^* d_f)}. \qquad (16)$$

Then, with the help of Eq. (11), we may easily obtain the pair-breaking parameter β. Close to T_c, the Usadel equation may be linearized over F_0, and the normal Green function is $G_0=\mathrm{sgn}(\omega)$; thus Eq. (8) is simply written in first order of F_0 as

$$\left[|\omega|-\frac{D_s\beta}{2}\right]F_0=\Delta. \qquad (17)$$

Using this relation and the self-consistency equation [Eq. (4)], we can write down the expression for the critical temperature of the S layer in the general form

$$\ln\frac{T_c}{T_{c0}}=\Psi\left(\frac{1}{2}\right)-\mathrm{Re}\,\Psi\left\{\frac{1}{2}+\frac{1}{\tau(d_f)\pi T_c}\right\}, \qquad (18)$$

where T_{c0} is the critical temperature of the S layer without any proximity effect. This type of expression reminds one of the corresponding formula for the critical temperature of a superconductor with magnetic impurities,[23] though the "magnetic scattering time" τ may be complex in our system. It is easy to verify that in the parallel case, the effective magnetic scattering rate τ^{-1} is indeed complex and given by the expression

$$\tau^P(d_f)^{-1}=\tau_0^{-1}\frac{(1+i)\tanh(\tilde{d}_f)}{1+\tilde{\gamma}_B\tanh(\tilde{d}_f)}, \qquad (19)$$

and τ^{-1} in the antiparallel case is real,

$$\tau^A(d_f)^{-1}=\mathrm{Re}[\tau^P(d_f)^{-1}]; \qquad (20)$$

here $\tilde{d}_f=(1+i)(d_f/\sqrt{2}\xi_f)$, $\tilde{\gamma}_B=(1+i)(\gamma_B/\sqrt{2})$, and $\tau_0^{-1}=(\gamma T_{c0}/\sqrt{2})(\xi_s/d_s)(\xi_s/\xi_f)$. Note that for the parallel orientation case, the critical temperature must be the same as for an S/F bilayer with an S-layer thickness equal to $d_s/2$. The critical temperature of S/F bilayers was recently studied in Ref. 24 and in the limit $d_s \ll \xi_s$ the expression for T_c in Ref. 24 is indeed the same as Eq. (18), with $\tau=\tau^P$ and d_s replaced by $d_s/2$. In the limit of infinite F layers ($d_f\to\infty$) and infinite transparency of the interfaces ($\gamma_B\to 0$), expression (18) reproduces the results for T_c found previously in Refs. 16 and 17. If the proximity effect is weak, the parameter τ_0^{-1} goes to zero. Expanding the Digamma function about 1/2 yields the following result in this limit:

$$T_c^A=T_c^P=T_{c0}-\frac{\pi}{2}\tau^A(d_f)^{-1}. \qquad (21)$$

Thus for a weak proximity effect, the shift of the transition temperature is a linear function of τ_0^{-1} (here we find the same result as in the study of a superconducting alloy with magnetic impurities) and the difference between the critical temperatures of parallel and antiparallel orientation appears only at the order τ_0^{-3}.

The different kinds of obtained $T_c(d_f)$ curves, depending on the parameters of the trilayers, are presented in Fig. 2 for illustration. We plot several curves for various values of γ_B assuming that the parameter $\pi T_{c0}\tau_0$ is constant and equals to 1. We may notice four characteristic types of $T_c(d_f)$ behavior. The first one Fig. 2(a), at a small interface transparency T_c, decays slightly nonmonotonically to a finite value, and the critical temperature difference between both orientations is very small. The decay presents a minimum at a particular value of d_f of the order of magnitude of ξ_f. The second one [Fig. 2(b)], at a moderate interface transparency T_c exhibits a reentrant behavior; this means that the superconductivity vanishes in a certain interval of d_f. For special values of the parameter γ_B the reentrant behavior can be observed only for the parallel orientation. The reentrance of the superconductivity was been observed recently in Fe/V/Fe trilayers a with parallel orientation of the ferromagnetic moments by Tagirov *et al.*[25] The experiments[25] were performed for rather thick S-layers ($d_s \gtrsim \xi_s$), so a quantitative comparison with our theory is impossible. Unfortunately, even the qualitative comparison is difficult due to the absence of experimental data in Ref. 25 on the S/F boundary resistance, which could provide the estimate of the important parameter γ_B. We may, however, state that the characteristic thickness which corresponds to the minimum of T_c is of the order of magnitude of ξ_f and only slightly depends on the boundary transparency when the oscillatory behavior of $T_c(d_f)$ is present. In the third model [Fig. 2(c)], at a moderately high interface transparency, the critical temperature decays monotonically and vanishes at finite value of d_f. The last type of $T_c(d_f)$ behavior Fig. 2(d) is observed at an extremely high interface transparency and rather thin F layers with parallel orientations. Under these conditions the phase transition between the normal and superconducting a states presents a triple point at which the transition switch to a first-order one. This behavior is similar to the $T_c(h)$ dependance in bulk superconductor (see, for example, Ref. 26), where at low temperature the S/N transition becomes first order. Schematically, the line of the first order transition is presented in Fig. 2 by a dotted line.

In order to observe a significant spin-valve effect experimentally, it is crucial to choose the right materials and thicknesses of superconductor and F layers to maximize $\Delta T_c = T_c^P - T_c^A$. Equation (18) shows that the important parameters are γ_B, d_f, and τ_0^{-1}. The value of τ_0^{-1} is directly related to the choice of the superconductor and of the ferromagnet since it is proportional to γ the ratio of the conductivities. This parameter does not play a crucial role in the spin-valve effect, and a choice of τ_0^{-1} of around 1 should permit an easy observation of the effect. The choice of the thickness of the F layer can be rather important, as shown by the curves $T_c(d_f)$. Indeed, due to the additional boundary condition at the interface between the ferromagnet and the vacuum, there are some interferences between incoming and reflected Copper pairs in the F layer. Depending on the value of d_f, these interferences can be destructive or constructive, leading to a maximum or minimum of ΔT_c. Finally, the curves $T_c(d_f)$ (see Fig. 2) show that the key factor of the spin-valve effect is the transparency of the interface. For values of γ_B of around 1 the effect can be easily observed, whereas, if γ_B is an order of magnitude stronger, the effect almost disappears.

In our case, both F layers have the same thickness. The generalization to the case of F layers of arbitrary thickness d_{f1} (for $x<0$) and d_{f2} (for $x>0$) is straightforward using Eq. (16). In the parallel case we have to make the substitution $\tau^P(d_f)^{-1} \to \tau^P(d_{f1})^{-1} + \tau^P(d_{f2})^{-1}$, and in the antiparallel case we have to make the substitution $\tau^A(d_f)^{-1} \to [\tau^P(d_{f1})^{-1}]^* + \tau^P(d_{f2})^{-1}$.

V. THERMODYNAMIC PROPERTIES OF THE STRUCTURE

In this section, we will consider the temperature dependance of the superconducting order parameter and the superconducting condensation energy in $F/S/F$ systems. For simplicity, we concentrate on the case of F layers of thickness $d_f \gg \xi_f$, which corresponds in practice to $d_f \gg 50$ Å. Using the classical parametrization of the Usadel equation by $F = \sin\theta$ and $G = \cos\theta$, we may easily find the complex angle $\theta(x)$ in our limit of infinite F layers for a parallel orientation[14]

$$\theta_f^P(x>d_s/2) = 4\arctan\{\tan(\theta_0^P/4)\exp[-k_n(x-d_s/2)]\},$$

$$\theta_f^P(x<-d_s/2) = 4\arctan\{\tan(\theta_0^P/4)\exp[k_n(x+d_s/2)]\}, \tag{22}$$

and for the antiparallel one,

$$\theta_f^A(x>d_s/2) = 4\arctan\{\tan(\theta_0^A/4)\exp[-k_n(x-d_s/2)]\},$$

$$\theta_f^A(x<-d_s/2) = 4\arctan\{\tan(\theta_0^A/4)\exp[k_n^*(x+d_s/2)]\}, \tag{23}$$

where θ_0 is the complex angle describing the superconducting order parameter in an F layer at the S/F boundary. Note that we have assumed in the previous equations that ω is positive (the case $\omega<0$ is obtained by the substitution $k_n \to k_n^*$). These solutions give us immediately the ratios $(F_f'/F_f)_{\pm d_s/2}$ and so via boundary conditions (12) the ratios $(F_s'/F_s)_{\pm d_s/2}$ determining the pair-breaking parameter in the Usadel equations for the S layer:

$$(F_s'/F_s)_{d_s/2} = -\frac{\gamma k_n \cos\theta_0}{\cos\theta_0/2 + \widetilde{\gamma_B}\cos\theta_0},$$

$$(F_s'/F_s)^P_{-d_s/2} = -(F_s'/F_s)_{d_s/2},$$

$$(F_s'/F_s)^A_{-d_s/2} = \frac{\gamma k_n^* \cos\theta_0^A}{\cos\theta_0^A/2 + \widetilde{\gamma_B^*}\cos\theta_0^A}. \tag{24}$$

Using Eq. (11), we can deduce the coefficient β for the effective Usadel equations for both orientations ($\omega>0$):

$$\beta^P = -\frac{2\gamma k_n \cos\theta_0^P}{d_s(\cos\theta_0^P/2 + \widetilde{\gamma_B}\cos\theta_0^P)}, \tag{25}$$

$$\beta^A = -\left(\frac{\gamma k_n \cos\theta_0^A}{d_s(\cos\theta_0^A/2 + \widetilde{\gamma_B}\cos\theta_0^A)} + \text{c.c.}\right). \tag{26}$$

Equation (9) combined with Eqs. (25) and (26) gives in an implicit form the angle θ (and so the Usadel Green functions) as a function of the Matsubara frequencies and the superconducting order parameter Δ. Together with the self-consistency equation [Eq. (4)], this permits one, in principle to find the dependence of the superconducting order parameter on the temperature and all the thermodynamics of the $F/S/F$ system. Below, we will discuss two limiting cases which can be handled analytically: the low-temperature limit and temperatures close to T_c.

A. Low-temperature behavior

When the temperature goes to zero, we may substitute the integration by a summation over Matsubara frequencies ($\pi T\Sigma \rightarrow \int d\omega$) in the Usadel self-consistency equation for order parameter (4),

$$\Delta = \lambda N(0)\int_{-\omega_D}^{\omega_D} F(\omega)d\omega = \lambda N(0)\int_{-\omega_D}^{\omega_D}\sin\theta d\omega, \tag{27}$$

where ω_D of the order of magnitude of the Debye frequency is the usual cutoff in the BCS model (it will not enter in the final expressions), and λ is the BCS coupling constant. The integration over the Matsubara frequencies can be performed analytically when the transparency of the S/F interface is small and when it goes to infinity.

1. High transparency limit

In the high transparency limit ($\gamma_B \rightarrow 0$), the angle θ, characterizing superconductivity in the S layer, is the same as at the S/F interface, i.e., θ_0; see Eq. (5). Thus the Usadel equations, for parallel and antiparallel cases, become ($\omega > 0$)

$$\left(\omega + \frac{2(1+i)\tau_0^{-1}}{\cos\theta_0^P/2}\right)\sin\theta_0^P = \Delta\cos\theta_0^P, \tag{28}$$

$$\left(\omega + \frac{(1+i)\tau_0^{-1}}{\cos\theta_0^A/2} + \text{c.c.}\right)\sin\theta_0^A = \Delta\cos\theta_0^A. \tag{29}$$

Note that these equations are quite different from the corresponding expressions found in the case of a superconductor with magnetic impurities[23] and the analogy which worked for T_c is no longer applicable. Let us first consider the parallel case. Integral (27) can be performed analytically by changing the integration over ω by integration over θ,

$$\Delta^P = \lambda N(0)\int_{\Delta^P/\omega_D}^{\tilde{\theta}^P}\left[\frac{\Delta^P}{\sin^2\theta} + 2\tau_0^{-1}(1+i)\frac{\sin\theta/2}{\cos^2\theta/2}\right]\sin\theta d\theta + \text{c.c.}, \tag{30}$$

where τ_0^{-1} is given by $(\gamma T_{c0}/\sqrt{2})(\xi_s/d_s)(\xi_s/\xi_f)$, and $\tilde{\theta}^P$ is the solution of equation

$$\Delta^P\cos\tilde{\theta}^P = 4\tau_0^{-1}(1+i)\sin(\tilde{\theta}^P/2). \tag{31}$$

In the absence of F layers and at zero temperature, the order parameter Δ_0 verifies Eq. (30) with $\tau_0^{-1}=0$, i.e.,

$$\Delta_0 = 2\lambda N(0)\int_{\Delta_0/\omega_D}^{\pi/2}\frac{\Delta_0}{\sin\theta}d\theta = 2\lambda N(0)\Delta_0\left[-\ln\left(\tan\frac{\Delta_0}{2\omega_D}\right)\right]. \tag{32}$$

Combining Eqs. (30) and (32), we may eliminate the diverging terms when θ goes to zero and, finally, performing the remaining integration, we obtain the following explicit relation for the ratio Δ^P/Δ_0:

$$\ln\left(\frac{\Delta^P}{\Delta_0}\right) = \text{Re}\left\{\ln\tan(\tilde{\theta}^P/2) + 4\tau_0^{-1}\frac{(1+i)}{\Delta^P}\times\left[\ln\tan\left(\frac{\tilde{\theta}^P+\pi}{4}\right) - \sin(\tilde{\theta}^P/2)\right]\right\}. \tag{33}$$

Performing the same kind of calculation in the antiparallel case we have, for the ratio Δ^A/Δ_0,

$$\ln\left(\frac{\Delta^A}{\Delta_0}\right) = \ln\tan(\tilde{\theta}^A/2) + \frac{4\tau_0^{-1}}{\Delta^A}\left[\ln\tan\left(\frac{\tilde{\theta}^A+\pi}{4}\right) - \sin(\tilde{\theta}^A/2)\right], \tag{34}$$

where $\tilde{\theta}^A$ is the solution of

$$\Delta^A\cos\tilde{\theta}^A = 4\tau_0^{-1}\sin(\tilde{\theta}^A/2). \tag{35}$$

The density of states for one direction of spin is given by $N_\uparrow(\omega) = \frac{1}{2}N(0)\text{Re}[G(\omega\rightarrow i\omega)]$, where $N(0)$ is the total density of state in the normal state. Considering the limit $\omega=0$, this relation becomes $N_\uparrow(\omega) = \frac{1}{2}N(0)\text{Re}(\cos\tilde{\theta}^{A,P})$, where $\tilde{\theta}^P$ is given by Eq. (31) and $\tilde{\theta}^A$ by Eq. (35). An analytical study of Eqs. (31) and (35) shows that the real part of the solutions $\tilde{\theta}^{A,P}$ always exists; thus $N_\uparrow(\omega)$ is finite at $\omega=0$. As a result, at low temperatures and in both configurations, the superconductivity in $F/S/F$ systems should be a gapless one. Note however that all our calculations correspond to the case of strong ferromagnets ($h \gg T_c$). In the case of a weak exchange field ($h \ll T_c$), the gap in the density of states will be restored.

In Fig. 3, we plot the order parameter in both parallel and anti-parallel case as a function of the pair-breaking parameter $(\Delta_0\tau_0)^{-1}$. At a small exchange field or at a small conductivity ratio, there is almost no difference between Δ^P and Δ^A, and their evolution with $(\Delta_0\tau_0)^{-1}$ is linear as in the case of superconducting alloys containing magnetic impurities; however, the overall behavior in the whole temperature region is different. Naturally, the superconducting order parameter is always larger in the antiparallel case due to the partial compensation of the exchange field effect.

The thermodynamic potential (per unit area) for both orientations can be found by integrating Eqs. (33) and (34)

$$\Omega^P(h,\Delta)=d_s N(0)\left(\frac{\Delta^2}{2}\ln\frac{\Delta^2}{e\Delta_0^2}-\tau_0^{-2}\mathrm{Re}[if(X^P)]\right), \quad (36)$$

$$\Omega^A(h,\Delta)=d_s N(0)\left(\frac{\Delta^2}{2}\ln\frac{\Delta^2}{e\Delta_0^2}-\tau_0^{-2}f(X^A)\right), \quad (37)$$

where the function $f(X)$ is defined by

$$\frac{1}{2(2(X)^2-1)^2}\left[\begin{array}{c}-(X+1)^2(2X-1)^2\ln(1+X)-(X-1)^2(2X+1)^2\ln(1-X)\\ +2X^2\ln X+6X^2(2X^2-1)\end{array}\right], \quad (38)$$

and while $X^{A,P}=\sin(\widetilde{\theta}^{A,P}/2)$. Minimizing Eqs. (36) and (37) in respect to the order parameter at fixed exchange field gives back the self-consistency equations (33) and (34) determining $\Delta(h)$. Keeping in mind the fact that the free energy F of the system is equal to the thermodynamic potential when the order parameter is minimized, we have determined the difference of the free energy between the parallel and antiparallel configurations $F^P-F^A=\Omega^P(h,\Delta^P)-\Omega^A(h,\Delta^A)$. The analysis of Eq. (18) in the case of infinite F layers and a high transparency of the interfaces immediately shows that the superconducting transition temperature is going to zero for

$$(\Delta_0\tau_0)^{-1}=0.25 \quad \text{in the parallel case,} \quad (39)$$

$$(\Delta_0\tau_0)^{-1}=0.175 \quad \text{in the antiparallel case.} \quad (40)$$

These values naturally correspond to the order parameter vanishing in Fig. 3. As a result, F^P is equal to zero for $(\Delta_0\tau_0)^{-1}>0.25$ and F^A for $(\Delta_0\tau_0)^{-1}>0.175$. In Fig. 4, we plot the normalized expression of (F^P-F^A), by the free energy in the antiparallel configuration, as a function of the parameter $(\Delta_0\tau_0)^{-1}$. The expression (F^P-F^A) is always positive, and, in conclusion, the antiparallel configuration is always more stable than the parallel configuration.

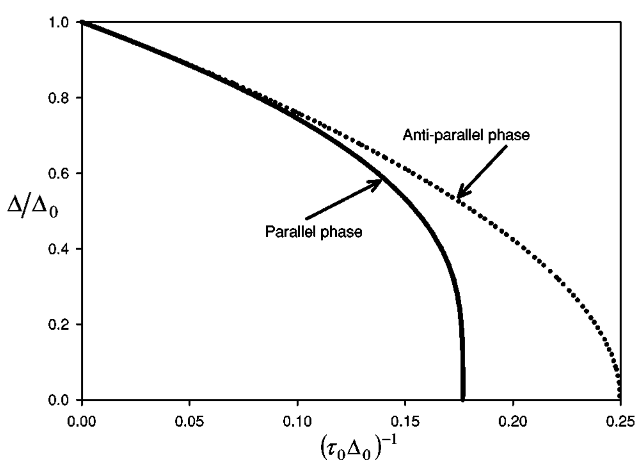

FIG. 3. The order parameter Δ normalized by its value in the absence of a proximity effect Δ_0 in both parallel and antiparallel cases as a function of the pair-breaking parameter $(\tau_0\Delta_0)^{-1}$.

2. Low transparency limit

In this limit, an expansion of Eqs. (25) and (26) with respect to $1/\gamma_B$ can be made. In the limit of low transparency $(\gamma_B\to\infty)$, the order parameter in the F layer almost completely disappears; thus the angle θ_0 describing the superconducting order parameter in the F layer at the S/F boundary is small $(\theta_0\ll 1)$. So, with the help of boundary conditions (5), we find that the angle θ, characterizing superconductivity in the S layer, is given in this limit by $\sin\theta=\theta_0\widetilde{\gamma_B}/2$. With this relation and Eq. (11), we can easily find the expression of the coefficient β in both configurations:

$$\beta^P=-\frac{\gamma}{\xi_f d_s\gamma_B}(1-\widetilde{\gamma_B}^{-1}+2\widetilde{\gamma_B}^{-2}), \quad (41)$$

$$\beta^{AP}=(\beta^P+\beta^{P*})/2. \quad (42)$$

The stability of both parallel and anti-parallel configurations of the FSF trilayer in the low transparency limit can also be studied by performing the integration over ω in the Usadel self-consistency equation (27). Here we only give the results of the corresponding calculations:

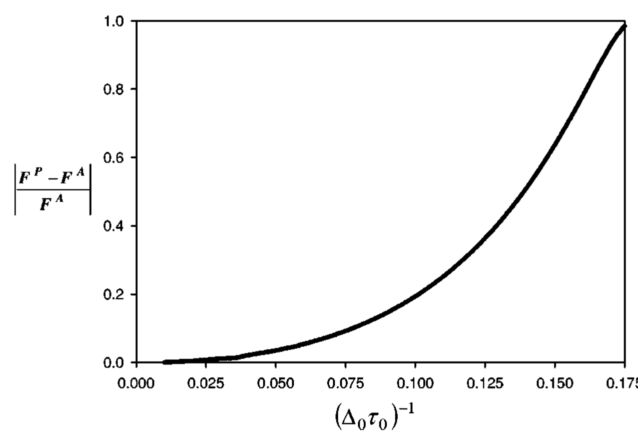

FIG. 4. Normalized value of the difference of free energy between the parallel and antiparallel configurations plotted as a function of the parameter $(\tau_0\Delta_0)^{-1}$. For $(\tau_0\Delta_0)^{-1}\geqslant 0.175$, the superconducting transition temperature is equal to zero in the parallel configuration.

$$\ln\left(\frac{\Delta^P}{\Delta_0}\right) = -\sqrt{\frac{2}{\Delta^P\tau_0\gamma_B}}\left(1-\frac{1}{2\sqrt{2}\gamma_B}\right)+\frac{\pi}{4\Delta^P\tau_0\gamma_B}\left(1-\frac{1}{\sqrt{2}\gamma_B}\right), \tag{43}$$

$$\ln\left(\frac{\Delta^A}{\Delta_0}\right) = -\sqrt{\frac{2}{\Delta^A\tau_0\gamma_B}}\left(1-\frac{1}{2\sqrt{2}\gamma_B}-\frac{1}{16\gamma_B^2}\right)+\frac{\pi}{4\Delta^A\tau_0\gamma_B}\left(1-\frac{1}{\sqrt{2}\gamma_B}\right). \tag{44}$$

Following the method presented in the previous paragraph, we obtain the expression for the thermodynamic potential,

$$\Omega^{A,P}(h,\Delta)=d_sN(0)\left(\frac{\Delta^2}{2}\ln\frac{\Delta^2}{e\Delta_0^2}+a^{A,P}\Delta^{3/2}-b^{A,P}\Delta\right), \tag{45}$$

where the coefficients

$$a^P=\frac{4}{3}\sqrt{\frac{2}{\tau_0\gamma_B}}\left(1-\frac{1}{2\sqrt{2}\gamma_B}\right),$$

$$a^A=\frac{4}{3}\sqrt{\frac{2}{\tau_0\gamma_B}}\left(1-\frac{1}{2\sqrt{2}\gamma_B}-\frac{1}{16\gamma_B^2}\right),$$

and

$$b^{A,P}=\frac{\pi}{2\tau_0\gamma_B}\left(1-\frac{1}{\sqrt{2}\gamma_B}\right).$$

The term containing γ_B^{-2} in a^A contributes to the stabilization of the antiparallel configuration compared to the parallel configuration. Although, as follows from Eqs. (43)–(45), the orientation dependent relative variation of the order parameter and condensation energy is very small:

$$\frac{\Delta^P-\Delta^A}{\Delta_0}\sim\frac{F^P-F^A}{F_0}\sim\frac{\gamma_B^{-5/2}}{\sqrt{\Delta_0\tau_0}}. \tag{46}$$

B. Free energy, entropy, and specific heat of the trilayer close to T_c

At the transition temperature, the order parameter Δ goes to zero, and the Green functions F and G go, respectively, to 0 and $\mathrm{sgn}(\omega)$. In the limit of high S/F interface transparency, we may use Eqs. (28) and (29) and develop all the quantities around T_c to obtain an expansion of F in powers of Δ,

$$F=\frac{\Delta}{|\omega|+\epsilon(\omega)}-\frac{\Delta^3}{2[|\omega|+\epsilon(\omega)]^3}-\frac{\Delta^3\epsilon(\omega)}{8[|\omega|+\epsilon(\omega)]^4}+o(\Delta^5), \tag{47}$$

where for the parallel case $\epsilon=\epsilon^P(\omega)=[2(1+i)\tau_0^{-1}]$ while in the parallel case $\epsilon=\epsilon^A(\omega)=[\epsilon^P(\omega)+\epsilon^P(\omega)^*]/2$. Thus, using expression (47) and the self-consistency equation, we may directly obtain the dependance of the order parameter with the temperature. In the antiparallel case we have

$$-\ln\left(\frac{T}{T_{c0}}\right)=\Psi\left(\frac{1}{2}+\frac{1}{\pi\tau_0T}\right)-\Psi\left(\frac{1}{2}\right)+\left(\frac{\Delta}{2\pi T}\right)^2g_1\left(\frac{1}{\pi\tau_0T}\right), \tag{48}$$

and in the parallel case

$$-\ln\left(\frac{T}{T_{c0}}\right)=\mathrm{Re}\,\Psi\left(\frac{1}{2}+\frac{(1+i)}{\pi\tau_0T}\right)-\Psi\left(\frac{1}{2}\right)+\left(\frac{\Delta}{2\pi T}\right)^2\mathrm{Re}\,g_1\left[\frac{(1+i)}{\pi\tau_0T}\right], \tag{49}$$

where the function $g_1(x)=-\frac{1}{4}\Psi^{(2)}(\frac{1}{2}+x)+(x/48)\Psi^{(3)}(\frac{1}{2}+x)$. It is important to note that the function $g_1(x)$ is positive for all values of $(\pi\tau_0T)^{-1}$, so the superconducting phase transition is always a second-order one for $d_f\gg\xi_f$. The transition temperature of the superconductor in contact with the F layers is determined by putting $\Delta=0$ in the previous equations, and gives back the results of Sec. IV and of Refs. 16 and 17 in the limit of a large F layer and a large transparency of the interfaces. Simplifying Eqs. (48) and (49) using Eq. (18), and defining the function $g_2(x)=1-x\Psi^{(1)}(\frac{1}{2}+x)$, we obtain

$$\Delta_A^2=(2\pi T_c^A)^2\frac{g_2[(\pi\tau_0T_c^A)^{-1}]}{g_1[(\pi\tau_0T_c^A)^{-1}]}\left(1-\frac{T}{T_c^A}\right), \tag{50}$$

$$\Delta_P^2=(2\pi T_c^P)^2\frac{\mathrm{Re}\,g_2[(\pi\tau_0T_c^P)^{-1}]}{\mathrm{Re}\,g_1[(\pi\tau_0T_c^P)^{-1}]}\left(1-\frac{T}{T_c^P}\right). \tag{51}$$

This shows that the order parameters increases as $(1-T/T_c)^{1/2}$ when the temperature is sufficiently low. The free energy of the system is simply given by (see Ref. 20)

$$F_s-F_n=\Delta F=-\int_0^\lambda\frac{\Delta^2}{\lambda_1^2}d\lambda_1=\int_0^\Delta\Delta_1^2\frac{d\left(\frac{1}{\lambda}\right)}{d\Delta_1}d\Delta_1. \tag{52}$$

Using the relation $\delta(1/\lambda)=-N(0)\delta T_{c0}/T_{c0}$ (see Ref. 20), and Eqs. (50) and (51), we obtain

$$\frac{d(\lambda^{-1})}{d\Delta}=-\frac{\Delta N(0)}{2\pi^2T_c^2}g_1[(\pi\tau_0T_c)^{-1}].$$

Thus the calculation of the free energy is straightforward [Eq. (52)]:

$$\Delta F^A=-2N(0)\pi^2(T_c^A)^2\frac{g_2^2[(\pi\tau_0T_c^A)^{-1}]}{g_1[(\pi\tau_0T_c^A)^{-1}]}\left(1-\frac{T}{T_c^A}\right)^2, \tag{53}$$

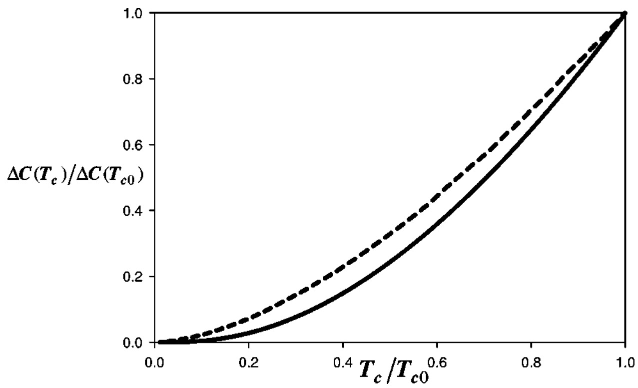

FIG. 5. Discontinuity of the specific heat at the critical temperature vs T_c/T_{c0}. The full line corresponds to the antiparallel case, and the dashed line to the parallel case.

$$\Delta F^P = -2N(0)\pi^2 (T_c^P)^2 \frac{\{\mathrm{Re}\, g_2[(\pi\tau_0 T_c^P)^{-1}]\}^2}{\mathrm{Re}\, g_1[(\pi\tau_0 T_c^P)^{-1}]}\left(1-\frac{T}{T_c^P}\right)^2. \tag{54}$$

From the above equations, the entropy and the heat capacity are obtained using $S=-(\partial F/\partial T)_h$ and $C=-T[(\partial^2 F/\partial T^2)]_h$. We present the results obtained for the heat capacity only:

$$\Delta C^P(T_c^P) = 4\pi^2 N(0) T_c^P \frac{\{\mathrm{Re}\, g_2[(\pi\tau_0 T_c^P)^{-1}]\}^2}{\mathrm{Re}\, g_1[(\pi\tau_0 T_c^P)^{-1}]}, \tag{55}$$

$$\Delta C^A(T_c^A) = 4\pi^2 N(0) T_c^A \frac{g_2^2[(\pi\tau_0 T_c^A)^{-1}]}{g_1[(\pi\tau_0 T_c^A)^{-1}]}. \tag{56}$$

The jump of the specific heat at the transition decreases monotonically as T_c decreases (i.e., as the pair-breaking effect of the F layers increases). The corresponding results are plotted in Fig. 5, where the jump of the specific heat at T_c is normalized by the jump of the specific heat at T_{c0} the critical temperature without any proximity effect.

VI. CONCLUSION

We have considered the properties of $F/S/F$ spin-valve systems, and presented their general theoretical description for the most interesting case of a thin superconducting layer. The spin-valve effect occurs to be very strongly dependent on the S/F interface transparency. So, to observe it in an experiment, it is necessary to choose superconductor-ferromagnet systems with a low barrier at the interface. The oscillatorylike T_c dependence on the F-layer thickness d_f gives the optimum condition for spin-valve observation for $d_f \sim \xi_f$ but the situation also remains qualitatively the same for higher thicknesses. The characteristic length ξ_f can be estimated in some ferromagnetic compounds: for example, in cobalt, using $v_f \sim 2.10^6$ m.s^{-1} and $h \sim 120$ meV, ξ_f should be around 17 Å, in gadolinium,[27] $v_f \sim 2.10^6$ m.s^{-1} and $h \sim 250$ meV, ξ_f should be around 18 Å.

The maximum gain in the superconducting energy corresponds to the antiparallel configuration, and this gain may be of the same order of magnitude as the superconducting condensation energy itself. So we may expect that, without an external applied field, the parallel configuration will be unstable. Therefore, with the decrease of the temperature below T_c, the transition from parallel to antiparallel configurations may be observed. Since it would depend on the magnetic cohercitivity force, thin F layers would be *a priori* more suitable to observe such an effect. A very interesting situation can be also observed when the Curie temperature is lower than the superconducting critical temperature. In such a case we may expect the spontaneous appearance of an antiparallel configuration by decreasing the temperature. It is worth noting that in the case when the domain wall energy is small, the formation of short length-scale magnetic domains could occur at the contact of the ferromagnet and the superconductor.[28,29]

In conclusion, $F/S/F$ trilayer systems reveal strong interferences between superconducting and magnetic effects. They could be quite interesting for applications, as a very small magnetic field may strongly influence the superconducting characteristics via the spin-valve effect.

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[1] A.I. Buzdin and M.Y. Kuprianov, JETP Lett. **53**, 321 (1991).
[2] Z. Radovic, M. Ledvij, L. Dobrosavljevic-Grujic, A.I. Buzdin, and J.R. Clem, Phys. Rev. B **44**, 759 (1991).
[3] L.R. Tagirov, Physica C **307**, 145 (1998).
[4] Y.N. Proshin and M.G. Khusainov, Zh. Éksp. Teor. Fiz. **113**, 1708 (1998) [JETP **86**, 930 (1998)]; **116**, 1887 (1999) [**89**, 1021 (1999)].
[5] J.S. Jiang, D. Davidovic, D.H. Reich, and C.L. Chien, Phys. Rev. Lett. **74**, 314 (1985).
[6] T. Mühge, N.N. Garif'yanov, Yu.V. Goryunov, G.G. Khaliullin, L.R. Tagirov, K. Westerholt, I.A. Garifullin, and H. Zabel, Phys. Rev. Lett. **77**, 1857 (1996).
[7] L. Lazar, K. Westerholt, H. Zabel, L.R. Tagirov, Yu.V. Goryunov, N.N. Garif'yanov, and I.A. Garifullin, Phys. Rev. B **61**, 3711 (2000).
[8] Y.A. Izyumov, Y.N. Proshin, and M.G. Khusainov, Phys. Usp. **45**, 109 (2002).
[9] A.I. Buzdin, L.N. Bulaevskii, and S.V. Panyukov, JETP Lett. **35**, 178 (1982).
[10] A.I. Buzdin and M.Y. Kuprianov, JETP Lett. **52**, 487 (1990).
[11] V.V. Ryazanov, V.A. Oboznov, A.Yu. Rusanov, A.V. Veretennikov, A.A. Golubov, and J. Aarts, Phys. Rev. Lett. **86**, 2427 (2001).
[12] T. Kontos, M. Aprili, J. Lesueur, F. Genet, B. Stephanidis, and R. Boursier, cond-mat/0201104 (unpublished).

[13] A. Buzdin, Phys. Rev. B **62**, 11 377 (2000).
[14] I. Baladié and A. Buzdin, Phys. Rev. B **64**, 224514 (2001).
[15] T. Kontos, M. Aprili, J. Lesueur, and X. Grison, Phys. Rev. Lett. **86**, 304 (2001).
[16] A.I. Buzdin, A.V. Vedyayev, and N.V. Ryzhanova, Europhys. Lett. **48**, 686 (1999).
[17] I. Baladié, A. Buzdin, N. Ryzhanova, and A. Vedyayev, Phys. Rev. B **63**, 054518 (2001).
[18] L.R. Tagirov, Phys. Rev. Lett. **83**, 2058 (1999).
[19] L. Usadel, Phys. Rev. Lett. **95**, 507 (1970).
[20] A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinskii, *Methods of Quantum Field Theory in Statistical Physics* (Dover, New York, 1963).
[21] M.Y. Kuprianov and V.F. Lukichev, Zh. Éksp. Teor. Fiz. **94**, 139 (1988) [Sov. Phys. JETP **67**, 1163 (1988)].
[22] J. Aarts, J.M.E. Geers, E. Brück, A.A. Golubov, and R. Coehoorn, Phys. Rev. B **56**, 2779 (1997).
[23] A.A. Abrikosov and L.P. Gor'kov, Zh. Éksp. Teor. Fiz. **39**, 1781 (1960) [Sov. Phys. JETP **12**, 1243 (1961)].
[24] Ya.V. Fominov, N.M. Chtchelkatchev, and A.A. Golubov, Phys. Rev. B **66**, 014507 (2002).
[25] L.R. Tagirov, I.A. Garifullin, N.N. Garif'yanov, S.Ya. Khlebnikov, D.A. Tikhonov, K. Westerholt, and H. Zabel, J. Magn. Magn. Mater. **240**, 577 (2002).
[26] D. Saint-James, G. Sarma, and E. J. Thomas, *Type II Superconductivity* (Pergamon, New York, 1969).
[27] C. Strunk, C. Sürgers, U. Paschen, and H.V. Löhneysen, Phys. Rev. B **49**, 4053 (1994).
[28] A.I. Buzdin and L.N. Bulaevskii, Zh. Éksp. Teor. Fiz. **94**, 256 (1988) [Sov. Phys. JETP **67**, 576 (1988)].
[29] F.S. Bergeret, K.B. Efetov, and A.I. Larkin, Phys. Rev. B **62**, 11 872 (2000).