Theory of "excitonic" ferromagnetism

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A model proposed earlier [1] for the ferromagnetic state of band electrons is investigated in detail. It is shown that the ferromagnetic ordering of electrons in a system that is unstable against electron-hole pairing arises by way of a second-order phase transition, for arbitrarily small coupling constants, if there exists an excess concentration of electrons or holes. The model is used to explain the magnetic properties of certain narrow-band semiconductors and also of some metals of the iron group.

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I. INTRODUCTION

The idea of ferromagnetic ordering of the band electrons in systems with a one-electron spectrum that is unstable against electron-hole (excitonic) pairing was put forward earlier. In the present article this model is considered in more detail.

The known models of the ferromagnetic state of band electrons, obtained from "first" principles (of the type of the Stoner model [2]), lead to a criterion for the appearance of magnetic order, namely, that the potential energy of the interaction of the electrons should become of the order of their kinetic energy. The model we are considering leads to ferromagnetism even for arbitrarily small electron-electron interaction, provided that the one-electron spectrum satisfies certain requirements. Namely, the substance in the initial phase is a semimetal with electron and hole Fermi surfaces that almost coincide on translation by a certain vector $\mathbf{P}$ [3], or it is a metal with narrow forbidden bands, when, for certain values of the vector $\mathbf{P}$, the condition $\epsilon(\mathbf{k}) = -\epsilon(\mathbf{k}+\mathbf{P})$ is fulfilled [4], or it is metal with planar parts of the Fermi surface [5], or, finally, it is a semiconductor for which the exciton binding energy is greater than the width of the forbidden band [6].

In all the cases enumerated, the system is unstable against the formation of excitons (electron-hole pairing) in a singlet or a triplet state. If a singlet instability develops first the substance goes over into an insulating state with a charge-density wave, i.e., structural transformations occur at the phase-transition point, and, possibly, ferroelectric properties arise. But if a triplet instability develops first, an antiferromagnet with a spin-density wave is formed [7].

It can be shown [1] that the simultaneous existence of singlet (Δ_s) and triplet (Δ_t) order parameters is not only accompanied by the appearance of charge- and spin-density waves but also lifts the degeneracy in the spin of the electron and hole bands. If the number of electrons is not equal to the number of holes (the chemical potential μ lies above (or below) the insulating gap formed), the number of carriers in the bands with oppositely oriented spins will be different. The total spin of such a system is not equal to zero, but is proportional to the difference in the concentrations of electrons and holes. Consequently, ferromagnetism appears, which we shall call excitonic in contrast to Stoner ferromagnetism [2].

Thus, the problem of excitonic ferromagnetism reduces to ascertaining the range of values of the coupling constants in which coexistence of triplet and singlet pairings in the presence of a difference in the electron and hole concentrations is energetically favorable.

Below, for definiteness, this question will be studied using the example of an isotropic semi-metal with almost coincident electron and hole Fermi surfaces.

The reason that a ferromagnetic state appears in such systems even when the interaction is weak is as follows. For singlet pairing alone or triplet pairing alone, the non-coincidence of the electron and hole Fermi surfaces leads to a decrease of the dielectric gap Δ (cf. (30)), and, consequently, to a free-energy loss (31) on account of the appearance of free carriers above the gap at $T = 0$. When the spin degeneracy is lifted in the ferromagnetic state there occurs a redistribution of these carriers over the spin sub-bands, leading to an increase of Δ (cf. (39)). In this case the kinetic-energy loss, which arises on account of the redistribution of the carriers above the gap (cf. (42)) and which makes a ferromagnetic state impossible in the case of weak interaction and a spherical Fermi surface [2], is compensated by the energy gain (41) resulting from the states below the gap. As the coupling constant increases, dielectric pairing and a ferromagnetic state are possible for electron and hole Fermi surfaces that are less and less similar in shape and size, i.e., a larger and larger fraction of the electrons will make a contribution to the magnetization. Thus, the Bloch-Stoner model, in which the electrons are completely spin-polarized, is the limiting case, for sufficiently strong interaction, of the model considered in the present work. It may be hoped, therefore, that the nature of the appearance of ferromagnetism in our model with weak interaction has an extremely general character.

II. MODEL HAMILTONIAN

The one-particle spectrum of the electrons of an isotropic semi-metal is described by the Hamiltonian

$$\mathcal{H}_0 = \sum_{\mathbf{k}\alpha} \left(\frac{\hbar^2\mathbf{k}^2}{2m} + \frac{\epsilon_g}{2} \right) \left(a_{1\alpha}^+(\mathbf{k}) a_{1\alpha}(\mathbf{k}) - a_{2\alpha}^+(\mathbf{k}) a_{2\alpha}(\mathbf{k}) \right). \qquad (1)$$

Here $\hbar\mathbf{k}$ is the electron quasi-momentum, 1 and 2 are the indices of the electron and hole bands respectively, m is the effective mass, which for simplicity we assume to be the same for electrons and holes, ϵ_g is the width of the forbidden band, which is negative ($\epsilon_g < 0$) for a semimetal, and $a_{1\alpha}(\mathbf{k})$, $a_{2\alpha}(\mathbf{k})$ are the Fermi annihilation operators for an electron with spin $\alpha/2 = \pm\frac{1}{2}$ in bands 1 and 2. Terms of the type $a_1^+ a_2$ describing interband transitions are not taken into account. It is known [8] that allowance for interband transitions in the problem of an excitonic insulator leads to a change in the character of the phase transformation. The phase transition to the excitonic-insulator state becomes a first-order transition.

In the Hamiltonian $\mathscr{H}_{int}$ of the electron-electron and electron-phonon interactions we retain only the terms responsible for the excitonic instability:

$$H_{int}=\sum_{\substack{\mathbf{k},\mathbf{k}',\mathbf{q};\\ \alpha,\beta}}\{V_1(\mathbf{q})a_{1\alpha}^+(\mathbf{k}+\mathbf{q})a_{2\beta}^+(\mathbf{k}'-\mathbf{q})a_{2\beta}(\mathbf{k}')a_{1\alpha}(\mathbf{k})$$
$$+V_2[a_{1\alpha}^+(\mathbf{k}+\mathbf{q})a_{1\beta}^+(\mathbf{k}'-\mathbf{q})a_{2\beta}(\mathbf{k}')a_{2\alpha}(\mathbf{k})+a_{1\alpha}^+(\mathbf{k}+\mathbf{q})a_{2\beta}^+(\mathbf{k}' \quad (2)$$
$$-\mathbf{q})a_{1\beta}(\mathbf{k}')a_{2\alpha}(\mathbf{k})+\text{c.c.}]\}+\sum_{\mathbf{k},\alpha}g(b^++b)(a_{1\alpha}^+(\mathbf{k})a_{2\alpha}(\mathbf{k})+\text{c.c.}),$$

where α and β are, as before, the spin indices, taking the two values ±1, and b is the Bose annihilation operator for the phonon of the unstable mode. Generally speaking, the interaction Hamiltonian ought to contain terms of the type $a_1^+a_1^+a_1a_2$, which, as is shown in the paper by Guseĭnov and Keldysh [9], determine the character of the phase transformation (as do the interband transitions).

Finally, the free phonon mode is described by the Hamiltonian

$$\mathscr{H}_{ph}=\hbar\omega_0(b^+b+1/2), \quad (3)$$

where ω_0 is the bare phonon frequency.

Thus, the total Hamiltonian of the system under investigation has, for a given number of electrons, the form

$$\mathscr{H}=\mathscr{H}_0+\mathscr{H}_{int}+\mathscr{H}_{ph} \quad (4)$$

If the chemical potential μ is fixed, we must add to the Hamiltonian $\mathscr{H}$ the term

$$-\mu\sum_{\mathbf{k},\alpha}\{a_{1\alpha}^+(\mathbf{k})a_{1\alpha}(\mathbf{k})+a_{2\alpha}^+(\mathbf{k})a_{2\alpha}(\mathbf{k})\}.$$

The subsequent treatment of the problem will be carried out in the high-density approximation, when the inequalities

$$|\varepsilon_g|\gg e^2\varkappa/\varepsilon, \quad \mu, \quad (5)$$

are fulfilled, where e is the electron charge, ϵ is the dielectric permittivity of the lattice, and κ is the inverse screening length. In this case the potentials $V_1(\mathbf{q})$ and $V_2(\mathbf{q})$ can be replaced in (2) and (4) by constants g_1 and g_2 that do not depend on the momentum. This replacement is possible for the potential V_1 because it is screened, while the potential V_2 is short-range by its very definition. Its matrix element are calculated between Bloch functions of different bands, and the latter are practically orthogonal to each other. Below we shall assume that the coupling constants g_1 and g_2 are real. We can ensure that they are real in the case of simple bands (degenerate only with respect to spin) by means of a gauge transformation.

III. SELF-CONSISTENT SYSTEM OF EQUATIONS FOR THE ORDER PARAMETERS

1. Definitions

The problem of excitonic ferromagnetism, describable by the Hamiltonian (4), is conveniently analyzed by means of the diagram technique for time-dependent Green functions [10]. The Green function is defined by the equality

$$G_{\alpha\beta}(\mathbf{r},\mathbf{r}';t)=-i\langle T\psi_\alpha(\mathbf{r},t)\psi_\beta^+(\mathbf{r}',0)\rangle. \quad (6)$$

Here $\psi_\alpha(\mathbf{r},t)$ and $\psi_\alpha^+(\mathbf{r},t)$ are operators annihilating and creating an electron with spin $\alpha/2$ at the point $\mathbf{r}$, in the Heisenberg picture. In the Schrödinger picture, these operators can be expressed in terms of the creation and annihilation operators $a_{i\alpha}^+(\mathbf{k})$ and $a_{i\alpha}(\mathbf{k})$ for band electrons with quasi-momentum $\hbar\mathbf{k}$. In the two-band model (1), we obtain

$$\psi_\alpha(\mathbf{r})\approx\sum_{\mathbf{k}}\{\varphi_{1\mathbf{k}}(\mathbf{r})a_{1\alpha}(\mathbf{k})+\varphi_{2\mathbf{k}}(\mathbf{r})a_{2\alpha}(\mathbf{k})\},$$
$$\psi_\alpha^+(\mathbf{r})\approx\sum_{\mathbf{k}}\{\varphi_{1\mathbf{k}}^*(\mathbf{r})a_{1\alpha}^+(\mathbf{k})+\varphi_{2\mathbf{k}}^*(\mathbf{r})a_{2\alpha}^+(\mathbf{k})\}, \quad (7)$$

where $\varphi_{1\mathbf{k}}(\mathbf{r})$ and $\varphi_{2\mathbf{k}}(\mathbf{r})$ are the Bloch functions, with quasimomentum $\hbar\mathbf{k}$, of an electron in the electron and hole bands respectively. We now introduce the matrix elements of the exact Green function (6), calculated in the Bloch-function basis,

$$G_{ij}^{\alpha\beta}(\mathbf{k},t)=-i\langle Ta_{i\alpha}(\mathbf{k})a_{j\beta}^+(\mathbf{k})\rangle, \quad (8)$$

where i, j are the band indices, taking the values 1 and 2. In the diagrams the exact Green function will be depicted by a thick line. The free Green functions $G_1^{(0)}(\mathbf{k},t)$ and $G_2^{(0)}(\mathbf{k},t)$ corresponding to the Hamiltonian (1) are diagonal in the band and spin indices; they are depicted by a thin solid line.

The temporal Fourier components of these functions have the usual form:

$$G_1^{(0)}(\mathbf{k},\omega)=\left[\omega+\mu+\frac{\hbar^2\mathbf{k}^2}{2m}+\frac{\varepsilon_g}{2}\right]^{-1}, \quad (9)$$

$$G_2^{(0)}(\mathbf{k},\omega)=\left[\omega+\mu-\frac{\hbar^2\mathbf{k}^2}{2m}-\frac{\varepsilon_g}{2}\right]^{-1}. \quad (10)$$

The electron-electron Coulomb interaction contained in $\mathscr{H}_{int}$ will be depicted by a dashed line, and the interaction of the electrons via the phonons by a wavy line, to which corresponds the free phonon Green function

$$D=\omega_0/(\omega^2-\omega_0^2). \quad (11)$$

With the aid of the Green function (6) we can find the mean values of any one-particle operator [10]. In the following we shall be interested in the mean values of the total magnetization $\mathbf{M}$, the local magnetic moment (spin-density wave) $\mathbf{M}(\mathbf{R})$, and the local charge density $Q(\mathbf{R})$. The matrices of the corresponding operators have the form

$$\mathbf{M}=m\sigma_{\alpha\beta}, \quad \mathbf{M}(\mathbf{R})=m\sigma_{\alpha\beta}\delta(\mathbf{r}-\mathbf{R}),$$
$$Q(\mathbf{R})=e\hat{I}\delta(\mathbf{r}-\mathbf{R}), \quad (12)$$

where m is the Bohr magneton, $\delta(\mathbf{r})$ is the Dirac delta-function, $\sigma_{\alpha\beta}$ is a vector whose components are the Pauli matrices, and $\hat{I}$ is the unit matrix. Using the expression for the Green function (6) in terms of its matrix elements (8)

$$G_{\alpha\beta}(\mathbf{r},\mathbf{r}';t)=\sum_{i,j,\mathbf{k}}G_{ij}^{\alpha\beta}(\mathbf{k},t)\varphi_{j\mathbf{k}}(\mathbf{r})\varphi_{i\mathbf{k}}^*(\mathbf{r}'),$$

with the aid of (12) we obtain

$$\mathbf{M}=-im\sum_{i,\mathbf{k}}\mathrm{Sp}\,\hat{\sigma}\hat{G}_{ii}(\mathbf{k},-0),$$
$$\mathbf{M}(\mathbf{R})=-im\sum_{i,j,\mathbf{k}}\varphi_{i\mathbf{k}}(\mathbf{R})\varphi_{j\mathbf{k}}^*(\mathbf{R})\mathrm{Sp}\,\hat{\sigma}\hat{G}_{ij}(\mathbf{k},-0), \quad (13)$$
$$Q(\mathbf{R})=-ie\sum_{i,j,\mathbf{k}}\varphi_{i\mathbf{k}}(\mathbf{R})\varphi_{j\mathbf{k}}^*(\mathbf{R})\mathrm{Sp}\,\hat{G}_{ij}(\mathbf{k},-0).$$

In these formulas the trace is calculated only over the spin indices. It can be seen from the expressions (13) that if all the anomalous Green functions (functions with non-coincident spin or band indices) are equal to zero, there is no magnetization or spin-density wave, and the charge-density wave has the period and symmetry of the lattice. When there are nonzero anomalous Green functions, additional terms of the type $\varphi_1(\mathbf{R})\varphi_2^*(\mathbf{R})$, associated

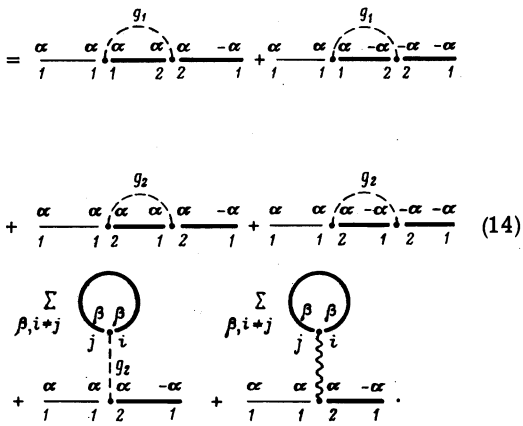
with the interference of Bloch waves from different bands, appear in the formulas (13).

In the case under consideration in which the extrema of the conduction and valence bands coincide, the presence of such terms leads to the result that the point symmetry group of the waves formed may differ from the point group of the lattice. This occurs if the functions $\varphi_{1\mathbf{k}}(\mathbf{R})$ and $\varphi_{2\mathbf{k}}(\mathbf{R})$ belong to inequivalent irreducible representations. In the general case when the extrema are displaced relative to each other by a vector $\mathbf{P}$, a new period, determined by this vector, also arises.

2. Properties of the Self-Consistent Potential

In order to find the anomalous Green functions in the high-density case (5), it is sufficient in the corresponding equations to retain only the direct and exchange diagrams containing interband Green functions $G_{ij}^{\alpha\beta}$ with $i \neq j$. This corresponds to the Hartree-Fock approximation.

One can immediately convince oneself that the simultaneous existence of interband anomalous functions of the "singlet" type $G_{ij}^{\alpha\alpha}(i \neq j)$ and "triplet" type $G_{ij}^{\alpha,-\alpha}(i \neq j)$ leads automatically to the existence of intraband anomalous Green functions $G_{ij}^{\alpha,-\alpha}$. Indeed, as shown in [1], the equation for the function $G_{11}^{\alpha,-\alpha}$ in the Hartree-Fock approximation has the following form



(14)

or, in analytic form,

$$G_{11}^{\alpha,-\alpha}(\mathbf{k},\omega)=-iG_1^{(0)}(\mathbf{k},\omega)\Big\{\sum_{\mathbf{k}',\omega'}[g_1G_{12}^{\alpha\alpha}(\mathbf{k}',\omega') + g_2G_{21}^{\alpha\alpha}(\mathbf{k}',\omega')]+\left(\frac{g^2}{\omega_0}-g_2\right)\sum_{\mathbf{k}',\omega',\beta}[G_{12}^{\beta\beta}(\mathbf{k}',\omega') + G_{21}^{\beta\beta}(\mathbf{k}',\omega')]\Big\}G_{21}^{\alpha,-\alpha}(\mathbf{k},\omega)-iG_1^{(0)}(\mathbf{k},\omega) \times\sum_{\mathbf{k}',\omega'}[g_1G_{12}^{\alpha,-\alpha}(\mathbf{k}',\omega')+g_2G_{21}^{\alpha,-\alpha}(\mathbf{k}',\omega')]G_{21}^{-\alpha,-\alpha}(\mathbf{k},\omega). \tag{15}$$

Therefore, in accordance with formula (13), the magnetization $\mathbf{M}$ (either M_x or M_y) is not equal to zero, e.g.,

$$M_x \sim -i\sum_{i,\mathbf{k},\alpha}G_{ii}^{\alpha\alpha}(\mathbf{k},-i0),$$

and, consequently, ferromagnetism arises.

It is necessary to draw attention to one more circumstance following from Eq. (15). Because of the presence in this equation of Hartree terms and terms containing the coupling constant g_2, it is not gauge-invariant. The phase of the left-hand side of Eq. (15) does not depend on the relative phase shift between the basis wave-functions

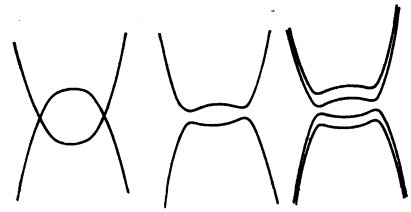


FIG. 1

of the conduction and valence bands, while the right-hand side changes its phase under such a transformation. It is easy to convince oneself that Eq. (15) is consistent in only two cases: when there is a purely real or a purely imaginary phase factor associated with the interband Green function.

As already noted above, we use the Hartree-Fock approximation, which only takes into account diagrams containing interband anomalous Green functions. This is equivalent to the statement that only the off-diagonal (in the band indices) components of the self-consistent Hartree-Fock potential V are nonzero. Inasmuch as the initial Hamiltonian (2) is invariant under the operation of rotation, and the spin-orbit interaction is not taken into account, the self-consistent one-particle potential V should depend only on the scalar product of the spin operator σ with some axial vector. In this case the structure of V is uniquely determined and, in the space of the band indices, the potential V has the form of the matrix:

$$V=\begin{pmatrix}0 & \Delta_s I+(\Delta_t\sigma)\\ \Delta_s^* I+(\Delta_t\sigma)^* & 0\end{pmatrix}. \tag{16}$$

In this formula the triplet (Δ_t) and singlet (Δ_s) order parameters are either both real or both purely imaginary, as is obvious from what was said about the gauge-noninvariance (of the second kind) of Eq. (15).

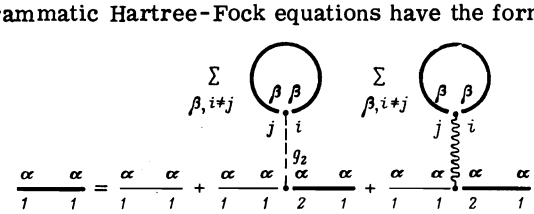
It follows from the expressions (16) and (1) that in the self-consistent potential V the one-particle excitation spectrum $\omega(\mathbf{k})$ has the form

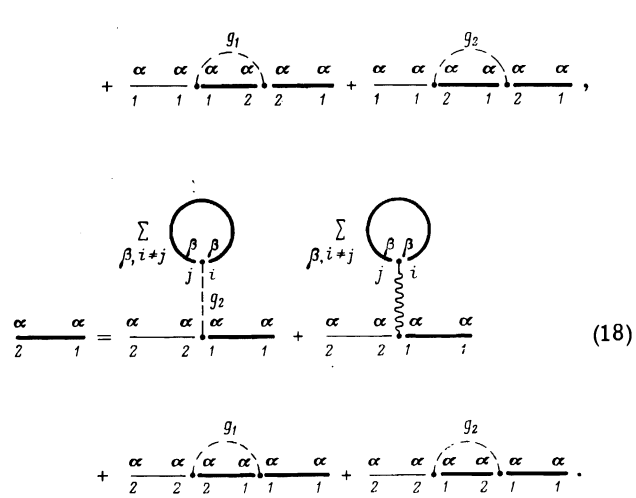
$$\omega(\mathbf{k})=\pm\{(\hbar^2\mathbf{k}^2/2m+\varepsilon_g/2)^2+|\Delta_s\pm\Delta_t|^2\}^{1/2} \tag{17}$$

and consists of four branches, distinguished by the band indices (the sign ± in front of the square root in (17)) and by the projection of the spin on the vector Δ_t (the sign ± under the square root). Fig. 1 illustrates the spectra described by formula (17) for the cases: a) spectrum of the unchanged phase ($\Delta_s = \Delta_t = 0$), b) spectrum of an excitonic insulator (either $\Delta_s = 0$, $\Delta_t \neq 0$ or $\Delta_t = 0$, $\Delta_s \neq 0$), and c) spectrum of an excitonic ferromagnet $\Delta_s \neq 0$, $\Delta_t \neq 0$).

3. Self-Consistency Equations

It follows from the structure of the self-consistent potential V (16) that, with no loss of generality, the di direction of the vector Δ_t can be chosen arbitrarily. It is convenient to point it along the z axis. Then the system of equations for the Green functions is simplified, since it does not contain anomalous Green functions with different spin indices. In this representation, the diagrammatic Hartree-Fock equations have the form





(18)

In analytic form, the system of equations (18) can be written as:

$$G_{11}^{\alpha\alpha}(\mathbf{k},\omega)=G_1^{(0)}(\mathbf{k},\omega)+G_1^{(0)}(\mathbf{k},\omega)(\Delta_s\pm\Delta_t)G_{21}^{\alpha\alpha}(\mathbf{k},\omega),$$
$$G_{21}^{\alpha\alpha}(\mathbf{k},\omega)=G_2^{(0)}(\mathbf{k},\omega)(\Delta_s\pm\Delta_t)G_{11}^{\alpha\alpha}(\mathbf{k},\omega), \tag{19}$$

where the sign ± in the brackets coincides with the sign of the spin index α. The self-consistency equations have the form

$$\Delta_s=-ig_s\int\frac{d\omega}{4\pi}\sum_{\mathbf{k}}[G_{21}^{11}(\mathbf{k},\omega+i0)+G_{21}^{-1,-1}(\mathbf{k},\omega+i0)],$$
$$\Delta_t=-ig_t\int\frac{d\omega}{4\pi}\sum_{\mathbf{k}}[G_{21}^{11}(\mathbf{k},\omega+i0)-G_{21}^{-1,-1}(\mathbf{k},\omega+i0)]. \tag{20}$$

Here g_s and g_t are the singlet and triplet coupling constants, defined by the expressions

$$g_s=g_1+g_2+4\left(\frac{g^2}{\omega_0}-g_2\right),$$
$$g_t=g_1+g_2. \tag{21}$$

Eqs. (19) and the self-consistency conditions (20) are valid in the case of real order parameters Δ_s and Δ_t. The case of imaginary order parameters, being energetically less favorable, will not be considered. It can be seen from (18) that the constant $g_{1m} = g_1 - g_2$, which is smaller than either g_s or g_t for either sign of g_2 and $g_1 > 0$, corresponds to this case.

We now find the functions $G_{21}^{\alpha\alpha}$ from the system (19). Substituting them into the self-consistency conditions (20), we obtain equations for the order parameters Δ_s and Δ_t. After integration over the frequency ω and summation over $\mathbf{k}$ (with truncation of the sum at large $\mathbf{k}$) in (20), these equations take the form

$$[g_sN(0)]^{-1}\Delta_s=(\Delta_s+\Delta_t)\ln\left[\frac{\tilde\omega}{\mu+\sqrt{\mu^2-(\Delta_s+\Delta_t)^2}}\right]$$
$$+(\Delta_s-\Delta_t)\ln\left[\frac{\tilde\omega}{\mu+\sqrt{\mu^2-(\Delta_s-\Delta_t)^2}}\right]$$
$$[g_tN(0)]^{-1}\Delta_t=(\Delta_s+\Delta_t)\ln\left[\frac{\tilde\omega}{\mu+\sqrt{\mu^2-(\Delta_s+\Delta_t)^2}}\right] \tag{22}$$
$$+(\Delta_s-\Delta_t)\ln\left[\frac{\tilde\omega}{\mu+\sqrt{\mu^2-(\Delta_s-\Delta_t)^2}}\right],$$

where $N(0) = mk_F/2\pi^2\hbar^2$ is the density of states at the Fermi level, $\hbar k_F = \sqrt{m|\epsilon_g|}$ is the Fermi momentum, $\tilde\omega$ is the characteristic energy cutoff, and $\mu > 0$[1)]. Eqs. (22) are written for the case when $|\Delta_s \pm \Delta_t| < \mu$. In the opposite case, e.g., when $|\Delta_s - \Delta_t| < \mu < |\Delta_s + \Delta_t|$, in Eqs. (22) it is necessary to replace the quantity $\mu + \sqrt{\mu^2 - (\Delta_s + \Delta_t)^2}$ in the logarithm by $|\Delta_s + \Delta_t|$. But if $\mu < |\Delta_s \pm \Delta_t|$, the analogous replacement is also performed in the logarithm containing $|\Delta_s - \Delta_t|$.

In the absence of doping ($\mu = 0$) the following solutions of the system of equations (22) correspond to the minimum energy:

$$\Delta_t=0,\quad \Delta_s=\Delta_{s0}\equiv\tilde\omega\exp[-1/g_sN(0)] \tag{23}$$

or

$$\Delta_s=0,\quad \Delta_t=\Delta_{t0}\equiv\tilde\omega\exp[-1/g_tN(0)]. \tag{24}$$

The solution (23) produces a singlet gap Δ_{s0} and only a charge-density wave, and the solution (24) produces a triplet gap Δ_{t0} and only a spin-density wave.

Using the relations (23) and (24), by means of straightforward transformations we can bring Eqs. (22) to the form, for $|\Delta_+|, |\Delta_-| < \mu$:

$$\Delta_+\ln\delta+\Delta_-\ln\gamma=\Delta_-\ln(\mu+\sqrt{\mu^2-\Delta_-^2}),$$
$$\Delta_-\ln\delta+\Delta_+\ln\gamma=\Delta_+\ln(\mu+\sqrt{\mu^2-\Delta_-^2}); \tag{25}$$

for $|\Delta_+| < \mu < |\Delta_-|$:

$$\Delta_+\ln\delta+\Delta_-\ln\gamma=\Delta_-\ln(\mu+\sqrt{\mu^2-\Delta_-^2}).$$
$$\Delta_-\ln\delta+\Delta_+\ln\gamma=\Delta_+\ln|\Delta_+|; \tag{26}$$

for $|\Delta_+|, |\Delta_-| > \mu$:

$$\Delta_+\ln\delta+\Delta_-\ln\gamma=\Delta_-\ln|\Delta_-|,$$
$$\Delta_-\ln\delta+\Delta_-\ln\gamma=\Delta_+\ln|\Delta_+|. \tag{27}$$

Here we have introduced the notation:

$$\delta=(\Delta_{s0}/\Delta_{t0})^{1/2},\quad \gamma=(\Delta_{s0}\Delta_{t0})^{1/2},\quad \Delta_{\pm}=\Delta_s\pm\Delta_t.$$

The parameters Δ_+ and Δ_- have the meaning of the energy gaps for states of electrons with positive and negative spin projections on the z axis, respectively.

Since we wish to study Eqs. (25), (26), (27) for a given number of electrons, we shall need an expression connecting the chemical potential μ and the excess-electron concentration N, which is determined by the level of doping. It is easy to show [11] that this relationship looks like:

$$(\mu^2-\Delta_+^2)^{1/2}+(\mu^2-\Delta_-^2)^{1/2}=2n,\ \mu>|\Delta_-|,\ |\Delta_-|,$$
$$(\mu^2-\Delta_-^2)^{1/2}=2n,\ |\Delta_-|<\mu<|\Delta_+|, \tag{28}$$

where $N = 4N(0)n$, and n is the concentration expressed in energy units. In addition, we shall need the formula for the energy change δE per unit volume that occurs in the phase transition of the semi-metal to an excitonic-insulator or ferromagnetic state. By direct averaging of the Hamiltonian (4), we find

$$\delta E=-N(0)\{(\Delta_+^2+\Delta_-^2)/2-2\mu n+2n^2\}. \tag{29}$$

IV. PARAMAGNETIC SUSCEPTIBILITY OF THE EXCITONIC INSULATOR

It is not difficult to see that Eqs. (25), (26) and (27) are symmetric with respect to replacement of Δ_{s0} by Δ_{t0} (with a simultaneous change of the sign in Δ_-). It is therefore sufficient to consider only the case $\Delta_{s0} \geq \Delta_{t0}$, and the formulas for the case $\Delta_{t0} > \Delta_{s0}$ will be analogous to those obtained below when, in the latter, the indices $s \rightleftarrows t$ are interchanged and the sign in Δ_- is changed.

For $\Delta_{s0} \geq \Delta_{t0}$ one of the solutions of Eqs. (25) and (28) is known [11]. It describes a metal-excitonic insulator phase transition accompanied by structural distortion. In the excitonic-insulator phase we have

$$\Delta_+^2=\Delta_-^2=\Delta_s^2=\Delta_{s0}(\Delta_{s0}-2n). \tag{30}$$

The energy gain in the formation of this phase is equal to

$$\delta E_{ex}=-N(0)(\Delta_{s0}-2n)^2. \quad (31)$$

We can determine the spin susceptibility of the excitonic insulator. For this we must write down self-consistency equations of the type (25) in an external magnetic field H acting on the electron spins, and then pass to the limit H → 0. Adding to the Hamiltonian (4) the Zeeman term

$$\mathcal{H}_z=-mH\sum_{i,\mathbf{k}}\{a^+_{i,+1}(\mathbf{k})a_{i,+1}(\mathbf{k})-a^+_{i,-1}(\mathbf{k})a_{i,-1}(\mathbf{k})\}, \quad (32)$$

it is straightforward to obtain, in place of (25) and (28), the following self-consistency and electrical-neutrality equations:

$$\begin{aligned}&\Delta_+\ln\delta+\Delta_-\ln\gamma=\Delta_-\ln(\mu_-+\sqrt{\mu_-^2-\Delta_-^2}),\\ &\Delta_-\ln\delta+\Delta_+\ln\gamma=\Delta_+\ln(\mu_++\sqrt{\mu_+^2-\Delta_+^2}),\\ &\sqrt{\mu_-^2-\Delta_-^2}+\sqrt{\mu_+^2-\Delta_+^2}=2n,\end{aligned} \quad (33)$$

where

$$\mu_\pm=\mu\pm mH.$$

Such a form for the equations in an external field is associated with the fact that, effectively, the magnetic field simply leads to the moving-apart of the Fermi surfaces of electrons with opposite spins. It must be noted that Eqs. (33) are valid only if $|\Delta_\pm| < \mu_\pm$. The magnetic moment **M** is directed along the field **H** and, according to (14), is equal to

$$M=2mN(0)\{\sqrt{\mu_-^2-\Delta_-^2}-\sqrt{\mu_+^2-\Delta_+^2}\}. \quad (34)$$

Solving Eq. (33) to terms linear in H and using (30) and (34), we obtain the following expression for the susceptibility of a doped excitonic insulator:

$$\chi=\frac{M}{H}=4m^2N(0)\left\{\frac{\sqrt{n^2+\Delta_s^2}}{n}+\frac{\Delta_s^2}{n(n\ln\delta-\Delta_{s0}+2n)}\right\}. \quad (35)$$

The first term in this formula corresponds to the susceptibility of the free carriers, and the second is associated with the fact that the external field induces a triplet gap. This term increases with increasing Δ_{t0}, becoming infinite when the equality

$$\Delta_{t0}=\Delta_{s0}\exp\{2-\Delta_{s0}/n\}. \quad (36)$$

is fulfilled. Thus, an excitonic insulator in a singlet state becomes unstable with respect to triplet pairing Δ_t when Δ_{t0} exceeds the value given by the equality (36), and a spontaneous magnetic moment arises in the system. An analogous picture arises when $\Delta_{t0} > \Delta_{s0}$. Then, when the inequality

$$\Delta_{s0}\geqslant\Delta_{t0}\exp\{2-\Delta_{t0}/n\} \quad (37)$$

is fulfilled, the triplet state of the excitonic insulator turns out to be unstable against the formation of a small singlet order parameter Δ_s. Thus, in the $(\Delta_{s0}, \Delta_{t0})$-plane there exists a region in which excitonic ferromagnetism appears. In Fig. 2 this region lies above the solid line defined by the relations (36) and (37).

As regards an undoped excitonic insulator (n = 0) in a weak magnetic field H (mH < $|\Delta_+|$, $|\Delta_-|$), the self-consistency equations for it do not depend on H, and coincide with (27). Therefore, at T = 0 an undoped excitonic insulator has susceptibility $\chi = 0$ and is always stable. The case with n = 0 is considered in more detail in [1], where it is shown that the coexistence in it of a triplet (Δ_t) and a singlet (Δ_s) order parameter cannot be realized even as metastable coexistence.

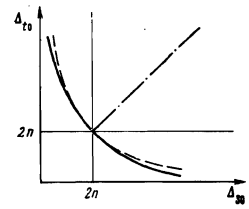


FIG. 2

V. SOLUTIONS OF THE EQUATIONS IN THE CASE OF AN EXCITONIC FERROMAGNET

Unlike the nonmagnetic solution (30), a solution of the equations for the order parameters in the general case $|\Delta_+| \neq |\Delta_-|$ cannot be obtained in the entire region of coupling constants $g_s(\Delta_{s0})$ and $g_t(\Delta_{t0})$. Therefore, we shall consider the behavior of the ferromagnetic solution only in the vicinity of characteristic values of Δ_{s0} and Δ_{t0}, namely, near the line (34) where weak ferromagnetism first appears in the $(\Delta_{s0}, \Delta_{t0})$-plane, on the line where $\Delta_+ = \mu$ and where the transition from weak to strong ferromagnetism occurs, and on the diagonal $\Delta_{s0} = \Delta_{t0}$.

1. Weak Ferromagnetism $|\Delta_+|, |\Delta_-| < \mu$

As can be seen from the expression (35) for the susceptibility of the dielectric phase, when the triplet constant Δ_{t0} exceeds the value given by the equality (36) the system is unstable against the appearance of a small triplet parameter Δ_t. Therefore, we shall seek the solution of Eqs. (25) near the line on which the ferromagnetic phase originates, to terms of fourth order in Δ_t. This makes it possible to calculate the energy gain associated with the formation of the excitonic ferromagnet, relative to the energy (31) of the nonmagnetic excitonic phase. The necessity of taking fourth-order terms into account is due to the fact that it follows from the Landau expansion [12] for the free energy near a second-order phase transition point that the energy gain on formation of the new phase is proportional to the fourth power of the equilibrium value of the order parameter.

The solution proceeds as follows. When the condition (28) is taken into account, the initial system of equations can be represented in the form

$$\begin{aligned}&2\Delta_s\ln\delta=\Delta_s\ln\left(\frac{f_+f_-}{\gamma^2}\right)-\Delta_t\ln\left(\frac{f_+}{f_-}\right),\\ &2\Delta_t\ln\delta=\Delta_s\ln\left(\frac{f_+}{f_-}\right)-\Delta_t\ln\left(\frac{f_+f_-}{\gamma^2}\right),\\ &f_\pm=n\pm n^{-1}\Delta_s\Delta_t+n^{-1}[(\Delta_s^2+n^2)(\Delta_t^2+n^2)]^{1/2}.\end{aligned} \quad (38)$$

Expanding the first equation up to terms $\sim\Delta_t^4$, we obtain the relationship between the singlet (Δ_s) and triplet (Δ_t) gaps:

$$\Delta_s^2\approx\Delta_{s0}(\Delta_{s0}-2n)+\frac{\Delta_{s0}-n}{n}\Delta_t^2-\frac{\Delta_{s0}^2-4n^2}{12\Delta_{s0}n^3}\Delta_t^4. \quad (39)$$

It follows from Eq. (39) that the appearance in the system of triplet pairing against the background of the singlet pairing Δ_s induces an increase of the latter as compared with the situation in which triplet pairing is absent, i.e., as compared with the non-magnetic phase (see (30)). In order to calculate the equilibrium value of the parameter Δ_t, it is necessary to use an expansion of the second equation of the system (38) to fifth order in Δ_t. As a result, we obtain

$$\Delta_t^2=6\Delta_{s0}n^3(\Delta_{s0}-2n)^{-2}[(\Delta_{s0}-2n)n^{-1}-2\ln\delta]. \quad (40)$$

As we should expect, the order parameter Δ_t is nonzero inside the region (36) of instability of the excitonic-insulator phase. If we now write the expression for the energy in the presence of a small triplet parameter Δ_t, then, to terms of order Δ_t^4, we obtain from (29) and (39):

$$\delta E_f = -N(0)\{(\Delta_{s0}-2n)^2 + 6\Delta_t^4\Delta_{s0}^{-1}n^{-3}(\Delta_{s0}-2n)^2\}. \quad (41)$$

Thus, it turns out that, first, the energy of the excitonic ferromagnet is lower than the energy (31) of the nonmagnetic phase, and, secondly, the expression (41) for the energy starts from terms of fourth order in Δ_t, in agreement with the Landau theory of second-order phase transitions.

To conclude this Section it must also be noted that, in the transition to the ferromagnetic state in a system with a given number of particles, the chemical potential μ increases relative to its value μ_0 in the excitonic-insulator phase:

$$\mu \approx \mu_0 + \Delta_{s0}^2 n^{-2}\Delta_t^2 + \ldots \quad (42)$$

In the quadratic approximation in Δ_t, this exactly cancels the energy gain produced by the increase in Δ_s (39) and the appearance of Δ_t. Obviously, complete cancellation does not occur if an electron reservoir of arbitrarily small capacity is introduced into the electrical-neutrality equation and terms quadratic in Δ_t appear in the energy. This indicates clearly the possibility of a first-order phase transition in systems with a reservoir [13].

2. Transition from Weak to Strong Ferromagnetism

This transition occurs when the triplet coupling constant Δ_{t0}, and with it the triplet gap Δ_t, become so large that the chemical potential μ moves out of one of the spin sub-bands (i.e., $|\Delta_+| \geq \mu$). The line of such a transition in the $(\Delta_{s0}, \Delta_{t0})$-plane is determined parametrically by the system of equations (26), if in these we put $\mu = |\Delta_+|$ and make use of the electrical-neutrality condition (28).

It is convenient to introduce the notation

$$\Delta_+ = 2n \operatorname{ch}\varphi, \quad \Delta_- = 2n \operatorname{sh}\varphi, \quad (43)$$

or

$$\Delta_s = n\exp\varphi, \quad \Delta_t = n\exp\{-\varphi\},$$

which automatically ensure conservation of the number of particles on the line $\Delta_+ = \mu$. By means of (26), the equation $\Delta_+ = \mu$ can be represented in the parametric form

$$\ln\{\Delta_{s0}/2n\} = \ln\operatorname{ch}\varphi + \exp\{-\varphi\}\ln\{1+\operatorname{ch}^{-1}\varphi\},$$
$$\ln\{\Delta_{t0}/2n\} = \ln\operatorname{ch}\varphi - \exp\{\varphi\}\ln\{1+\operatorname{ch}^{-1}\varphi\}. \quad (44)$$

This dependence is illustrated by the dashed line in Fig. 2. On this line the energy gain of the purely excitonic phase is known (see (31)). The corresponding expression (29) for the energy of the ferromagnetic phase can be written in the form

$$\delta E_f = -4N(0)n^2(\operatorname{ch}\varphi - 1). \quad (45)$$

The case of small values of φ corresponds to the region near the point $\Delta_{s0} \approx \Delta_{t0} \approx 2n$. Then, according to (44), we have $\Delta_{s0} \approx 2n(1+\varphi\ln 2)$. Consequently ($\varphi \ll 1$),

$$\delta E_{ex} = -N(0)(2n)^2\varphi^2/2,$$
$$\delta E_f = -N(0)(2n)^2\varphi^2/\ln 2, \quad (46)$$

i.e., the ferromagnetic phase is favored relative to the purely excitonic phase ($|\delta E_f| > |\delta E_{ex}|$).

In an analogous way we can also convince ourselves that this statement is valid for large values of φ, i.e., when the condition $\Delta_{s0} \gg n$ is fulfilled. Omitting the tedious intermediate calculations, we only give the result:

$$\delta E_{cx} = \delta E_f + n^2 N(0)\exp\{-3\varphi\}/6,$$

or, in the original notation,

$$(\delta E_{cx} - \delta E_f)/\delta E_{cx} = n^3/24\Delta_{s0}^3. \quad (47)$$

3. Solution on the Diagonal

For $\Delta_{s0} = \Delta_{t0}$, i.e., on the diagonal in the $(\Delta_{s0}, \Delta_{t0})$-plane (the dashed-dotted line in Fig. 2), the solution can be obtained exactly. On the diagonal, $\ln\delta = 0$, and therefore it can be seen immediately that Eqs. (25) cannot give a ferromagnetic solution. It remains to study Eqs. (26). Taking into account the electrical-neutrality condition (28), we obtain

$$0 = \Delta_-[\ln(2n + \sqrt{4n^2 - \Delta_-^2}) - \ln\Delta_{s0}],$$
$$\Delta_+ = \Delta_{s0}. \quad (48)$$

There exist two types of solutions of these equations:

$$\Delta_- = 0, \quad \Delta_+ = \Delta_{s0} \quad (49)$$

and

$$\Delta_-^2 = \Delta_{s0}(\Delta_{s0} - 4n), \quad \Delta_+ = \Delta_{s0}. \quad (50)$$

In the first case, (49), the energy gain is

$$\delta E_1 = N(0)[\Delta_{s0}^2 - 4n^2]/2. \quad (51)$$

In the second case, (50), we have

$$\delta E_2 = N(0)[(\Delta_{s0}-2n)^2 - 4n^2]. \quad (52)$$

A solution of the first type ((49), (51)) exists for $\Delta_{s0} > 2n$ and, as is easily seen, is more favorable than the nonmagnetic solution (30), (31) for $2n < \Delta_{s0} < 6n$. The solution of the second type ((50), (52)) exists for $\Delta_{s0} > 4n$ and, starting from this value, is favorable both in comparison with the nonmagnetic solution (30), (31) and in comparison with the solution of the first type ((49), (51)). Thus, as we should expect, everywhere along the diagonal $\delta = 1$ the most favorable phase is that of a strong excitonic ferromagnet.

All the solutions of the ferromagnetic type obtained in this paper require that the number of particles be (at least approximately) constant. If we study the other situation, with a fixed chemical potential, it can be shown that a ferromagnetic state will not arise. This is simply understood from the following arguments. Eqs. (25) have nontrivial solutions when $\mu < \Delta_{s0}$. But in this case, for μ = const and T = 0, the nontrivial solution (23), (24) of the system (27) immediately becomes possible, and this is certainly energetically more favorable, and is not ferromagnetic.

VI. DISCUSSION

We have shown that the transition to the excitonic-ferromagnetic state from the excitonic-diamagnetic ($\Delta_{s0} > \Delta_{t0}$) and excitonic-antiferromagnetic ($\Delta_{t0} > \Delta_{s0}$) states occurs by way of the second-order phase transition, for arbitrarily small coupling constants, provided that relations of the type (37) between these coupling constants are fulfilled. In conditions of thermodynamic equilibrium this transition exists only when the semimetal is doped, i.e., when there exists a small relative displacement of the electron and hole Fermi surfaces,

and the difference in the electron and hole concentrations is fixed. Incidentally, it is evident, because of the presence of a reservoir, that chromium is not ferromagnetic even though it is an antiferromagnet with unequal concentrations of electrons and holes and is well described by the excitonic-insulator model[13].

Near the phase-transition point from the excitonic insulator to the ferromagnetic state, according to the equations (18) for the Green functions and the formulas (14) for the magnetization waves and charge waves, the following effects should arise. If the parent phase contained a charge-density wave ($\Delta_{s0} > \Delta_{t0}$), then, as the magnetization M increases, a spin-density wave (the antiferromagnetic component of the magnetization density) will be formed in the system. If the parent phase was antiferromagnetic, then, in the ferromagnetic state, as the ferromagnetic component of M increases, a charge-density wave will build up in it, and, consequently, structural distortions will arise.

Thus, in the model we are discussing for excitonic ferromagnetism, there exists a coupling, via the uniform component of the magnetization, between the deformation of the lattice (the charge-density wave) and the spin-density wave. Effectively, there is a coupling between the phonons and magnons which does not contain a relativistically small parameter. Therefore, external fields should substantially alter the state of an excitonic insulator inclined toward ferromagnetism. Application of a uniform external magnetic field will induce magnetostriction effects. A constant external electric field in the case of allowed band-band transitions will induce a singlet gap Δ_S and, even in the excitonic-antiferromagnetic state, will induce spin-splitting of the bands, i.e., spin polarization. The role of an external uniaxial deformation can turn out to be equally important.

From this standpoint we can interpret the results of the work of Strakhov[14], who discovered ferromagnetic properties in powders and mechanically strongly-stressed crystals of the narrow-band semiconductor PbS. He noted that unstressed PbS samples were ordinary diamagnets, whereas in the stressed or crushed state they were ferromagnetic, and associated this with the well-known[15] property of PbS of going over from the symmetric cubic phase to an orthorhombic phase under mechanical stresses. For an excitonic insulator in an antiferromagnetic phase, such behavior under deformation is entirely natural. The formation of a charge-density wave as a result of a forced structural transition leads to the result that this wave, together with the antiferromagnetic spin-density wave, induces a magnetic moment as predicted by Eq. (15). The fact that lead sulfide in the cubic phase is an excitonic insulator is indicated by the experiments on the determination of the optical width of the forbidden band in it[16]. It is found that the optical width of the forbidden band increases as the level of doping decreases, i.e., it behaves as required by the relation (30).

In the work of Ivanov-Omskiĭ and co-workers[17], magnetic-susceptibility anomalies have been observed in HgTe compounds at low temperatures. The paramagnetic component of the susceptibility increased on decrease of the external magnetic field. This phenomenon is easily understood if we assume that at these temperatures the substance becomes an excitonic ferromagnet with a small magnetic moment.

As other examples of ferromagnetic compounds constructed from nonmagnetic elements we can cite $ZrZn_2$[18], Sc_3In[19] and the trichalcogenides of molybdenum with added Al and Ga[20]. These latter compounds are also superconductors with anomalously large values of the upper critical field H_{c2} for the comparatively low critical temperatures T_{cs}. From the recent data of[21], for the compound $Pb_{1.0}Mo_{5.1}S_6$ these values are: $T_{cs} = 14.4$ K, $H_{c2}(0) \approx 600$ kOe. Such high values of $H_{c2}(0)$ can be understood only by assuming the existence of planar sections on the Fermi surface. As follows from the theory developed above, under certain conditions this can lead to the appearance of conduction-electron ferromagnetism.

For the rare-earth metals[22], as the temperature is lowered there occurs first a transition to the antiferromagnetic phase, and then a structural transition arises at the ferromagnetic Curie point. From a theoretical point of view, this behavior can be described in the framework of the two-band model; the f-electrons can be regarded simply as a reservoir of spins which are aligned under the magnetization of the conduction electrons.

To conclude we shall make a few comments concerning the ferromagnetism in iron. It is known that the body-centered cubic modification of Fe is ferromagnetic. In this phase, according to the neutron-scattering data of[23], the spin-density distribution in the unit cell, although periodic with the period of the bcc structure, is alternating in sign, viz., a large positive spin density is concentrated at the lattice sites, and a negative spin density in the middle of the cube edges.

The absence of structural distortions in the ferromagnetic phase can be understood if we assume that the "excitonic" instability in iron is due to the interaction of two bands of the same symmetry, overlapping in momentum space. Then, as already noted at the end of Sec. III, subsection 1, the symmetry of the spin- and charge-density waves that arise coincides with the symmetry of the original lattice. Therefore, changes in the structure do not occur at the ferromagnetic transition point, although gaps appear, corresponding to the triplet and singlet order parameters. There are experimental indications of the formation of energy gaps on part of the Fermi surface in the ferromagnetic phase of iron[24]. On the basis of this, the authors propose that there is a possible analogy between the mechanism of the formation of the gap in Fe and the corresponding situation in Cr[13]. This picture agrees qualitatively with the model that we have considered, although it is difficult to expect quantitative agreement, inasmuch as the ferromagnetic interaction in iron is not weak.

[1)] The system of equations (22) differs from the system given in [1], because of a calculational error committed there; this error, however, had no effect on the qualitative conclusions drawn in [1].

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205