The dielectric constant of magnetic semiconductors

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The magnetization dependence of the dielectric constant $e(\omega,q)$ of a ferromagnetic semiconductor is calculated. Since the semiconductor gap narrows rapidly with increasing magnetization, $\varepsilon(\omega,q)$ may increase considerably as the temperature is lowered or on the application of a strong enough external magnetic field. This entails a number of physical consequences (the possibility of an insulator-metal phase transition in donor-doped crystals, etc.). The light absorption coefficient in the spin-wave region is calculated. The temperature dependence of the absorption coefficient indicates that an electron that appears in the conduction band as a result of interaction with light is not bare, but is dressed because of its interaction with magnons. Thus, the Franck-Condon principle, according to which there should be no change in the state of the magnetic subsystem when an electron goes from the valence band to the conduction band, turns out to be violated in this case.

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INTRODUCTION

Experiment shows that the width of the energy gap of a ferromagnetic semiconductor depends strongly oh the temperature and the magnetic field strength. On approaching the region in which ferromagnetic order becomes established from the high-temperature side, the gap width decresses by some 0.2-0.5 eV. In many cases the decrease in the gap width Eg is comparable with its high-temperature value. In HgCr₂Se₄ for example, the gap width decreases by a factor of three as the temperature is lowered.^{1,2} This suggests that the electron part of the dielectric constant of a ferromagnetic semiconductor, which is determined by the gap width, should also depend strongly on the temperature and magnetic field strength. Such a dependence could lead to interesting physical phenomena.

Although the situation is fairly clear in a qualitative way, the question of how to carry through a correct calculation of the temperature dependent deilectric constant of a ferromagnetic semiconductor arises. For a magnetic semiconductor, the electron part of the polarizability is given, as is well known, by the expression

$$\alpha_{e}(\omega,q) \approx \frac{2e^{2}}{q^{2}} \sum_{\mathbf{k}(\pm)} \frac{\left| \langle \mathbf{k}, 1 | e^{iqr} | \mathbf{k} + q, 1 \rangle \right|^{2}}{E_{1,k} - E_{2,k+q} \pm \omega},\tag{I}$$

where **k** is the quasimomentum and the indices 1 and 2 refer to the conduction band and the valence band, respectively. To estimate $\alpha_e(0,0)$ it may be assumed that the energy denominator in (I) may be set equal to the gap width E_g and that the square of the modulus of the interband matrix element will be inversely proportional to E_g . As a result, when $\omega = 0$ and q = 0 expression (I) is approximated by $(\omega_g / E_g)^2$, where ω_q , is the plasma frequency of the valence electrons (see, e.g., Ref. 3).

The temperature dependence of the spectrum of the charge carriers in ferromagnetic semiconductors is due to the interaction of the conduction electrons with magnons. At finite temperatures the charge carrier, a "spin poiaron",² is a quasiparticle of a more complex type than the ordinary band electron; a cloud of localized f- or d-spin deviations moves through the crystal together with the electron. At first glance it would seem that in treating the case under

considertion it would be sufficient to replace the electron energies in CD by spin-polaron energies, thus renormalizing the interband matrix element. In other words, it would seem that the static polarizability of a ferromagnetic semiconductor should be inversely proportional to $E_g^2(T)$.

There is actually no basis for such a conclusion, for the real part of the dielectric constant, unlike the imaginary part, is due to virtual electron transitions, rather than to real ones, and the real and virtual transitions do not necessarily lead to states of the same type. The analysis presented below shows that the temperature dependence of $\varepsilon(\theta, q)$ is actually much more complicated.

We shall not present the expressions obtained for $\varepsilon(\omega, q)$ by treating the problem in its most general form because of their complexity, but shall discuss only two important special cases: In Sec. 1 we take account of the renormalization of both the spectrum and the interband matrix element due to a change in the magnetization of the crystal, but obtain an expression for $\varepsilon(\theta, q)$ only for the limiting cases $T=\theta$ and $T \rightarrow \infty$ In Sec. 2 we calculate $\varepsilon(\omega, q)$ throughout the entire spin-wave region and at $T = \infty$ for the case in which the renormalization of the matrix element can be neglected. ¹⁾ Finally, in Sec. 3 we obtain an expression for the dielectric constant of a semiconductor with helicoidal magnetic ordering at $\Phi = \theta$. The physical consequences of the results obtained in the paper are analyzed in Sec. 4.

1. STATIC DIELECTRIC CONSTANT OF A FERROMAGNETIC SEMICONDUCTOR IN THE TWO-BAND MODEL

To obtain the most general idea of the nature of the change in the static dielectric constant of a crystal when magnetic ordering becomes established in it, we shall examine a model that makes it possible to take account of the rxx renormalization of both the energy denominator and the interband matrix element in an expression such as (I) at the same time.

In the model examined below the crystal is assumed to consist of magnetic cations and nonmagnetic anions. The conduction band consists mainly of s orbits on cations, and the valence band, of p orbits on anions, while the degree of hybridization of the cation and anion states depends on the magnetization of the crystal. A similar assertion holds for the valence band too. To calculate the magnetization dependence of the dielectric constant, we must therefore use the two-band model of a magnetic semiconductor (as far as we know, this model has never been used before in the theory of magnetic semiconductors).

For definiteness we shall consider a crystal which has a structure of the NaCl type and consists of doubly charged cations and anions. For simplicity we do not take account of the spatial degeneracy of the anion states (i.e., we attribute s orbits rather than ρ orbits to the anions). We shall assume that the exchange energy between the conduction electrons and the localized f spins of the cations is small as compared with the width W of the conduction band (the exchange of holes with f spins is much weaker than that of the conduction electrons, since they move mainly along the nonmagnetic anions).

In this section we shall examine the electron states for the two limiting cases in which T=0, and $T=\infty$. In the first case the Hamiltonian for the crystal can be written in the form

$$H = B_c \sum_{g,\Delta_1} a_{g\sigma}^+ a_{g+\Delta_1,\sigma} + B_a \sum_{\mathbf{f},\Delta_1} a_{\mathbf{f}\sigma}^+ a_{\mathbf{f}+\Delta_1,\sigma} + E_{c\sigma} \sum_g a_{g\sigma}^+ a_{g\sigma} + \sum_{\mathbf{f}} E_a a_{\mathbf{f}\sigma}^+ a_{\mathbf{f}\sigma} + B_{ca} \sum_{g,\Delta_2} a_{g\sigma}^+ a_{g+\Delta_2,\sigma} + B_{ca} \sum_{\mathbf{f},\Delta_2} a_{\mathbf{f}\sigma}^+ a_{\mathbf{f}+\Delta_2,\sigma},$$
(1)

where $a_{g\sigma}^{+}$ and $a_{g\sigma}(a_{f\sigma}^{+}$ and $a_{f\sigma})$ are the creation and destruction operators for an electron with spin projection y on the magnetic cation (nonmagnetic anion) specified by the index g(f); B_c , B_a ., and B_{ca} are the cation-cation, anion-anion, and cation-anion Bloch integrals, respectively; E_c (E_a) is the energy of an electron localized on a cation (anion); $E_{c\sigma} = E_c - AS\sigma$, A is the s-f exchange integral; **S** is the spin of the unfilled f shell of a magnetic cation; the index Δ_1 enumerates the nearest neighbor cations (anions) to a given cation (anion); and Δ_2 enumerates the nearest neighbor cations (anions) to a given anion (cation).

On transforming the electron operators in (1) from the lattice representation to the momentum representation, we obtain the following Hamiltonian from (1):

$$H = \sum E_{c\mathbf{k}\sigma} a_{c\mathbf{k}\sigma}^{+} a_{c\mathbf{k}\sigma} + \sum E_{c\mathbf{k}} a_{c\mathbf{k}\sigma}^{+} a_{c\mathbf{k}\sigma} + Z_{1} B_{ce} \sum \gamma_{\mathbf{k}}^{ce} \left(a_{c\mathbf{k}\sigma}^{+} a_{e\mathbf{k}\sigma} + a_{e\mathbf{k}\sigma}^{+} a_{c\mathbf{k}\sigma} \right),$$

$$E_{c\mathbf{k}\sigma} = E_{c\sigma} + Z_{1} B_{c} \gamma_{\mathbf{k}}, \quad E_{e\mathbf{k}} = E_{e} + Z_{1} B_{e} \gamma_{\mathbf{k}},$$

$$\gamma_{\mathbf{k}} = \frac{1}{Z_{1}} \sum_{\Delta_{1}} e^{i\mathbf{k}\Delta_{1}}, \quad \gamma_{\mathbf{k}}^{ce} = \frac{1}{Z_{2}} \sum_{\Delta_{2}} e^{i\mathbf{k}\Delta_{2}},$$
(2)

where $Z_1(Z_2)$ is the number of nearest neighbor cations to a cation (anion). For a lattice of the NaCl type we have

$$\gamma_{\mathbf{k}} = \cos k_x a + \cos k_y a + \cos k_z a$$

and

$$\gamma_{\mathbf{k}}^{ce} = \cos k_x \frac{a}{2} \cos k_y \frac{a}{2} \cos k_z \frac{a}{2}$$

where a is the shortest distance between like ions. With the aid of a canonical transformation to the new operators

$$c_{i\mathbf{k}\sigma} = x_{i\mathbf{k}\sigma}a_{c\mathbf{k}\sigma} + y_{i\mathbf{k}\sigma}a_{c\mathbf{k}\sigma}$$

we can diagonalize the quadratic form (2) and obtain

$$H = \sum \left\{ E_{1k\sigma} c_{1k\sigma}^{+} c_{1k\sigma} + E_{2k\sigma} c_{2k\sigma}^{+} c_{2k\sigma} \right\},$$

$$E_{ik\sigma} = \frac{1}{2} \left(E_{ck\sigma} + E_{ak\sigma} \right) - \left(-1 \right)^{i} \left[\frac{1}{4} \left(E_{ck\sigma} - E_{ak} \right)^{2} + \left(Z_{2} B_{ca} \gamma_{k}^{ca} \right)^{2} \right]^{1/2},$$

$$x_{1k\sigma} = y_{1k\sigma} = \left[1 + \frac{\left(Z_{2} B_{ca} \gamma_{k}^{ca} \right)^{2}}{\left(E_{1k\sigma} - E_{ak} \right)^{2}} \right]^{-1/2}, \qquad y_{1k\sigma} = -x_{2k\sigma} = x_{1k\sigma} \frac{Z_{2} B_{ca} \gamma_{k}^{ca}}{E_{1k\sigma} - E_{ak}},$$
(3)

(i = 1 corresponds to the conduction band, and i = 2, to the valence band). In the adopted model, the only Fourier components of the density operator that do not vanish in the dipole approximation will be those that contain the product of two atomic wave functions of which one belongs to a cation and the other to an anion. In view of this, the Fourier transform p_q of the density operator takes the form

$$\rho_{q} = \sum_{ij\mathbf{k}\sigma} B(i\mathbf{k}\sigma; j, \mathbf{k} - q, \sigma) c^{\dagger}_{i\mathbf{k}\sigma} c_{j,\mathbf{k}-q,\sigma},$$

$$B(i\mathbf{k}\sigma; j, \mathbf{k} - q, \sigma) = \int \varphi_{c\mathbf{k}} e^{iqr} \varphi_{c\mathbf{k}-q} d^{3}r y_{i\mathbf{k}\sigma} x_{j,\mathbf{k}-q,\sigma} + \int \varphi_{c\mathbf{k}} e^{iqr} \varphi_{c\mathbf{k}-q} d^{3}r y_{i,\mathbf{k}-q,\sigma} x_{j\mathbf{k}\sigma}, \qquad (4)$$

$$\varphi_{c\mathbf{k}}(r) = \frac{1}{N^{1/2}} \sum_{g} e^{-ig\mathbf{k}} \varphi_{g}(r), \qquad \varphi_{a\mathbf{k}} = \frac{1}{N^{1/2}} \sum_{f} e^{-if\mathbf{k}} \varphi_{f}(r),$$

where N is the number of unit cells, and φ_g and φ_f are atomic wave functions for the cations and anions, respectively. Taking Eqs. (3) and (4) into account in a standard calculation of the static dielectric constant leads to the expression

$$\varepsilon(0,q) = \varepsilon_L + \frac{4\pi e^2}{q^2} \sum_{\mathbf{k},\mathbf{k}',i,j,\sigma} \frac{\left|B\left(i\mathbf{k}\sigma;j\mathbf{k}'\sigma\right)\right|^2 \left(x_{i\mathbf{k}\sigma}^2 - x_{j\mathbf{k}'\sigma}^2\right)}{E_{i\mathbf{k}'\sigma} - E_{j\mathbf{k}\sigma}}.$$
(5)

Here ε_L is the lattice part of the dielectric constant and is assumed to be known. Taking account of the fact that $\langle \mathbf{g} | \mathbf{r} | \mathbf{g} + \Delta_2 \rangle = -\langle \mathbf{g} | \mathbf{r} | \mathbf{g} - \Delta_2 \rangle$, we obtain the following expression for the matrix element B(ij) that occurs in (5) in the long-wave limit $(q \rightarrow 0)$:

$$B(\mathbf{lk}, 2\mathbf{k}) = B(2\mathbf{k}, \mathbf{lk}) = iqZ_2 l\Gamma_{\mathbf{k}} (y_{\mathbf{lk}} x_{2\mathbf{k}} + y_{2\mathbf{k}} x_{\mathbf{lk}}),$$

$$l = |\langle g | r | g + \Delta_2 \rangle|, \quad \Gamma_{\mathbf{k}} = \sin k_x \frac{a}{2} \sin k_y \frac{a}{2} \sin k_z \frac{a}{2}.$$
 (6)

As follows from (3) and (4), the diagonal elements of the matrix B(ij) tend to zero more rapidly as $q \rightarrow 0$ than do the off-diagonal ones. Thus, we obtain the following expression for the dielectric constant from Eqs. (3)-(6):

$$\varepsilon_{o}\left(T=0\right) = \varepsilon\left(0,0\right) = 8\pi e^{2} \sum \frac{\left(Zl\Gamma_{\mathbf{k}}\right)^{2}}{E_{1\mathbf{k}\sigma} - E_{2\mathbf{k}\sigma}} \left(\frac{\left(E_{1\mathbf{k}\sigma} - E_{a\mathbf{k}}\right)^{2} - \left(Z_{2}B_{ca}\gamma_{\mathbf{k}}^{ca}\right)^{2}}{\left(E_{1\mathbf{k}\sigma} - E_{a\mathbf{k}}\right)^{2} + \left(Z_{2}B_{ca}\gamma_{\mathbf{k}}^{ca}\right)^{2}}\right) + \varepsilon_{L}.$$
(7)

The expression for $\varepsilon_0(T = \infty)$, the dielectric constant at $T = \infty$, may be obtained from (7) by setting AS = 0 in it we shall consider only the case in which both the top of the valence band and the bottom of the conduction band lie at the point

$$Q = (\pi/2, \pi/2, \pi/2)$$

when $E_{ck\sigma}$ - $E_{ak} \ge Z_2 B_{ca}$ in (7) we can carry through an expansion in the quantities

$$Z_2 B_{ca} \gamma_{\mathbf{k}}^{ca} / (E_{c\mathbf{k}\sigma} - E_{a\mathbf{k}}).$$

This makes it possible to estimate the change in the dielectric constant when ferromagnetic order is established in the crystal. Taking (3) and (6) into account, assuming that $\varepsilon_L <<\varepsilon_0(\infty)$, and setting $AS/E_g = 1/2$ and $B_{ca} \approx E_g = E_{c\theta} - E_{a\varrho}$, we find from (7) that the relative change η in the dielectric constant when ferro-magnetic order is established amounts to 40%. If we assume that $AS/E_g = 4/3$, however (in this case the gap width decreases by a factor of almost three as in $HgCr_2Se_2$), we find that the dielectric constant is several times larger. In this case, however, $AS/W \sim I$, and the estimate of η obtained under the assumption that AS << W is no longer accurate. Under less favorable conditions, in EuO, for example, the dielectric constant should change by only a few percent.

2. THE DIELECTRIC CONSTANT WITH ALLOWANCE FOR SPACE AND TIME DISPERSION IN THE ONE-BAND MODEL

Let us examine the dielectric constant in detail for the case in which the interband coordinate matrix elements are independent of the magnetization, e.g. when B_{ca} in (1) is negligible. Taking account of the limitations mentioned above, we can express the total Hamiltonian for the system under consideration within the limitations of the s-f model in the form

$$\begin{split} H &= H_0 + H_1 + H_2 + H_{el-el} \\ H_0 &= \sum \omega_q b_q^+ b_q + \sum_{i\mathbf{k}\sigma} E_{i\mathbf{k}\sigma} a_{i\mathbf{k}\sigma}^+ a_{i\mathbf{k}\sigma} \\ H_1 &= -A \bigg(\frac{S}{2N} \bigg)^{1/2} \sum_{\mathbf{k},q} \Big(a_{1\mathbf{k}\uparrow}^+ a_{1\mathbf{k}+q\downarrow} b_q^+ + a_{1\mathbf{k}\downarrow}^+ a_{1\mathbf{k}-q\uparrow} b_q \Big), \\ H_2 &= \frac{A}{2N} \sum_{\mathbf{k},q,r} \Big(a_{1\mathbf{k}\uparrow}^+ a_{1\mathbf{k}-r\uparrow} - a_{1\mathbf{k}\downarrow}^+ a_{1\mathbf{k}-r\downarrow} \Big) b_q^+ b_{q+r}, \\ H_{el-el} &= 2\pi e^2 \sum_{q\neq 0} \frac{\rho_q^+ \rho_q}{q^2}, \end{split}$$

Here b_q^+ and b_q , are the magnon creation and destruction operators. We use Bloch functions for the one-electron wave functions:

$$\varphi_{i\mathbf{k}}\left(r\right) = u_{i\mathbf{k}}\left(r\right)e^{i\mathbf{k}r}.$$
(9)

For definiteness, the s~f exchange integral A is assumed to be positive. Only interband terms that are essential for the problem are included in the electron-electron interaction Hamiltonian H_{el-el} .

The electron part of the dielectric constant is calculated for wide bands (AS/W << 1) to the first order in 1/S, where S is the magnitude of the f spin and is assumed to be large. The calculation in the random phase approximation can be reduced by standard techniques to the summation of polarization graphs as described by the equation

in which the smooth lines represent electron Green's functions-the upper ones are for the conduction band and the lower ones, for the valence band, while the fine lines represent the bare, and the heavy ones, the renormalized functions; the wavy lines represent magnon Green's functions; and the single and double dashes represent one- and two-magnon interactions, respectively. As a result, we obtain the following expression for the dielectric constant from Eqs. (8)-(10):

$$\varepsilon(\omega,q) = \varepsilon_{L}(\omega,q) + \frac{4\pi e^{2}}{q^{2}} \sum_{\mathbf{k}',\sigma} \left\langle 2\mathbf{k}' \left| e^{-iqr} \right| \mathbf{l}\mathbf{k} \right\rangle^{2} \\ \times \left\{ \frac{1}{\omega + E_{\mathbf{l}\mathbf{k}'\sigma}(-\omega,q) - E_{2\mathbf{k}} + i\delta} - \frac{1}{\omega + E_{2\mathbf{k}} - E_{\mathbf{l}\mathbf{k}'\sigma}(\omega,q) + i\delta} \right\},$$

$$\mathbf{k}' = k + q$$
(11)

Here $\varepsilon_L(\alpha, q)$ is the lattice part of the dielectric constant, which is assumed to be known, and E is given by

$$E_{\mathbf{lk}\sigma}(\omega,q) = E_{\mathbf{lk}\sigma} + \frac{A\sigma}{N} \sum_{p} m_{p} - \frac{A^{2}S}{2N} \sum_{p} \frac{m_{p}\delta_{\sigma\uparrow} + (m_{p}+1)\delta_{\sigma\downarrow}}{E_{\mathbf{lk}+p,-\sigma} - E_{2,\mathbf{k}-q} - \omega - i\delta},$$
(12)

in which m_p is the magnon distribution function. The magnon frequencies are omitted from the energy denominators in the last sum in (12) because they are small as compared with the electron energies.

First we shall discuss the absorption of light. As in Sec. I, we shall assume that the bottom of the conduction and the top of the valence bands lie at the same wave vector Q. It follows from (11) and (12) in the first order in 1/5 that near the absorption edge, light of frequency ω can stimulate optical transitions between valence-band and conduction-band states with small **k**, whose energies satisfy the condition

$$\omega = E_{1\mathbf{k}\uparrow}^{\bullet} - E_{2\mathbf{k}-q}, \quad E_{1\mathbf{k}\uparrow}^{\bullet} = E_{1\mathbf{k}\uparrow} + \frac{A}{2N} \sum_{q'} \frac{\left(E_{1\mathbf{k}+q'} - E_{1\mathbf{k}}\right) m_{q'}}{AS + E_{i\mathbf{k}+q'} - E_{1\mathbf{k}}}.$$
(13)

Expression (13) agrees precisely with the energies of spin-polaron states⁷. Under the action of light, therefore, an electron can actually make a transition from the valence band directly to a spin-polaron state. As the temperature rises, the absorption edge $E_g = E^*_{1,0}$, $-E_{2,0}$ shifts upwards, the shift being at first proportional to $T^{5/2}$, and then [when $T > T_0 = ISa^2Q^2_0$ where $Q_0 = (m^*AS)^{1/2}$,, I is the Heisenberg exchange integral, and m^* is the effective electron mass] to $T^{5/2}$.

According to (11) and (12), the absorption coefficient near the optical absorption edge at frequency $E_g + \Delta \omega$ is proportional to the density of spin-polaron levels of energy $\Delta \omega$, i.e., to $\mathbf{m} *_{\mathbf{qp}} {}^{3/2}(\Delta \omega))^{1/2}$, where the effective mass of the spin polaron is related to that of the electron by the formula:

$$m_{sp}^{\bullet} = m^{\bullet} \left(1 + \frac{1}{2NS} \sum \left(\gamma_q - 1 \right) m_q \right)^{-1}.$$

When $\omega >> E_g$, the last term in (12) is of order *AS/W* with respect to the preceding term and may therefore be neglected. Physically, this means that when the electron momenta **k** are large enough the electrons must undergo transitions with fixed f spins, i. e., in the final states the spins of the atoms are not adjusted to the electron and no spin-polaron state is realized. This is essentially a manifestation of the Franck-Condon principle; in this case the electron transitions are "fast." Actually, one must take account of the fact that according to (12) the electron states turn out to be damped when $[2m^*(\omega \cdot E_{1,0}\downarrow + E_{2,0})]^{1/2} \ge \kappa$, i. e. when the absorption of a magnon by an electron from the lower subband and its transition to the upper subband becomes possible. (At T=0 such processes are forbidden by conservation of the total angular momentum of the system. Transitions from the upper subband to the lower one with magnon emission according to (12) are possible even when $\Phi = 0$.) In this case the absorption coefficient is proportional to the level density of the damped electron states. The frequency dependence of the absorption coefficient becomes different from the radical type and represents a monotonically increasing function of $\Delta \omega$.

Now let us discuss the behavior of the static dielectric constant $\varepsilon(0,0) = \varepsilon_0$ It follows from (12) that

$$\tilde{E}_{lk\sigma}(0) \approx E_{lk} - A\sigma \left[S - \frac{1}{N} \frac{E_g - AS\sigma}{E_g + AS\sigma} \sum m_g \right], \qquad T < T_0,$$

$$\tilde{E}_{lk\sigma}(0) \approx E_{lk} - A\sigma \left[S - \frac{1}{N} \sum m_g \right] - \frac{A^2 S}{2} \frac{m^* a^2}{2\pi a q_\sigma} \frac{T}{SI}, \qquad T > T_0,$$
(14)

where

$$q_{\sigma} = \left[2m^{\bullet}(E_{g} + AS\sigma)\right]^{1/2}, \qquad E_{g} = E_{1,0} - E_{2,0}$$

When $AS << E_g$, the following expression for the energy of a conduction electron corresponding to fixed *f* spins follows from (14):

$$\tilde{E}_{1\mathbf{k}\sigma}(0) = E_{1\mathbf{k}} - A\tilde{S}\sigma, \qquad \tilde{S} = S - \frac{1}{N}\sum m_q.$$
(15)

Thus, in the static case virtual electron transitions take place between states in which the atomic spins are not adjusted to the electron. From the standpoint of the Franck-Condon principle, one may speak of such transitions, as of real transitions with ω E_g , as "fast." In the following we shall consider only the temperature region $T > T_0$, which is the most interesting. Then the last term in (14) is of order *AS/W* with respect to the term preceding it and may therefore be dropped. In this case the change 3 in the dielectric constant is determined by formula (7) with $B_{ac} = 0$ and S replaced by S.

3. DIELECTRIC CONSTANT OF A MAGNETIC SEMICONDUCTOR WITH HELICOIDAL MAGNETIC STRUCTURE

In Secs. 1 and 2 we obtained general expressions for the susceptibility of a dielectric with uniform magnetization. Expression (15), which gives $E_{lk\sigma}$ to the first order in AS/W, is not valid if the direction of the local magnetic moment varies slowly from place to place. For example, the energy of an electron in a helicoid with helicoidal vector Q is given by the accurate expression⁴

$$E_{1\mathbf{k}\pm}(Q) = \frac{1}{2} \left(E_{1\mathbf{k}} + E_{1,\mathbf{k}+Q} \right) \pm \left[\left(E_{1\mathbf{k}} - E_{1,\mathbf{k}+Q} \right)^2 + A^2 S^2 \right]^2$$
(16)

and for the limiting case in which $k \rightarrow 0$ and $Q << Q_0$, the gain in the s-f exchange energy is -AS/2, although S = 0. Expression (16) is similar to the expression [formula (3)] obtained in Sec. 1 for the energy of an electron in the two-band model, the only difference being that in (16) the s-f exchange energy AS plays the part of the cation-anion Bloch integral B_{ca} . We shall not be concerned with this analogy any more in what follows.

To clarify the behavior of the dielectric constant in crystals in which the magnetization direction varies from place to place, let us consider a crystal with helicoidal spin ordering at T=0. In this case the Hamiltonian of the system has the form

$$H = H_{1} + H_{el-el\uparrow},$$

$$H_{1} = \sum_{i\mathbf{k}\sigma} \mathbf{E}_{i\mathbf{k}\sigma} a_{i\mathbf{k}\sigma} - \frac{AS}{2} \sum \left(a_{1\mathbf{k}\uparrow}^{+} a_{1\mathbf{k}+q\downarrow} + a_{1\mathbf{k}\downarrow}^{+} a_{1\mathbf{k}-q\uparrow} \right).$$
(17)

The relation between the dielectric constant of the electron system and the Fourier component of the retarded Green's function is given by the formula⁵

$$\frac{1}{\varepsilon(q,\omega)} = 1 + \frac{4\pi e^2}{q^2} << \rho_q \mid \rho_{-q} >>_{\omega},$$
(18)

where $\langle \rho_q | \rho - q \rangle_{\omega}$, is the Fourier component with respect to energy of the two-time Green's function. To find the Green's function $\langle \rho_q | \rho_{-q} \rangle_{\omega}$ one formulates the equations of motion for $\langle a^+_{ik'} \uparrow a_{jk} \uparrow | p_{-q} \rangle_{\omega}$ From these one obtains the following set of equations:

$$\left(\omega + E_{2,k+q} - E_{ik} \right) \left\langle \left\langle a_{2,k+q\uparrow}^{+} a_{ik\uparrow} \middle| \rho_{-q} \right\rangle \right\rangle_{\omega} = \left\langle 2k \uparrow \middle| e^{iqr} \middle| \uparrow, \mathbf{k} - \mathbf{q}, 1 \right\rangle \left(1 / \varepsilon(\omega, q) \right)$$

$$- 1 / 2AS \left\langle \left\langle a_{2,k+q\uparrow}^{+} a_{i,k+Q\downarrow} \middle| \rho_{-q} \right\rangle \right\rangle_{\omega}$$

$$(19)$$

$$\left(\omega + E_{2,k+q} - E_{i,k+Q}\right) \left\langle \left\langle a_{2,k+q\uparrow}^{+} a_{i,k+Q\downarrow} \left| \rho_{-q} \right\rangle \right\rangle_{\omega} = -1/2AS \left\langle \left\langle a_{2,k+q\uparrow}^{+} a_{i,k\uparrow} \left| \rho_{-q} \right\rangle \right\rangle_{\omega} \right\rangle$$
(20)

By substituting the Green's function from (20) into (19), making use of (16) and (18), and neglecting the dependence of the interband coordinate matrix element on the degree of magnetic ordering, we obtain the following expression for the static dielectric constant in the limit $q \rightarrow 0$.

$$\varepsilon_{0} = \varepsilon_{L} + 8\pi e^{2} \sum_{k} \frac{\left| \langle k, 1 | x | k, 2 \rangle \right|^{2}}{E_{\tau k} - E_{ik}} \left\{ 1 - \frac{A^{2} S^{2} \wp_{k}}{4 (E_{\tau k} - E_{ik})} \right\}^{-1},$$

$$\wp_{k} = \left(E_{1k} - E_{2k} \right) / \left(E_{i,k+Q} - E_{2k} \right).$$
(21)

In the limit $\wp_k \to 1$, expression (21) reduces to the expression obtained from (11) for a ferromagnetic at T = 0, and in the opposite limit $\wp_k \to 0$, it reduces to expression (I) for a nonmagnetic crystal. Assuming a simple quadratic dispersion law with the same effective mass for both bands,

$$E_{1\mathbf{k}} = E_{g} + k^{2} / 2m^{\bullet}, \qquad E_{2\mathbf{k}} = -k^{2} / 2m^{\bullet}$$

we find that the condition $\wp_k \ll 1$ is satisfied provided

$$Q > Q_g = \left(2m^{\bullet}E_g\right)^{1/2}$$

i. e. for realistic values of m^* and E_g in the case of a helicoid whose period is of the order of the lattice constant. In the case of a helicoid with a fairly large period, however, the dielectric constant at T = 0 is virtually the same as in the case of ferromagnetic ordering. Hence the temperature dependence of ε_0 should be of the same type for a helicoid with $Q > Q_g$ as for a ferromagnet.

The last circumstance is of special interest in connection with the fact that many crystal with helicoidal ordering exhibit a temperature shift of the absorption edge with the same sign and of the same order of magnitude as is observed in ferromagnets [e. g., in HgCr₂S₄ the red shift of the edge on lowering the temperature exceeds 0. 4 eV, while for most ferromagnets in amounts to 0. 2-0. 3 eV (Ref. 2)]. As follows from (16), the edge shift is large if $Q < (2m^*AS)^{1/2}$, and then e₀ may be strongly temperature dependent.

4. DISCUSSION

It follows from (8), (14), and (15) that the dielectric constant depends on the temperature and on the strength H of the external magnetic field. The decrease in the width of the forbidden gap in ferromagnetic semiconductors on lowering the temperature should lead to a substantial increase in the dielectric constant, and the same effect should be observed in a magnetic field. The dependence of e_0 on Φ and 3 has many consequences. In particular, one such consequence should be a decrease in the depth of the donor level and an increase of the radius n_B of the donor orbit. A decrease in the difference between the local magnetization in the vicinity of the donor and the average magnetization of the crystal leads to the same thing.² Because of this, a transition of the crystal from the insulating to the conducting state on lowering the temperature (a Mott transition) is possible if the impurity concentration 3 lies in a certain range: if the criterion $a_Bn^{1/3}>0$. 25 donor collectivization is not satisfied at high temperatures, it may be satisfied at low temperatures.

The above mentioned increase in a_B due to the increase in e_0 facilitates the insulatormetal transition and may even turn out to be sufficient by itself to cause such a transition. Such a transition may also be observed in a magnetic field; for example, if the impurity concentration 3 in an antiferromagnet lies within the interval $n_0 < n < n_1$, where

$$n_1 = n_0 (1+\eta)^3$$
, $n_0 = (4a_B)^{-3}$, $\eta = \left[\varepsilon_0(0) - \varepsilon_0(\infty)\right] / \varepsilon_0(\infty)$

then in a magnetic field of strength $H >> H_F$ (where H_F is the field in which the sublattices collapse) the semiconductor will undergo a transition from the insulating to the metallic state simultaneously with its transition from antiferromagnetic to ferromagnetic order.

The phase transition in a magnetic field has been discussed in Ref. 2 for the case in which the electrons in the antiferromagnet lie on collective or individual ferronic states, i. e., when they are self trapped in regions of the ferromagnetic phase. The metal-insulator transition in an antiferromagnet due to the change in the dielectric constant is not connected with the formation of ferronic states in the crystal and may take place not only in the case in which the formation of ferronic states in the crystal is possible, but also in the case in which such states cannot be formed, e. g., in aniso-tropic antiferromagnetic semiconductors with laminar magnetic ordering.² As another example we may note the dependence of the frequency of optical phonons on the magnetization of the crystal. It is obvious from the Lyddane-Sachs-Teller relation, according to which the frequency of longitudinal phonons differs from that of transverse phonons by the factor $(e_0/e_{\infty})^{1/2}$, where e_{∞} is the static dielectric constant at frequencies above the phonon frequencies but small as compared with the width of the forbidden gap. As was pointed out above, in this case one must take account of the fact that not only electron polarization, but also lattice polarization, contributes to ε_0 .

Formula (21) makes it possible to carry through a qualitative analysis of the behavior of the dielectric constant of crystals having domain structure. The spin ordering in a Bloch domain wall may be regarded as locally helicoidal with wave vector Q. The thinner the wall (thickness \sim 1/Q), the greater the difference between the values of the dielectric constant within the wall and within the ferromagnetic domain. Thus, a crystal with thin domain walls should behave toward light as a periodic structure with a modulated refractive index. As another example of materials in which a periodic variation of the dielectric constant is possible, we may mention crystals having a large anisotropy constant when the magnetization vector does not rotate within the domain wall, but changes in magnitude while keeping its direction fixed.

¹⁾It is interesting to note that, in accordance with the results of Sec. 2, the imaginary part of the dielectric constant describes transitions of the electrons from the valence band to spin-polaron states — not to bare conduction-band states. The situation should be similar

in the case of weak-coupled polarons in ionic crystals. This means essentially that the Franck-Condon principle is violated, since in this case the electron transition is accompanied by a change in the coordinates of the low-energy subsystem (the magnon or phonon system), which adjusts itself to the charge carriers that appear as a result of the transition. The results obtained in the paper indicate that a more careful analysis of the limits of applicability of the Franck-Condon principle is necessary.

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