Ferromagnetism of electrons of a filled band

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A model is proposed of the ferromagnetic state of valence electrons of a narrow-band semiconductor (semimetal) with low symmetry of the extremal point in the Brillouin zone, when dipole interband transitions are forbidden. It is shown that in a system with imaginary order parameter the ferromagnetic ordering of electrons is produced to a degree determined by the interband transitions with respect to the angular momentum. The uncompensated magnetic moment of the electrons of a completely filled band is produced even in an undoped semiconductor (semimetal), in contrast to an excitonic ferromagnetism. It must be emphasized that in systems with imaginary order parameter, states with spontaneous currents arise in the case of allowed interband dipole transitions. The expressions obtained for the magnetic moment and for the magnetic susceptibility are determined by the modulus of the imaginary gap and by the square of the matrix element of the angular-momentum operator. Allowance for the spin–orbit interaction does not alter qualitatively the results obtained neglecting this interaction. The temperature dependence of the susceptibility at temperatures close to 0°K and close to the transition temperature T_c is calculated. It is shown that the susceptibility diverges in accordance with the Curie–Weiss law at the transition point.

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INTRODUCTION

The problem of the orbital magnetism of electrons is one of the fundamental problems of solid-state physics. A theoretical investigation of magnetism calls for a cautious analysis of the dynamics of the electron in the presence of a periodic potential and of a magnetic field, and this is a rather complicated problem. The conduction electrons in some metals have a paramagnetic behavior, since the Pauli paramagnetism in them is larger than the Landau diamagnetism, while substances such as bismuth and graphite are known to be anomalously diamagnetic. The variety of magnetic properties of the band electrons finds its explanation in the manifold of factors that influence in one way or another the behavior of the electrons. For example, the periodic potential of a crystal can change the relative values of the paramagnetic and the diamagnetic effects, thereby coupling these two phenomena via the spin–orbit interaction.

A qualitative explanation of many magnetic properties of band electrons can be found in the Landau–Peierls theory. This theory is based on two assumptions: first, the interband effects are neglected; second, no account is taken of the collective effects. It is clear that allowance for these effects should lead to a more adequate explanation of the known experimental data. The collective effects, in particular, are of very great importance for the explanation of strong magnetism phenomena. Interband effects in orbital magnetism of electrons of a crystal can be divided into two types. First, if the band is degenerate, as, for example, in the case of a p band or d band, the interband effects lead to orbital paramagnetism, which is analogous to the Van Vleck paramagnetism. Second, interband effects lead to changes of the Landau diamagnetism. The first type of these interband effects is investigated in Ref. 1. The influence of interband effects on the Landau diamagnetism was investigated by Fukuyama and Kubo,[2] where the static magnetic susceptibility was calculated for an ideal crystal in which the periodic potential is assumed to be one-dimensional. The following cases are considered in Ref. 2: 1) when the number of electrons in the upper band is small and all electrons are under the influence of the Bragg scattering, and 2) when the upper band contains many electrons and only a small fraction of them is under the influence of the periodic potential. Allowance for the interband effects leads to substantial changes of the properties when the Fermi level lands in the region of the forbidden energy band.

The effect of the spin–orbit interaction on the magnetic susceptibility of the conduction-band electrons was taken into account in Ref. 3. The experimental and theoretical investigations show that the spin–orbit interaction in crystals with very small energy gaps leads to substantial changes in the mixing of the spin and orbital properties of the electrons. The large effective g-factor is evidence that the spin–orbit interaction exerts a substantial influence on the magnetisation of the system on account of the orbital motion of the electrons.

In Ref. 4, in the language of Green's functions, a theory was developed of the orbital magnetism of Bloch electrons, with account taken not only of the interband effects but also of the interelectron interactions. The onset of varied singularities of different characteristics of the orbital magnetism attests to the important role of collective and interband effects in the investigation of this problem.

It is well known that the dielectric gap in the state of an excitonic insulator has a collective character. In addition, since the electron and hole states in an excitonic dielectric are intermixed, the influence of both the lower (filled) and upper (free) band of the initial model on the different properties of the excitonic dielectric is automatically taken into account. When the system goes over into the state of an excitonic insulator, new magnetic properties also appear, based on collective and interband effects. In particular, the dependence of the magnetic susceptibility on the temperature in the excitonic-dielectric state was obtained in Ref. 5. Below the

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transition temperature T_c but not far from it, the paramagnetic character of the susceptibility of a semimetal assumes the diamagnetic character of a dielectric.

It was shown in Ref. 6 that in systems that are unstable to electron–hole pairing, ferromagnetic ordering of the band electrons sets in at arbitrarily small coupling constants, provided an excess electron or hole density is present. In contrast to other models, this model leads to ferromagnetism at arbitrarily small electron-electron interactions, provided only that the interelectron spectrum meets certain requirements. Namely, the substance in the initial phase should be a semimetal whose electron and hole Fermi surfaces are almost the same, apart from translation by a certain vector $\mathbf{p}$, or a metal with narrow allowed bands and for certain values of the vector $\mathbf{p}$ the condition $\varepsilon(\mathbf{k}) = -\varepsilon(\mathbf{k}+\mathbf{p})$ is satisfied, or a metal with flat Fermi sections, or a semiconductor whose exciton binding energy is larger than the band gap.

The variety of magnetic properties of systems in the state of an excitonic insulator is not limited to the foregoing examples. All these systems, which are unstable to electron–hole pairing, are characterized by the fact that all the phenomena they exhibit are connected with the onset of an ordering that is determined by a real order parameter. However, in addition to states with real order parameters, which lead to charge-density waves (ChDW), i.e., to structural transitions, or to spin-density waves (SDW), that lead to antiferromagnetism of the system, there exist also states with imaginary order parameters, which lead to current density waves (CuDW) and to spin–current density waves (SCDW).[7] These may turn out to be the ground states if the spin–orbit interaction in such systems is strong enough (as indicated in Ref. 8), or when scattering by charged impurities takes place. An idea was advanced earlier that an equilibrium system with direct bands acquires an interesting property in the case of dielectric pairing with imaginary order parameter Δ. Namely, if only transitions with change of total angular momentum j are allowed in the system (as in HgTe), then the ordering in this case is accompanied by the appearance of ferromagnetic properties.

In the two-band model, in contrast to the single-band model with flat Fermi surface sections, owing to the additional degree of freedom connected with the presence of two bands with different symmetries, the choice of the phase of the order parameter leads to new physical effects (current in the gound state or ferromagnetism).

It should be noted that for systems with current in the ground state, at a temperature above the transition point, an interesting temperature dependence of the diamagnetic susceptibility is obtained, namely in accordance with the Curie law.[9]

The present paper is devoted to an investigation of this new model of ferromagnetism, which is produced in systems with a single-electron spectrum unstable to electron–hole pairing. The ferromagnetic ordering of the band electrons is due to the fact that the order parameter is imaginary and is governed by the number of allowed interband transitions with respect to the orbital angular momentum.

We note that usually in the band-ferromagnetism model the ferromagnetic ordering is due to the electron spin. In our model, ordering takes place of the orbital magnetic moment of the electrons of a fully filled valence band, for which the total spin is equal to zero.

We note here an important property of the orbital angular momentum, namely its quenching ability.[10] According to this property, if the crystal–field symmetry is low enough to lift all the orbital degeneracy, then in first-order approximation the orbital angular momentum is equal to zero and it is said that the crystal field quenches the orbital momentum. Nevertheless, in our model a ferromagnetic state is produced because the order parameter is imaginary.

1. FORMULATION OF PROBLEM AND HAMILTONIAN OF THE SYSTEM

We consider a semiconductor or semimetal with conduction-band and valence-band extrema that coincide in momentum space. The dispersion is isotropic and parabolic, and the corresponding energy branches are nondegenerate. Such a picture can arise in a system whose crystal-field symmetry is low enough to lift all or part of the orbital degeneracy. We note that in the case of incomplete lifting of the degeneracy, the subsequent splitting is realized by other distortions. For example, the field of a cubic crystal does not lift fully the orbital degeneracy of the branches of the $3d$ electrons, and further splitting takes place as a result of additional tetragonal distortions. Such a picture can result from a rather strong crystal field, and therefore this field turns out to be one of the largest terms in the Hamiltonian. One can expect the contributions to the Hamiltonian to decrease in the following sequence[10]

$$\mathcal{H} = \mathcal{H}_{ia} + \mathcal{H}_{cf} + \mathcal{H}_Z + \mathcal{H}_{so},$$

where $\mathcal{H}_{ia}$ is the contribution of intra-atomic Coulomb interaction, $\mathcal{H}_{cf}$ is the contribution of the crystal field, $\mathcal{H}_Z$ is the contribution of the Zeeman energy, and $\mathcal{H}_{so}$ is the contribution of the spin–orbit interaction.

Thus, $\mathcal{H}_{ia} + \mathcal{H}_{cf}$ provide the corresponding band picture of the semiconductor or semimetal considered by us.

We note also that the symmetry and parity of the corresponding nondegenerate modes are such that interband transitions in the angular orbital momenta are allowed but dipole transitions are forbidden. For example, for systems having the symmetry O_h, with conduction- and valence-band extrema located at the X points of the Brillouin zone with point group D_{4h}, and with an electronic-state symmetry described by the spinor representations X_2^+ and X_3^- or X_2^- and X_3^+, or else X_1^+ and X_4^- or X_1^- and X_4^+ of the D_{4h} group, the foregoing conditions are satisfied. Interband orbital–angular-momentum transitions are due to the Zeeman term in the Hamiltonian.

We break up the Hamiltonian $\mathcal{H}$ of the investigated system into three parts: $\mathcal{H}_0$, $\mathcal{H}_Z$, and $\mathcal{H}_{int}$, representing respectively the Hamiltonians of the system of the electrons that do not interact with one another and are in

the periodic field of the lattice, the Zeeman Hamiltonian, and the Hamiltonian describing the electron-electron interaction, in which we retain only the terms responsible for the excitonic instability of the density-density type from the different bands. To simplify the calculations we shall not write out terms of the type $a_1^+a_1^+a_2a_2$, so that the phase of the order parameter is fixed.

The interband electron-phonon interaction is disregarded, since it does not affect qualitatively the results that follow. We start out with one more simplifying assumption, namely we neglect the spin-orbit interaction, and thus separate completely the spin contribution to the magnetism from the orbital contribution. We note that later on the calculation will be carried out with this interaction taken into account.

The sum of the single-particle Hamiltonian and of the Zeeman term is

$$\mathcal{H}_0+\mathcal{H}_Z=p^2/2m+\mu_B(\mathbf{L}+2\mathbf{S})\mathbf{H}, \tag{1}$$

and if the magnetic field $\mathbf{H}$ is parallel to the z axis, which is itself directed along the axis of higher symmetry of the crystal, then

$$\mathcal{H}_0+\mathcal{H}_Z=p^2/2m+\mu_B(L_z+\sigma_z)H, \tag{1a}$$

where μ_B is the Bohr magneton, L_z is the orbital angular momentum, and σ_z is a Pauli matrix.

We obtain the form of the Hamiltonian $\mathcal{H}_0+\mathcal{H}_Z$ in the representation of Luttinger and Kohn. In this representation, the term of the interband transitions, which is linear in the electron creation and annihilation operators is explicitly separated and the interaction Hamiltonian is written in the simplest form. The total and orthonormal system of the basis functions of this representation is

$$\psi_{n\mathbf{k}}=\psi_{n\mathbf{k}_0}\exp(i\mathbf{kr})\equiv u_{n\mathbf{k}_0}(\mathbf{r})\exp\{i(\mathbf{k}+\mathbf{k}_0)\mathbf{r}\}, \tag{2}$$

where

$$\psi_{n\mathbf{k}_0}=u_{n\mathbf{k}_0}(\mathbf{r})\exp(i\mathbf{k}_0\mathbf{r}) \tag{3}$$

is the exact solution of the Schrödinger equation in the given periodic potential of the lattice, and is known for a fixed value of the wave vector k_0 in all the bands. Here n is the number of the band, k is an arbitrary vector of the Brillouin zone reckoned from the end of the vector k_0. The wave functions of type (2) correspond to the electron creation and annihilation operators $a^+_{n\mathbf{k}\sigma}$ and $a_{n\mathbf{k}\sigma}$.

In the basis (2) the Schrödinger equation in a specified periodic potential corresponds to the Hamiltonian (we leave out here the spin indices)

$$\sum_{n,\mathbf{k}}\{(\hbar^2/2m_0)\mathbf{k}^2+\varepsilon_n(\mathbf{k}_0)\}a_{n\mathbf{k}}^+a_{n\mathbf{k}}+\sum_{n,n',\mathbf{k}}(\hbar/m_0)\mathbf{P}_{nn'}\mathbf{k}a^+_{n'\mathbf{k}}a_{n\mathbf{k}}, \tag{4}$$

where m_0 is the mass of the free electron, $\varepsilon_n^{(\mathbf{k}_0)}$ is the energy of the state with wave function (3), and the vector $\mathbf{P}_{nn'}$ is defined as

$$\mathbf{P}_{nn'}=-i\hbar\int d\mathbf{r}\psi^*_{n\mathbf{k}_0}(\mathbf{r})\nabla_{\mathbf{r}}\psi_{n'\mathbf{k}_0}(\mathbf{r}).$$

Bearing in mind the indicated symmetry, we obtain $P_{12}=0$, and as the vector $\mathbf{k}_0$ we choose the vector corresponding to the band extremum.

In the Zeeman Hamiltonian we have for L_z in this basis the two matrix elements

$$\hbar\int d\mathbf{r}\psi^*_{n\mathbf{k}_0}(\mathbf{r})[\mathbf{r}\times\mathbf{k}]_z\psi_{n'\mathbf{k}_0}(\mathbf{r}), \tag{5}$$

$$-i\hbar\int d\mathbf{r}\psi^*_{n\mathbf{k}_0}(\mathbf{r})[\mathbf{r}\times\nabla_{\mathbf{r}}]_z\psi_{n'\mathbf{k}_0}(\mathbf{r}). \tag{6}$$

It should be noted here that it is most convenient to direct the z axis, and consequently also the magnetic field $\mathbf{H}$, along the direction of the highest symmetry of the crystal. In this case for a crystal with forbidden dipole transitions the first matrix element drops out ($n=1$, $n'=2$). We confine ourselves to a two-band model, assuming that the influence of the remaining bands on the spectrum near the given point X reduces only to a renormalization of the effective masses of the electrons. The renormalized mass m for both bands will be assumed to be the same in absolute value and isotropic. The Zeeman Hamiltonian is only slightly modified by σ_z. Thus

$$\mathcal{H}_0+\mathcal{H}_Z=\sum_{\mathbf{k},\alpha}\{[\varepsilon(\mathbf{k})+\alpha J\mu_BH][a_{1\alpha}^+(\mathbf{k})a_{1\alpha}(\mathbf{k})-a_{2\alpha}^+(\mathbf{k})a_{2\alpha}(\mathbf{k})]+\mu_BLHa_{1\alpha}^+(\mathbf{k})a_{2\alpha}(\mathbf{k})+\text{H.c.}\}, \tag{7}$$

where $\varepsilon(k)=\hbar^2k^2/2m-\varepsilon_F$; $L=L_{12}^z$ and L is an imaginary quantity; by J we have designated the g factor. The energy is reckoned from a point halfway between the band extrema, while the subscrips 1 and 2 pertain respectively to the conduction and valence bands.

In the basis (2), the interaction Hamiltonian takes the form

$$\mathcal{H}_{int}=\sum_{\mathbf{k},\mathbf{k}',\mathbf{q}}V(\mathbf{q})a_{1\alpha}^+(\mathbf{k}+\mathbf{q})a_{2\beta}^+(\mathbf{k}'-\mathbf{q})a_{2\beta}(\mathbf{k}')a_{1\alpha}(\mathbf{k}), \tag{8}$$

where $V(\mathbf{q})$ is the Fourier transform of the Coulomb interaction of the electrons. We note that in $\mathcal{H}_{\text{int}}$ we have retained only the interaction of the density–density type, and neglected all others as being inessential for the calculation of this effect, but in the final expression for Δ we shall include them in the effective coupling constant g, since they fix the phase of the order parameter.

The total Hamiltonian of the system takes the form

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_Z+\mathcal{H}_{int}. \tag{9}$$

2. SYSTEM OF BASIC EQUATIONS. EQUATION FOR THE GAP

We consider the situation that arises in the case when the system described by the Hamiltonian (9) is unstable with respect to electron pairing. We shall also assume that the indicated instability is connected only with one of the rays of the vector $\mathbf{k}_0$, i.e., we shall not take into account the interaction of the electrons located at different points X of the Brillouin zone.

Using the standard technique for the time-dependent Green's functions,[11] we can obtain in the momentum representation a system of equations for the electron Green's functions:

$$[\omega-(\varepsilon(\mathbf{k})+\alpha J\mu_BH)]G_{11}^{\alpha\alpha}(\omega,\mathbf{k})-[\mu_BL^*H+\Sigma^{\alpha\alpha}(\mathbf{k})]G_{21}^{\alpha\alpha}(\omega,\mathbf{k})=1,$$
$$[\omega+\varepsilon(\mathbf{k})+\alpha J\mu_BH]G_{21}^{\alpha\alpha}(\omega,\mathbf{k})-[\mu_BLH+\Sigma^{\alpha\alpha*}(\mathbf{k})]G_{11}^{\alpha\alpha}(\omega,\mathbf{k})=0 \tag{10}$$

with the self-consistency condition

$$\Sigma^{\alpha\alpha}(\mathbf{k})=\frac{i}{(2\pi)^4}\int d\omega d\mathbf{q}\, V(\mathbf{k}-\mathbf{q})G_{21}^{\alpha\alpha}(\omega,\mathbf{q}), \tag{11}$$

where $\alpha=1$ or $\alpha=-1$, depending on the spin direction. Solving the system (10), we determine the function $G_{21}^{\alpha\alpha}$, which must be substituted in (11) in order to obtain the self-consistency equation for the gap

$$\Sigma^{\alpha\alpha}(\mathbf{k})=\frac{1}{2(2\pi)^3}\int d\mathbf{q}\, V(\mathbf{k}-\mathbf{q})\frac{\mu_B L\cdot H+\Sigma^{\alpha\alpha}(\mathbf{q})}{\{[\varepsilon(\mathbf{q})+\alpha J\mu_B H]^2+|\mu_B L\cdot H+\Sigma^{\alpha\alpha}(\mathbf{q})|^2\}^{1/2}}. \tag{12}$$

The integral equation (12) cannot be solved in general form. We therefore confine ourselves to the limiting case of a short-range potential. We are interested in states with imaginary order parameter, and therefore assume henceforth that $\Sigma^{\alpha\alpha}$ is imaginary, i.e., it takes the form $i\Sigma^{\alpha\alpha}$, and we shall work with the modulus of this parameter, which we shall also designate by $\Sigma^{\alpha\alpha}$. Subject to these stipulations, Eq. (12) becomes

$$\Sigma^{\alpha\alpha}(\mathbf{k})=\frac{1}{2(2\pi)^3}\int d\mathbf{q}\, V(\mathbf{q}-\mathbf{k})\times\frac{\mu_B|L|H+\Sigma^{\alpha\alpha}(\mathbf{q})}{\{[\varepsilon(\mathbf{q})+\alpha J\mu_B H]^2+[\mu_B|L|H+\Sigma^{\alpha\alpha}(\mathbf{q})]^2\}^{1/2}}. \tag{12a}$$

For a short-range potential we obtain after integration and linearization with respect to μ_B^H, and also taking into account the conditions $\Sigma^{\alpha\alpha}\ll\varepsilon_F$ and $\mu_B H\ll\Sigma^{\alpha\alpha}$,

$$\Sigma^{\alpha\alpha}=gN(0)(\mu_B|L|H+\Sigma^{\alpha\alpha})\ln(2\varepsilon_F/\Sigma^{\alpha\alpha}), \tag{13}$$

where g is the effective but momentum-dependent coupling constant that replaced $V(\mathbf{k}-\mathbf{q})$ and the remaining interaction is not included in $\mathscr{H}_{\text{int}}$, and is equal to zero at momentum transfers $\hbar|\mathbf{q}|>\mathbf{k}_F$; $\Sigma^{\alpha\alpha}$ is likewise independent of the wave vector; $N(0)=2m\mathbf{k}_F/(2\pi)^2\hbar^2$. Next, defining the order parameter Σ in the form

$$\Sigma={}^1/_2(\Sigma^{1,1}+\Sigma^{-1,-1}),$$

we obtain an expression for the modulus of the gap

$$\Sigma=\Sigma_0+\mu_B|L|H \tag{14}$$

or, recognizing that this is a pure imaginary quantity, we can write

$$\Sigma^{im}=i(\Sigma_0+\mu_B|L|H). \tag{14a}$$

where

$$\Sigma_0=2\varepsilon_F\exp(-1/gN(0)). \tag{15}$$

Taking into account the expression for the gap we can write down also the anomalous Green's function

$$G_{21}^{\alpha\alpha}(\mathbf{k},\omega)=i\frac{\Sigma+\mu_B|L|H}{\omega^2-[\varepsilon(\mathbf{k})+\alpha J\mu_n H]^2-(\Sigma+\mu_n|L|H)^2}. \tag{16}$$

3. EXPRESSION FOR THE SUSCEPTIBILITY $T=0$

The orbital-momentum operator is of the form

$$\hat{\mathbf{L}}=-i\hbar[\mathbf{r}\times\nabla], \tag{17}$$

and consequently its matrix elements in the basis chosen by us are imaginary. Since the order parameter is imaginary, the orbital magnetic moment acquires a nonzero mean value

$$M=-i\mu_B|L|\sum_{i\neq j;\mathbf{k}}\operatorname{Sp}G_{ij}(\mathbf{k},-0), \tag{18}$$

where L is given by (6) and $i\neq j$, since the intraband matrix elements are equal to zero. Changing over in (18) to summation over $\mathbf{k}$ to integration, and also substituting the expressions for the Green's functions and summing over i, we get

$$M_L=\mu_B|L|\frac{1}{(2\pi)^3}\int d\mathbf{q}\left(\frac{\Sigma+\mu_n|L|H}{[(\varepsilon(\mathbf{q})+J\mu_B H)^2+(\Sigma+\mu_n|L|H)^2]^{1/2}}+\frac{\Sigma+\mu_n|L|H}{[(\varepsilon(\mathbf{q})-J\mu_B H)^2+(\Sigma+\mu_n|L|H)^2]^{1/2}}\right). \tag{18a}$$

Integrating with respect to q in the same approximation as for the gap, i.e., accurate to terms linear in H, we obtain the expression for the moment

$$M_L=2\mu_B|L|N(0)(\Sigma+\mu_B|L|H)\ln\frac{2\varepsilon_F^2+(\Sigma+\mu_B|L|H)^2}{(\Sigma+\mu_n|L|H)^2}, \tag{19}$$

or, taking into account the expression for Σ,

$$M_L=2\mu_B|L|N(0)(\Sigma_0+2\mu_B|L|H)\ln\frac{2\varepsilon_F^2+\Sigma_0(\Sigma_0+4\mu_B|L|H)}{\Sigma_0(\Sigma_0+4\mu_B|L|H)}. \tag{19a}$$

Having now an expression for the magnetic moment, we obtain the following expression for the magnetic susceptibility, using the connection between the magnetic moment and the susceptibility $\chi_L=\partial M_L/\partial H$ and neglecting the small terms $\Sigma_0^2/\varepsilon_F^2$:

$$\chi_L=4\mu_B^2|L|^2N(0)[\ln(2\varepsilon_F^2/\Sigma_0^2)-2]. \tag{20}$$

We note that χ_L in our approximation $\varepsilon_F/\Sigma_0\gg1$ is always larger than zero, since

$$\ln(2\varepsilon_F^2/\Sigma_0)>2.$$

It is of interest to compare χ_L with the Landau dimagnetic susceptibility

$$\chi_L=-mk_F\mu_B^2/3\pi^2.$$

Since $|L|^2=\hbar^2|\tilde{L}|^2$, it follows that

$$\chi_L/|\chi_L|=6|\tilde{L}|^2[\ln(2\varepsilon_F^2/\Sigma_0^2)-2]. \tag{21}$$

From this we see that $\chi_L>|\chi_L|$, if

$$|L|^2>\{6[\ln(2\varepsilon_F^2/\Sigma_0^2)-2]\}^{-1}. \tag{22}$$

4. ALLOWANCE FOR THE SPIN–ORBIT INTERACTION

We analyze this problem with account taken of the spin–orbit interaction within the framework of the method used in Ref. 8, but instead of the generalized Bloch functions we use the generalized basis of Luttinger and Kohn. The wave eigenfunctions of the zeroth Hamiltonian, which includes the spin–orbit interaction, which does not giving the splitting, have a Kohn form and spinors, i.e.,

$$\psi_{n\mathbf{k}_0\uparrow}=\begin{pmatrix}u_{n\mathbf{k}_0}(\mathbf{r})\\ v_{n\mathbf{k}_0}(\mathbf{r})\end{pmatrix}\exp\{i(\mathbf{k}+\mathbf{k}_0)\mathbf{r}\}. \tag{23}$$

Using the invariance of the Hamiltonian to space inversion I and to time reversal, we have

$$\psi_{n\mathbf{k}_0\downarrow}=KI\psi_{n\mathbf{k}_0\uparrow}, \tag{24}$$

where $K=-i\sigma_y K_0$ is the time reversal operator after Kramers, K_0 is the complex-conjugation operator, and

σ_y is a Pauli matrix. Regarding from the very outset $u_{nk}(\mathbf{r})$ and $v_{nk}(\mathbf{r})$ as real functions, something that can always be done for band extrema, we obtain also an expression for $\psi_{nk\downarrow}$ in terms of these functions:

$$\psi_{nk\downarrow}=\begin{pmatrix}-v_{nk_0}(\mathbf{r})\\ u_{nk_0}(\mathbf{r})\end{pmatrix}\exp\{i(\mathbf{k}-\mathbf{k}_0)\mathbf{r}\}. \tag{25}$$

The matrix elements of the type (6) take the form

$$L_{12}^{\uparrow\uparrow}=-i\hbar\int\{u_{1k_0}(\mathbf{r})[\mathbf{r}\times\nabla_r]_z u_{2k_0}+v_{1k_0}(\mathbf{r})[\mathbf{r}\times\nabla_r]_z v_{2k_0}(\mathbf{r})\}d\mathbf{r}$$
$$=L_{12}^{\downarrow\downarrow}=-L_{21}^{\uparrow\uparrow}=-L_{21}^{\downarrow\downarrow}=L, \tag{26a}$$

$$L_{12}^{\uparrow\downarrow}=-i\hbar\int\{v_{1k_0}(\mathbf{r})[\mathbf{r}\times\nabla_r]_z u_{2k_0}(\mathbf{r})-u_{1k_0}(\mathbf{r})[\mathbf{r}\times\nabla_r]_z v_{2k_0}(\mathbf{r})\}$$
$$\times\exp\{-2i\mathbf{k}_0\mathbf{r}\}d\mathbf{r}=-L_{21}^{\uparrow\downarrow}=-L(-\mathbf{k}_0), \tag{26b}$$

$$L_{12}^{\downarrow\uparrow}=-i\hbar\int\{u_{1k_0}(\mathbf{r})[\mathbf{r}\times\nabla_r]_z v_{2k_0}(\mathbf{r})$$
$$-v_{1k_0}(\mathbf{r})[\mathbf{r}\times\nabla_r]_z u_{2k_0}(\mathbf{r})\}\exp\{2i\mathbf{k}_0\mathbf{r}\}d\mathbf{r}=-L_{21}^{\downarrow\uparrow}=L(\mathbf{k}_0). \tag{26c}$$

Recognizing that as a rule the extremal points are symmetrically arranged in Wigner–Seitz cells, we obtain

$$L(-\mathbf{k}_0)=L(\mathbf{k}_0). \tag{27}$$

In the spinor basis (23), (24) the changes of $\mathcal{H}_0$ reduce only to renormalizations of the constants. The Zeeman Hamiltonian connected with the orbital angular momentum takes the form

$$\mathcal{H}_Z=\sum_{k;\alpha=\beta}\{[\mu_B LH a_{1\alpha}^{+}(\mathbf{k})a_{2\alpha}(\mathbf{k})+\text{H.c.}]$$
$$-\alpha[\mu_B L(\mathbf{k}_0)H a_{1\alpha}^{+}(\mathbf{k})a_{2\beta}(\mathbf{k})-\text{H.c.}]\}. \tag{28}$$

The interaction Hamiltonian in the basis (23) and (24) takes the form indicated in Ref. 8.

Using the technique of time-dependent Green's functions, we can write down in matrix form a system of equations for the matrices $G_{11}^{\alpha\beta}(\mathbf{k},\omega)$ and $G_{21}^{\alpha\beta}(\mathbf{k},\omega)$:

$$\hat{\Omega}_-\hat{G}_{11}=I+\hat{\Delta}\hat{G}_{21},\quad \hat{\Omega}_+\hat{G}_{21}=\hat{\Delta}^{+}\hat{G}_{11}, \tag{29}$$

where

$$\Omega_{(\mp)}=\begin{pmatrix}\omega\mp(\varepsilon(\mathbf{k})+J\mu_B H) & \omega-\varepsilon(\mathbf{k})\\ \omega\mp\varepsilon(\mathbf{k}) & \omega\mp(\varepsilon(\mathbf{k})-J\mu_B H)\end{pmatrix}, \tag{30}$$

I is a unit two-dimensional matrix and $\hat{\Delta}$ is the order-parameter matrix.

We are interested in states with imaginary order-parameter order-parameter matrix. For the moduli of the components of the order-parameter matrix $\hat{\Delta}$ we have the following self-consistency conditions:

$$\Sigma_{11}+\Sigma_{-1,-1}+2\mu_B|L|H=(g_1-g_2+g_2')(\Sigma_{11}-\Sigma_{-1,-1}+2\mu_B|L|H)K$$
$$-2g_2''(\Sigma_{1,-1}-\Sigma_{-1,1}+2\mu_B|\tilde{L}|H)K,$$
$$\Sigma_{11}-\Sigma_{-1,-1}=(g_1-g_2-g_2')(\Sigma_{11}-\Sigma_{-1,-1})M,$$
$$\Sigma_{1,-1}+\Sigma_{-1,1}=(g_1-g_2-g_2')(\Sigma_{1,-1}+\Sigma_{-1,1})M,$$
$$\Sigma_{1,-1}-\Sigma_{-1,1}+2\mu_B|\tilde{L}|H=(g_1-g_2-3g_2')(\Sigma_{1,-1}-\Sigma_{-1,1}+2\mu_B|\tilde{L}|H)K$$
$$-2g_2''(\Sigma_{11}+\Sigma_{-1,-1}+2\mu_B|L|H)K, \tag{31}$$

where

$$K=-i\sum_{k,\omega}\frac{\omega^2-\varepsilon^2(\mathbf{k})}{\text{Det}}-\frac{1}{\text{Det}}[(\Sigma_{11}+\mu_B H|L|)(\Sigma_{-1,-1}+\mu_B H|L|)$$
$$-(\Sigma_{1,-1}+\mu_B|\tilde{L}|H)(\Sigma_{-1,1}-\mu_B|\tilde{L}|H)],$$
$$M=-i\sum_{k,\omega}\frac{\omega^2-\varepsilon^2(\mathbf{k})}{\text{Det}}+\frac{1}{\text{Det}}[(\Sigma_{11}+\mu_B|L|H)(\Sigma_{-1,-1}+\mu_B H|L|)$$
$$-(\Sigma_{1,-1}+\mu_B|\tilde{L}|H)(\Sigma_{-1,1}-\mu_B|\tilde{L}|H)].$$

The symbol Det denotes the expression that determines the spectrum of the single-particle excitations:

$$\text{Det}=[\omega^2-\varepsilon^2(\mathbf{k})]^2-[\omega^2-\varepsilon^2(\mathbf{k})][(\Sigma_{11}+\mu_B|L|H)^2+(\Sigma_{-1,-1}+\mu_B|L|H)^2$$
$$+(\Sigma_{1,-1}+\mu_B|\tilde{L}|H)^2+(\Sigma_{-1,1}-\mu_B|\tilde{L}|H)^2]+[(\Sigma_{11}+\mu_B|L|H)(\Sigma_{-1,-1}$$
$$+\mu_B|L|H)-(\Sigma_{1,-1}+\mu_B|\tilde{L}|H)(\Sigma_{-1,1}-\mu_B|\tilde{L}|H)]^2. \tag{32}$$

We have not written out in the self-consistency equations (31) terms of the type $J\mu_B H$, which, as we have seen in Sec. 2, do not enter in the expression (14) for the gap. We have therefore eliminated them.

In the system (31), the matrix $\hat{\Sigma}$ is connected with the matrix of the order parameter in the following manner:

$$\hat{\Delta}=\hat{\Sigma}+\hat{O}_L,$$

where

$$\hat{O}_L=\begin{pmatrix}\mu_B|L|H & \mu_B|\tilde{L}|H\\ -\mu_B|\tilde{L}|H & \mu_B|L|H\end{pmatrix}.$$

The system (31) reduces to two equations relative to the new unknowns Δ_s and Δ_t, defined by the relations

$$(\Sigma_{11}+\Sigma_{-1,-1}+2\mu_B|L|H)\left[2\left(1+\frac{g_2'}{(g_2'^2+g_2''^2)^{1/2}}\right)\right]^{-1/2}=\Delta_s,$$
$$^1/_2(\Sigma_{11}-\Sigma_{-1,-1})=\Delta_t \tag{33}$$

under the additional condition

$$\Sigma_{1,-1}-\Sigma_{-1,1}+2\mu_B|\tilde{L}|H=-\frac{g_2''}{g_1'+(g_2'^2+g_2''^2)^{1/2}}(\Sigma_{1,1}+\Sigma_{-1,1}+2\mu_B|L|H),$$
$$\Sigma_{1,-1}=-\Sigma_{-1,1}, \tag{34}$$

where g_1 is an interaction constant of the density-density type, g_2 is the unrenormalized interaction connected with the transition of the pair from one band to another, and g_2' and g_2' are unrenormalized interactions connected also with the transition of electrons from one band to another, but now the electron reverses spin direction in the transition, on account of the spin-orbit interaction. The singlet and triplet coupling constants take the form

$$g_s=g_1-g_2-g_2'+2(g_2'^2+g_2''^2)^{1/2},\quad g_t=g_1-g_2-g_2'. \tag{35}$$

The singlet order parameter is connected with the orbital magnetic moment.

We note that the new system of equations with respect to Δ_s and Δ_t coincides with the system obtained in Ref. 6. In the absence of doping and at $H=0$, the energy minimum corresponds to the following solutions of the new system:

$$\Delta_t=0,\quad \Delta_s=\Delta_{s0}=2\varepsilon_F\exp[-1/g_s N(0)], \tag{36}$$

or

$$\Delta_s=0,\quad \Delta_t=\Delta_{t0}=2\varepsilon_F\exp[-1/g_t N(0)]. \tag{37}$$

The solution (36) is more convenient, as was noted in Ref. 8 and as is seen from relations (35). Performing the same calculations as in the preceding section, we obtain for the orbital moment the following expression:

$$M_L=2\mu_B N(0)|L|(\tilde{\Delta}_{s0}+2\mu_B|L|H)\ln\frac{2\varepsilon_F^2+\tilde{\Delta}_{s0}(\tilde{\Delta}_{s0}+4\mu_B|L|H)}{\tilde{\Delta}_{s0}(\tilde{\Delta}_{s0}+4\mu_B|\tilde{L}|H)}, \tag{38}$$

where

$$\tilde{\Delta}_{s0}=\frac{\Delta_{s0}}{2}\left[2\left(1+\frac{g_2'}{(g_2'+g_2'')^{1/2}}\right)\right]^{1/2}. \tag{39}$$

As seen from (38), the influence of the spin-orbit interaction reduces to a renormalization of the order param-

eter, and the form remains the same as in (19) and (19a). In this case, too, M_L is determined only by $|L|$, i.e., by the interband matrix element of the orbital moment diagonal in the spin.

We obtain similarly an expression for the susceptibility:

$$\chi_L = 4\mu_B^2 |L|^2 \left[\ln \frac{2\varepsilon_F^2}{\tilde{\Delta}_{s0}^2} - 2 \right], \tag{40}$$

where $\tilde{\Delta}_{s0}$ is determined by (39) and (36).

5. SUSCEPTIBILITY AT FINITE TEMPERATURES

In the preceding section we have seen that the spin-orbit interaction does not lead to new qualitative changes of the orbital ferromagnetism obtained neglecting this interaction. Therefore the calculation of the magnetic susceptibility at finite temperatures will be carried out within the framework of a simple interaction Hamiltonian of the density-density type for carriers from different bands, neglecting the spin-orbit interaction, i.e., in analogy with the calculation carried out in Sec. 2, taking into account the same remarks made with respect to the interaction of the type $a_1^+ a_1^+ a_2 a_2$. In this case, to describe this effect we use the temperature Green's functions[11]

$$\begin{aligned} G_{11}^{\alpha\beta}(x, x') &= \langle T\tilde{\psi}_{1\alpha}(x)\tilde{\psi}_{1\beta}^+(x')\rangle, \\ G_{21}^{\alpha\beta}(x, x') &= \langle T\tilde{\psi}_{2\alpha}(x)\tilde{\psi}_{1\beta}^+(x')\rangle, \end{aligned} \tag{41}$$

where $x \equiv \mathbf{r}, \tau$. Here $\tilde{\psi}_{1\alpha}(x)$ and $\tilde{\psi}_{2\alpha}(x)$ are operators expressed in the Heisenberg representation

$$\begin{aligned} \psi_{1\alpha}(\mathbf{r}) &= \sum_{\mathbf{k}} u_{1\mathbf{k}_0}(\mathbf{r}) \exp\{i(\mathbf{k}+\mathbf{k}_0)\mathbf{r}\} a_{1\alpha}(\mathbf{k}), \\ \psi_{2\alpha}(\mathbf{r}) &= \sum_{\mathbf{k}} u_{2\mathbf{k}_0}(\mathbf{r}) \exp\{i(\mathbf{k}+\mathbf{k}_0)\mathbf{r}\} a_{2\alpha}(\mathbf{k}). \end{aligned} \tag{42}$$

From the equations of motion for the operators $\tilde{\psi}_{1\alpha}$ and $\tilde{\psi}_{2\alpha}$, we obtain, after taking Fourier transforms with respect to the coordinates $\mathbf{r}$ and the imaginary time τ, the following system of equations for the Green's functions:

$$\begin{aligned} &[i\omega_n - (\varepsilon(\mathbf{k}) + \alpha J\mu_B H)] G_{11}^{\alpha\alpha}(\omega_n, \mathbf{k}) - [\mu_B L^* H + \Sigma^{\alpha\alpha}(\mathbf{k})] G_{21}^{\alpha\alpha}(\omega_n, \mathbf{k}) = 1, \\ &[i\omega_n + \varepsilon(\mathbf{k}) + \alpha J\mu_B H] G_{21}^{\alpha\alpha}(\omega_n, \mathbf{k}) - [\mu_B L H + \Sigma^{\alpha\alpha *}(\mathbf{k})] G_{11}^{\alpha\alpha}(\omega_n, \mathbf{k}) = 0 \end{aligned} \tag{43}$$

with the self-consistency condition

$$\Sigma^{\alpha\alpha}(\mathbf{k}) = \frac{iT}{(2\pi)^3} \sum_n \int d\mathbf{q}\, V(\mathbf{k}-\mathbf{q}) G_{21}^{\alpha\alpha}(\omega_n, \mathbf{q}), \tag{44}$$

where $\omega_n = (2n+1)\pi T$; $n = 0, \pm 1, \pm 2, \ldots$, and T is the temperature.

We are interested in states with imaginary order parameter, and we therefore obtain for the modulus of this parameter from (43) and (44)

$$\Sigma^{\alpha\alpha}(\mathbf{k}) = \frac{iT}{(2\pi)^3} \sum_n \int d\mathbf{q} \frac{V(\mathbf{k}-\mathbf{q})(\mu_B |L| H + \Sigma^{\alpha\alpha}(\mathbf{q}))}{\omega_n^2 - (\varepsilon(\mathbf{q}) + \alpha J\mu_B H)^2 - (\mu_B |L| H + \Sigma^{\alpha\alpha}(\mathbf{q}))^2}.$$

In the short-range-potential approximation and after summation over n, we get the following equation for Σ:

$$\Sigma = gN(0)(\mu_B |L| H + \Sigma) \int_0^{\varepsilon_F} \frac{1 - 2n(\varepsilon, T)}{[\varepsilon^2 + (\mu_B |L| H + \Sigma)^2]^{1/2}} d\varepsilon, \tag{45}$$

where Σ was defined earlier in terms of $\Sigma^{\alpha\alpha}$, and

$$n(\varepsilon, T) = \left[\exp\left\{ \frac{[\varepsilon^2 + (\mu_B |L| H + \Sigma)^2]^{1/2}}{T} \right\} + 1 \right]^{-1}. \tag{46}$$

It is known that analytic integration of (45) is impossible, all that can be obtained is the behavior of the gap in the spectrum at $T \ll T_c$ and at $(T_c - T)/T_c \ll 1$. In the case of low temperatures, $T \ll T_c$, we can obtain an asymptotic formula for $\Sigma = \Sigma(T)$ at $\Sigma \gg T$, similar to the formula for superconductors[11]:

$$\begin{aligned} \Sigma(T) = \Sigma_0 + \mu_B |L| H - [2\pi T(\Sigma_0 + \mu_B |L| H)]^{1/2} \\ \times \left[1 - \frac{T}{8(\Sigma_0 + \mu_B |L| H)} \right] \exp\left[-\frac{\Sigma_0 + \mu_B |L| H}{T} \right], \end{aligned} \tag{47}$$

where Σ_0 is defined in (15).

Substituting (47) in (19) and neglecting the temperature correction under the logarithm sign as inessential, we obtain

$$\begin{aligned} M_L(T) = 2\mu_B |L| N(0) \Big\{ \Sigma_0 + 2\mu_B |L| H - [2\pi T(\Sigma_0 + \mu_B |L| H)]^{1/2} \\ \times \left[1 - \frac{T}{8(\Sigma_0 + \mu_B |L| H)} \right] \exp\left[-\frac{\Sigma_0 + \mu_B |L| H}{T} \right] \\ \times \ln \frac{2\varepsilon_F^2 + \Sigma_0(\Sigma_0 + 4\mu_B |L| H)}{\Sigma_0(\Sigma_0 + 4\mu_B |L| H)} \Big\}. \end{aligned} \tag{48}$$

From this we get the temperature dependence of the susceptibility at low temperatures:

$$\begin{aligned} \chi_L(T) = \chi_L(T=0) + 4\mu_B^2 |L|^2 N(0) \exp(-\Sigma_0/T) \\ \times \left\{ \left[\left(\frac{\pi\Sigma_0}{2T} \right)^{1/2} - \frac{5}{8} \left(\frac{\pi T}{2\Sigma_0} \right)^{1/2} - \frac{1}{16} \left(\frac{\pi T^3}{2\Sigma_0^3} \right)^{1/2} \right] \ln \frac{2\varepsilon_F^2}{\Sigma_0^2} \right. \\ \left. + 2 \left(\frac{2\pi T}{\Sigma_0} \right)^{1/2} \left(1 - \frac{T}{8\Sigma_0} \right) \right\}. \end{aligned} \tag{49}$$

Since $\Sigma_0 \gg T$, the more important term in (49) for the T-dependent part is $(\pi\Sigma_0/2T)^{1/2}$. Retaining only this term, we get

$$\chi_L(T) = \chi_L(T=0) + 4\mu_B^2 |L|^2 N(0) \ln(2\varepsilon_F^2/\Sigma_0^2) \exp(-\Sigma_0/T) (\pi\Sigma_0/2T)^{1/2}. \tag{49a}$$

From the expression for $\chi_L(T)$ we can see that at small T the susceptibility increases with increasing temperature, since $d\chi_L/dT > 0$ at $T \ll \Sigma_0$.

Neglecting the influence of the field on the transition temperature T_c, we can determine T_c in standard fashion from the equation for the gap (4)

$$T_c = \frac{\gamma}{\pi} \Sigma_0 = \frac{2\gamma}{\pi} \varepsilon_F \exp\left(-\frac{1}{gN(0)} \right), \tag{50}$$

where $\gamma = C = 0.577$.

Near T_c the gap is small, therefore we can expand in (45) in powers of $(\Sigma + \mu_B |L| H)^2/T^2$; as a result we get the following expression for the gap:

$$\begin{aligned} \Sigma(T, H) = gN(0)(\Sigma(T, H) + \mu_B |L| H) \\ \times \left[\ln \frac{2\varepsilon_F \gamma}{\pi T} - \frac{(\Sigma + \mu_B |L| H)^2}{\pi T^2} \frac{7\zeta(3)}{8} \right], \end{aligned} \tag{51}$$

where $\zeta(3)$ is the Riemann zeta function.

We obtain a third-order equation for Σ. It can be solved to yield an exact expression for Σ, but within the framework of our approximations we can confine ourselves to substituting in the right-hand side of (51) the expression for Σ at $H = 0$

$$\Sigma(T, H=0)=\pi T_c[8/7\zeta(3)]^{1/2}(1-T/T_c)^{1/2}. \quad (52)$$

Using the same expansion in powers of $[\Sigma(T, H)+\mu_B|L|H]^2/T^2$, we easily obtain an expression for the magnetic moment:

$$M_L(T)=2\mu_B|L|N(0)\Sigma(T, H). \quad (53)$$

Substituting in (53) Σ from (51), we see that with increasing T the moment decreases. At $T>T_c$ we have

$$M_L(T)=2\mu_B^2|L|^2H\frac{\ln(2\varepsilon_F\gamma/\pi T)}{\ln(2\varepsilon_F\gamma/\pi T_c)}, \quad (54)$$

from which we see that the magnetic moment continues to decrease.

From the expression for the moment we easily obtain the susceptibility of the system:

$$\chi_L=\frac{2\mu_B^2|L|^2N(0)T^2T_c\ln(2\varepsilon_F\gamma/\pi T)}{(T^2+3\pi T_c)(T_c-T)}. \quad (55)$$

As expected, at the transition point the system susceptibility becomes infinite. The obtained magnetic susceptibility also satisfied the Curie–Weiss law.

6. CONCLUSION

Thus, in a system with direct bands, in which only transitions with change of the orbital angular momentum L are allowed, dielectric pairing in a state with imaginary order parameter Σ results in ferromagnetic ordering of the valence electrons of the completely filled bands. It should be recalled that in systems with imaginary parameter, for allowed interband dipole transitions, states with spontaneous currents are produced.[7]

Even though the crystal field quenches the intraband orbital angular momentum, an uncompensated magnetic moment is produced in a system with imaginary order parameters, in proportion to the interband transitions with respect to the angular momentum. It must be emphasized that this ferromagnetism arised in systems with completely filled bands, i.e., the ferromagnetic ordering arises even in undoped semiconductors (semimetals), in contrast to excitonic ferromagnetism.

We can compare the obtained magnetic moment with the saturation moment $M_{sat}\sim\mu_BN(0)\varepsilon_p$, where ε_p is the width of the filled band:

$$\frac{M_L}{M_{sat}}\sim\frac{\Sigma_0\ln(2\varepsilon_F^2/\Sigma_0^2)}{\varepsilon_p}.$$

We see therefore that the ferromagnetism of the electrons of the filled band can exceed the saturation magnetism in systems with narrow filled bands. It must be emphasized that we have neglected the spin splitting of the bands because of the smallness of this effect compared with the obtained effect.

We note in conclusion that suitable materials in which ferromagnetic ordering of valence electrons of completely filled bands can occur may be systems with narrow forbidden bands having low symmetry at the external Brillouin points. It appears that they include also solid solutions (see Refs. 12 and 13).

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