Effect of indirect exchange via conduction electrons on magnetic ordering of the layer type

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Degenerate magnetic semiconductors possess highly specific magnetic properties, inasmuch as the indirect exchange interaction via the conduction electrons is essentially of a non-Heisenberg type. If the initial ordering in the crystal is helicoidal and the period of the structure is not very large, then at a sufficiently high electron concentration the simple spiral is transformed into a ferromagnetic spiral, which is theoretically impossible in the framework of the Heisenberg model. The concentration phase transition to the magnetized state is second-order. With further increase in concentration the moment of the spiral gradually grows until the magnetization of the crystal reaches saturation. If the period of the initial helix is large, the spiral does not acquire a moment with increase of the electron concentration, but its period increases. At a certain concentration the period becomes infinite, i.e., the crystal goes over into a magnetized state. In an analogous way the conduction electrons increase the Bloch-wall thickness in ferromagnetic semiconductors.

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1. INTRODUCTION

It is well known that for degenerate magnetic semiconductors a situation in which the criterion for applicability of the Rudermann-Kittel-Kasuya-Yosida (RKKY) theory is not fulfilled is typical, i.e., the ratio of the s—d exchange energy AS to the Fermi energy μ is not small. At the concentrations typical for them, indirect exchange via the conduction electrons always facilitates the establishment of long-range ferromagnetic order. If the direct exchange between the magnetic atoms is antiferromagnetic, then, because of the competition between the direct and indirect exchange, magnetic structures that are forbidden in the Heisenberg model, to which the RKKY theory leads [1], turn out to be possible in degenerate magnetic semiconductors.

When direct antiferromagnetic exchange with nearest neighbors only is taken into account, in a certain range of carrier concentrations the energy minimum corresponds to a noncollinear antiferromagnetic structure, or, under certain conditions, a state with magnetization and electron density that are distributed nonuniformly over the crystal [1-3].

In real substances one not infrequently observes magnetic ordering of the layer type, in which the lattice consists of layers of ferromagnetically ordered magnetic atoms and the moments of the layers form a one-dimensional magnetic structure, e.g., an antiferromagnetic structure in layer antiferromagnets (see the book by Vonsovskiĭ [4]) or a helicoidal structure.

Usually such ordering arises as a consequence of the anisotropy of the exchange interaction in crystals with low symmetry, but sometimes it is also possible in isotropic substances, e.g., in certain magnetic semiconductors with cubic symmetry. Thus, in the chromium spinels $HgCr_2S_4$ and $ZnCr_2Se_4$ helicoidal ordering is observed [5, 6] while in EuSe at $2.2 < T < 4.6$ K a four-sublattice antiferromagnetic structure of the layer type is established [7].

The present paper is devoted to elucidating the character of the magnetic order in such materials when conduction electrons are present in them. We investigate uniform magnetic structures in a magnetic semiconductor with anisotropic exchange, consisting of layers of magnetic atoms that are ferromagnetically ordered within a layer, taking into account the direct exchange between nearest neighbors and between next-nearest neighbors and the indirect exchange via the conduction electrons. It will be shown that if the moments of the layers form a simple spiral a smooth transition of the spiral to ferromagnetic order on increase of the carrier concentration is possible only for a large value of the period of the spiral. Otherwise, an intermediate state that is theoretically impossible in the Heisenberg model can be formed, namely, a ferromagnetic spiral, a particular case of which is a noncollinear antiferromagnetic structure.

Qualitatively, these results can be understood from the following arguments. As it moves through the crystal, for maximum gain in s-d exchange energy an electron should align its spin along the direction of the local magnetic moment. Thus, in the case of a semiconductor with a narrow conduction band ($AS \gg W$, where W is the width of the conduction band), the spin of a conduction electron is rigidly coupled to the spin of the magnetic atom at which it is situated [8]. However, in the case of a broad conduction band ($AS \ll W$), which is investigated in the present paper, the loss in kinetic energy on rotation of the spin leads to the result that alignment of the electron spin parallel to the local moment is energetically favorable only if the latter changes its direction in space sufficiently slowly. For illustration, we shall consider the well-known formula for the electron energy for helicoidal ordering in the case of a broad band [9, 10]:

$$\varepsilon_{\mathbf{k}}^{\pm} = \frac{\varepsilon_{\mathbf{k}}^{0} + \varepsilon_{\mathbf{k}-\mathbf{q}}^{0}}{2} \pm \left[\left(\frac{\varepsilon_{\mathbf{k}}^{0} - \varepsilon_{\mathbf{k}-\mathbf{q}}^{0}}{2} \right)^2 + \left(\frac{AS}{2} \right)^2 \right]^{1/2}. \tag{1}$$

Here $\epsilon_{\mathbf{k}}^0$ is the energy when the s—d exchange is neglected, and is such that $\epsilon_{-\mathbf{k}}^0 = \epsilon_{\mathbf{k}}^0 \geq \epsilon_0^0$. Alignment of the electron spin along the direction of the local magnetic moment corresponds to the case when $\mathbf{k} = \mathbf{q}/2$ and the minus sign in front of the root in (1) is chosen (in this case only, the average value of the s—d exchange energy $\langle H_{s-d} \rangle = -AS/2$). Since, in (1), $\epsilon_{\mathbf{k}}^- = \epsilon_{\mathbf{q}-\mathbf{k}}^-$, the quantity $\epsilon_{\mathbf{k}}^-$ has an extremum at $\mathbf{k} = \mathbf{q}/2$, and for small $\mathbf{q}$ this extremum corresponds to a minimum. From this it is clear that if the helix vector $\mathbf{q}$ is small the introduction of electrons into the conduction band should lead not to

magnetization of the crystal, since almost the full gain in s–d exchange energy has already been obtained, but only to a decrease of the quantity q.

However, for $q > q_c$ (for $\epsilon_k^0 = k^2/2m$, q_c is defined by the condition $\epsilon_{q_c}^0 = AS/2$), this extremum corresponds to a saddle point and alignment of the spin is energetically unfavorable. For large values of q the magnetic ordering weakly alters the character of the motion of the slow electron, and its energy near the bottom of the conduction band is analytic in AS and can be sought by perturbation theory. Since, in this case, the dependence of the energy on the period of the spiral appears only in second order in $AS/(\epsilon_0^0 - \epsilon_q^0)$, and the dependence on the magnetization only in first order, it is natural that at a certain carrier concentration a magnetized state—the ferromagnetic spiral—will correspond to a lower energy than the simple spiral. Obviously, the spiral period corresponding to the energy minimum should also vary, but relatively little, as electrons appear.

A domain wall in a ferromagnetic semiconductor can also be regarded as order of the layer type. The presence of conduction electrons leads to an increase in the thickness of the wall, just as they decrease the magnitude of the helix vector.

2. THE MODEL

In the absence of conduction electrons the state of the system is determined entirely by direct exchange, which is described by the usual Heisenberg Hamiltonian:

$$H_M = -\frac{J_0}{2}\sum (\mathbf{S}_g\mathbf{S}_{g+\Delta}) - \frac{J_1}{2}\sum (\mathbf{S}_g\mathbf{S}_{g+\Delta_1}) - \frac{J_2}{2}\sum (\mathbf{S}_g\mathbf{S}_{g+\Delta_2}), \qquad (2)$$

$\mathbf{S}_g$ is the spin operator of the magnetic atom with label g. The lattice is assumed to be simple-tetragonal. The index Δ labels the nearest neighbors of the atom under consideration in the same plane, and Δ_1 and Δ_2 label those in the neighboring plane and in the plane beyond that, these planes being perpendicular to the special direction in the crystal (the C axis).

The integral within each plane (J_0) is assumed to be positive, and J_2 negative. Here we shall assume the quanquantity J_1 to be such that $|J_1| < -4J_2$, and the lowest energy corresponds to ordering of the simple-spiral type in which the angle of rotation of the moment from one ferromagnetic layer to another is $aq_0 = \arccos(-J_1/4J_2)$, where a is the distance between the layers [11].

In the presence of conduction electrons, we use the Hamiltonian of the s–d model:

$$H = H_e + H_{s-d} + H_M,$$
$$H_e = B_0\sum a_{g\sigma}^{\dagger} a_{g+\Delta\sigma} + B_1\sum a_{g\sigma}^{\dagger} a_{g+\Delta_1\sigma}, \quad H_{s-d} = -A\sum (\mathbf{S}_g\mathbf{s})_{\sigma\sigma'} a_{g\sigma}^{\dagger} a_{g\sigma'}. \qquad (3)$$

Here $a_{g\sigma}^*$ and $a_{g\sigma}$ are the creation and annihilation operators for a conduction electron with spin projection σ at the magnetic atom with label g. The signs of the parameters B_0, B_1 and A are unimportant, but it is convenient to assume that B_0 and B_1 are negative and A positive.

The Bloch integrals B_0 and B_1 and the s–d exchange energy AS are considerably greater than the magnetic-ordering energy $|J_i|S^2z_i$. Therefore, the crystal becomes ferromagnetic at carrier concentrations so low that the condition $AS \gg \mu$ is fulfilled, even though we are considering the case of a broad conduction band ($AS/4|B_1| \equiv \alpha \ll 1$). With such a relationship between the parameters it is possible to introduce a separation (characteristic for the adiabatic approximation) of the system into rapid (the conduction electrons) and slow (the spins) subsystems. In the spirit of this approximation it is necessary first of all to find the electron spectrum for an arbitrary state of the slow subsystem and fill the lower electronic levels with a given number of charge carriers, and only then seek the ground state corresponding to the minimum of the total energy expressed in terms of the variables of the spin subsystem.

3. THE ELECTRON SPECTRUM

In accordance with what has been said, we first study the electron subsystem; however, we consider not an arbitrary ordering of the spins, but (since this is precisely what is needed in the following) a ferromagnetic spiral—a magnetic structure with two parameters: the magnitude of the helix vector **q** directed along the C axis, and the magnetization S cos θ per atom.

Regarding the operators $\mathbf{S}_g$ as classical vectors, we put

$$\mathbf{S}_g = (S\sin\theta\cos qg,\ S\sin\theta\sin qg,\ S\cos\theta). \qquad (4)$$

Substituting (4) into $\tilde{H}_e = H_e + H_{s-d}$ from (3) and going over to the momentum representation, we obtain a Hamiltonian $\sum_{\mathbf{k}} H_{\mathbf{k}}$ quadratic in the operators $a_{\mathbf{k}\sigma}^*$, $a_{\mathbf{k}\sigma}$, the diagonalization of which reduces to diagonalizing the quadratic form

$$H_{\mathbf{k}} = \left(\varepsilon_{\mathbf{k}}^0 - \frac{AS}{2}\cos\theta\right)a_{\mathbf{k}\uparrow}^{\dagger} a_{\mathbf{k}\uparrow} + \left(\varepsilon_{\mathbf{k}-\mathbf{q}}^0 + \frac{AS}{2}\cos\theta\right)a_{\mathbf{k}-\mathbf{q}\downarrow}^{\dagger} a_{\mathbf{k}-\mathbf{q}\downarrow} \qquad (5)$$
$$-\frac{AS}{2}(a_{\mathbf{k}\uparrow}^{\dagger} a_{\mathbf{k}-\mathbf{q}\downarrow} + a_{\mathbf{k}-\mathbf{q}\downarrow}^{\dagger} a_{\mathbf{k}\uparrow})\sin\theta.$$

From (5) it is not difficult to obtain the electron spectrum for an arbitrary dispersion law:

$$\varepsilon_{1,2} = \frac{\varepsilon_{\mathbf{k}}^0 + \varepsilon_{\mathbf{k}-\mathbf{q}}^0}{2} \pm \left[\left(\frac{\varepsilon_{\mathbf{k}}^0 - \varepsilon_{\mathbf{k}-\mathbf{q}}^0}{2} - \frac{AS}{2}\cos\theta\right)^2 + \left(\frac{AS}{2}\sin\theta\right)^2\right]^{1/2}, \qquad (6)$$

or, for the cosine dispersion law used here,

$$\varepsilon_{\mathbf{k}}^0 = -4|B_0|\gamma_{\mathbf{k}0} - 2|B_1|\gamma_{\mathbf{k}1},$$
$$\gamma_{\mathbf{k}\lambda} = \frac{1}{z_\lambda}\sum_{\Delta_\lambda}\exp(i\mathbf{k}\Delta_\lambda), \qquad (7)$$
$$\varepsilon_{1,2} = -4|B_0|\gamma_{\mathbf{k}0} - 2|B_1|\{c\cos\varkappa \pm [(s\sin\varkappa + \alpha\cos\theta)^2 + (\alpha\sin\theta)^2]^{1/2}\},$$

where $s = \sin(aq/2)$, $c = \cos(aq/2)$, $\varkappa = ak_{||} - aq/2$, and $k_{||}$ is the projection of the quasi-momentum **k** on the C axis. The expression (6) generalizes the result of [9,10] for a simple spiral to the case of a ferromagnetic spiral.

First we discuss the character of the low-lying part of the electron spectrum for cos θ = 0. For sufficiently small q, the state with $\varkappa = 0$ corresponds to the bottom of the conduction band. The expansion of (7) (with the plus sign) in a series in powers of $\varkappa$ has the form

$$\varepsilon = -4|B_0|\gamma_{\mathbf{k}0} - 2|B_1|\left[\alpha + c + \frac{s^2 - \alpha c}{\alpha}\frac{\varkappa^2}{2} - \frac{(3s^4 + 4\alpha^2 s^2 - \alpha^3 c)}{\alpha^3}\frac{\varkappa^4}{24} + \ldots\right]. \qquad (8)$$

According to (8), the energy has a minimum at $\varkappa = 0$ only if $q \le q_c$, where the quantity q_c is defined by the equality

$$\sin^2\frac{aq_c}{2} = \alpha\cos\frac{aq_c}{2}$$

(for $\alpha^{1/2} \ll 1$ the quantity $aq_c = 2\alpha^{1/2}$, and for $\alpha = 1/5$ the period of the critical spiral is $2\pi/q_c \approx 7a$). For $q > q_c$ the minimum at the point $\varkappa = 0$ goes over into a local maximum ($\partial^2\epsilon/\partial\varkappa^2 < 0$) and from (7) we find that the energy is a minimum at the point

$$\varkappa = \varkappa_0 \equiv \arcsin\frac{(s^4-\alpha^2c^2)^{1/2}}{s},$$

and also, since the energy is even in κ in the absence of magnetization ($\cos\theta = 0$), at the point $\kappa = -\kappa_0$. Near the minima $\kappa = \pm\kappa_0$, the expansion of the energy is given by the expression

$$\varepsilon_{1,2} = -4|B_0|\gamma_{k_0} - 2|B_1|\left[\left(1+\frac{\alpha^2}{s^2}\right)^{1/2} - \left(1-\frac{\alpha^2c^2}{s^4}\right) \times\left(1+\frac{\alpha^2}{s^2}\right)^{-1/2}\frac{(\varkappa\pm\varkappa_0)^2}{2} + \ldots\right]. \tag{9}$$

If the spiral becomes ferromagnetic, the positions of the energy minima change. In the case of a weakly ferromagnetic spiral for $q < q_c$, expanding (7) in a series in powers of $\cos\theta$ and κ we find that, in the leading approximation in $\cos\theta$, the energy is a minimum not at $\kappa = 0$ but at

$$\varkappa = -\frac{\alpha s\cos\theta}{s^2-\alpha c},$$

and the energy at the bottom of the band is smaller than that from (8) for $\kappa = 0$ by an amount

$$|B_1|\frac{\alpha s^2\cos^2\theta}{\alpha c - s^2}$$

For $q > q_c$ the degeneracy in the sign of κ is lifted. As compared with the energy of the minima from (9), in the leading approximation in $\cos\theta$ the energy at one of the minima is increased by an amount D and at the other is decreased by an amount D, where

$$D = 2|B_1|\alpha\cos\theta\left(1-\frac{c^2\alpha^2}{s^4}\right)^{1/2}\left(1+\frac{\alpha^2}{s^2}\right)^{-1/2}.$$

The change in the quantity κ leads to small corrections $\sim\cos^2\theta$ and is not taken into account here. Because of the smallness of effects $\sim\mu/AS \ll 1$ we also neglect the change in the effective mass, so that, for $0 < \cos\theta \ll 1$, instead of the expression (9) we shall use below a dispersion law of the form

$$\varepsilon_{1,2} = -4|B_0|\gamma_{k_0} - 2|B_1|\left[\left(1+\frac{\alpha^2}{s^2}\right)^{1/2} - \left(1-\frac{\alpha^2c^2}{s^4}\right) \times\left(1+\frac{\alpha^2}{s^2}\right)^{-1/2}\frac{(\varkappa\pm\varkappa_0)^2}{2}\right] \pm D. \tag{10}$$

In the opposite case of states that are almost ferromagnetic, irrespective of the magnitude of q it is sufficient to consider only one subband. Expanding (7) (with the plus sign) in a series in powers of $k_\parallel$ and $y = \sin\theta$, it is not difficult to show that for $\sin\theta \ll 1$ the minimum of ϵ is attained at

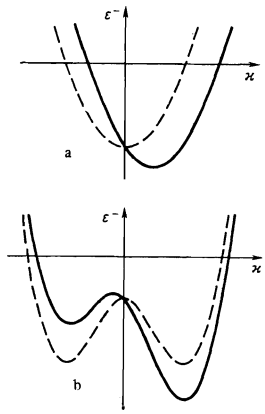
$$k_\parallel = \frac{sc\alpha^2}{2(s^2+\alpha)}\sin^2\theta, \quad k = k_\parallel$$

and the expansion of ϵ_{min} in powers of the small parameter y has the form

$$\varepsilon_{min} = -4|B_0|\gamma_{k_0} - 2|B_1|\left\{\left[1-\frac{2\alpha s^2}{s^2+\alpha}\right]y^2 + \frac{\alpha^4}{8}\left[\frac{2}{\alpha(s^2+\alpha)^2} - \frac{1}{\alpha^3} - \frac{1}{(s^2+\alpha)^3} + \frac{s^2c^2}{(s^2+\alpha)^4}\right]y^4 + \ldots\right\}. \tag{11}$$

4. THE FERROMAGNETIC STATE

As can be seen from (8)–(11), an electron at the bottom of the conduction band has the lowest energy for ferromagnetic ordering, since then the maximum possible gain in s–d exchange energy ($-AS/2$) is achieved. Thus, for a sufficiently high carrier concentration, when the magnetic ordering is determined entirely by the indirect exchange via the conduction electrons, the crystal should be ferromagnetic. At lower concentrations the variation of the direct-exchange energy becomes important, and so the magnetic order is determined by the competition of the two types of exchange and is no longer ferromagnetic, although it is not necessarily the same as the ordering in the absence of current carriers.



Lower part of the electron spectrum for helicoidal ordering (by formula (7)). a) for $q<q_c$; b) for $q>q_c$. Solid curve—$\cos\theta\neq 0$ (ferromagnetic spiral); dashed curve—$\cos\theta=0$ (simple spiral).

In order to elucidate the character of the magnetic ordering at not too high conduction-electron concentrations, we shall trace how the stability of the ferromagnetic order is lost on decrease in the number of current carriers. For this we shall analyze the magnon spectrum and find the conditions for which magnon frequencies with nonzero momentum vanish.

Since the direct exchange between the layers is nonferromagnetic, for a given value of q the frequency ω_q is a minimum if the vector is directed along the C axis. For such vectors $\mathbf{q} \parallel C$, with the aid of the formulas from [1], to within terms $\sim\mu/AS$ and $\sim 1/2S$ an expression for the magnon frequency is obtained in the form ($\nu = n/N$ is the relative carrier concentration)

$$\omega_q = 4|I_2||B_1|\frac{1-\cos aq}{AS+2|B_1|(1-\cos aq)}\cdot\left\{\left[2\sin^2\frac{aq}{2} - \left(\frac{I_1}{4I_2}+1-\alpha\right)\right]^2 - \left(\frac{I_1}{4I_2}+1+\alpha\right)^2 + \frac{A\nu}{4|I_2|}\right\}, \tag{12}$$

$$I_i = J_i S z_i, \quad z_i = \sum_{\Delta_i} 1.$$

As can be seen from (12), the sign of ω_q coincides with the sign of the factor in the curly brackets and the frequency corresponding to the minimum of this factor is the first to vanish.

If the period of the original spiral is large, i.e., $aq_0 < 2\arcsin\sqrt{\alpha/2}$, the first frequencies to become negative are those of the longest-wavelength magnons, with

$$\nu < \nu_{M1} \equiv \frac{|I_1+4I_2|}{|B_1|}S.$$

But if, on the other hand, $aq_0 > 2\arcsin\sqrt{\alpha/2}$, the first to vanish is the frequency with

$$q = q_{M2} \equiv \frac{1}{\alpha}\arccos\left(\alpha - \frac{I_1}{4I_2}\right) \tag{13}$$

at the concentration

$$\nu_{M2} \equiv \frac{4|I_2|}{A}\left(\frac{I_1}{4I_2}+1+\alpha\right)^2.$$

As is well known from the theory of second-order

phase transitions [12], if the first frequency to vanish is that of a magnon with momentum $\mathbf{q}$, then, as a result of the transition, a structure with the same wave-vector should be established. This result also remains valid for concentration phase transitions. We shall confine ourselves to considering the simplest structures possessing the appropriate periodicity and having at the same time, generally speaking, a nonzero moment—ferromagnetic spirals.

In the framework of the variational principle used below, the replacement of the spin operators $\mathbf{S}_g$ by the classical vectors (4) corresponds to a certain choice of a trial function, in which the parameters q and θ of the ferromagnetic spiral play the role of variational parameters. The structure is determined from the minimum, with respect to q and θ, of the total energy of the system, which is obtained from the Hamiltonian (3) with the equality (4) taken into account:

$$E=E_e+E_M=\sum \varepsilon_i\Theta(\mu-\varepsilon_i)-(I_1\cos aq+I_2\cos 2aq)\times\frac{NS\sin^2\theta}{2}-(I_1+I_2)\frac{NS\cos^2\theta}{2}; \tag{14}$$

here $\Theta(x) = 1$ for $x > 0$ and $\Theta(x) = 0$ for $x < 0$.

If the state of the system is almost ferromagnetic ($\sin\theta \ll 1$), one sub-band is filled by electrons at all values of q and we can assume in the calculations that all the carriers are at the bottom of the band (the kinetic energy $3\mu n/5$ is assumed to be a constant in this case, since the corresponding corrections are small $\sim\mu/AS \ll 1$), i.e.,

$$\frac{E_e}{N}=\frac{1}{N}\sum \varepsilon_i\Theta(\mu-\varepsilon_i)=\text{const}+\varepsilon_{min}\nu. \tag{15}$$

In this case, the expansion of the energy (14) in powers of the small parameter $y = \sin\theta$ has, when (11) is taken into account, the form

$$\frac{E_e}{N}=\text{const}+\frac{S\omega_q}{2}y^2-\frac{|B_1|\nu\alpha^4}{4}\left[\frac{2}{\alpha(s^2+\alpha)^2}-\frac{1}{\alpha^3}-\frac{1}{(s^2+\alpha)^3}+\frac{s^2c^2}{(s^2+\alpha)^4}\right]y^4+\ldots \tag{16}$$

We note that for $y = 0$ the energy cannot depend on q, since then the ordering is purely ferromagnetic. Therefore, the constant in (16) does not depend on q.

Since, for $\nu_{Mi} > \nu$, because of the negative sign of $\omega_{q_{Mi}}$, the ferromagnetic order is replaced by order with $q \sim q_{Mi}$, for $(\nu_{Mi}-\nu)/\nu_{Mi} \ll 1$ it is convenient to represent (16) in the form of an expansion in powers of $(q-q_{Mi})$:

$$\frac{E}{N}=\text{const}+A_0(q_{Mi})y^2+A_2(q_{Mi})(q-q_{Mi})^2y^2+C_0(q_{Mi})y^4+C_1(q_{Mi})(q-q_{Mi})y^4+\ldots \tag{17}$$

We note that a term $\sim y^2$ linear in $(q-q_{Mi})$ is absent, since q_{Mi} has been chosen from the condition for the minimum of $\omega_{q_{Mi}}$ from (12). We do not give the detailed form of the coefficients A_λ, C_λ, etc., since for the investigation of the phase transition only the signs of these coefficients are important.

5. THE FERROMAGNETIC-STATE–SIMPLE-SPIRAL TRANSITION

First of all we shall consider the case of small values of q_0, when the first frequencies to become negative are those of the longest-wavelength magnons.

Using the expansion (17), it can be shown that here an energy minimum with small $y = \sin\theta$ does not exist, and a smooth transition from ferromagnetic order to a ferromagnetic spiral is impossible. We note now that, for small values of $\cos\theta$, here only one electron sub-band is filled (it follows from the condition $aq_0 < 2\arcsin\sqrt{\alpha/2}$ on the period of the spiral that q is certainly $< q_c$, and then, according to (8), the minimum at $\kappa = 0$ is nondegenerate) and it is possible, by discarding corrections $\sim\mu/AS$, to disregard the dependence of the kinetic energy on q and θ.

Substituting into (14) the expression (15) with ϵ_{min} from (8) with corrections $\sim\cos^2\theta$, it is not difficult to show that the coefficient of $\cos^2\theta$ in the total energy will be positive for all $\nu \le \nu_{M1}$. This means that in the case of a long-wavelength spiral a second-order concentration phase transition to a ferromagnetic spiral is impossible not only from the ferromagnetic state but also from a simple spiral, and in the expression for the energy we can put $\cos\theta = 0$:

$$\frac{E}{N}=-\frac{I_1S}{2}\cos aq-\frac{I_2S}{2}\cos 2aq-2|B_1|\nu\cos\frac{aq}{2}. \tag{18}$$

Expanding (18) in a series in powers of q, we see that the coefficient of q^2 is proportional to $\nu-\nu_{M1}$ and that of q^4 is positive. In accordance with the Landau theory of second-order phase transitions a concentration phase transition from the ferromagnetic state to a simple spiral occurs at $\nu = \nu_{M1}$, and for $\nu \lesssim \nu_{M1}$

$$a^2q^2=\frac{8|B_1|(\nu-\nu_{M1})}{S(I_1+2I_2)}. \tag{19}$$

It should be noted that in real substances the long-range dipole-dipole interaction makes ferromagnetic ordering of the whole crystal unfavorable, and the crystal breaks down into ferromagnetic domains. Therefore, a phase transition from helicoidal ordering should be calculated with allowance for the factors determining the parameters of the domain structure, so that formula (19) has meaning only if the period of the helix is much smaller than the period of the domain structure.

The domain-structure parameters in a degenerate ferromagnetic semiconductor also depend on the carrier concentration. We shall estimate the contribution of the conduction electrons to the domain-wall energy, following the paper by Landau and Lifshitz [13]. To the appropriate functional, containing the parameters of the magnetic subsystem, we add only the energy densities of the conduction electrons and of the s-d exchange. We shall assume that the electron concentration is sufficiently great ($n \sim 10^{19}-10^{20}\ \text{cm}^{-3}$), so that the screening length is small and we can neglect the Coulomb energy.

The ordering of the spins in the Bloch domain-wall can be assumed to be locally helicoidal with a small wave-vector $\mathbf{q}(\mathbf{r})$, varying slowly in space. Therefore, for the energy of an electron in the domain wall we can use the expression (8) in the limit $a^2q^2 \ll AS/|B_1|$:

$$\varepsilon=|B_1|\frac{a^2q^2}{4}-\frac{AS}{2}+|B_1|\nu^2. \tag{20}$$

Taking into account (20), the condition $\mu < AS$, and also the condition that the domain wall is electrically neutral to within terms of second order in the deviation of the electron density from the average, we find that the contribution of the electrons to the energy of the wall is given by the expression

$$\int\sigma_e\,d\mathbf{r}=\tfrac{1}{4}|B_1|na^2\int q^2(\mathbf{r})\,d\mathbf{r}. \tag{21}$$

According to (21), the contribution of the electrons does not depend on the s–d exchange energy AS, but is proportional to the width of the electron band. Naturally, an analogous result is obtained when the contribution of indirect exchange to the energy of the long-wavelength ferromagnons [1] is taken into account (this can also be seen from the expansion of (12) in powers of q), inasmuch as we can speak of qualitatively the same violation of the ferromagnetic order in the two cases.

It is well-known [13] that the contribution of the direct exchange to the wall energy has the same structure as (21):

$$\int \sigma_M \, d\mathbf{r} = \xi T_C a^2 \int q^2(\mathbf{r}) \, d\mathbf{r} \tag{22}$$

(here T_C is the Curie temperature and ξ is a constant of order unity). Therefore, we can state that the Bloch-wall thickness δ increases as electrons appear in the conduction band. In accordance with the results of [13], this change in thickness is described by the formula

$$\delta(\nu) = \delta(0)\left[\frac{|B_1|\nu}{4\xi T_C} + 1\right]^{1/2}. \tag{23}$$

Taking, for the estimates, $|B_1| \sim 1$ eV and $T_C \sim 100$ K, we obtain from (23) that the increase $\delta(\nu) - \delta(0)$ in the wall thickness becomes of the same order as $\delta(0)$ itself at carrier concentrations $\sim 10^{20}$ cm^{-3}.

6. FERROMAGNETIC SPIRAL

In the case when the period of the original spiral is not large ($aq_0 > 2\arcsin\sqrt{\alpha/2}$), for $\nu \lesssim \nu_{M2}$ ordering of the ferromagnetic-spiral type corresponds to the minimum of the energy (17). In fact, for $0 < q_{M2} < \pi/a$, we find from (16) that, in (17),

$$A_0 \sim \nu - \nu_{M2}, \quad A_2 > 0, \quad C_0 > 0, \quad C_1 > 0 \tag{24}$$

and for the parameters q and θ corresponding to the minimum of E this leads, for $\nu \lesssim \nu_{M2}$, to dependences that are characteristic for second-order phase transitions:

$$\sin\theta \sim (1 - \nu/\nu_{M2})^{1/2}, \quad q - q_2 \sim -(1 - \nu/\nu_{M2}). \tag{25}$$

We note that the total change in the period of the spiral when the concentration ν changes from 0 to ν_{M2} is small: from (13) it can be seen that

$$\cos aq_{M2} - \cos aq_0 = \alpha.$$

For small magnetizations ($\cos\theta \ll 1$), we confine ourselves for simplicity to studying ferromagnetic spirals spirals with $q > q_c$, such that $1 - c^2\alpha^2 s^{-4} \sim 1$. In this case conduction electrons are in both subbands, which are describable by a quadratic dispersion law. Neglecting terms $\sim \mu/AS$, according to (10) the electron energy can then be written in the form

$$E_e/N = \text{const} - 2|B_1|\sqrt{1+\alpha^2/s^2}\,\nu + (\nu_+ - \nu_-)D. \tag{26}$$

We determine the difference $\nu_+ - \nu_-$ of the carrier concentrations in the sub-bands from the equation $\mu(\nu_-) - \mu(\nu_+) = 2D$, which, for $\cos\theta \to 0$ (and, consequently, $D \to 0$), has the solution $\nu_- - \nu_+ = 2\nu D[3\mu(\nu/2)]^{-1}$. For the Fermi energy we use the usual formula

$$\mu(\nu) = \frac{(6\pi^2\nu)^{2/3}}{(8m_x m_y m_z)^{1/3}}. \tag{27}$$

Here, according to (10), $2m_x = 2m_y = |B_0|^{-1}$, and $2m_z = |B_1|^{-1}(1 - \alpha^2 c^2 s^4)^{-1}(1 + \alpha^2/s^2)^{1/2}$.

Substituting the expression (26) for E_e into (14), we find that in a total energy of the form

$$E = E_1(q) + K_2(q)\cos^2\theta + K_4\cos^4\theta + \ldots \tag{28}$$

the coefficient $K_2(q)$ vanishes at $\nu = \nu_A(q)$, where ν_A is determined by the equality

$$\nu_A = \frac{|I_1 + 4I_2c^2|s^2S(1+\alpha^2/s^2)}{3\alpha^2|B_1|^2(1 - c^2\alpha^2/s^4)}\mu\left(\frac{\nu}{2}\right). \tag{29}$$

It can be shown that the coefficient K_4 is positive, i.e., at $\nu = \nu_A(q_A)$ a second-order phase transition occurs from the simple spiral to a ferromagnetic spiral with

$$\cos\theta \sim [\nu - \nu_A(q_A)]^{1/2}, \quad q - q_A \sim \nu - \nu_A(q_A).$$

However, before the appearance of the magnetization, the period of the spiral changes very little with increase of the electron concentration and, in the leading approximation in μ/AS, we can seek the quantity ν_A by putting $K_2(q_0) = 0$, i.e., using formula (29) in which q is replaced by q_0. We shall estimate ν_A, assuming that $1 - c^2\alpha^2 s^{-4} \sim 1$. Eliminating μ from (29), using (27) it is not difficult to obtain for ν_A the expression

$$\nu_A^{1/3} \approx 10\frac{|I_2 S|}{\alpha^2|B_1|}\left|\frac{B_0}{B_1}\right|^{2/3} s^4. \tag{30}$$

For the estimate we put $|B_0| = |B_1| = 0.5$ eV, $\alpha = 1/5$, $|I_2 S| = 30$ K and put the spiral period equal to six lattice constants (i.e., $s^2 = \frac{1}{2}(1 + I_1/4I_2) = \frac{1}{4}$). If we assume that there are $\sim 10^{22}$ magnetic atoms in one cubic centimeter, then from (30) we obtain in this case $\nu_A \sim 10^{19}$ cm^{-3}.

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[1] É. L. Nagaev, Fiz. Tverd. Tela **11**, 2779 (1969) [Sov. Phys.-Solid State **11**, 2249 (1970)].

[2] É. L. Nagaev, Zh. Eksp. Teor. Fiz. **57**, 1274 (1969) [Sov. Phys.-JETP **30**, 693 (1970)]; Fiz. Tverd. Tela **14**, 773 (1972) [Sov. Phys.-Solid State **14**, 658 (1972)].

[3] É. L. Nagaev, ZhETF Pis. Red. **16**, 558 (1972) [JETP Lett. **16**, 394 (1972)]; V. A. Kashin and É. L. Nagaev, Zh. Eksp. Teor. Fiz. **66**, 2105 (1974) [Sov. Phys.-JETP **39**, 1036 (1974)].

[4] S. V. Vonsovskiĭ, Magnetizm (Magnetism), "Nauka", M., 1971.

[5] R. J. Plumier, J. Appl. Phys. **37**, 964 (1966).

[6] J. M. Hastings and L. M. Corliss, J. Appl. Phys. **39**, 632 (1968).

[7] R. Griesson, M. Landolt and H. R. Ott, Solid State Commun. **9**, 2219 (1971).

[8] É. L. Nagaev, Zh. Eksp. Teor. Fiz. **56**, 1013 (1969) [Sov. Phys.-JETP **29**, 545 (1969)]; Fiz. Metal. Metalloved. **29**, 905 (1970) [Phys. Metals Metallog. **29**, No. 5, 10 (1970)].

[9] I. E. Dzyaloshinskiĭ, Zh. Eksp. Teor. Fiz. **47**, 336, 992 (1964) [Sov. Phys.-JETP **20**, 223, 665 (1965)].

[10] R. J. Elliott and F. A. Wedgwood, Proc. Phys. Soc. **81**, 846 (1963); **84**, 63 (1964).

[11] S. V. Tyablikov, Metody kvantovoi teorii magnetizma (Methods in the Quantum Theory of Magnetism), "Nauka", M., 1965 (English translation published by Plenum Press, N. Y., 1967).

[12] L. D. Landau and E. M. Lifshitz, Statisticheskaya fizika (Statistical Physics), "Nauka", M., 1964 (English translation published by Pergamon Press, Oxford, 1969).

[13] L. D. Landau and E. M. Lifshitz, Phys. Z. Sowjet. **8**, 153 (1935) (English translation in "Collected Works of L. D. Landau", Gordon and Breach, N.Y., 1965).

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231