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Possibility of periodically reentrant superconductivity in ferromagnet/superconductor layered structures

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We develop the theory of the proximity effect in layered ferromagnetic metal/superconductor (F/S) structures taking into account finite transparency of the F/S interface as well as depression of the Cooper pairing and diffusionlike motion of conduction electrons by a strong exchange field of the ferromagnet. It is shown that the oscillatory dependence of the critical temperature on the F-layer thickness is due to a periodic modulation of the F/S boundary transparency by the oscillations of pair amplitude within the F layer. It is possible not only in the F/S multilayers, but in the F/S bilayers as well. The phenomena of reentrant and periodically reentrant superconductivity in the F/S contacts and superlattices are predicted. The competition between the "0" phase and " π " phase types of superconductivity in the F/S multilayers is also discussed. [S0163-1829(97)51546-3]

The analysis of experiments¹⁻⁵ on ferromagnet/ superconductor (F/S) multilayers testifies that there is various qualitative behavior of the critical temperature T_c dependence on the ferromagnetic layer thickness d_f for the same F/S structures. In particular, in the experiments on systems Fe/V (Ref. 1) and Gd/Nb (Ref. 2) the fast initial T_c decrease and the subsequent output on a plateau were observed with increasing d_f . In other experiments on the same systems (Refs. 3, 4, and 5, accordingly) the oscillatory behavior $T_c(d_f)$ have preceded the same output on a plateau. The theoretical interpretation of oscillations $T_c(d_f)$ in Refs. 6 and 7 was reduced to the periodic "switching" from the traditional "0" phase type superconductivity to the " π " phase type, in which the sign of the order parameter Δ under the transition through F layers changes. However, as it will be shown below, the theories^{6,7} are valid only in the case of high transparency of the F/S boundary and are limited to the extremely dirty ferromagnetic metal case, when $2I\tau_f \ll 1$, where I is a exchange field and τ_f^{-1} is a frequency of the electrons scattering on nonmagnetic impurities. Therefore two different types of experimental $T_c(d_f)$ dependences cannot be described in the context of the existing theory.^{6,7} Moreover, recently it was revealed that the oscillation $T_c(d_f)$ takes place in the trilayered structure Fe/Nb/Fe,⁸ where the " π " phase type superconductivity is impossible. Hence the question about the origin of nonmonotonic dependence $T_c(d_f)$ in the F/S systems remains unsolved and a theory adequately describing an available set of the experimental data is required.

In this paper we present the complete theory of the proximity effect in the layered F/S structures taking into account finite transparency of the F/S boundaries. We also take into account the depression of the Cooper pairing by a strong exchange field I and a competition between a diffusionlike and spin-wave-like motion of quasiparticles in a ferromagnet. The present theory allows us not only to explain available experimental data, but also to predict a number of effects, the pronounced peculiarity of which might be periodically reentrant superconductivity appearance. The oscillatory behavior of $T_c(d_f)$ can be possible not only in the multilayers, but also in F/S bilayers and F/S/F trilayers.

First let us consider a planar contact between a ferromagnetic metal occupying the region $-d_f < z < 0$ and a superconductor occupying the region $0 < z < d_s$. In a vicinity of the second order phase transition point the critical temperature T_c of the F/S contact is determined from the Gor'kov integral equation for the order parameter $\Delta(z)$ of a nonuniform system. For convenience we write it down in the terms of anomalous Usadel function $F_{\alpha\beta}(z,\omega)$ (see, for example, Refs. 6 and 7)

$$\Delta(z) = \frac{1}{2}\lambda(z)\pi T \sum_{\omega}' \sum_{\alpha \neq \beta} F_{\alpha\beta}(z,\omega), \qquad (1)$$

$$F_{\alpha\beta}(z,\omega) = \frac{1}{\pi N(z)} \int_{-d_f}^{d_s} H_{\alpha\beta}(z,z',\omega) \Delta(z') dz', \quad (2)$$

where $\lambda(z>0) = \lambda_s$ and $\lambda(z<0) = \lambda_f$ are dimensionless parameters of the electron-electron interaction, N(z) is the density of states at the Fermi level, α and β are the spin indices and the prime on the summation denotes the cutoff at the Debye frequency ω_D ; T is the temperature, $\omega = \pi T(2n + 1)$ is the Fermi frequency. It can be shown by the diagram technique (see Ref. 9 and Fig. 1) that in the presence of the exchange field and conduction electrons scattering on non-magnetic impurities the correlator $H_{\alpha\beta}(z,z',\omega)$ is a solution of the other integral equation

$$H_{\alpha\beta}(z,z',\omega) = K_{\alpha\beta}(z,z',\omega) + \int_{-d_f}^{d_s} \frac{K_{\alpha\beta}(z,z_1,\omega)H_{\alpha\beta}(z_1,z',\omega)}{2\pi N(z_1)\tau(z_1)} dz_1;$$
(3)

see for a comparison Ref. 10. Here we use the following notations:

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FIG. 1. Diagrammatic representation of Eq. (2) for vertex part $H_{\alpha\beta}(z,z',\omega)$. Solid lines correspond to the normal state Green functions averaged on the nonmagnetic impurities configurations.

$$K_{\alpha\beta}(z,z',\omega) = \frac{1}{(2\pi)^2} \int G_{\alpha\alpha}(\mathbf{p},z,z',\omega) G_{\beta\beta}$$
$$\times (\mathbf{p},z,z',-\omega) d^2p, \qquad (4)$$

where $G_{\alpha\alpha}(\mathbf{p},z,z',\omega)$ is the Green function of conduction electrons in a normal phase, \mathbf{p} is the two-dimensional momentum in the plane of contact, and $\tau^{-1}(z)$ is the impurity scattering rate which stepwiselike changes its value with transition through the F/S boundary z=0 as well as N(z). Solving the problem with a potential barrier at the F/S interface for value $K_{\alpha\beta}(z,z',\omega)$ (see, for example, Ref. 10), it can be shown that the integral equation set (2), (3) is reduced to a boundary problem for function $F_{\alpha\beta}(z,\omega)$ in the dirty metal case, when the mean free path $l_j=v_j\tau_j$ (j=f,s) becomes the least among all characteristic scales. This boundary problem contains the diffusionlike type differential equation

$$\left[\left| \omega \right| + iI(z)g_{\alpha\beta} \operatorname{sgn} \omega - \frac{1}{2}D_{\alpha\beta}(z)\frac{\partial^2}{\partial z^2} \right] F_{\alpha\beta}(z,\omega) = \Delta(z)$$
(5)

and the boundary conditions relating a flux of the function $F_{\alpha\beta}(z,\omega)$ with its jump on the flat F/S interface z=0

$$D_{s\alpha\beta} \frac{\partial F_{\alpha\beta}(z,\omega)}{\partial z} \bigg|_{z=+0} = \frac{\sigma_s v_s}{4} [F_{\alpha\beta}(+0,\omega) - F_{\alpha\beta}(-0,\omega)];$$

$$D_{f\alpha\beta} \frac{\partial F_{\alpha\beta}(z,\omega)}{\partial z} \bigg|_{z=-0} = \frac{\sigma_f v_f}{4} [F_{\alpha\beta}(+0,\omega) - F_{\alpha\beta}(-0,\omega)];$$
(6)

where $g_{\uparrow\downarrow} = -g_{\downarrow\uparrow} = 1$, and the *F/S* contact transparencies σ_f and σ_s are connected by the detailed balance relation $\sigma_s v_s N_s = \sigma_f v_f N_f$.¹¹ The presence of the ferromagnet spin stiffness leads to the complex diffusion coefficient,¹² i.e.,

$$D_{\alpha\beta}(z) = \frac{D(z)}{1 + 2i\tau(z)I(z)g_{\alpha\beta}\operatorname{sgn}\omega}; \quad D(z) = D_j = v_j l_j/3,$$
(7)

where I(z<0)=I and I(z>0)=0. Equations (5) and (6) are right on the condition $l_j < \xi_j$ ($\xi_j = \text{Re}\sqrt{D/2\pi T + 2iI_j}$ is the coherence length) and in addition the parameter l_f should be less than the ferromagnet spin stiffness length $a_f = v_f/2I$.

The received boundary problem (1), (5), and (6) for the proximity effect in the F/S contact differs from the former one^{6,7} in two respects. Firstly, the boundary conditions, which were used in Refs. 6 and 7, consist of a continuity of the Usadel function at the F/S interface (z=0). It is a special case of Eqs. (6) which corresponds to the high transmission limit $\sigma_j \ge l_j/\xi_j$ or to the neglect of the $F_{\alpha\beta}(z,\omega)$ flux through the F/S boundary. However, the σ_j value determines the rate at which electrons are transferred between F and S layers (see Ref. 11) and it strongly depends on the F/S

boundary manufacturing conditions and the sample preparation technique. Because of this, the σ_j value remains a fitting parameter of theory or has to be measured experimentally. Secondly, the depression of diffusionlike motion of conduction electrons by the exchange field *I* of ferromagnet leads to the occurrence of an imaginary part in the effective diffusion coefficient $D_f^* = D_f / (1 + 2iI\tau_f)$.¹² Due to this the quasiparticles motion in the ferromagnet has the mixed diffusionlike and spin-wave-like character. The spin-wave contribution dominates with the increase of parameter $2I\tau_f$ and the penetration depth of the pair amplitude into a ferromagnet becomes larger than a period of its oscillations, as will be shown below.

Strong pairbreaking by the exchange field $I \ (I \gg \pi T_{cs})$ where T_{cs} is the critical temperature of the isolated S layer) is the main mechanism of the superconductivity destruction in the F/S systems. For the sake of simplicity in the following calculations we neglect the order parameter induced in a F layer $\Delta(z < 0) \approx 0$ ($\lambda_f \approx 0$) and we search the solutions of Eqs. (1), (5), and (6) in the form excluding the electrons flux through external boundaries of the contact, i.e., $F_s(z,\omega)$ $\propto \cos k_s(z-d_s)$ for z>0 and $F_f(z,\omega) \propto \cos k_f(z+d_f)$ for z <0. We also assume that k_s and k_f do not depend on the frequency ω . Here we use the symmetry of the Usadel function $F_{\alpha\beta}(\omega) = F^*_{\beta\alpha}(\omega) = F_{\beta\alpha}(-\omega)$ and, turning to positive frequencies ω in Eq. (1), we drop spin indices for convenience. Then we receive the closed set of equations for the reduced temperature $t = T_c/T_{cs}$ of the F/S contact superconducting transition

$$\ln t = \Psi\left(\frac{1}{2}\right) - \operatorname{Re} \Psi\left(\frac{1}{2} + \frac{D_s k_s^2}{4\pi T_{cs} t}\right),$$
$$D_s k_s \tan k_s d_s = \frac{\sigma_s v_s}{4 - (\sigma_f v_f / D_f^* k_f) \cot k_f d_f},$$
$$k_f^2 = -\frac{2iI}{D_f^*} = -\frac{2iI(1 + 2iI\tau_f)}{D_f},$$
(8)

where $\Psi(x)$ is the digamma function. As follows from Eq. (8), pair-breaking parameter $D_s k_s^2$ and critical temperature T_c strongly depend on the layer's thickness, the *F/S* boundary transparency, and a relation between the Fermi level exchange splitting 2*I* and the collision's frequency of electrons τ_f^{-1} with nonmagnetic impurities in the ferromagnet.

The spatial changes of the Usadel function in a *F* layer are characterized by a wave number $k_f = 1/\xi'_f - i/\xi''_f$, where ξ'_f responds for an oscillation period, and ξ''_f responds for the penetration depth of the Cooper pairs in the ferromagnet. In a dirty limit $(l_f \ll \xi'_f, \xi''_f)$ these two lengths differ only by small corrections of the order $I\tau_f$

$$\xi_{f}' = \sqrt{\frac{D_{f}}{I}} (1 - I\tau_{f}); \quad \xi_{f}'' = \sqrt{\frac{D_{f}}{I}} (1 + I\tau_{f}); \quad 2I\tau_{f} \ll 1,$$
(9)

as it is seen from (8). Owing to a strong attenuation, the oscillations of the function $F_f(z,\omega)$ are not displayed and in the last equation in (8) $\tan k_f d_f \approx -i$ at $d_f \geq \xi'_f$, that corresponds to an output of the pair-breaking factor $D_s k_s^2$ and the



FIG. 2. The dependence of reduced transition temperature for F/S bilayer on the reduced Flayer thickness d_f/a_f for various values of parameters ($N_s v_s = N_f v_f$, $d_s = 500$ Å and ξ_{so} = 400 Å), where ξ_{so} is the BCS coherence length: (a) critical temperature flattens out; (b) reentrant superconductivity; (c) critical temperature oscillations; and (d) periodically reentrant superconductivity. The tricritical points line (t^*) is shown by the dashed line.

dependence $T_c(d_f)$ on a constant at small transparencies σ_j , as shown in Fig. 2(a). This behavior was observed in Gd/Nb bilayers.² At the sufficiently large transparency of F/S boundary and the strong exchange field *I* the phenomenon of the reentrant superconductivity is possible, as shown in Fig. 2(b).

In the opposite limit $2I\tau_f > 1$ (this is possible since $I \sim 10^3$ K for Gd and Fe) the diffusion description of the conduction electrons motion in F layer employed above fails (see Ref. 12), whereas the traditional condition of the dirty limit $\pi T \tau_f \ll 1$ can be held. In this case the asymptotic behavior of $F_f(z, \omega)$ is described by a wave number k_f which is slightly distinct from that in Eqs. (8), namely

$$k_f^2 \approx -\frac{2iI(1+2iI\tau_f)}{v_f l_f}; \quad \xi_f' \simeq a_f = \frac{v_f}{2I}; \quad \xi_f'' \simeq l_f; \quad 2I\tau_f \gg 1.$$
(10)

It follows from joint solution of Eqs. (2)-(4) and (8).

After the replacement $D_f \Rightarrow 3D_f$ the set of Eq. (8) becomes suitable for the description of the dependence $T_c(d_f)$ in the F/S contacts with relatively pure layers of ferromagnet and/or with a strong exchange field. The Usadel function, the pair-breaking factor $D_s k_s^2$, and critical temperature oscillate as the thickness d_f increases with a period determined by the spin stiffness length a_f . These oscillations are damped if $d_f > 2l_f (\gg a_f)$ and the dependence $T_c(d_f)$ flattens out, as shown in Fig. 2(c). The similar behavior of $T_c(d_f)$ was observed in trilayered contact Fe/Nb/Fe.⁸ Note that Eqs. (8) can be used for F/S/F trilayers with a substitution $d_s \Rightarrow d_s/2$. It is interesting that for rather large meanings of parameters σ_s and $2I\tau_f$ the superconductivity of the F/S contact has periodically reentrant character for low temperatures, as shown in Fig. 2(d). The number of maxima of the dependence $T_{c}(d_{f})$ is defined by the number of halfwaves of function $\cos k_f(z+d_f)$ fitted on the F layer thickness. Every time, when this number becomes an integer, there is effective "self-locking" of the F/S boundary and critical temperature passes through the maximum. The minima points correspond to odd numbers of wave quarters within F layer.

Hence the physical reason of oscillatory behavior of the function $T_c(d_f)$ in F/S systems is the periodic compensation of the exchange field paramagnetic effect by oscillations of the pair amplitude inside the F layer. It leads to the periodic modulation of the S/F boundary transparency σ_s for $2I\tau_f > 1$ according to expressions (8) and Figs. 2(c) and 2(d). The nonmagnetic scattering presence leads to the fact that this exchange field compensation becomes incomplete, and modulations σ_s and T_c are damped. Thus the superconductivity in F/S bilayers has a combined character. In S layers the BCS type of pairing is realized with zero pairs momentum. In F layers pairing conditions correspond to the Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) mechanism¹³ with nonzero pairs momentum $k \approx a_f^{-1}$ and, hence, with pair amplitude oscillations with period πa_f .

It is easy to show that Eqs. (8) for the critical temperature of the F/S contacts are generalized by replacement $d_j \Rightarrow d_j/2$ in the case of the "0" phase type F/S superlattices with a period $d_f + d_s$. However, in the F/S multilayer's the possible competition between "0" and " π " phase types of superconductivity^{6,7} should be taken into account.

In the case of the " π " phase type superconductivity the critical temperature T_c is found from Eqs. (8), where function cot x has to be replaced by $-\tan x$ on the right-hand side of the second equation in addition to the above-mentioned replacement $d_j \Rightarrow d_j/2$. As shown in Fig. 3(a), the weak transparency σ_s and small parameter $2I\tau_f$ lead to a very insignificant difference between "0" phase type and " π " phase type solutions for T_c in the "switching" area. Therefore the experimentalists have probably observed on the multilayers Fe/V (Ref. 1) and Gd/Nb (Ref. 2) fast initial decrease T_c and the subsequent output on a plateau with an increase of d_f instead of oscillations. If the parameters σ_s and $2I\tau_f$ are

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FIG. 3. The phase diagrams $T_c/T_{cs}(d_f/a_f)$ for F/S superlattices $(N_s v_s = N_f v_f, d_s = 500 \text{ Å}$ and $\xi_{so} = 400 \text{ Å}$): (a) critical temperature flattens out; (b) local maximum of critical temperature; (c) critical temperature oscillations with a competition "0" phase type and " π " phase type of superconductivity; and (d) periodically reentrant superconductivity with alternation of "0" phase type and " π " phase type of peaks. The marks "0" and " π " near the curves correspond to "0" phase type and " π " phase type of superconductivity, respectively.

slightly larger than the preceding ones, there is an individual burst (local maximum) of critical temperature $T_c(d_f)$ as a result of the transition from "0" phase type branch of function $T_c(d_f)$ to " π " phase type one [see Fig. 3(b)]. That behavior of $T_c(d_f)$ is likely to have been observed in Refs. 4, 5 on the same Gd/Nb multilayers. Hence the difference between the results in Refs. 2, 4, and 5 may be explained by distinctions in the Gd/Nb boundary transparency and the Gd layer's purity, which may arise at the F/S multilayer's formation. At the large value of transparency ($\sigma_s = 5$, $2I\tau_f$ =0.2) on the phase diagram $T_c(d_f)$ two superconductivity regions corresponding to the "0" and " π " types are in the neighborhood and are separated by a nonsuperconducting region of thicknesses d_f . Thus there occurs the reentrant superconductivity in the F/S multilayers that resembles the one in Fig. 1(b).

The oscillations of $T_c(d_f)$ are essential for the higher val-

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ues of $2I\tau_f$. Their period in the F/S multilayers for either "0" phase type of superconductivity or " π " phase type separately is twice more than that in appropriate F/S contact. The competition of the "0" and " π " states shown in Figs. 3(c) and 3(d) leads to their alternation on the phase diagrams and to formal coinciding of the oscillations period in superlattices and contacts. The availability of $T_c(d_f)$ oscillations does not mean an imperative presence in the " π " phase superconductivity in F/S multilayers, but the experiment here plays a crucial role. Thus to be certain that a " π " phase state is present on the $T_c(d_f)$ experimental phase diagram of F/S multilayers it is necessary to take parallel measurements on F/S contacts. This is important, since " π " phase superconductivity with the modulation of the order parameter and pair amplitude is the one-dimensional LOFF state¹³ over the whole F/S superlattice in contrast to the "0" phase one in which the LOFF state is realized in F layers only.

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