

Indirect RKKY exchange and magnetic states of ferromagnet–superconductor superlattices

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(Submitted 27 March 1995; resubmitted 31 July 1995)
Zh. Éksp. Teor. Fiz. **109**, 524–545 (February 1996)

It is shown that antiferromagnetic RKKY correlations between localized spins, caused by the transition of a metal to the superconducting state, increase in strength as one approaches the surface of the sample, and also as one decreases its dimensionality. On this basis we propose a model of exchange interactions between localized spins in ferromagnetic insulator/superconductor (F/S) junctions and superlattices. This model takes account of indirect RKKY exchange between localized spins located at the F/S boundaries through conduction electrons of the S layers as well as direct exchange of nearest neighbors in the F layers. By using this model we have examined the ground states of F/S systems, and also determined possible variants of the mutual accommodation of the superconducting and magnetic order parameters. We find that F/S systems, depending on the relation between the antiferromagnetic and ferromagnetic molecular fields acting on each localized spin of the F/S boundary, divide into two types. F/S systems of the first type allow only a homogeneous ferromagnetic ordering in the F layers to coexist with superconductivity in the S layers. In this case the magnetizations of the neighboring F layers in the superlattices are antiparallel. In F/S systems of the second type the superconducting layers can induce a cryptoferrimagnetic modulation in the spin structure of the F layers. Depending on the magnitude of the exchange field induced by the localized spins of the F/S boundary at the conduction electrons of the S layer, the modulation period can be both larger than the coherence length of the superconductor ξ and smaller. The phases of the magnetic order parameters in neighboring F layers of the superlattice in this case are shifted by π . These results explain the nature of the extra splitting of the density of states of the conduction electrons in EuO/Al and EuS/Al contacts in a magnetic field, and also the weak suppression of superconductivity in EuO/V multilayers.
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1. INTRODUCTION

Superconductivity and ferromagnetism are two competing types of long-range order and their coexistence in one crystal is practically impossible. The point here is that the strong exchange field of the localized spins of a ferromagnet leads to a separation of the Fermi surfaces of the conduction electrons with antiparallel spins and thereby destroys the Cooper pairs (the paramagnetic effect, see Ref. 1). In turn, the singlet pairing of the conduction electrons in a superconductor gives rise to the appearance of a long-range antiferromagnetic contribution to the Ruderman–Kittel–Kasuya–Yosida (RKKY) indirect exchange between the localized spins² and thereby renders the purely ferromagnetic state unstable.³ It is important to note that the efficiency of both mechanisms, reflecting the mutual influence that superconductivity and ferromagnetism have on each other, is determined by the difference of the spin susceptibilities of the conduction electrons $\chi_n(q) - \chi_s(q)$ in the normal and superconducting states. This difference is greatest for $q=0$, which makes the coexistence of superconductivity and ferromagnetism energetically unfavored. At the same time, for values of q greater than ξ^{-1} , where ξ is the coherence length of the superconductor, the difference $\chi_n(q) - \chi_s(q)$ is small, and a compromise becomes possible, i.e., the mutual accommodation of the superconducting and magnetic ordering predicted

in Ref. 3 and investigated in detail in Ref. 4 (see also Ref. 5 and the references therein). This phase, called the cryptoferrimagnetic phase, is characterized by a long-wave (in comparison with the period of the magnetic lattice a) and, at the same time, small-scale (in comparison with the size of the Cooper pair ξ) modulation of the magnetic order of the localized spins. This leads, on the one hand, to conservation of the short-range ferromagnetic order of the localized spins and a not-too-large loss in the exchange energy, and on the other hand, to an effective averaging of the spin polarization of the conduction electrons and conservation of superconducting pairing. The main deficiency of homogeneous materials in achieving of the coexistence phase is the obvious requirement that the Curie temperature Θ be lower than the superconducting transition temperature T_c (Ref. 5). Therefore it is not surprising that superconductivity and inhomogeneous magnetic ordering has been observed to coexist only in a few ternary compounds of the types ErRh_4B_4 and HoMo_6S_8 (see references in Refs. 4 and 5).

In connection with the search for alternative systems in which such competing phenomena may be combined, the superconducting and magnetic properties of F/S superlattices formed by the alternation of layers of a ferromagnet (F) and a superconductor (S) are now being actively examined (see the review in Ref. 6). In such systems the regions in which the magnetic and superconducting order parameters

act are spatially separated, and the paramagnetic effect can be compensated to a significant degree both by the choice of the thicknesses of the F and S layers and as a result of mutual accommodation of the two antagonistic types of long-range order. A variant of such accommodation with the formation of a banded domain structure in a thin metallic ferromagnet on the surface of a pure, massive superconductor was discussed in Ref. 7. The authors of Ref. 7 suggested that the appearance of such a domain structure may explain the coexistence of superconductivity and the phenomenon of surface ferromagnetism, discovered recently in massive and thin-film samples of vanadium.^{8,9} At the same time, the coexistence of ferromagnetism and superconductivity, the unexpected growth of T_c with increase of the thickness of the Fe layers, and the transition from two-dimensional (2D) to three-dimensional (3D) behavior with increase of temperature or decrease of the thickness of the vanadium layers, detected in Fe/V superlattices,⁶ apparently cannot be explained solely on the basis of the “ π -phase” character of superconductivity in neighboring vanadium layers separated by a layer of iron.¹⁰ For a more complete analysis of the different types of mutual accommodation of superconductivity and ferromagnetism in F/S -systems, it is necessary to answer the question, by what mechanism do the magnetic layers interact through the superconducting layers. This question, by the way, is very important for very thin vanadium films with ferromagnetically ordered surfaces. Similar problems, connected with the competition of superconductivity with ferromagnetism, also arise in other F/S -systems, where, for example, F is a ferromagnetic insulator (see Refs. 6 and 11). In particular, the nature of the internal fields causing the splitting of the BCS peak in the density of states of the conduction electrons of aluminum in EuO/Al/Al₂O₃/Al (Ref. 12), EuS/Al/Al₂O₃/Ag (Ref. 13), and Au/EuS/Al (Ref. 14) tunnel contacts, where EuO and EuS are ferromagnetic insulators, is not clear. This splitting is observed as an extra splitting (in addition to the Zeeman splitting) in the presence of an external magnetic field and saturates with growth of this field, while in contacts with EuS^{13,14} it takes place in zero field. With increase of the magnetic field in F/S -contacts¹²⁻¹⁴ a first-order phase transition to the normal state occurs, although theory predicts a second-order transition for this region.¹¹

In the present paper we propose a simple model of exchange interactions between localized spins in junctions and F/S (ferromagnetic insulator/superconductor) superlattices. Together with the direct exchange of nearest neighbors in the ferromagnetic layers, it also allows for RKKY indirect exchange between localized spins, located on the F/S boundaries, through the conduction electrons of the superconducting layers. Using this model we examine the ground states of F/S systems, and also determine the possible types of inhomogeneous magnetic structures and the conditions of their coexistence with superconductivity. In Sec. 2 we find the dependence of RKKY exchange on the distance between the localized spins and on their positions relative to the boundaries of the superconducting sample for three different geometries: a half-space, a lamina, and a wire. On the basis of the results obtained in Sec. 2 and Sec. 3 we examine different

types of mutual accommodation of the superconducting and magnetic order parameters for two types of flat F/S junctions between a thin film of some ferromagnetic insulator and a superconducting substrate. An analogous problem is solved in Sec. 4 for F/S superlattices with allowance for RKKY exchange between the localized spins of neighboring F layers through the interlaid S layers. Section 5 is the conclusions and discussion of results.

2. THE EFFECT OF BOUNDARIES ON RKKY INDIRECT EXCHANGE IN SUPERCONDUCTORS

In BCS theory a dirty superconductor of finite dimensions with conduction electrons scattering off nonmagnetic impurities can be described by the Hamiltonian

$$H_S = \sum_{\alpha} \int d^3r \Psi_{\alpha}^{\dagger}(\mathbf{r}) \left[\frac{\hat{\mathbf{p}}^2}{2m} + \sum_i V \delta(\mathbf{r}-\mathbf{r}_i) \right] \Psi_{\alpha}(\mathbf{r}) - \lambda \int d^3r \Psi_{\uparrow}^{\dagger}(\mathbf{r}) \Psi_{\downarrow}^{\dagger}(\mathbf{r}) \Psi_{\downarrow}(\mathbf{r}) \Psi_{\uparrow}(\mathbf{r}), \quad (1)$$

where the integration is over the volume bounded by the surface σ of the superconductor-insulator (vacuum) interface, $\Psi_{\alpha}^{\dagger}(\mathbf{r})$ and $\Psi_{\alpha}(\mathbf{r})$ are the electron field operators, $\alpha = \uparrow, \downarrow$ are the projections of the electron spin on the quantization axis, V is the potential of the nonmagnetic impurities, the index i runs over the positions of these impurities, and, finally, λ is the interelectron interaction constant responsible for the superconducting correlations of the conduction electrons.

The dependence of the RKKY exchange integral on the distance between the localized spins \mathbf{S}_r and $\mathbf{S}_{r'}$ is determined, as is well known,¹⁵ by the spatial dispersion of the spin susceptibility of the conduction electrons $\chi(\mathbf{r}, \mathbf{r}')$, and the Hamiltonian of the indirect exchange has the form

$$H_{ex} = -\frac{1}{8} I^2 \sum_{\mathbf{r}, \mathbf{r}'} \chi(\mathbf{r}, \mathbf{r}') (\mathbf{S}_r \mathbf{S}_{r'}), \quad (2)$$

where I is the s - d -exchange integral; hence, in this paper we set $\hbar = k_B = \mu_B = 1$. In the normal phase the dependence $\chi_n(\mathbf{r}, \mathbf{r}')$ has the form of the characteristic Friedel oscillations, and its integral over all space gives the homogeneous Pauli susceptibility.

It was shown in Ref. 2 that in a dirty superconductor described by the Hamiltonian (1), the local spin polarization corresponding to the normal phase is compensated by the long-range contribution of the antiferromagnetic term. This additional contribution to the RKKY exchange results from excluding the paired electron contribution from the homogeneous spin polarization. The superconducting contribution to the susceptibility $\delta\chi_s(\mathbf{r}, \mathbf{r}')$ can be represented in the form

$$\delta\chi_s(\mathbf{r}, \mathbf{r}') = \chi_s(\mathbf{r}, \mathbf{r}') - \chi_n(\mathbf{r}, \mathbf{r}') = -2T \sum_{\omega} \Lambda_s(\mathbf{r}, \mathbf{r}', \omega). \quad (3)$$

For the case considered in Ref. 2 of an infinite, dirty superconductor the two-particle correlator $\Lambda_s(\mathbf{r}, \mathbf{r}', \omega)$ in the hydrodynamic limit, i.e., at distances $R = |\mathbf{r} - \mathbf{r}'|$ exceeding the mean free path l , is given by

$$\Lambda_s(\mathbf{r}, \mathbf{r}', \omega) = \frac{N(0)\Delta^2}{2DR(\omega^2 + \Delta^2)} \exp\left(-\frac{R}{\xi_\omega}\right). \quad (4)$$

Here $N(0)$ is the density of states of the conductivity electrons on the Fermi surface, Δ is the superconducting order parameter, D is the diffusion coefficient,

$$\xi_\omega = \sqrt{D/2} \sqrt{\omega^2 + \Delta^2}$$

is the radius of action of the correlator $\Lambda_s(\mathbf{r}, \mathbf{r}', \omega)$ and depends on the frequency $\omega = \pi T(2n + 1)$, T is the temperature, and $n = 0, \pm 1, \pm 2, \dots$. Since the homogeneous spin polarization in a superconductor should vanish at $T = 0$, in analogy with Ref. 16 we may derive a sum rule for $\Lambda_s(\mathbf{r}, \mathbf{r}', \omega)$. In the case of a homogeneous superconductor with order parameter Δ and density of states $N(0)$ independent of the coordinates, this sum rule has the form

$$\int d^3r' \Lambda_s(\mathbf{r}, \mathbf{r}', \omega) = \frac{\pi N(0)\Delta^2}{(\omega^2 + \Delta^2)^{3/2}}. \quad (5)$$

To find the long-range part of the RKKY exchange in the case of a superconductor bounded by the surface σ , it is convenient to represent $\Lambda_s(\mathbf{r}, \mathbf{r}', \omega)$ in the form of the solution of a boundary-value problem. It can be shown that expression (4) is the solution of a differential equation of diffusion type:

$$(2\sqrt{\omega^2 + \Delta^2} - D\nabla_r^2)\Lambda_s(\mathbf{r}, \mathbf{r}', \omega) = 2\pi N(0) \times \frac{\Delta^2}{\omega^2 + \Delta^2} \delta(\mathbf{r} - \mathbf{r}'). \quad (6)$$

The boundary conditions on this equation are found by integrating Eq. (6) over \mathbf{r} using the sum rule (5); they have the form

$$D\mathbf{n} \cdot \nabla_r \Lambda_s(\mathbf{r}, \mathbf{r}', \omega)|_\sigma = 0, \quad (7)$$

where \mathbf{n} is the normal to the superconductor–vacuum (insulator) interface σ . Physically, Eq. (7) corresponds to the absence of a Cooper pair flux through the superconductor surface.

Solving Eq. (6) together with the boundary condition (7) under the assumption that the density of states $N(0)$ and the order parameter Δ are constant and vanish discontinuously at the superconductor surface σ (Ref. 17), we can obtain the spatial dependence of the correlator $\Lambda_s(\mathbf{r}, \mathbf{r}', \omega)$ for all geometries of practical interest. We will consider three of them here: a superconducting half-space, a superconducting lamina, and a superconducting wire.

Thus, in the case of a superconducting half-space $z, z' \geq 0$, we have

$$\Lambda_s(\mathbf{r}, \mathbf{r}', \omega) = \frac{\pi N(0)\Delta^2}{\omega^2 + \Delta^2} \int \frac{d^2q_\perp \exp[i\mathbf{q}_\perp(\boldsymbol{\rho} - \boldsymbol{\rho}')] }{(2\pi)^2 Dk} \times \{\exp(-k|z - z'|) + \exp(-k(z + z'))\}, \quad (8)$$

where $k^2 = q_\perp^2 + \xi_\omega^{-2}$, $\mathbf{q}_\perp = i\mathbf{q}_x + j\mathbf{q}_y$, and $\boldsymbol{\rho} = i\mathbf{x} + j\mathbf{y}$. It is interesting to note that a spin pair on the surface of the superconductor ($z = z' = 0$) interacts twice as strongly as in its volume for $z = z' > \xi$ (here

$$\xi = \xi_{\omega 0} = \sqrt{D/2} \sqrt{\pi^2 T^2 + \Delta^2}$$

is the coherence length of the superconductor), where the formula (4) is valid for $\Lambda_s(\mathbf{r}, \mathbf{r}', \omega)$. Thus, elastic reflection of Cooper pair from the surface of the superconductor leads to its own kind of interference with amplification.

For a superconducting lamina of thickness L , i.e., when $0 \leq z, z' \leq L$, where $L \gg l$, we obtain

$$\Lambda_s(\mathbf{r}, \mathbf{r}', \omega) = \frac{2\pi N(0)\Delta^2}{\omega^2 + \Delta^2} \times \int \frac{d^2q_\perp \exp[i\mathbf{q}_\perp \cdot (\boldsymbol{\rho} - \boldsymbol{\rho}')] }{(2\pi)^2 Dk \sinh(kL)} \cosh(kz) \times \cosh[k(z' - L)] \quad (9)$$

for $z < z'$. If, on the contrary, $z > z'$, then it is necessary for them to change places in Eq. (9). It is not hard to convince oneself that in the case of a massive lamina ($L > \xi$) the antiferromagnetic coupling between the localized spins on each of the surfaces is two times stronger than in the interior. In the limit $L \rightarrow \infty$, expression (9) goes over to expression (8) for a half-space. However, the above result (9) is more graphically illustrated in the quasi-two-dimensional situation in which the film thickness L is small in comparison with the coherence length ξ . Then the interaction of the localized spins is practically independent of z and z' and is determined only by the projection of the radius vector \mathbf{R} on the $z = 0$ plane, i.e., by $R_\perp = |\boldsymbol{\rho} - \boldsymbol{\rho}'|$:

$$\Lambda_s(\mathbf{r}, \mathbf{r}', \omega) = \frac{N(0)\Delta^2}{DL(\omega^2 + \Delta^2)} K_0\left(\frac{R_\perp}{\xi_\omega}\right). \quad (10)$$

Here, as follows from the asymptotic behavior of the modified Bessel function

$$K_0\left(\frac{R_\perp}{\xi_\omega}\right) \propto \ln \frac{\xi_\omega}{R_\perp}, \quad R_\perp < \xi_\omega, \\ K_0\left(\frac{R_\perp}{\xi_\omega}\right) \propto \sqrt{\frac{\xi_\omega}{R_\perp}} \exp\left(-\frac{R_\perp}{\xi_\omega}\right), \quad R_\perp > \xi_\omega,$$

the power-law falloff of the RKKY potential with distance between the localized spins is weaker in comparison with the three-dimensional case (4), and antiferromagnetic correlations of localized spins at distances $R_\perp \leq \xi$ are enhanced. From a physical point of view, such a modification of RKKY exchange is due, apparently, to a “simplification” of the wave function of the Cooper pairs in a quasi-two-dimensional film with conservation of the phase volume they occupy.

For a superconducting wire of radius L , i.e., for $0 \leq \rho, \rho' \leq L$, ($L \gg l$), we have

$$\Lambda_s(\mathbf{r}, \mathbf{r}', \omega) = \frac{4N(0)\Delta^2}{D(\omega^2 + \Delta^2)} \sum_{m=0}^{\infty} \frac{\cos m\varphi}{1 + 3\delta_{m,0}} \times \int_{-\infty}^{\infty} \frac{dq_\parallel \exp[iq_\parallel(z - z')]}{2\pi I'_m(kL)} I_m(k\rho) \times [K_m(k\rho')I'_m(kL) - I_m(k\rho')K'_m(kL)] \quad (11)$$

for $\rho < \rho'$. If, on the contrary, $\rho > \rho'$ holds, then it is necessary for them to change places in Eq. (11). The symbol φ denotes the angle between the radius vectors $\boldsymbol{\rho}$ and $\boldsymbol{\rho}'$; we have set $q_{\parallel} = q_z$, $k^2 = q_{\parallel}^2 + \xi_{\omega}^{-2}$; and $I_m(x)$, $K_m(x)$ and $I'_m(x)$, $K'_m(x)$ are the modified Bessel functions of first and second order and their derivatives. In the quasi-one-dimensional case, when the radius of the wire satisfies $L \ll \xi$, expression (11) substantially simplifies:

$$\Lambda_s(\mathbf{r}, \mathbf{r}', \omega) = \frac{N(0)\Delta^2}{\omega^2 + \Delta^2} \frac{\xi_{\omega}}{DL^2} \exp\left(-\frac{|z-z'|}{\xi_{\omega}}\right), \quad (12)$$

and the energy of the antiferromagnetic interaction between the localized spins at distances $|z-z'| \leq \xi$ is practically constant.

Thus, the antiferromagnetic correlations of the localized spins, induced by the transition of the metal to the superconducting state, strengthen as one approaches the surface of a massive, dirty superconductor and as one lowers the dimensionality of the sample. It is not hard to convince oneself by direct integration that all of the above results (8)–(12) satisfy the sum rule (5), thereby ensuring that the homogeneous spin susceptibility χ_s equals zero at $T=0$.

3. MUTUAL INFLUENCE OF SUPERCONDUCTIVITY AND FERROMAGNETISM IN F/S CONTACTS

As an application of the results obtained in Sec. II, let us consider a flat junction between a thin film of some ferromagnetic insulator (F) occupying the region $0 < z < L$. The subsystems of localized spins of the ferromagnet and the conduction electrons of the superconductor, coupled with each other across the F/S boundary, can be described by the Hamiltonian

$$\begin{aligned} H &= H_F + H_S + H_{F/S}, \\ H_F &= -J \sum_{\mathbf{r}, \mathbf{a}} (\mathbf{S}_{\mathbf{r}} \mathbf{S}_{\mathbf{r}+\mathbf{a}}) + D \sum_{\mathbf{r}} (S_{\mathbf{r}}^z)^2, \\ H_{F/S} &= \frac{I}{2} \sum_{\mathbf{r}, \alpha, \beta} \Psi_{\alpha}^+(\mathbf{r}) (\mathbf{S}_{\mathbf{r}} \sigma_{\alpha\beta}) \Psi_{\beta}(\mathbf{r}), \end{aligned} \quad (13)$$

where the Hamiltonian H_F takes account of direct exchange of the localized spins of the F film, and the exchange integral J is greater than zero only for nearest neighbors located at the sites \mathbf{r} of a simple cubic lattice with period a . In addition, we assume that the localized spins of the F film are ordered as in an "easy plane," i.e., the magnetic anisotropy constant is greater than zero: $D > 0$. The Hamiltonian H_S in Eq. (13) [see Eq. (1)] describes the Cooper pairing of the conduction electrons scattering off the nonmagnetic impurities in the S layer. Finally, the last term $H_{F/S}$ describes the s - d -exchange interaction of the localized spins of the ferromagnet with the conduction electrons of the superconductor, and the prime on the summation sign indicates that \mathbf{r} runs only over the localized spins located on the F/S boundary. Here I is the s - d -exchange constant, σ is the Pauli matrix, and α and β are the spin indices of the conduction electrons. The proposed mechanism of the $H_{F/S}$ interaction is as follows. The overlap of the conduction electron wave functions with the s and d orbitals of the first atomic layer of the F

film leads to virtual transport of electrons out of the superconductor into the insulator and *vice versa*. The corresponding effective s - d -exchange integral I can be approximately

$$I \approx I_0 \frac{t_s^2}{(E_s - E_0)^2} + \frac{t_d^2}{E_0 - E_d}, \quad (14)$$

where I_0 is the Hund intra-atomic s - d exchange, and t_s and t_d are the transport integrals between the metal conduction band with characteristic Fermi energy E_0 and the s and d shells of the magnetic atom with energies E_s and E_d , respectively. Clearly, I is substantially less than I_0 and to a significant degree is determined by the quality of the F/S boundary, i.e., it depends strongly on the conditions and technique of preparation. The interaction $H_{F/S}$ leads, on the one hand, to a splitting of states of the conduction electrons in the S layer by the exchange field of the localized spins of the F/S boundary, and, on the other, to the indirect exchange (2) of these same localized spins through the conduction electrons of the superconducting substrate.

For definiteness in what follows, we will assume that the Curie temperature Θ is higher than the superconducting transition temperature T_c and that for $T_c < T < \Theta$ the ferromagnetism of the surface of the F film is not spoiled by the oscillations of the normal part of the RKKY exchange. This latter assumption in turn assumes that direct exchange at of nearest-neighbor distances is stronger than indirect exchange, i.e., $J > I^2 N(0)$. In addition, we will assume in what follows that the thickness of the F film d is much less than δ , where δ is the penetration depth of the surface distortions of the magnetic ordering in the ferromagnet.¹⁸ In this limit the long-wave modulations of the magnetic ordering of the localized spins generated at the F/S surface will be communicated over the entire thickness of the F film as a result of the strong interatomic exchange J . For simplicity, we also assume that the thickness of the S layer is small, i.e., $L \ll \xi$. In this case suppression of the order parameter Δ by the exchange field of the localized spins bordering on the superconductor occurs uniformly over the entire thickness of the S layer.¹¹

Averaging the Hamiltonian (3) over the electron and spin variables in the self-consistent field approximation in the spirit of Refs. 4 and 5, and assigning the magnetic order in the F film in the form

$$\langle S_{\mathbf{r}}^{\pm} \rangle = \langle S_{\mathbf{r}}^z \pm i S_{\mathbf{r}}^y \rangle = S \exp(\pm i \mathbf{q}_{\perp} \cdot \boldsymbol{\rho}), \quad \langle S_{\mathbf{r}}^z \rangle = 0, \quad (15)$$

for the surface density of the free energy of the F/S contact at $T=0$, we obtain the following functional:

$$\begin{aligned} f &= f_F^0 + f_N^0 + JS^2 q_{\perp}^2 \frac{d}{a} - I(q_{\perp}, 0, 0) \frac{S^2}{a^2} \\ &\quad - \frac{LN(0)}{2a^3} \Delta^2 \ln \frac{e\Delta_0^2}{\Delta^2}, \end{aligned} \quad (16)$$

where f_F^0 and f_N^0 are the free energies per unit area of the F/S junction for the F film and the S layer, respectively, in the normal phase. The third term describes the loss of the direct exchange energy due to the long-wave ($q_{\perp} a \ll 1$) modulation of the ferromagnetic ordering of the localized spins. The fourth term is the two-dimensional Fourier trans-

form of the superconducting contribution $I(\rho-\rho',z,z')$ to the RKKY potential, which is found by substituting expression (9) in Eqs. (3) and (2), i.e.,

$$I(q_{\perp},z,z') = -\frac{a}{2}N(0)I^2\pi T \times \sum_{\omega} \frac{\Delta^2}{\omega^2+\Delta^2} \frac{\cosh(kz)\cosh[k(z'-L)]}{Dk \sinh(kL)}. \quad (17)$$

After summing over ω , we can represent the low-temperature ($\pi T \ll \Delta$) asymptotic limits of the exchange integral $I(q_{\perp},0,0)$ for the various regions of variation of the wave vector q_{\perp} in the form

$$I(q_{\perp},0,0) = -\frac{a}{4L}N(0)I^2 \left(1 + \frac{\pi L^2}{6\xi^2} - \frac{\pi}{4}q_{\perp}^2\xi^2 + \frac{2}{3}q_{\perp}^4\xi^4 \right), \quad q_{\perp} < \xi^{-1},$$

$$I(q_{\perp},0,0) = -\frac{\pi a}{8L}N(0)I^2(q_{\perp}\xi)^{-2}, \quad \xi^{-1} < q_{\perp} < L^{-1},$$

$$I(q_{\perp},0,0) = -\frac{\pi a}{8\xi}N(0)I^2(q_{\perp}\xi)^{-1}, \quad L^{-1} < q_{\perp} < l^{-1}, \quad (18)$$

where $\xi = \sqrt{D/2\Delta}$. The term proportional to $I(q_{\perp},0,0)$ in expression (16) plays a double role. On the one hand, it describes the long-range antiferromagnetic correlations of the localized spins on the boundary ($z=z'=0$) via the Cooper pairs of the superconductor, and, on the other, it takes account of suppression of the order parameter Δ due to the paramagnetic effect of these spins. Finally, the last term in expression (16) describes the gain in the condensation energy (see Refs. 4 and 5) associated with the transition of the S layer to the superconducting state. Here $\Delta_0 = 1.76T_{c0}$, $e = 2.718$, and we neglect the difference in the lattice constants of the F and S layers ($a_F = a_S = a$).

Minimization of functional (16) over Δ and q_{\perp} using the first of expressions (18) for $I(q_{\perp},0,0)$ for $q_{\perp}\xi \ll 1$ leads to three different ground states or phases of the F/S junction, whose realization depends on the values of the parameters A and h :

$$A = \frac{N(0)h^2\pi\xi^2L}{JS^2 \cdot 4a^2d}, \quad h = IS \frac{a}{2L}. \quad (19)$$

The parameter A is the ratio of the absolute values of the antiferromagnetic and ferromagnetic molecular fields per localized spin of the F/S boundary due respectively to RKKY exchange via the superconducting electrons of the S layer and to direct exchange in the F layer. The quantity h is the mean exchange field acting on the conduction electrons coming from the localized spins at the F/S boundary.

In the case $A < 1$ ferromagnetic ordering is stable with respect to long-wave modulation, the tendency toward which arises thanks to RKKY exchange. In this case, if the exchange field h is not too large, superconductivity in the S layer arises against a background of ferromagnetism in the

F film and they coexist (the FS phase). In the FS phase the equilibrium values of the order parameter Δ and the magnetic structure wavevector Q_{\perp} are given by

$$\Delta^2 \ln \frac{\Delta_0}{\Delta} = \frac{\pi h^2}{12} \left(\frac{L}{\xi} \right)^2, \quad Q_{\perp} = 0. \quad (20)$$

The surface density of the free energy in this phase is equal to

$$f_{FS} = f_F^0 + f_N^0 - \frac{LN(0)}{2a^3} \left[\Delta^2 - 2h^2 \left(1 + \frac{\pi L^2}{12\xi^2} \right) \right]. \quad (21)$$

For $A > 1$, ferromagnetic ordering is unstable with respect to long-wave modulation and the free energy minimum (16) corresponds to the cryptoferromagnetic superconducting phase CFS , the parameters of which, Δ and Q_{\perp} are found from the self-consistent equilibrium conditions

$$\Delta^2 \ln \frac{\Delta_0}{\Delta} = \frac{\pi h^2}{4} \left[\frac{L^2}{3\xi^2} + \frac{(Q_{\perp}\xi)^2}{2A} \right],$$

$$Q_{\perp} = \frac{1}{\xi} \sqrt{\frac{3\pi}{16} \left(1 - \frac{1}{A} \right)}. \quad (22)$$

The free energy density in the CFS phase is given by

$$f_{CFS} = f_F^0 + f_N^0 - \frac{LN(0)}{2a^3} \left[\Delta^2 - 2h^2 \left(1 + \frac{\pi L^2}{12\xi^2} - \frac{\pi}{8}Q_{\perp}^2\xi^2 \right) \right]. \quad (23)$$

Comparing the free energies (21) and (23), it follows that the transition from the FS ground state to the CFS ground state takes place at $A = 1$ and is accompanied by a mutual accommodation of the superconducting and magnetic order parameters. Here the neighborhood $A - 1 \ll A$ of the transition point corresponds to large-scale sinusoidal modulation of the magnetic order of the localized spins in the F film. On the one hand, it leads to a partial compensation of the paramagnetic effect, and, on the other, to minimal loss of the direct exchange energy, since $Q_{\perp}^{-1} \gg \xi \gg a$.

With increase of the exchange field h , when the gain associated with the condensation energy is compensated by the paramagnetic effect and the loss in the direct exchange energy, the F/S junction transitions to the ferromagnetic normal state FN with $\Delta = Q_{\perp} = 0$ and free energy $f_{FN} = f_F^0 + f_N^0$. This transition can originate in either the CFS phase or the FS phase.

From the above low-temperature ($T=0$) analysis it follows that as the magnitude of the exchange field h grows and the ratio A of the molecular fields given by (19) correspondingly increases, the F/S junctions may behave quite differently. The two possible forms of the dependence of the superconducting order parameter Δ and magnetic structure wave vector Q_{\perp} on h are schematically depicted in Fig. 1.

In the case of F/S junctions of the first type (see Fig. 1a) the quantity A remains smaller than unity up to the critical value of the exchange field h_c . The critical parameters (h_c, Δ_c) of the first-order phase transition from the FS state to the FN state are found by equating the free energies of these phases and are given by

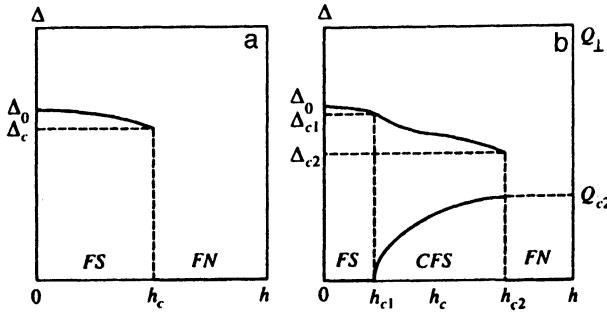


FIG. 1. Dependence of the superconducting order parameter Δ and wave vector Q_{\perp} of the magnetic structure on the exchange field h for F/S junctions a) of the first type with $A_c < 1$ ($Q_{\perp} = 0$) and b) of the second type with $A_c > 1$.

$$h_c \approx \frac{\Delta_0}{\sqrt{2}} \left(1 - \frac{\pi L^2}{12 \xi_a^2} \right), \quad \Delta_c \approx \Delta_0 \left(1 - \frac{\pi L^2}{24 \xi_0^2} \right), \quad (24)$$

where $\xi_0 = \sqrt{D/2\Delta_0}$ is the coherence length at $T=0$ and $h=0$. Small corrections of the order of $(L/\xi)^2$ to the well-known result for homogeneous ferromagnetic superconductors¹ $h_c = \Delta_0/\sqrt{2}$ and $\Delta_c = \Delta_0$ arise because the exchange field destroying the Cooper pairs is created by the localized spins located on the surface, and not in the interior of the sample.

For F/S junctions of the second type (see Fig. 1b) the ratio of the molecular fields is less than unity ($A < 1$) only up to some lower critical value of the exchange field h_{c1} . For $h = h_{c1} (< h_c)$ the second-order phase transition $FS \rightarrow CFS$ takes place, accompanied by a more abrupt decrease (break) than in the FS phase, of the superconducting order parameter Δ in the S layer and the appearance of a nonzero wave vector Q_{\perp} of modulation of the ferromagnetic ordering in the F film. The critical parameters of the transition (h_{c1}, Δ_{c1}), found by equating the energies of these phases, are

$$h_{c1} \approx \frac{h_c}{\sqrt{A_c}} \left[1 + \frac{\pi L^2}{48 \xi_0^2} \left(1 - \frac{1}{A_c} \right) \right],$$

$$\Delta_{c1} \approx \Delta_c \left[1 + \frac{\pi L^2}{24 \xi_0^2} \left(1 - \frac{1}{A_c} \right) \right]. \quad (25)$$

Here A_c is the value of the parameter A at $h = h_c$ and $\Delta = \Delta_c$ (see formula (27) below), where $A_c > 1$. With further increase of the exchange field a first-order phase transition from the cryptoferromagnetic superconducting state (CFS) to the ferromagnetic normal state (FN) takes place at $h = h_{c2}$. The upper critical value of the exchange field h_{c2} and the corresponding values of the order parameter Δ_{c2} and the modulation wave vector Q_{c2} in the case $A_c - 1 \ll A_c$ are given by

$$h_{c2} \approx h_c \left[1 + \frac{\pi^2}{256} \left(1 - \frac{1}{A_c} \right)^2 \right],$$

$$\Delta_{c2} \approx \Delta_c \left[1 - \frac{3\pi^2}{256} \left(1 - \frac{1}{A_c} \right) \right],$$

$$Q_{c2} \approx \frac{1}{\xi_0} \sqrt{\frac{3\pi}{16} \left(1 - \frac{1}{A_c} \right)}. \quad (26)$$

It can be shown that the quantity A_c plays a role for the F/S junctions analogous to the role of the Ginzburg–Landau parameter κ in dividing type-I and type-II superconductors. Indeed, the ratio of molecular fields A , which plays a decisive role in assigning the F/S junctions to the first or the second type, can be expressed in terms of A_c as

$$A = A_c \left(\frac{h}{h_c} \right)^2 \frac{\Delta_c}{\Delta}, \quad A_c = \frac{N(0)h_c^2 \pi \xi_c^2 L}{JS^2 \cdot 4a^2 d}. \quad (27)$$

Hence it follows that F/S junctions with $A_c < 1$ belong to the first type (Fig. 1a), and junctions with $A_c > 1$, to the second type (Fig. 1b). In addition, F/S junctions with $A_c - 1 \ll A_c$ are characterized by a wide coexistence region of ferromagnetism and superconductivity (FS) and a narrow region of the cryptoferromagnetic superconducting phase (CFS) since the critical values h_{c1} and h_{c2} are close to h_c in this situation.

In the case of lower stability of the ferromagnetism of the F film to long-wave modulation, i.e., when $1 \ll A_c \ll (\xi_0/L)^2$ holds, the region occupied by the CFS phase is substantially broadened, and the region corresponding to the FS phase, in contrast, narrows significantly since in this case $h_{c1} \ll h_c \ll h_{c2}$. Formulas (25) for h_{c1} and Δ_{c1} now substantially simplify since $A_c \gg 1$. The upper exchange field h_{c2} and the corresponding values of Δ_{c2} and Q_{c2} in this case are found from the new self-consistent equilibrium conditions

$$\Delta^2 \ln \frac{\Delta_0}{\Delta} = \frac{\pi h^2}{4Q_{\perp}^2 \xi^2}, \quad Q_{\perp} = \frac{(2A)^{1/4}}{\xi}, \quad (28)$$

which are obtained by minimizing the free energy (16) using the RKKY polarization (18) for the range of magnetic structure wave vectors $\xi^{-1} \ll q_{\perp} \ll L^{-1}$. The ratio of molecular fields A in this case obeys the inequalities $1 \ll A \ll (\xi/L)^4$. The free energy density of the F/S junction corresponding to conditions (28) has the form

$$f_{CFS} = f_F^0 + f_N^0 - \frac{LN(0)}{2a^3} \left(\Delta^2 - \frac{3\pi h^2}{2Q_{\perp}^2 \xi^2} \right). \quad (29)$$

The characteristic parameters of the first-order transition from the CFS phase to the FN phase in the given case are given by

$$h_{c2} = \frac{4\sqrt{2A_c}}{3\pi e^{1/4}} h_c, \quad \Delta_{c2} = \frac{\Delta_0}{e^{1/6}}, \quad Q_{c2} = \frac{1}{\xi_0} \sqrt{\frac{8A_c}{3\pi e^{1/2}}}. \quad (30)$$

Clearly, in the region of strong exchange fields $h \gg h_c$ superconductivity in the S layer can be preserved only under the condition that it be effectively averaged as a result of small-scale (in comparison with the coherence length ξ_0) modulation of the magnetic order in the F film. At the same time, in order to ensure that the loss in the direct exchange energy will be small, the modulation should remain relatively long-wave in comparison with the period of the magnetic lattice a , i.e., the condition $a \ll L \ll Q_{\perp}^{-1} \ll \xi_0$ should hold. The not

overly strong decrease in the superconducting order parameter Δ in the *CFS* phase is due to the optimal nature of the mutual accommodation of the two competing phenomena.

Finally, in the presence of even greater instability of ferromagnetic ordering, when $A_c \gg (\xi_0/L)^2$ holds, there can be an even finer-scale ($a \ll Q_{\perp}^{-1} \ll L \ll \xi_0$) modulation of the ferromagnetic ordering of the localized spins in the *F* film, characteristic for exchange fields $h \geq h_c(\xi_0/L)$. The equilibrium values of Δ and Q_{\perp} for this region of exchange fields in the *CFS* phase are found from the equations

$$\Delta^2 \ln \frac{\Delta_0}{\Delta} = \frac{\pi h^2 L}{4 Q_{\perp} \xi^2}, \quad Q_{\perp} = \left(\frac{AL}{\xi} \right)^{1/3} \frac{1}{\xi}, \quad (31)$$

obtained by minimizing the free energy (16) using the RKKY exchange potential (18) for the range of wave vectors $L^{-1} \gg q_{\perp} \ll L^{-1}$. This corresponds to quite large values of the ratio of molecular fields A such that $A \ll (\xi/L)^4$. The surface density of the free energy of the *F/S* junction corresponding to Eq. (31) is

$$f_{CFS} = f_F^0 + f_N^0 - \frac{LN(0)}{2a^3} \left(\Delta^2 - \frac{\pi h^2 L}{Q_{\perp} \xi^2} \right). \quad (32)$$

At the second-order phase transition *CFS* \rightarrow *FN* ($h = h_{c2}$) we have

$$h_{c2} = \left(\frac{8A_c \xi_0^2}{e \pi^3 L^2} \right)^{1/4} h_c, \quad \Delta_{c2} = \frac{\Delta_0}{e^{1/4}}, \quad Q_{c2} = \frac{1}{\xi_0} \sqrt{\frac{2A_c}{\pi \sqrt{e}}}. \quad (33)$$

In the last of the cases considered ($A_c \ll \xi_0^2/L^2$) the *FS* phase has its smallest range of values of the exchange field since $h_{c1} = h_c / \sqrt{A_c} \ll h_c L / \xi_0$. On the contrary, the *CFS* phase has its largest range of values of the exchange field since $h_{c2} \gg h_c \xi_0 / L$.

The inequality $A \gg (\xi/L)^4$ (or $A_c \gg (\xi/L)^2$ and $h \gg h_c \xi_0 / L$), which is necessary for the realization of the finest-scale ($Q_{\perp}^{-1} \ll L$) modulation of ferromagnetic ordering in an *F/S* junction is seen to be quite severe if one takes account of the assumption that we made, namely that $I^2 N(0) < J$. However, the smallness of the factor (itself a ratio) $N(0)h^2 / JS^2$ in the definition (19) of the ratio of molecular forces A is to a large extent compensated by the ratio $\pi \xi^2 L / 4a^2 d$ of the region of action of the superconducting part of the RKKY polarization and the region of action of direct exchange. As a result, A is bounded from above by a magnitude of the order of $I^2 N(0) \xi^2 / J L d$. Consequently, our results for the strongly modulated *CFS* phase in *F/S* junctions are valid for very thin layers of ferromagnetic insulator and relatively thick layers of superconductor such that the inequalities $J \xi^2 d / L^3 \ll I^2 N(0) < J$ are fulfilled.

4. COMPETITION BETWEEN FERROMAGNETISM AND SUPERCONDUCTIVITY IN *F/S* SUPERLATTICES

Let us consider an *F/S* superlattice obtained by alternately laying down layers of insulator, of thickness d , and superconductor, of thickness L , where $L \ll \xi$, as before. To examine the mutual accommodation of the superconducting and magnetic order parameters in such a system, it is sufficient to study the free energy density f^* of a unit cell con-

sisting of two magnetic half-layers (*F*) $-d/2 < z < 0$ and $L < z < L + d/2$, separated by a superconducting layer (*S*) $0 < z < L$. The functional f^* in this case will differ from the previous case [Eq. (16)] in that, in addition to the term $I(q_{\perp}, 0, 0)$, it should include an analogous term due to surface RKKY exchange $I(q_{\perp}, L, L)$ between the localized spins of the neighboring ferromagnetic layer ($z = z' = L$), and also a term $I(q_{\perp}, 0, L)$ accounting for RKKY exchange between the localized spins belonging to the magnetic surfaces $z = 0$ and $z' = L$ separated by the intervening superconducting layer *S*. In this light, we seek the magnetic order in the *F/S* superlattice in the form

$$\langle S_r^{\pm} \rangle = S \exp[\pm i(\mathbf{q}_{\perp} \cdot \boldsymbol{\rho} + q_{\parallel} z)], \quad \langle S_r^z \rangle = 0, \quad (34)$$

where q_{\parallel} is the component of the wave vector parallel to the superlattice axis. Below, as before, we assume that the thickness of the ferromagnetic layers d is much less than the penetration depth δ of the surface distortions of the magnetic ordering. This ensures that the magnetic order parameter will not vary within the limits of the thickness of any given *F* layer and that as a result of the translational invariance of the *F/S* superlattice, it will at most be multiplied by a constant phase factor as one goes from one *F* layer to the neighboring *F* layer, i.e.,

$$\langle S^{\pm}(\boldsymbol{\rho}, z + L + d) \rangle = \langle S^{\pm}(\boldsymbol{\rho}, z) \rangle \exp(\pm i q_{\parallel} L). \quad (35)$$

At the same time, we will temporarily neglect tunneling by the conduction electrons from one *S* layer to another through an intervening *F* layer. Therefore we will treat the phases of the superconducting order parameters Δ in neighboring *S* layers as uncoupled, and we will not consider the “ π -phase” variant (in the superconductivity sense, see Ref. 10) of the mutual accommodation.

Noting further that $I(q_{\perp}, L, L) = I(q_{\perp}, 0, 0)$ and $I(q_{\perp}, L, 0) = I(q_{\perp}, 0, L)$, we obtain the following expression for the free energy density f^* per unit cell:

$$f^* = f_F^0 + f_N^0 + JS^2 q_{\perp}^2 \frac{d}{a} - 2I(q_{\perp}, 0, 0) \left(\frac{S}{a} \right)^2 - 2I(q_{\perp}, 0, L) \left(\frac{S}{a} \right)^2 \cos(q_{\parallel} L) - \frac{LN(0)}{2a^3} \Delta^2 \ln \frac{e \Delta_0^2}{\Delta^2}, \quad (36)$$

where $I(q_{\perp}, 0, L)$ is found from Eq. (17) and has asymptotic limits similar to those for $I(q_{\perp}, 0, 0)$ [Eqs. (18)]. They only differ for wave vectors q_{\perp} comparable to L^{-1} . Indeed, for $q_{\perp} L \ll 1$ we obtain

$$I(q_{\perp}, 0, L) = I(q_{\perp}, 0, 0) + \frac{\pi L a}{16 \xi^2} N(0) I^2 \times \left(1 - \frac{q_{\perp}^2 L^2}{12} + \frac{q_{\perp}^4 L^4}{120} \right), \quad (37)$$

from which it can be seen that the corrections to $I(q_{\perp}, 0, 0)$ are small of order $(L/\xi)^2$. In this case, RKKY exchange between the localized spins belonging to the different magnetic surfaces $z = 0$ and $z' = L$ are of the same order of mag-

nitude as RKKY exchange between the localized spins on each of the surfaces $z = z' = 0$ and $z = z' = L$. In the opposite limit ($q_{\perp}L \gg 1$) we have

$$I(q_{\perp}, 0, L) \approx 2I(q_{\perp}, 0, 0) \exp(-q_{\perp}L), \quad (38)$$

i.e., the exchange coupling between magnetic surfaces through the intervening superconducting layer S is exponentially small in comparison with the exchange between the localized spins on each of the surfaces (although $L \ll \xi$, as before). This is due to extensive averaging of the spin polarization of the electrons in the superconductor with strong modulation of the ferromagnetic ordering in the F layers, thanks to which RKKY exchange in this case has a purely surface character.

Minimizing the free energy (36) over Δ , q_{\perp} , and q_{\parallel} using expression (37) for $I(q_{\perp}, 0, L)$ leads to two possible variants of the ground state of the F/S superlattice combining magnetic and superconducting types of long-range order.

If $A < 6(\xi/L)^4$ holds, then the mutual accommodation of superconductivity and ferromagnetism will take the form of a layered antiferromagnetic superconducting phase AFS , for which the equilibrium values of Δ , q_{\perp} , and q_{\parallel} are given by

$$\Delta^2 \ln \frac{\Delta_0}{\Delta} = \pi h^2 \left(\frac{L}{2\xi} \right)^2, \quad Q_{\perp} = 0, \quad Q_{\parallel} = \frac{\pi}{L}. \quad (39)$$

The free energy density corresponding to this phase, f_{AFS}^* , is equal to

$$f_{AFS}^* = f_F^0 + f_N^0 - \frac{LN(0)}{2a^3} \left(\Delta^2 - h^2 \frac{\pi L^2}{2\xi^2} \right). \quad (40)$$

In the AFS phase, which corresponds to three-dimensional behavior of the F/S superlattice, the localized spins of each of the F layers are ordered ferromagnetically, and the magnetizations of neighboring layers are antiparallel. Such a configuration of the localized spins ensures against any loss in the direct exchange energy. In addition, by virtue of the small thickness of the intervening superconducting layer ($L \ll \xi$), the exchange polarization induced in it by the localized spins of one of the F layers is almost completely canceled by the polarization of opposite sign induced by the neighboring F layer. The uncompensated part of the spin polarization of the conduction electrons, as can be seen from Eq. (40), is small of order L/ξ .

Note that the strong averaging of the spin polarization of the conduction electrons in the AFS phase leads to a substantial increase in the values of the exchange field h and the parameter A (19) that are required for the transition to the cryptoferrimagnetic phase CFS , in comparison with F/S junctions. Indeed, to realize the CFS phase in F/S superlattices it is necessary that the new (effective) ratio of molecular forces $A^* = AL^4/6\xi^4$ be greater than unity. The parameters of this state Δ , Q_{\perp} , and Q_{\parallel} are found from the self-consistent equilibrium conditions

$$\Delta^2 \ln \frac{\Delta_0}{\Delta} = h^2 \frac{\pi L^2}{4\xi^2} \left(1 - \frac{Q_{\perp}^2 L^2}{12} + \frac{Q_{\perp}^4 L^4}{120} \right),$$

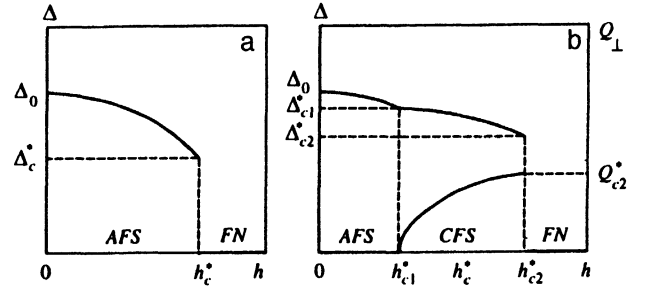


FIG. 2. Dependence of the superconducting order parameter Δ and wave vector Q_{\perp} of the magnetic structure on the exchange field h for F/S junctions a) of the first type with $A_c^* < 1$ ($Q_{\perp} = 0$) and b) of the second type with $A_c^* > 1$.

$$Q_{\perp} = \sqrt{5 \left(1 - \frac{1}{A^*} \right)} \frac{1}{L}, \quad Q_{\parallel} = \frac{\pi}{L}, \quad A^* = A \frac{L^4}{6\xi^4} > 1. \quad (41)$$

The free energy density in such a layered CFS phase has the form

$$f_{CFS}^* = f_F^0 + f_N^0 - \frac{LN(0)}{2a^3} \left[\Delta^2 - h^2 \frac{\pi L^2}{2\xi^2} \times \left(1 + \frac{Q_{\perp}^2 L^2}{12} - \frac{Q_{\perp}^4 L^4}{40} \right) \right]. \quad (42)$$

As follows from a comparison of Eqs. (40) and (42), the transition from the AFS state to the layered CFS state takes place at $A^* = 1$. In this case the neighborhood $A^* - 1 \ll A^*$ of the transition point is associated with a large-scale ($Q_{\perp}^{-1} \ll L$) sinusoidally modulated structure of the localized spins in the F layers. At the same time, the phases of the magnetic order parameters in neighboring F layers, as can be seen from Eqs. (35) and (41), are shifted by π . Such a three-dimensional (3D) “ π -phase” behavior of the magnetic order parameter in the AFS state and immediately after the transition to the CFS state is due to RKKY exchange, which sets up a long-range antiferromagnetic coupling between the localized spins of neighboring ferromagnetic layers through the conduction electrons of the intervening superconducting layers.

The above analysis shows that with growth of h and A^* the F/S superlattices, like F/S junctions, exhibit two kinds of behavior. The corresponding dependences of Δ and Q_{\perp} on h are schematically depicted in Fig. 2, where the critical exchange fields h_c^* , h_{c1}^* , and h_{c2}^* , and the corresponding order parameters Δ_c^* , Δ_{c1}^* , and Δ_{c2}^* for the superlattices are marked with asterisks to distinguish them from the corresponding values for the junctions.

For superlattices of the first type (see Fig. 2a), as h grows, the modified ratio of molecular forces A^* remains less than unity up to the critical value h_c^* , at which the first-order phase transition $AFS \rightarrow FN$ occurs. At this point the superlattice in the magnetic sense becomes quasi-two-dimensional (2D) since the long-range exchange coupling between the F layers disappears simultaneously with the destruction of superconductivity in the S layers. The critical

values h_c^* and Δ_c^* , corresponding to the $AFS \rightarrow FN$ transition point, are found by equating the energies of these phases:

$$h_c^* = \Delta_0 \frac{\xi_0}{L} \sqrt{\frac{2}{\pi\sqrt{e}}}, \quad \Delta_c^* = \frac{\Delta_0}{\sqrt{e}}. \quad (43)$$

In the case of F/S superlattices of the second type (see Fig. 2b) the ratio of molecular forces A^* is less than unity up to the lower critical value of the exchange field h_{c1}^* , where the second-order phase transition $AFS(3D) \rightarrow CFS(3D)$ takes place in the F/S system. Equating the free energies of these phases [Eqs. (40) and (42)], we obtain

$$h_{c1}^* \approx h_c^* \left(1 - \frac{1}{2} \sqrt{1 - \frac{1}{A_c^*}} \right),$$

$$\Delta_{c1}^* \approx \Delta_c^* \left(1 + \sqrt{1 - \frac{1}{A_c^*}} \right), \quad (44)$$

where $A_c^* - 1 \ll A_c^*$. Here the ratio of molecular forces is represented in a form normalized to the critical point (h_c^*, Δ_c^*), i.e.,

$$A^* = A_c^* \left(\frac{h}{h_c^*} \right)^2 \frac{\Delta}{\Delta_c^*}, \quad A_c^* = \frac{2}{3\pi e} \left(\frac{L}{\xi_0} \right)^2 A_c, \quad (45)$$

where A_c is the critical ratio of molecular forces (27) of the corresponding F/S junction. The $AFS \rightarrow CFS$ transition is accompanied by the appearance of a nonzero wave vector Q_\perp of the sinusoidal modulation of the spin order in the F layers, which with conservation of the phase shift by π between the sinusoids in neighboring layers leads to additional averaging of the spin polarization acting on the Cooper pairs in the intervening superconducting layers S . Therefore in the CFS state the falloff of the order parameter Δ in the region $h > h_{c1}^*$ is slower than in the AFS state in superlattices of the first type. Thus, at the point $h = h_{c1}^* + 0$ there is a positive jump in the first derivative $\Delta'(h)$, which is different from the case with F/S junctions, where this jump (at $h = h_{c1} + 0$) is negative. The upper critical exchange field h_{c2}^* of the superlattice and the parameters Δ_{c2}^* and Q_{c2}^* of the first-order transition $CFS \rightarrow FN$ for $A_c^* - 1 \ll A_c^*$ are given by

$$h_{c2}^* \approx h_c^* \left[1 + \frac{5}{28} \left(1 - \frac{1}{A_c^*} \right)^2 \right],$$

$$\Delta_{c2}^* \approx \Delta_c^* \left[1 + \frac{5}{7} \left(1 - \frac{1}{A_c^*} \right) \right],$$

$$Q_{c2}^* \approx \frac{1}{L} \sqrt{\frac{60}{7} \left(1 - \frac{1}{A_c^*} \right)}. \quad (46)$$

As follows from expressions (44)–(46), the critical ratio of molecular forces A_c^* plays the same role for the F/S superlattices as does the critical ratio of molecular forces A_c for the F/S junctions. Indeed, superlattices with $A_c^* < 1$ belong to the first type and, as can be seen from Fig. 2a, are characterized by homogeneous ordering of the localized spins in the F layers ($Q_\perp = 0$) both in the normal (FN) and superconducting (AFS) states. At the same time, superlat-

tices with $A_c^* > 1$, belonging to the second type, can possess one more intermediate superconducting phase (CFS) with sinusoidal modulation ($Q_\perp \neq 0$) of the spin structure in the F layers (Fig. 2b). As $A_c^* \rightarrow 1$, the region occupied by the cryptoferromagnetic state contracts into a point since $h_{c1}^*, h_{c2}^* \rightarrow h_c^*$ and, correspondingly, $\Delta_{c1}^*, \Delta_{c2}^* \rightarrow \Delta_c^*$ and $Q_{c2}^* \rightarrow 0$.

If $A_c^* \ll 1$ holds, then for the lower critical parameters h_{c1}^* and Δ_{c1}^* we obtain from Eqs. (40)–(42)

$$h_{c1}^* \approx h_c^* / \sqrt{A_c^* \sqrt{e}}, \quad \Delta_{c1}^* \approx \Delta_0 (1 - 1/2eA_c^*). \quad (47)$$

To find the upper critical parameters h_{c2}^* , Δ_{c2}^* , and Q_{c2}^* in this case, new equilibrium conditions, valid for the high exchange fields $h \gg h_c^*$ and, correspondingly, the strong modulation of the magnetic ordering in the F layers ($q_\perp L \gg 1$), are necessary. Minimizing the free energy density (36) over Δ and q_\perp while neglecting, by virtue of relation (38), the exponentially small interlayer F – F exchange gives

$$\Delta^2 \ln \frac{\Delta_0}{\Delta} = h^2 \frac{\pi L}{2Q_\perp \xi^2}, \quad Q_\perp = \frac{(12A^*)^{1/3}}{L}, \quad (48)$$

where $A^* \gg 1$. The free energy density corresponding to the so strongly modulated CFS phase is

$$f_{CFS}^* = f_F^0 + f_N^0 - \frac{LN(0)}{2a^3} \left(\Delta^2 - h^2 \frac{2\pi L}{Q_\perp \xi^2} \right). \quad (49)$$

For the case under consideration ($A_c^* \gg 1$ and $h \gg h_c^*$, i.e., $A^* \gg 1$), invoking Eqs. (48) and (49) for the parameters of the first-order phase transition $CFS \rightarrow FN$ we obtain

$$h_{c2}^* = h_c^* \left(\frac{3eA_c^*}{16} \right)^{1/4}, \quad \Delta_{c2}^* = \frac{\Delta_0}{e^{1/4}}, \quad Q_{c2}^* = \frac{\sqrt{3A_c^* \sqrt{e}}}{L}. \quad (50)$$

For superlattices with $A_c^* \gg 1$, as can be seen from relations (47) and (50), the span of the AFS state is minimal, while that of the CFS state, on the contrary, is maximal, since $h_{c1}^* \ll h_c^* \ll h_{c2}^*$. It is interesting to note that the origin of the CFS state, i.e., the region $h_{c1}^* \leq h \leq h_c^*$, corresponds to 3D behavior of the magnetic order parameter with “ π -phase” matching of the large-scale ($Q_\perp^{-1} \gg L$) modulations of the spin structure in the neighboring F layers. At the same time, the region $h_c^* \leq h \leq h_{c2}^*$ near the transition to the FN state already corresponds to quasi-two-dimensional behavior of the superlattice with small-scale ($Q_\perp^{-1} \ll L$) oscillations of the spin ordering in the practically non-interacting F layers, although superconductivity in the intervening S layers is still preserved. Such a quasi-two-dimensional variant of the coexistence of the two competing phenomena is due precisely to the surface character of RKKY exchange in the region of large wave vectors q_\perp . The F/S superlattice in this case decays into a system of weakly coupled $S/F/S$ sandwiches. In this case, whereas the phases of the magnetic order parameter on opposite sides of the same F layer are synchronized by strong direct exchange, the correlation between the phases of the order parameter in neighboring F layers is absent with exponential accuracy and Q_\parallel is arbitrary.

5. CONCLUSIONS AND DISCUSSION OF RESULTS

The work presented in this paper can be summarized as follows.

1. A simple model of exchange interactions between localized spins in F/S junctions and superlattices (ferromagnetic insulator/superconductor) has been proposed. In addition to the direct exchange between nearest neighbors in the F film, it also takes account of the indirect interaction between the localized spins on the F/S boundary by way of the conduction electrons of the superconductor. This latter exchange is due to the effective $s-d$ exchange I , which arises as a result of virtual electron transport from the superconductor to the insulator and *vice versa* as a consequence of the overlap of the corresponding wave functions.

2. The dependence of the RKKY potential on the distance between the localized spins and their positions relative to the surface of the superconductor for three geometries of practical interest: a half-space, a lamina (quasi-two-dimensional film), and a wire (quasi-one-dimensional whisker). It has been shown that the antiferromagnetic correlations between the localized spins grow as one approaches the surface of a massive superconductor or as one decreases the dimensionality of the sample.

3. It has been shown that F/S junctions can be divided into two types depending on the magnitude of the critical ratio of the antiferromagnetic and ferromagnetic molecular fields A_c (27):

a) F/S junctions of the first type with $A_c < 1$ allow only a homogeneous ferromagnetic ordering in the F film, which for exchange fields $h < h_c$ coexists with superconductivity in the S sublayer (FS phase). For $h \geq h_c$ the F/S junctions pass into the normal state (the FN phase) by way of a first-order phase transition (see Fig. 1a).

b) In F/S junctions of the second type with $A_c > 1$ the FS phase exists only for $h < h_{c1}$. For $h_{c1} < h < h_{c2}$ the superconducting substrate S induces a nonuniform cryptoferrimagnetic modulation (the CFS phase) in the F film as a result of the long-range antiferromagnetic RKKY exchange between the localized spins. This phenomenon can apparently be considered as a magnetic aspect of the well-known proximity effect. The $FS \rightarrow CFS$ transition at $h = h_{c1}$ is a second-order phase transition (see Fig. 1b). The wave vector Q_{\perp} of the spin structure $\langle S_i^z \rangle = S \cos(\mathbf{Q}_{\perp} \cdot \mathbf{p})$ in the CFS phase varies from $Q_{\perp} = 0$ at $h = h_{c1}$ to $Q_{\perp} \gg L^{-1}$ at $h = h_{c2}$ if $A_c \gg (\xi/L)^2$, where L is the thickness of the superconductor and ξ is the coherence length ($L \ll \xi$). The first-order phase transition $CFS \rightarrow FN$ takes place at $h = h_{c2}$.

4. The variants of mutual accommodation of superconductivity and ferromagnetism in an F/S superlattice also depend on its type, which is determined by their critical ratio of molecular fields A_c^* (45), specifically:

a) For superlattices of the first type (Fig. 2a) with $A_c^* < 1$ the 3D behavior corresponding to the layered antiferromagnetic superconducting state (AFS) is characteristic for $h < h_c^*$. It is due to the long-range RKKY exchange between the localized spins of neighboring F layers through the intervening superconducting layers. For $h \geq h_c^*$ these superlattices pass into the normal state (the FN phase) by way of a first-

order phase transition. Simultaneous with the destruction of superconductivity, the long-range coupling between the F layers disappears, and the F/S superlattice, in the magnetic sense, becomes quasi-two-dimensional ($2D$).

b) In superlattices of the second type (Fig. 2b) with $A_c^* > 1$ the AFS state exists only for $h < h_{c1}^*$ ($< h_c^*$). For $h = h_{c1}^*$ it gives way to the layered cryptoferrimagnetic state $CFS(3D)$, which in turn gives way, by way of a first-order phase transition at $h = h_{c2}^*$, to the $FN(2D)$ state. If $A_c^* \gg 1$, then the region $h \geq h_{c1}^*$ corresponds to large-scale ($Q_{\perp}^{-1} \gg L$) modulation of the spin ordering with a phase shift of π between neighboring F layers ($CFS(3D)$ behavior). The region $h \leq h_{c2}^*$ corresponds to quasi-two-dimensional $CFS(2D)$ behavior of the F/S superlattice when small-scale ($Q_{\perp}^{-1} \ll L$) oscillations in the spin structure of the F layers lead to exponentially weak RKKY exchange between these layers.

The differences in the character of the mutual accommodation of the F/S junctions and the F/S superlattices are due mainly to the presence in the latter (in addition to RKKY exchange between the localized spins at the F/S boundary) of antiferromagnetic exchange between the localized spins of neighboring F layers through the intervening S layer. The idea itself of antiferromagnetic coupling of two ferromagnetic insulators through a superconductor was first discussed by de Gennes.¹⁹ Now it can be expanded substantially. The interlayer $F-F$ exchange $I(Q_{\perp}, 0, L)$ (37) is of the same order of magnitude as the exchange $I(Q_{\perp}, 0, 0)$ between the localized spins on one F layer if the period Q_{\perp}^{-1} of the modulation of the magnetic ordering due to the latter is greater than the thickness of the intervening superconducting layer L . In the opposite case ($Q_{\perp}^{-1} < L$) the antiferromagnetic coupling between the F layers is exponentially small [see Eq. (38)] in spite of the fact that $L \ll \xi$. This is manifested most clearly by a comparison of the lower and upper critical exchange fields of the junctions and superlattices for $A_c^* \gg 1$. Indeed, whereas the lower boundary of the CFS phase in the superlattices is shifted, in comparison with the junctions, toward higher fields, i.e., $h_{c1}^* = h_{c1}(\xi/L)^2$ due to 3D behavior ($Q_{\perp} = 0$), the upper boundaries are of the same order of magnitude, $h_{c2}^* = h_{c2}/\sqrt{2}$, reflecting 2D behavior in this case, i.e., decay of the superlattice into a system of almost independent $S/F/S$ sandwiches at $Q_{\perp} \approx \sqrt{A_c^*/L}$. In addition, the differences between F/S junctions and superlattices are also manifested in the behavior of the superconducting order parameter $\Delta(h)$ (see Figs. 1b and 2b). Whereas the transition in F/S junctions to the CFS state with inhomogeneous magnetic ordering is accompanied by a negative jump in the derivative $\Delta'(h)$ at $h = h_{c1}$, in superlattices this jump is positive and Δ falls with growth of h slower, not faster, than in the first case. As a consequence of the large difference between the critical exchange fields h_c and h_c^* ($h_c^* = h_c \xi/L$) one can expect that superconductivity, initially suppressed in the F/S junction of the first type for $h > h_c$, is restored in an F/S superlattice obtained by periodic repetition of this junction if $h < h_c^*$.

Note that the conditions for the coexistence and mutual accommodation of superconductivity and ferromagnetism in F/S junctions and superlattices with a ferromagnetic insula-

tor are more favorable than in analogous systems with a ferromagnetic metal. The point here is that in our case the exchange field h is created solely by the localized spins of the first atomic layer of the F/S boundary and has smallness of the order of a/L . In addition, the effective exchange integral I [Eq. (14)], which depends on the degree of overlap of the wave functions of the conduction electrons and of the s and d orbitals of the localized spins of the F/S boundary, is smaller by an order of magnitude or more than in a metallic ferromagnet. Its magnitude is determined in a substantial way by the conditions and technique of preparation of the F/S boundary.

As the temperature increases, the attendant decrease of the number of paired electrons weakens the antiferromagnetic RKKY correlations between the localized spins, thereby leading to a decrease in the ratio of molecular fields A [Eq. (27)] for F/S junctions and the ratio of molecular fields A^* [Eq. (45)] for superlattices from values $A, A^* \gg 1$ down to zero. Therefore, under certain conditions, as the temperature is increased from zero to T_c , in F/S junctions and superlattices of the second type one can expect a cascade of alternating magnetic and superconducting transitions: $CFS \rightarrow FS \rightarrow FN$ for the junctions, and $CFS(2D-3D) \rightarrow AFS(3D) \rightarrow FN(2D)$ for the superlattices. To obtain a more detailed picture of the phase transitions in F/S systems, it is necessary to analyze the high-temperature expansion of the free energy.

An analogous sequence of phase transitions can be produced at $T=0$ by applying an external magnetic field parallel to the plane of the F/S junction. In this case, for a thin film of superconductor with $L \ll \xi$ the orbital depairing effect is small, and the main role in the suppression of superconductivity in metals with weak spin-orbit scattering is played by the paramagnetic effect of the external field. Destruction of Cooper pairs with increase of the magnetic field H (the orbital effect) gradually decreases the ratio of molecular forces A and wave vector Q_\perp , smoothing out the oscillations of the spin structure in the ferromagnet. In addition, the action of the magnetic field on the subsystem of localized spins of the F film gives rise to a preferred orientation and transforms the CFS phase into a banded-domain phase analogous to the phase that was predicted in Ref. 7. Only the width of the domains aligned with the field will be larger than the width of the anti-aligned domains. Further growth of the external field causes the system to transition to the single-domain FS phase with $Q_\perp=0$. The Zeeman splitting of the BCS peak in the density of states of the conduction electrons observed in F/S junctions will contain an additional term created by the oscillating exchange field $h \cos(\mathbf{Q}_\perp \cdot \mathbf{r})$ of the localized spins of the F/S boundary. This extra splitting saturates at the $CFS \rightarrow FS$ phase transition, where Q_\perp vanishes. With further increase of the external field, a first-order phase transition from the FS state to the FN state ($FS \rightarrow FN$) takes place at $H^2 + h^2 \approx \Delta^2(H)$. Such extra splitting was observed in tunneling experiments with the F/S junctions EuO/Al (Ref. 12) and EuS/Al (Refs. 13 and 14). Indeed, it was found that a thin aluminum film behaves as a BCS superconductor with an internal field causing additional splitting of the peak in the quasiparticle density of states. This splitting depended

on the thickness of the Al film as L^{-1} . From its saturation it is possible to determine the effective exchange integral I and, consequently, to estimate the transport integrals t_s and t_f according to our model (14). The estimates show that for a EuO/Al junction¹² $I \approx 0.7$ meV, and for EuS/Al (Ref. 13) $I \approx 1.6$ meV ($S_{Eu} = 7/2$, $L_{Al} = 4$ nm, and $a_{Al} = 0.4$ nm). The hysteresis observed in Au/EuS/Al junctions,¹⁴ i.e., the extra splitting after removing a field in excess of the paramagnetic limit can be explained by the fact that as a result of pinning of the inhomogeneous spin structure, the ground state of the EuS/Al junction turns out to be the CFS phase with modulation period Q_\perp^{-1} comparable with ξ . Additional experiments on magnetic scattering of neutrons might give valuable information about spin ordering in EuO/Al and EuS/Al junctions and how it is affected by an external field.

In F/S superlattices, increasing an external field parallel to the surface of the sample also leads to the transitions $CFS(2D-3D) \rightarrow AFS(3D)$, after which the system can be in the layered ferromagnetic superconducting phase FS if it has not already undergone a phase transition into the FN state. For a complete picture, it is necessary to analyze the phase diagrams of the F/S systems in an external magnetic field. In principle, it should be possible to fabricate a superlattice combining superconductivity in the S layers with a ferromagnetic orientation of the magnetizations of the neighboring F layers even in the absence of an external magnetic field. To this end, it is necessary that the magnetic atoms of neighboring layers F and F^* have $s-d$ exchange integrals I and I^* of different sign. In this case, superconducting RKKY exchange (17) will lead to ferromagnetic coupling between the localized spins of neighboring F layers without disturbing the compensation of the exchange polarization in the intervening S layers.

Since all of the results obtained in this paper are based on the expression (4) for the RKKY exchange integral in a dirty superconductor, let us briefly discuss the nontrivial role of impurity scattering in this interaction. As is well known, in a clean superconductor the spin susceptibility is defined as the sum of two terms:² an oscillating term (associated with the quasiparticles) and a nonoscillating term (due to the Cooper pairs). It has recently been shown²⁰ that for the case of weak impurity scattering the oscillating part of the RKKY exchange acquires a random phase φ and does not cut off at one mean free path l , as was previously established for metals.^{21,22} At the same time, the non-oscillating part of the RKKY exchange $\Lambda_s(\mathbf{r}, \mathbf{r}', \omega) \propto R^{-2}$ remains unchanged, reflecting the smallness of the impurity fluctuations for that part of the interaction due to the contribution of the phase-coherent Cooper condensate. For large impurity concentrations, when the motion of the conduction electrons for $R > l$ ($< \xi$) is diffusive, it is necessary to allow for interference of the interelectron (BCS) interaction with the elastic scattering off the impurities, which necessitates² replacing the foregoing term by the also nonoscillating but still more slowly falling-off expression (4) $\Lambda_s(\mathbf{r}, \mathbf{r}', \omega) \propto (Rl)^{-1}$. Physically, this is due to an increase of the effective time of the pairwise correlations for diffusive motion of the conduction electrons through the region of the BCS interaction. The radius of action of the exchange in this case decreases from

$$\xi_{0\omega} = v / \sqrt{\omega^2 + \Delta^2}$$

to

$$\xi_{\omega} = \sqrt{D/2} \sqrt{\omega^2 + \Delta^2},$$

ensuring that the sum rule (5) will also be fulfilled in the case of a dirty superconductor. The role of the impurity fluctuations, which affect only the oscillating (normal) part of the RKKY exchange, can be important for a study of the coexistence of superconductivity and ordering of spin glass type, where the spin per site averaged over configurations is zero, $\langle S_r \rangle_c = 0$, but $\langle S_r^2 \rangle_c \neq 0$. In our case, $\langle S_r \rangle_c \neq 0$ and the oscillating part of the interaction can enter into the free energy only if it is first averaged over impurities (i.e., with the factor $\exp(-R/l)$, Ref. 23). However, since we assumed in Sec. III that the direct exchange is stronger at small distances than the indirect ($J > I^2 N(0)$), the contribution of the oscillating part can be neglected, leaving in place the more important short-range ferromagnetic (J) and long-range antiferromagnetic ($I^2 \delta \chi_s(\mathbf{r}, \mathbf{r}')$) parts of the exchange interaction.

In the present paper we have also not taken into consideration suppression of superconductivity due to magnetic scattering of the conduction electrons off the F/S boundary. It is a second-order effect in comparison with the exchange polarization h , with relative smallness of the order of $IN(0)$. In principle, it should be possible to take it into account by renormalization of the order parameter Δ in the cryptoferrimagnetic phase CFS , where it can be important, under the condition that the s - d exchange integral I be sufficiently large. In the experiments,^{12,13} however, the value of I turns out to be two orders of magnitude lower than in homogeneous materials, and $IN(0) \ll 1$. Strong spin-orbit scattering, which keeps the spin susceptibility of the conduction electrons from vanishing, can substantially weaken the antiferromagnetic coupling of the localized spins operating through the superconductor. Also, we have not considered effects associated with single-axis magnetic anisotropy, which is characteristic of thin films and interfaces of two media (see the review in Ref. 6). An account of this anisotropy will, apparently, lead to a transformation of the cryptoferrimagnetic phases of F/S systems into banded-domain phases similar to those suggested in Ref. 7 and observed in homogeneous materials of the type ErRh_4B_4 .^{4,5}

With the exception of “ π -phase” superconductivity effects,¹⁰ arising as a result of the transparency of the F -layers to the conduction electrons in metallic superlattices, the variants of mutual accommodation we have examined seem to reflect general tendencies in the behavior of thin-layered ferromagnet/superconductor multisystems. In particular, due to the coexistence of superconductivity and layered antiferromagnetism discovered recently in rare-earth boron-nickel carbides of the type $\text{HoNi}_2\text{B}_2\text{C}$ (Ref. 24), these

compounds are a natural microscopic analog of superconducting magnetic superlattices in the AFS phase. Finally, in passing, we might note that the weak suppression of superconductivity experimentally observed²⁵ in EuO/V multilayers may be explained by self-induced compensation of the exchange field in the vanadium interlayers or by antiferromagnetic alternation of the ferromagnetic layers of europium oxide in the AFS state, or by “ π -phase” magnetism which is characteristic of the layered CFS state.

ACKNOWLEDGMENTS

The author is grateful to I. A. Garifullin, B. I. Kochelaev, G. B. Teitel'baum, and G. G. Khaliullin for discussion of the results obtained here.

This work was partly supported by the International Scientific Fund.

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Translated by Paul F. Schippnick