Multicritical points in the phase diagrams of layered ferromagnet–superconductor structures

M. G. Khusainov

Kazan State University, 420008 Kazan, Russia
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Landau's theory of phase transitions is used to study the coexistence and mutual accommodation of superconductivity and ferromagnetism in F/S (ferromagnetic insulator/superconductor) junctions and superlattices. The dependence of the superconducting transition temperature T_c on the magnitude of the exchange field h of the localized spins in the F/S-boundary is established. The phase diagrams (T_c, h) of F/S-systems are found to contain a tricritical point t at which the superconducting phase transition changes from second order to first. The conditions are established for a Lifshitz point L_p to appear on the first-order transition line (two commensurate magnetically ordered phases and one incommensurate phase, with two phases being superconducting, meet at this point). As the point L_p is approached, the period q_0^{-1} of modulation of the incommensurate phase increases without limit, with the corresponding critical exponent β_q equaling 1/2. New critical behavior of the interphase boundaries in the vicinity of the Lifshitz point is predicted, with two different crossover exponents $\phi = 1/2$ and $\phi^* = 1$ characterizing such behavior. The presence or absence of a Lifshitz point in the phase diagrams makes it possible to assign a F/S-system to one of two distinct types. The feasibility of experimentally observing the competition between superconductivity and ferromagnetism near the multicritical points t and L_p by employing magnetic neutron scattering and by examining the spin-wave spectrum of the F/S-system are analyzed. The possible candidates for F/S-systems with phase diagrams that might have a Lifshitz point are EuO/Al and EuS/Al junctions and EuO/V multilayers, in which coexistence of superconductivity and inhomogeneous magnetic ordering has already been observed.
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1. INTRODUCTION

Crystal structures formed by alternating layers of different metals or a metal and a magnetic material constitute a new class of layered materials with unique electronic and magnetic properties that depend on the type of material comprising the superstructures, called superlattices, and the thicknesses of the layers.[1,2] Modern techniques used in fabricating layer structures (such as molecular beam epitaxy) make it possible to lay down layers of atomic thickness and thus "control" their properties. In this respect, artificially layered superlattices differ favorably from natural layered compounds, such as high-T_c superconductors, dichalcogenides of transition metals, and boride carbides. The decrease in the size of layered systems must lead to the emergence of competing interactions and phenomena whose simultaneous observation in homogeneous materials is sometimes simply impossible. In this connection, special attention is focused today on the problem of coexistence and mutual influence of superconductivity and ferromagnetism in layered structures manufactured by alternately depositing layers of ferromagnetic material (F) and superconductor (S) (see Refs. 1–8).

Notwithstanding the many studies done in this field, several important experimental observations remain without adequate theoretical interpretation. Such phenomena as the anomalously weak suppression of superconductivity in EuO/V multilayers[3] and the crossover from three-dimensional (3D) behavior to two-dimensional (2D) in Fe/V superlattices[1] as the temperature is lowered or the thickness of the vanadium layers is increased, just cannot be explained solely by the π-phase nature of superconductivity[4,5] in the layered structures. Moreover, the oscillations of T_c as a function of the thickness of the ferromagnetic layers predicted by Buzdin *et al.*[4] and Radović *et al.*[5] have not been observed in experiments involving Gd/Nb superlattice[6] and Fe/V superlattice.[7]

On the other hand, the nonmonotonic variation (or even a slight increase) in T_c as the Fe layer[1] or the Gd layer[8] gets thicker can be explained, for instance, by the logarithmic growth in interelectron attraction[9,10] that results from a decrease in the rate of electron exchange between F- and S-layers.

The point is that a similar increase in T_c with the thickness of the nonsuperconducting layers can also be observed in Cr/V, Cu/Nb, and Mo/V superlattices (see Ref. 1 and the literature cited therein), where Cr is an antiferromagnet, Cu a nonsuperconducting metal, and Mo a poor superconductor. This phenomenon must probably be attributed not to the paramagnetic effect of the exchange field, as assumed in Refs. 4 and 5, but to the proximity effect, whose modified theory[9,10] provides a natural explanation of such behavior of T_c.

It must also be noted that full analysis of the variants of mutual accommodation of superconductivity and ferromagnetism in F/S-systems is also impossible without solving the problem of how magnetic layers interact through the superconducting layers. A possible mechanism that explains the long-range coupling between localized spins belonging to the same F-layer and to neighboring F-layers is the indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange[11,12] via the conduction electrons belonging to the superconducting layers. Consequently, a meaningful and complete picture of the competition between superconductivity and ferromagnetism in bimetallic F/S-systems can be obtained only if we simultaneously allow for the proximity effect, the paramagnetic effect of the exchange field, and the interaction of the magnetic layers through the superconducting layers.

In pure form, the influence of the latter two effects on the superconducting transition temperature makes it possible to study F/S structures with a ferromagnetic insulator acting as the F-layer. In the author's previous work,[12] the possible ground states of such F/S-systems and the corresponding variants of mutual accommodation of the superconducting and magnetic order parameters were established. Special attention was paid in Ref. 12 to the emergence of superconducting magnetically ordered phases incommensurate with the period of the crystal lattice of the F- and S-layers. The results made it possible to explain the existence of a nonuniform internal field, which splits the BCS peak in the quasiparticle density of states, in EuO/Al contacts[13] and EuS/Al contacts[14] and the unexpectedly weak suppression of superconductivity in EuO/V multilayers.[3] The origin of the incommensurate magnetic phases in F/S-systems rests in the competition between the short-range direct ferromagnetic exchange of localized spins in the F/S-boundary and the long-range antiferromagnetic RKKY exchange between these spins via Cooper pairs.[11,12]

The state diagrams for materials that have incommensurate phases are characterized by a triple point, the Lifshitz point,[15] at which three phases meet: the initial, the commensurate, and the incommensurate. As the Lifshitz point is approached, the period of the incommensurate phase increases and finally becomes infinite. The fact that ferromagnets have a Lifshitz point has been experimentally corroborated only for MnP (see Ref. 16). The presence of an incommensurate phase in layered F/S-systems suggests that the phase diagrams of these systems can also contain such an unusual singularity as a Lifshitz point.

The present paper studies this problem within the framework of Landau's theory of phase transitions and the model of exchange interactions for layered ferromagnetic insulator/superconductor structures (F/S structures) suggested in Refs. 11 and 12. In Sec. 2 we find the dependence of the transition temperature of F/S junctions on the magnitude of the exchange field h generated by the localized spins in the F/S-boundary. We find that the presence or absence of a Lifshitz point in the (T_c,h) phase diagrams makes it possible to classify F/S-contact into two different types. A similar problem for F/S superlattices with allowance for interlayer $F-F$ RKKY exchange via the conduction electrons of the superconducting layers S is solved in Sec. 3. Finally, in Sec. 4 we discuss the results.

2. PHASE DIAGRAMS OF *F/S* JUNCTIONS

Let us examine a planar junction between a thin ferromagnetic insulator (F) occupying the region $-d\leqslant z\leqslant 0$ and a superconducting wafer (S) occupying the region $0\leqslant z\leqslant L$. We assume that the localized spins of the F-film, $\mathbf{S_r}$ and $\mathbf{S_{r'}}$, ordered according to the "easy plane" type, interact via direct exchange. Here the exchange integral J is positive only for the nearest neighbors, localized at the sites $\mathbf{r}$ of a simple cubic lattice with period a. In addition, at the F/S phase boundary ($z=z'=0$) the localized spins $\mathbf{S_r}$ and $\mathbf{S_{r'}}$ also interact indirectly by long-range RKKY exchange[11,12] via the conduction electrons of the superconductor. The latter effect results from effective $s-d(f)$ exchange (I), which emerges because of virtual electron transfer from superconductor to insulator and back due to the overlap of the corresponding wave functions at the F/S-boundary. We also assume that the Curie temperature Θ of the F-film is much higher than the transition temperature T_{c0} of the S-layer, and that for $T_{c0}<T<\Theta$, the ferromagnetism of the F/S-boundary is not destroyed by the oscillations of the normal part of the RKKY exchange. The latter assumption means that direct exchange over the distance between nearest neighbors is stronger than indirect exchange, i.e., $J>I^2N(0)$, where $N(0)$ is the density of states of conduction electrons at the Fermi surface. This will allow us to deal with only the most important short-range ferromagnetic and long-range antiferromagnetic (superconducting) parts of the exchange interaction (for more details see Ref. 12). We assume that the F- and S-layers are thin, i.e., $d<\delta$ and $L<\xi$, where δ is the depth of penetration of the surface distortions of magnetic ordering,[17] and $\xi=\sqrt{D/2\pi T_{c0}}$ is the superconductor's coherence length, with D the electron diffusion coefficient. This guarantees the homogeneity of the magnetic and superconducting order parameters along the z axis within the F and S layers, respectively.

Landau's theory of phase transitions[18] can be used to do an elementary qualitative analysis of the possible variants of the coexistence and mutual accommodation of superconductivity and ferromagnetism in F/S contacts. In the self-consistent field approximation, we define the magnetic order in the F-film as

$$\langle S_{\mathbf{r}}^{\pm}\rangle=\langle S_{\mathbf{r}}^{x}+iS_{\mathbf{r}}^{y}\rangle=\langle S\rangle\exp(\pm i\mathbf{q}_{\perp}\boldsymbol{\rho}),\qquad \langle S_{\mathbf{r}}^{z}\rangle=0, \tag{1}$$

where $\langle S\rangle$ stands for the thermodynamic average of the localized spin at the site with $\boldsymbol{\rho}=0$, where $\boldsymbol{\rho}=\mathbf{i}x+\mathbf{j}y$ and $\mathbf{q}_{\perp}=\mathbf{i}q_x+\mathbf{j}q_y$. Then at temperatures close to the transition temperature, we have the following functional for the free energy per unit area of the F/S junction:

$$f=f_F^0+f_N^0+J\langle S\rangle^2\frac{d}{a}q_{\perp}^2-\frac{I^2\langle S\rangle^2}{8a}\,\delta\chi_s(q_{\perp},0,0)$$
$$+\frac{L}{a^3}\left(\alpha_0\frac{\Delta^2}{2}+\beta_0\frac{\Delta^4}{4}+\gamma_0\frac{\Delta^6}{6}\right), \tag{2}$$

where f_F^0 and F_N^0 are densities of the free energies of the F-film and S-layer in the normal state. The third term describes the loss of direct exchange energy due to the long-wave ($q_{\perp}a\ll 1$) modulation of ferromagnetic ordering. The fourth term is proportional to the superconducting contribution to the spin susceptibility of the conduction electrons, $\delta\chi_s(q_\perp,z,z')$ (see Refs. 11 and 12):

$$\delta\chi_s(q_\perp,z,z')=-4\pi N(0)T \times\sum_\omega \frac{\Delta^2\cosh(kz)\cosh[k(z'-L)]}{(\omega^2+\Delta^2)Dk\sinh(kL)}, \tag{3}$$

where $k^2=q_\perp^2+\xi_\omega^{-2}$, with $\xi_\omega^2=D/2\sqrt{\omega^2+\Delta^2}$, and $\omega=\pi T(2n+1)$. This term describes both the long-range antiferromagnetic coupling of the localized spins at the F/S-boundary ($z=z'=0$) via the conduction electrons of the S-layer, and the suppression of the superconducting order parameter Δ due to the paramagnetic effect of these localized spins. The last term on the right-hand side of Eq. (2) is the Landau expansion in powers of Δ responsible for the gain in condensation energy due to the transition of the S-layer into the superconducting state. The coefficients α_0, β_0, and γ_0 of the given expansion are well known from the microscopic theory of superconductivity:[19]

$$\alpha_0=-2N(0)\left(1-\frac{T}{T_{c0}}\right),\qquad \beta_0=\frac{7\zeta(3)N(0)}{(2\pi T_{c0})^2},$$
$$\gamma_0=\frac{93\zeta(5)N(0)}{2(2\pi T_{c0})}, \tag{4}$$

where $\zeta(x)$ is Riemann's zeta function.

For subsequent analysis it is convenient to employ the high-temperature expansion of the RKKY potential (3) in powers of Δ and $q_\perp$ (both $\Delta/2\pi T_{c0}$ and $q_{\perp\xi}$ are much smaller than unity) and rewrite the functional (2) as

$$f=f_F^0+f_N^0+J\langle S\rangle^2\frac{d}{a}q_\perp^2+\frac{L}{a^3}\left(\alpha\frac{\Delta^2}{2}+\beta\frac{\Delta^4}{4}+\gamma\frac{\Delta^6}{6}\right). \tag{5}$$

Here the renormalized coefficients α, β, and γ are given by

$$\alpha=\alpha_0+2N(0)\eta\left(\frac{h}{h_t}\right)^2[1-b(q_\perp\xi)^2+g(q_\perp\xi)^4],$$
$$\beta=\beta_0\left[1-\left(\frac{h}{h_t}\right)^2\right],\qquad \gamma=\gamma_0\left[1+p\left(\frac{h}{h_t}\right)^2\right],$$
$$h=\frac{I\langle S\rangle a}{2L},\qquad h_t=\sqrt{\frac{7\zeta(3)}{186\zeta(5)}}2\pi T_{c0}\simeq1.312T_{c0}, \tag{6}$$

and the numerical values of the coefficients η, b, g, and p are

$$\eta=\frac{[7\zeta(3)]^2}{186\zeta(5)}\simeq0.367,\qquad b=\frac{\pi^4}{84\zeta(3)}\simeq0.963,$$
$$g=\frac{31\zeta(5)}{28\zeta(3)}\simeq0.955,\qquad p=\frac{4445\zeta(3)\zeta(7)}{[62\zeta(5)]^2}\simeq1.303.$$

Minimization of the functional (5) in Δ and $q_\perp$ indicates the presence of three phases:

(1) a ferromagnetic normal phase FN with $\Delta=q_\perp=0$;

(2) a ferromagnetic superconducting phase FS with $\Delta=\Delta_1$, $q_\perp=0$, and $A<1$: .

$$\Delta_1^2=\frac{-\beta+\sqrt{\beta^2-4\gamma\alpha_1}}{2\gamma},\qquad \alpha_1=\alpha_0+2N(0)\eta\left(\frac{h}{h_t}\right)^2; \tag{7}$$

(3) a cryptoferromagnetic superconducting phase CFS with $\Delta=\Delta_2$, $q_\perp=q_0$, and $A>1$:

$$\Delta_2^2=\frac{-\beta+\sqrt{\beta^2-4\gamma\alpha}}{2\gamma},\qquad q_0^2=\left(1-\frac{1}{A}\right)\frac{b}{2g\xi^2}. \tag{8}$$

The realization of each phase depends on the magnitude of three parameters: the temperature T, the exchange field $h=I\langle S\rangle a/2L$ generated by the localized spins at the F/S-boundary of the conduction electrons of the superconductor, and the ratio

$$A=\frac{\pi^2N(0)h^2\xi^2L}{12J\langle S\rangle^2a^2d}\left(\frac{\Delta}{2T_{c0}}\right)^2 \tag{9}$$

of the antiferromagnetic and ferromagnetic molecular fields acting on each localized spin at the F/S-boundary. Note that the smallness of antiferromagnetic and ferromagnetic polarizations in (9) is balanced by the exceptionally large ratio of their ranges of action (ξ^2L/a^2d).

The parameter A can therefore vary over a wide range. For $A>1$ the ferromagnetic state is unstable and the long-wave modulation originating at the FS-boundary is extended via the strong interatomic exchange J to the entire thickness of the F-film. Here the loss in direct-exchange energy proves to be smaller than the gain in energy caused by the transition of the S-layer to the superconducting state and the reduction in the paramagnetic effect of the exchange field. A rough sketch of the phase diagrams (T_c,h) for two types of F/S junctions is depicted in Figs. 1a and b.

F/S junctions of the first type (Fig. 1a) with $A_c<1$, where $A_c=A(h=h_c,T=0)$ and h_c is the critical exchange field, allow for the coexistence with superconductivity of only homogeneous ferromagnetic ordering. The transition temperature $T_c(h)$ on the $T_{c0}-T$ line of second-order phase transitions (solid curve, $\beta>0$) is given by $\alpha_1=0$, which implies that

$$T_c=T_{c0}(1-\eta x),\qquad x=\left(\frac{h}{h_t}\right)^2<1. \tag{10}$$

On the $t-h_c$ line of first-order phase transitions (dashed curve, $\beta<0$) the function $T_c'(h)$ is given by $\alpha_1=3\beta^2/16\gamma$, i.e.,

$$T_c'=T_{c0}\left[1-\eta x\left(1-\frac{3(x-1)^2}{8x(px+1)}\right)\right],\qquad x>1. \tag{11}$$

The behavior of the superconducting order parameter Δ_1 along this curve (the upper half of Fig. 1a) is described by

$$\Delta_1^2=-\frac{3\beta}{4\gamma}=3h_t^2\frac{x-1}{px+1}. \tag{12}$$

As illustrated by the lower part of Fig. 1a, T_c' tends to zero when the exchange field h reaches its critical value $h_c\simeq1.74h_t$, which can be found from Eq. (11). The value of Δ_1 corresponding to $h=h_c$ is $\Delta_c\simeq1.11h_t$. Clearly, the balance A of the molecular fields, which is conveniently written as

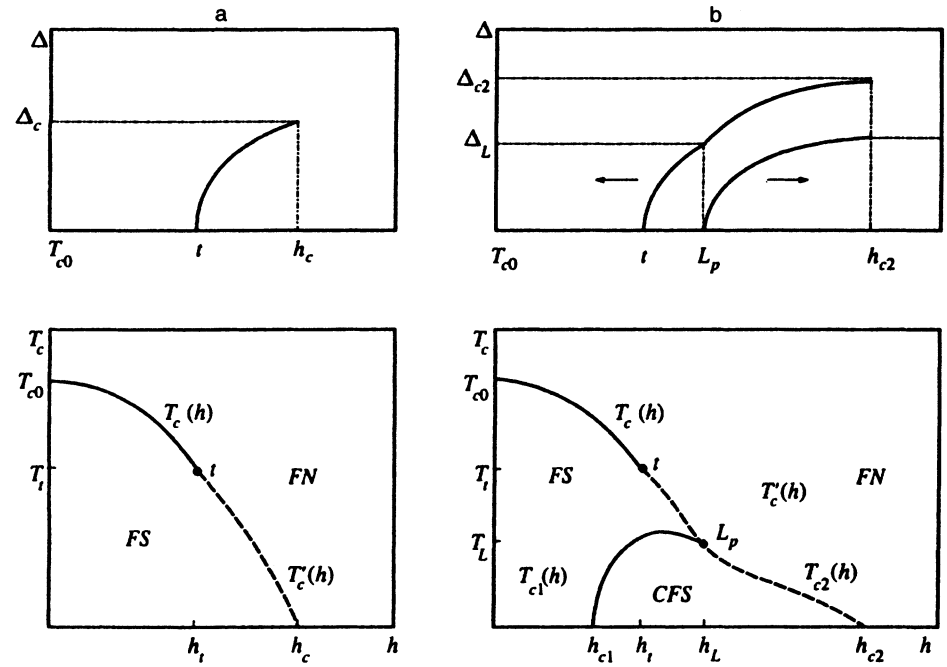

FIG. 1. Phase diagrams (T_c, h) of F/S junctions of (a) the first type, with $A_c<1$ $(q_0=0)$, and (b) the second type, with $A_c>1$. The solid curves depict the lines of second-order phase transitions, and the dashed curves the lines of first-order transitions. The upper half depicts the behavior of the equilibrium order parameter Δ and wave vector q_0 along (a) the T_{c0}–t–h_c transition line and (b) the T_{c0}–t–L_p–h_{c2} transition line.

$$A=A_c\left(\frac{h}{h_c}\right)^2\left(\frac{\Delta}{\Delta_c}\right)^2, \qquad A_c=\frac{\pi^2 N(0) h_c^2 \xi^2 L}{12 J\langle S\rangle^2 a^2 d}\left(\frac{\Delta_c}{2T_{c0}}\right)^2, \tag{13}$$

remains smaller than unity as we move along the line $T_{c0}-t-h_c$ of the $FS-FN$ phase transitions, since $A_c<1$. The coordinates $T=T_c=T_{c0}(1-\eta)$ of the tricritical point t, at which the order of the phase transition changes, can be found from the requirement that $\beta=0$ and $\alpha_1=0$ simultaneously.

It is the presence of the tricritical point t that requires retaining terms up to Δ^6 inclusive in the expansion (5). Note, however, that the phase diagrams and the values of the critical parameters obtained in this paper in the framework of Landau's theory are only qualitative, because the real temperature dependence of the coefficients α, β, and γ in the expansion (5) has been ignored. In particular, the value of h_c obtained here is roughly 1.7 times the value found in Ref. 12, where the problem of the ground state is solved more accurately.

A distinctive feature of F/S junctions of the second type (Fig. 1b) with $A_c>1$ is the presence of a Lifshitz point L_p (see Ref. 15) on the line of first-order phase transitions. All three possible phases, FN, FS, and CFS, meet at this point. The coordinates of the Lifshitz point (T_L, h_L) can be found from the requirement that $\alpha_1=3\beta^2/16\gamma$ and $A=1$ simultaneously, i.e.,

$$T_L=T_{c0}\left[1-\eta x_L\left(1-\frac{3(x_L-1)}{8Gx_L^2}\right)\right],$$

$$x_L=\left(\frac{h_L}{h_t}\right)^2=\frac{1}{2}+\frac{p}{2G}+\sqrt{\left(\frac{1}{2}+\frac{p}{2G}\right)^2+\frac{1}{G}}, \tag{14}$$

$$G=3A_c\left(\frac{h_t}{h_c}\right)^2\left(\frac{h_t}{\Delta_c}\right)^2\simeq 0.8A_c.$$

The latter condition $(A=1)$ corresponds to a situation in which the period q_0^{-1} of modulation of magnetic order in the F-layer becomes infinite. Together with the expression (7) for Δ_1 it determines the line $T_{c1}(h)$ of second-order transitions:

$$T_{c1}=T_{c0}\left[1-\eta x\left(1-\frac{3(x-1)}{2Gx^2}+\frac{9(px+1)}{8G^2x^3}\right)\right], \qquad x<x_L; \tag{15}$$

this line separates the commensurate superconducting magnetic phase (FS) from the incommensurate (CFS).

Note that the curve $T_{c1}(h)$ passes through a maximum, ensuring that the behavior of the system over a certain interval of exchange field values between h_{c1} and h_L at a fixed temperature is reciprocal, $FS-CFS-FS$. Near the Lifshitz point the line $T_{c2}(h)$ of first-order phase transitions, which separates the ferromagnetic normal phase FN from the superconducting phase CFS with a sinusoidally modulated magnetic order, is determined by the equality of the free energies of these phases, and we get (for $x>x_L$)

$$T_{c2}=T_{c0}\left[1-\eta x\left(1-\frac{3(x-1)^2}{8x(px+1)}\right.\right.$$
$$\left.\left.-\left(g+\frac{2b^2x(px+1)}{3(x-1)^2}\right)(q_0\xi)^4\right)\right], \tag{16}$$

where q_0 is defined in (7). Clearly, the lines $T_c'(h)$ and $T_{c2}(h)$ of first-order transitions described by Eqs. (11) and (16) have a common tangent at the Lifshitz point. At the same time, at point L_p the line $T_{c1}(h)$ of second-order transitions forms a finite angle with this tangent. For $A_c-1\ll A_c$ the Lifshitz point L_p is farthest from the tricritical point t: $T_L\propto h_c-h_t\propto 1-A_c^{-1}$ The lower and upper critical exchange fields h_{c1} and h_{c2}, which can be found by solving Eqs. (15) and (16) with $T_{c1}=T_{c2}=0$, differ from h_c only by small correction terms of order $1-A_c^{-1}$ and $(1-A_c^{-1})^2$, respectively. Here the maximum value of the modulation wave vector,

$$q_{c2}\simeq\frac{\sqrt{1-A_c^{-1}}}{\xi},$$

is also small ($q_{c2}\ll\xi^{-1}$). As A_c grows, the Lifshitz point asymptotically approaches the tricritical point, i.e.,

$$T_t-T_L\propto h_L-h_t\propto A_c^{-1},$$

and the range of exchange field values occupied by the CFS-phase widens.

Since the modulation wave vector q_0 continuously increases as we move along the line $T_{c2}(h)$ of first-order phase transitions (see the upper half of Fig. 1*b*), to find h_{c2} for $(\xi/L)^2\gg A_c\gg 1$ we must use the expansion of the RKKY potential (3) for $L^{-1}\gg q_\perp\gg\xi^{-1}$. In this case the coefficients α, β, and γ in the expansion (5) for the free energy have the form

$$\alpha=\alpha_0+2N(0)\nu y,\quad \beta=\beta_0(1-\sigma y),\quad \gamma=\gamma_0(1+\mu y),$$
$$y=\left(\frac{h}{q_\perp\xi h_t}\right)^2,\qquad \nu\simeq0.430,\qquad \sigma\simeq0.672,\qquad \mu\simeq0.694, \tag{17}$$

and we find that $h_{c2}\simeq h_c\sqrt{A_c}$.

For $A_c\gg(\xi/L)^2$ and $q_\perp\gg L^{-1}$ the expressions in (17) remain valid if y is replaced by $y'=yq_\perp L$ and yield $h_{c2}\simeq h_c(A_c\xi^2/L^2)^{1/4}$. The other critical parameters in both cases are determined by expressions of type $h_{c1}\simeq h_c/\sqrt{A_c}$ and $q_{c2}\simeq\sqrt{A_c}/\xi$. Since in the latter case ($A_c\gg\xi^2/L^2$) the lower critical field is weak, i.e., $h_{c1}\ll h_cL/\xi$, the FS-phase occupies the minimum range of values of exchange fields. The CFS-phase, on the contrary, occupies the maximum range of values of exchange fields, since $h_{c2}\gg h_c\xi/L$.

The above analysis shows that the emergence of the Lifshitz point L_p and the related incommensurate CFS-phase in the state diagrams of F/S junctions depends entirely on the value of the critical balance A_c of the molecular fields. The parameter A_c makes it possible to break down F/S-junctions into two types according to the magnitude of the paramagnetic effect of the exchange field h, just as the Ginzburg–Landau parameter κ separates type-I and type-II superconductors according to the orbital effect of an external magnetic field H.

This fact is best illustrated by the phase diagram (h,A_c) in Fig. 2, which schematically depicts the dependence of the critical exchange fields h_c, h_{c1}, and h_{c2} on A_c. The point with coordinates $h=h_c$ and $A_c=1$ corresponds to the Lifshitz point at $T=0$. Three curves meet at this point: the dashed curves h_c and h_{c2}, which are the lines of the first-order transitions $FS-FN$ and $CFS-FN$, respectively, and the solid curve h_{c1}, which is the line of second-order transitions $FS-CFS$.

3. PHASE DIAGRAMS OF *F/S* SUPERLATTICES

To study the competition between ferromagnetism and superconductivity in a F/S superlattice, it is necessary to

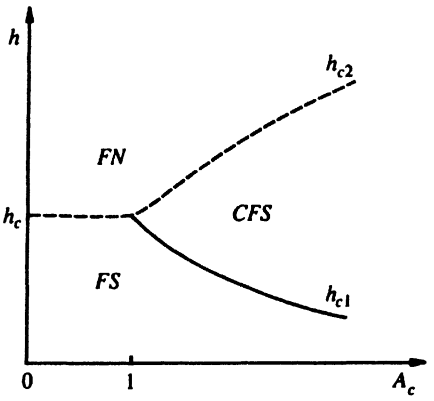

FIG. 2. Phase diagrams (h,A_c) of F/S junctions at $T=0$. Dashed curves represent interphase $FS-FN$ and $CFS-FN$ boundaries, and the solid curve h_{c1} separates the FS and CFS phases. The three curves meet at the Lifshitz point.

study only the density f^* of the free energy of the unit cell of this superlattice, which consists of two magnetic F-half-layers $-d/2\leq z\leq0$ and $L\leq z\leq L+d/2$ separated by the superconducting S-layer $0\leq z\leq L$. The functional f^* differs from (2) in that in addition to the term $\delta\chi_s(q_\perp,0,0)$, it must contain the term $\delta\chi_s(q_\perp,L,L)$ representing the surface RKKY exchange of the localized spin in the neighboring ferromagnetic layer ($z=z'=L$), and the term $\delta\chi_s(q_\perp,0,L)$ representing the exchange of localized spins at the magnetic surfaces $z=0$ and $z'=L$, which are by the superconducting S-layer.

We therefore seek to represent the magnetic order in the F/S superlattice in the form

$$\langle S_{\mathbf{r}}^{\mp}\rangle=\langle S\rangle\exp[\pm i(\mathbf{q}_\perp\boldsymbol{\rho}+q_\parallel z)],\qquad \langle S_{\mathbf{r}}^z\rangle=0, \tag{18}$$

where $q_\parallel$ is the component of the wave vector parallel to the z axis of the superlattice. We still assume that the F and S-layers are thin, i.e., $d<\delta$ and $L<\xi$. The translational invariance of the superlattice, with allowance for interlayer $F-F$ exchange, leads only to the multiplication of $\langle S_{\mathbf{r}}^{(\pm)}\rangle$ by a constant phase factor $\exp(\pm iq_\parallel L)$ as we move from one F-layer to the neighboring layer. Ignoring the tunneling of c<onduction electrons through the insulating magnetic layers, we can write for the functional f^*

$$f^*=f_F^0+F_N^0+\frac{J\langle S\rangle^2d}{a}q_\perp^2-\frac{I^2\langle S\rangle^2}{4a}[\delta\chi_s(q_\perp,0,0)+\delta\chi_s$$
$$\times(q_\perp,0,L)\cos(q_\parallel L)]+\frac{L}{a^3}\left[\alpha_0\frac{\Delta^2}{2}+\beta_0\frac{\Delta^4}{4}+\gamma_0\frac{\Delta^6}{6}\right]. \tag{19}$$

For the discussion that follows, it is important that only at values of the wave vector $q_\perp$ comparable with L^{-1} can the intralayer RKKY exchange term $\delta\chi_s(q_\perp,0,0)$ differ significantly from the interlayer RKKY exchange term $\delta\chi_s(q_\perp,0,L)$. Indeed, Eq. (3) with $q_\perp L\ll1$ yields

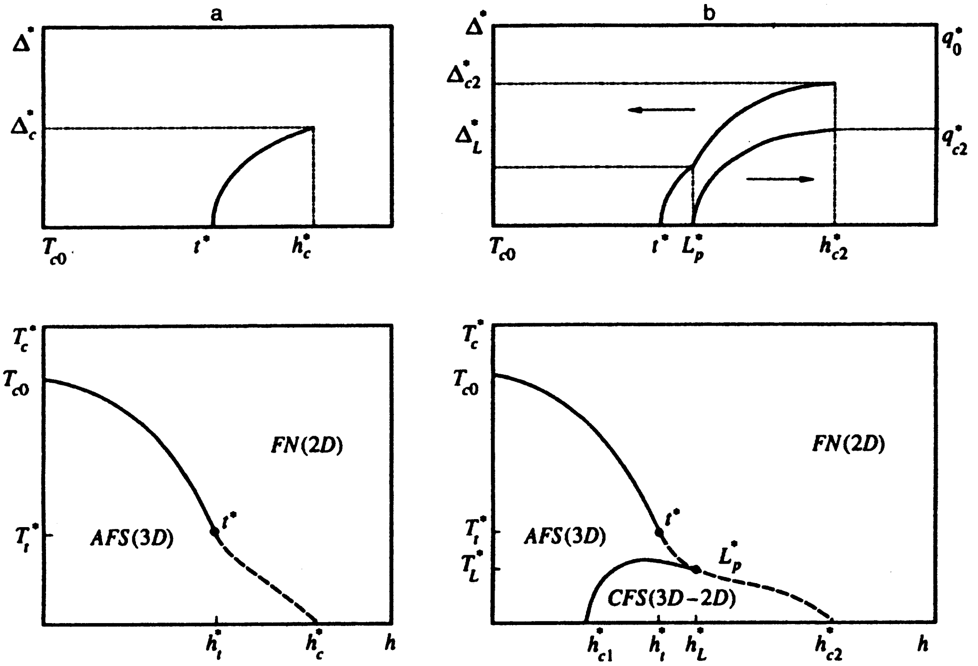

FIG. 3. Phase diagrams (T_c^*,h) of F/S superlattices of (a) the first type, with $A_c<1$ $(q_0=0)$, and (b) the second type, with $A_c>1$. The solid curves depict the lines of second-order phase transitions, and the dashed curves the lines of first-order transitions. The upper half depicts the behavior of the equilibrium order parameter Δ and wave vector q_0 along (a) the T_{c0}–t^*–h_c^* transition line and (b) the T_{c0}–t^*–L_p^*–h_{c2}^* transition line.

$$\delta\chi_s(q_\perp,0,L)\simeq\delta\chi_s(q_\perp,0,0)+\pi N(0)\frac{L}{D}\left[1-\frac{q_\perp^2L^2}{12}+\frac{q_\perp^4L^4}{120}\right]\Delta\tanh\frac{\Delta}{2T}, \tag{20}$$

and the corrections to $\delta\chi_s(q_\perp,0,0)$ are small in view of the smallness of $(L/\xi)^2$. In the opposite limiting case, $q_\perp L\gg 1$, we have

$$\delta\chi_s(q_\perp,0,L)\simeq 2\delta\chi_s(q_\perp,0,0)\exp(-q_\perp L), \tag{21}$$

i.e., the exchange coupling between neighboring F-layers through the superconducting S-layers is negligible, due to the surface nature of RKKY exchange in the event of strong modulation of the ferromagnetic ordering in the F-layers. Minimizing the functional (19) in Δ, $q_\perp$, and $q_\parallel$ leads to three possible superlattice states:

(1) a ferromagnetic normal phase FN with $\Delta=q_\perp=0$ and arbitrary $q_\parallel$;

(2) a layered antiferromagnetic superconducting phase AFS with $\Delta=\Delta_1^*$, $q_\perp=0$, $q_\parallel=\pi/L$, and $A^*<1$:

$$\Delta_1^{*2}=\frac{-\beta^*+\sqrt{\beta^{*2}-4\alpha_1^*\gamma^*}}{2\gamma^*},$$

$$\alpha_1^*=\alpha_0+2\eta^*N(0)\left(\frac{h}{h_t^*}\right)^2,$$

$$\beta^*=\beta_0\left[1-\left(\frac{h}{h_t^*}\right)^2\right],\quad \gamma^*=\gamma_0\left[1+p^*\left(\frac{h}{h_t^*}\right)^2\right],$$

$$h_t^*\simeq 1.6\frac{\xi}{L}T_{c0}, \tag{22}$$

the values of the numerical constants are $\eta^*\simeq 0.640$ and $p^*\simeq 1.032$;

(3) a layered cryptoferromagnetic superconducting phase CFS with $\Delta=\Delta_2^*$, $q_\perp=q_0^*$, $q_\parallel=\pi/L$, and $A^*>1$, where

$$\Delta_2^{*2}=\frac{-\beta^*+\sqrt{\beta^{*2}-4\alpha^*\gamma^*}}{2\gamma^*},\quad q_0^{*2}=\frac{5}{L^2}\left(1-\frac{1}{A^*}\right), \tag{23}$$

$$\alpha^*=\alpha_0+2\eta^*N(0)\left(\frac{h}{h_t^*}\right)^2\left(1-\frac{q_\perp^2L^2}{12}+\frac{q_\perp^4L^4}{120}\right),$$

$$A^*=\left(\frac{L}{\sqrt{\pi}\,\xi}\right)^4A,$$

with A defined in (9).

The corresponding phase diagrams (T_c^*,h) for two types of F/S superlattices are depicted schematically in Figs. 3a and b. Here and in what follows, in contrast to F/S junctions, all quantities characterizing the superlattice states are labeled by asterisks. Since the formulas determining the lines of phase transitions in the state diagrams of F/S lattices are similar to Eqs. (10)–(16) for F/S junctions, we do not give them here but only review the results.

We start with the phase diagram of superlattices of the first type, with $A_c^*<1$ (see Fig. 3a), where $A_c^*=A^*(h=h_c^*,T=0)$. The transition line consisting of the line T_{c0}–t^* of second-order transitions (with $\alpha_1^*=0$ and $b^*>0$) and the line t^*–h_c^* of the first-order transitions (with $\alpha_1^*=3\beta^{*2}/16\gamma^*$ and $\beta^*<0$) separates the quasi-two-dimensional (2D) magnetic behavior in the FN-phase from the three-dimensional (3D) in the AFS-phase. In the latter the antiferromagnetic alternation of the ferromagnetic layers due to by RKKY exchange through the superconducting layers largely balances the paramagnetic effect of localized spins. The critical and tricritical exchange fields, h_c^* and

h_t^*, therefore increase by a factor of ξ/L, but the effective parameter $A_c^* \simeq (L/\xi)^2 A_c$ decreases in comparison to the value for F/S-junctions.

The phase diagrams of F/S superlattices of the second type, with $A_c^* > 1$ (see Fig. 3b), also possess a well-defined feature, a Lifshitz point L_p^* at which three transition lines meet. Two of these are lines of the following first-order phase transitions: $t^*-L_p^*$ (with $\alpha_1^* = 3\beta^{*2}/16\gamma^*$), and $L_p^*-h_{c2}^*$. The latter is determined near L_p^* by Eq. (16) (after obvious changes in notation) and separates the FN(2D) state from the layered cryptoferromagnetic superconducting state CFS(3D), in which the phases of the sinusoidally modulated structures of localized spins in neighboring F-layers are shifted by π (the π-phase magnetism). The third line $L_p^*-h_{c1}^*$ (with $A^* = 1$), a line of second-order phase transitions (on it $q_0^* = 0$), separates the states AFS(3D) and CFS(3D). For $A_c^* - 1 \ll A_c^*$ the Lifshitz point (T_L^*, h_L^*) is located near the point $(0, h_c^*)$, and the lower and upper critical exchange fields, h_{c1}^* and h_{c2}^*, differ only slightly from h_c^*.

On the other hand, when $A_c^* \gg 1$, the Lifshitz point L_p^* shifts toward the tricritical point t^* and

$$h_{c1}^* \simeq \frac{h_c^*}{\sqrt{A_c^*}}, \quad h_{c2}^* = \frac{h_{c2}}{\sqrt{2}} \simeq h_c^* (A_c^*)^{1/4}, \quad q_{c2}^* = \frac{\sqrt{A_c^*}}{L},$$

where h_{c2} is the upper critical field of the F/S-junction. For the exchange field range $h_{c1}^* \leqslant h \leqslant h_c^*$ we have a CFS(3D)-state, while for $h_c^* < h < h_c^*$ we have the CFS(2D)-behavior, in which the strong modulation of the spin structures in neighboring F-layers ($q_\perp \gg L^{-1}$) leads to exponentially weak RKKY exchange between these layers (a 3D–2D crossover).[12] In the latter case the superlattice decays into a system of weakly coupled $S/F/S$ sandwiches, and there is no correlation between the phases of the modulated spin structures in neighboring F-layers, which means that the value of $q_\parallel$ can be chosen arbitrarily. Although the antiferromagnetic orientation of the magnetizations of the neighboring F-layers in the commensurate AFS-phase does shift the multicritical points t^* and L^* in the direction of higher exchange fields in comparison to those in F/S-junctions, there is no real increase in h_{c2}^* since 3D–2D crossover sets in earlier.

Thus, the emergence of a Lifshitz point L_p^* and of the corresponding incommensurate layered cryptoferromagnetic superconducting CFS-layers in the state diagrams of F/S superlattices is determined by the magnitude of the critical balance A_c^* of molecular fields. Just like F/S junctions, F/S superlattices can be divided into two types according to the value of the paramagnetic effect. As Fig. 4 implies, superlattices of the first type, with $A_c^* < 1$, have only one interphase boundary AFS(3D)–FN(2D), while superlattices of the second type, with $A_c^* > 1$, have two such boundaries: AFS(3D)–CFS(3D) and CFS(3D–2D)–FN(2D).

4. DISCUSSION

The possible ground states of F/S junctions and superlattices and the dependence of the critical parameters of the system, h_c, h_{c1}, h_{c2}, Δ_c, Δ_{c1}, Δ_{c2}, and q_{c2} on the quantities A_c and A_c^* which characterize the balance between the long-range antiferromagnetic interaction and the short-range ferromagnetic interaction, have been studied in the author's previous paper[12] (see also Figs. 2 and 4 in the present paper). Hence here we analyze the phase diagrams for nonzero temperatures and are interested primarily in the extraordinary behavior of F/S-systems near multicritical points, whose source is the competition between ferromagnetism and superconductivity.

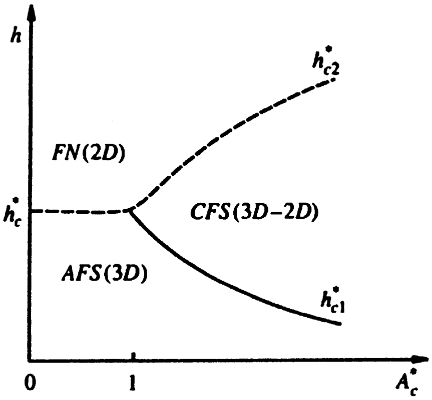

FIG. 4. Phase diagrams (h, A_c^*) of F/S superlattices at $T=0$. The dashed curves h_c^* and h_{c2}^* depict the interphase boundaries $AFS(3D)-FN(2D)$ and $CFS(3D-2D)-FN(2D)$, respectively, and the solid curve h_{c1}^* separates the $AFS(3D)$ and $CFS(3D)$ phases. The point at which all three curves intersect is the Lifshitz point.

The Lifshitz point in the phase diagrams of F/S-systems (see the lower halves of Figs. 1b and 3b), whose existence is predicted in the present work, is unusual, since it is preceded by a tricritical point at which the order of the phase transition changes from second to first. Physically such behavior is understandable and can be related to the specific properties of F/S-systems. The point here is that a transition to an incommensurate magnetically ordered phase is possible only if the balance of molecular fields, A or A^*, which is proportional to Δ^2, is greater than unity. However, this is impossible if we are moving along a line of second-order superconducting transitions with $\Delta = 0$. Only after the tricritical point t (t^*) has been passed, i.e., when the superconducting order parameter Δ on the line of first-order phase transitions, increasing from zero (see the upper halves of Figs. 1b and 3b), attains a value sufficient for A or A^* to become equal to unity, only then a Lifshitz point L_p or L_p^* appears on the line.

Usually the Lifshitz point in the state diagrams of substances that have incommensurate phases appears on the line of second-order phase transitions (see Refs. 15, 16, and 20). All three interphase boundaries (two lines of second-order transitions and one line of first-order transitions) that meet at an ordinary Lifshitz point have a common tangent at that point. In our case, however, the Lifshitz points L_p and L_p^* appear on the lines of first-order transitions that originate at the tricritical points t and t^*.

The lines of first- and second order transitions emerging at a Lifshitz point separate, respectively, the normal FN-phase from the incommensurate superconducting CFS-phase and the CFS-phase from the commensurate FS- or AFS-phase. Here the lines of first-order transitions parabolically merge at the Lifshitz point, while the line of second-order transitions forms a certain angle with their common tangent, an angle that decreases asymptotically like A_c^{-1} or $(A_c^*)^{-1}$ as the Lifshitz point approaches the tricritical point. A distinctive feature of the $FN-CFS$ interphase boundary is that interrelated first-order phase transitions (a superconducting and a cryptoferromagnetic) emerge when this boundary is crossed, with Δ and q_0 experiencing a jump.

This means that a new type of critical behavior (in comparison to the behavior discussed in Refs. 15 and 16) is to be expected in the vicinity of the Lifshitz points L_p and L_p^* For instance, for F/S junctions, this means that Eqs. (7)–(16) with $h \leqslant h_L$ yield

$$q_0=0,\ T_c'-T_{c1} \propto [A_c^{-1}(h_L-h)^{1/\phi^*}+\mathrm{const}\cdot(h_L-h)^{1/\phi}],$$

where in Landau's theory the critical crossover exponents ϕ and ϕ^* are equal to $\frac{1}{2}$ and 1, respectively. But if $h \geqslant h_L$, by introducing a new critical exponent β_q characterizing the variations of the modulation wave vector on the $T_{c2}(h)$ line we get

$$q_0 \propto (h-h_L)^{\beta_q}, \quad T_{c2}-T_c' \propto (h-h_L)^{1/\phi}.$$

In mean field theory $\beta_q=\frac{1}{2}$. Consequently, in contrast to the classical behavior of interphase boundary near a Lifshitz point,[15,16,20] in F/S junctions the behavior cannot be described by one universal crossover exponent ϕ. This becomes possible only when $A_c \rightarrow \infty$. Similar relationships can easily be derived for F/S superlattices.

Thus, the ratio A_c (for junctions) or A_c^* (for superlattices) of the antiferromagnetic and ferromagnetic molecular fields makes it possible to classify F/S-systems into two types, just as the Ginzburg–Landau parameter κ classifies type-I and type-II superconductors.

In F/S-systems of the first type, with A_c or A_c^* less than unity, only superconductivity in the S-layers can coexist with homogeneous ferromagnetic ordering in the F-layers (see Figs. 1a and 3b). The phase diagrams of F/S-systems of the second type (A_c or A_c^* greater than unity; see Figs. 1b and 3b) suggest that under certain conditions involving a change in temperature T or exchange field h, one can expect a cascade of alternating magnetic and superconducting transitions: $CFS \rightarrow FS \rightarrow FN$ for junctions, and $CFS(2D-3D) \rightarrow AFS(3D) \rightarrow FN(2D)$ for superlattices.

A similar chain of transitions generated by an external magnetic field parallel to the plane of the F/S-boundary explains[11,12] the increase and subsequent saturation of the exchange splitting of the BCS peak in the density of states of aluminum quasiparticles in EuO/Al junctions[13] and EuS/Al junctions.[14]

We also note that the weak suppression of superconductivity in EuO/V multilayers[3] can be explained by the large compensation of the exchange fields in vanadium layers at the expense of the π-phase compatibility of the magnetic structures of the localized spins in the neighboring F-layers in AFS or CFS states.

The presence of an incommensurate superconducting phase of the CFS type in EuO/Al and EuS/Al junctions and the likely presence of such a phase in EuO/V superlattices make these systems potential candidates for having a Lifshitz point, and hence possible objects of future experimental investigations.

Because of competition between superconductivity and ferromagnetism, changes in magnetic ordering in F-layers can probably only be directly observed by employing the magnetic neutron scattering method. A fact that might be used in experimental studies of state diagrams is that the vicinity of multicritical points can be passed by varying either the temperature T or the exchange field $h=I\langle S\rangle a/2L$, varying the thickness L of the superconducting layers or manufacturing these layers in the form of a wedge.

The dependence of the neutron elastic scattering cross section on the wave vector $\mathbf{k}$ is determined by the static spin susceptibility $\chi(k)$ of localized spins. In particular, for F/S junctions in the vicinity of the Lifshitz point L_p and the $FS-CFS$ transition line ($|A-1| \ll A$) at $\mathbf{k} \| \mathbf{q}_\perp$, the expansion (5) for the free energy yields

$$\chi^{-1}(k) \propto \{\mathrm{const}+\Delta^2[1-b(1-A^{-1})k^2\xi^2+gk^4\xi^4]\}.$$

The last two terms can also be shown to determine the spin-wave spectrum of the system near the $T_{c1}(h)$ line. In crossing this line ($h<h_L$) by varying the temperature, the factor of k^2 changes sign, passing through zero ($A=1$) continuously. Hence on the transition line $FS-CFS$ the dispersion curves of the spin waves at small k become unusually flat, and the χ vs k dependence becomes non-Lorentzian. At the same time, when the Lifshitz point L_p ($h=h_L$) is passed from high temperatures to low, the phase changes from FN to CFS, and the factor of k^2 for the first time vanishes suddenly. At other neighboring points of the $T_{c2}(h)$ line of first-order transitions the factor of k^2 suddenly changes sign.

In the incommensurate CFS-phase ($A>1$), the peak in the χ vs k dependence and, accordingly, the minimum in the spin-wave spectrum, are far from the Brillouin zone and are attained at $k=q_0$.

Figure 1b also suggests the possibility of reciprocal FS-behavior, since the T_{c1} vs h curve passes through a maximum, which is to the left of the Lifshitz point ($h_{c1}<h_{\max}$). Reciprocal behavior can be observed by varying the exchange field h at fixed temperature T ($T_L<T<T_{c1}^{\max}$). The tricritical point can be recorded at the moment when continuous broadening of the Lorentzian peak in the χ vs k dependence changes to sudden broadening along the $T_{c0}-t-L_p$ line in the $FN-FS$ transition.

A similar study can easily be done with F/S superlattices if one also allows for antiferromagnetic coupling between neighboring ferromagnetic layers through the superconducting layer. This leads to significant anisotropy in the system's response $\chi(k)$ and to radically different behavior in two directions, $\mathbf{k} \| \mathbf{q}_\perp$ and $\mathbf{k} \| \mathbf{q}_\|$.

Note, however, that the phase diagrams and the positions of the multicritical points t and L_p obtained in Landau's theory are of a qualitative nature, and require refining that would allow for fluctuations of the order parameters Δ and $\langle S_{\mathbf{r}}^{(\pm)}\rangle$, and a realistic temperature dependence of the coefficients α, β, and γ in the functional (5). For instance, ignoring the latter feature results in the reference critical fields h_c and h_c^* being a factor of approximately 1.5 times the values found in Ref. 12 in a more accurate solution for the ground state of F/S junctions and superlattices. The need to simultaneously describe two multicritical points, t and L_p, in the phase diagrams of F/S-systems of the second type considerably complicates an accurate microscopic analysis of the free-energy functional by requiring that terms up to Δ^6 and $q_\perp^4$ inclusive be present in the expansion, although such calculations are possible.

In principle, the model of exchange interactions employed here is probably applicable to boride nickel carbides of the $HoNi_2B_2C$ type, which because of alternation of nonconducting Ho–C planes and (super)conducting Ni_2B_2 layers,[21,22] are natural microscopic analogs of the F/S superlattice considered in this paper. Indeed, assuming that the $4f$-momenta of the Ho^{2+} ions are strongly coupled by the intralayer direct (or super-) exchange interaction, and the weak but long-range intra- and interlayer RKKY exchange via the conduction electrons of the Ni_2B_2 layer, we can describe the transformation of the helical structure into the layered antiferromagnetic structure,[23] which is dominant after the transition to the superconducting state.[21,22]

In thin insulator F-layers (of order 10 Å), allowing for the tunneling of conduction electrons leads to Josephson coupling between S-layers, and the π-phase magnetism, as a variant of mutual accommodation of superconductivity and ferromagnetism, will likely be augmented by π-phase superconductivity, suggested in Refs. 4 and 5 for metallic F/S superlattices. In turn, allowing for indirect exchange between localized spins through the superconducting layers in metallic F/S-multilayers leads to competition between the two competing (π-phase) variants of coexistence of two competing types of long-range order. Hence it is to be expected that, in principle, the phase diagrams of F/S superlattices can have other multicritical points (in addition to t^* and L_p^*) and can become even more nontrivial. The current problem, and the problem of allowing for uniaxial magnetic anisotropy and an external magnetic field in the state diagrams of F/S-systems, require new theoretical—and, more to the points,—experimental research.

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