

Nondissipative photoferrromagnetism of magnetic semiconductors

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A theory is developed of a change in the magnetization of ferromagnetic semiconductors as a result of illumination. In contrast to the usual situation when the frequency of light lies within an absorption band, an analysis is made of the frequency range in which a crystal is transparent. In this case the photoferrromagnetic effect is due to virtual transitions of electrons from the valence to the conduction band. Such transitions result in the mixing of the conduction- and valence-band states. This mixing enhances the exchange of electrons between the valence bands and localized d or f magnetic moments, and thus facilitates the establishment of the ferromagnetic order. The situation is treated as a nonlinear kinetic problem. An expression is obtained for the shift of the magnon frequency in the spin-wave range. Extrapolation of the calculated results to higher temperatures makes it possible to estimate the shift of the Curie temperature.

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1. Magnetic properties of semiconductor compounds of transition and rare-earth elements depend strongly on the carrier density because carriers provide the means for the indirect exchange between localized d or f magnetic moments. It was suggested some time ago^[1] that the photoexcitation of carriers should increase the Curie temperature of ferromagnetic semiconductors. However, the theoretical estimates of this effect^[1] have been found to be several orders of magnitude too high and this is why the phenomenon of photoferrromagnetism has not been discovered experimentally until many years later.^[2] Only very recently some very precise experiments have established the influence of light on the magnetization of the ferromagnetic semiconductor EuS.^[3] However, a parallel investigation of the photoconductivity has not been made because of considerable technical difficulties^[3] and, therefore, it is not yet clear whether the reported effect^[3] is associated with photoelectrons. The estimates^[3] obtained from the shift of the Curie point under the action of a laser beam indicate a carrier density of $\sim 10^{18}$ cm⁻³. The possibility of optical generation of such a high carrier density is doubtful if we carried out on highly defective crystals (films) in the vicinity of the Curie point and that a sharp photoconductivity minimum is observed in this temperature range.^[2]

We shall demonstrate the possibility of the photoferrromagnetic effect which is not associated with the optical generation of carriers, so that an increase in the degree of ferromagnetic order under the influence of light may occur even in the range of frequencies corresponding to the optical transparency of a crystal. This effect is due to virtual transitions of electrons from the valence to the conduction band producing virtual conduction electrons and holes (we shall use these terms for the changes in the valence-band states under the influence of illumination and we shall understand them to mean that the valence band acquires an admixture of the conduction-band states; it should be understood that electrons present in the valence band before illumination remain in that band although the band itself changes).

In many magnetic semiconductors (for example,

EuO^[2]) the valence-band states originate mainly from the p orbitals of the nonmagnetic anions and the conduction-band states from the outer orbitals of the magnetic cations. The overlap of the orbitals of the partly filled cation shells with the orbitals of the outer shells of the same cations is much stronger than with the anion orbitals. Therefore, the exchange interaction of the localized d and f moments with the conduction electrons is much stronger than with the valence-band electrons. Under these conditions the mixing of the conduction- and valence-band states under the influence of illumination enhances greatly the exchange of electrons between the valence band and the localized moments.

Depending on the orientation of the electron spin relative to the moment of a crystal, this exchange enhancement may either decrease or increase the electron energy. If the degree of mixing is independent of the electron spin, there is no net change in the energy of the exchange of electrons between the completely filled valence band and the localized moments. However, in ferromagnetic semiconductors the degree of mixing depends on the electron spin direction. This is due to the fact that in the case of a finite moment of a crystal the energy bands are spin-split (this is the Zeeman splitting of the electron energy in the molecular field of the crystal). Consequently, the spin orientation affects also the gap between the valence and conduction bands, which governs the degree of mixing. In terms of the virtual conduction electrons we may say that they are spin-polarized and that the dominant virtual transitions are those with the spin projection that ensures an increase in the energy of their exchange with the localized moments. This energy increases with the magnetization of the crystal. Therefore, the virtual electrons created by illumination tend to establish and maintain the ferromagnetic order in a crystal.

The mechanism described above operates also in those cases when carrier generation is possible under the influence of light. If the magnetization relaxation time is shorter than other characteristic times, the contributions of the virtual and real photoelectrons to the photoferrromagnetic effect may be separated in ac-

cordance with the relaxation times (the former contribution is instantaneous and the second has the same relaxation time as the photoconductivity).

The fact that the virtual electron transitions are not accompanied by the absorption of light makes it much easier to observe experimentally the photoferrromagnetic effect in the transparency range of a crystal: illumination does not cause heating which might destroy the ferromagnetic order.

We shall consider the photoferrromagnetic effect due to virtual electron transitions in the spin-wave range. Extrapolation of the results to higher temperatures makes it possible to estimate the shift of the Curie temperature under the influence of illumination. Since photoferrromagnetism is a nonequilibrium effect, we cannot use the standard methods of statistical physics and we have to treat the situation as a nonlinear kinetic problem. We shall show that when certain conditions of a dynamic electron equilibrium are satisfied and a sufficiently long time passes from the beginning of illumination, a magnon distribution is established and this distribution is described by the usual Bose function with a renormalized magnon frequency which depends on the intensity of light. The method of equations of motion for the occupation numbers with explicit allowance for the interaction of the system in question with the thermostat, which we shall use here, may be of interest also in other applications.

2. In a mathematical description of the photoferrromagnetic effect we must bear in mind that if we ignore the relativistic effects, the electron transitions are then due to the electric vector of an electromagnetic wave, i.e., the Hamiltonian of the interaction of electrons with light conserves the spin of the electron system. Therefore, without the interaction of a crystal with the thermostat there is no change in its magnetization under the action of light. It might seem that a change in the effective exchange interaction in the electron subsystem under the influence of illumination may, without the thermostat, alter its magnetization. In fact, this is not true: such a modification to the exchange does not alter the symmetry properties of the Hamiltonian of the system which are responsible (if the relativistic effects are ignored) for the conservation of its total spin. If we consider the difference between the Hamiltonian of the exchange renormalized under the action of light and the initial Hamiltonian as a perturbation we find that the perturbation Hamiltonian still conserves the total spin and, therefore, the only nonzero matrix elements of this Hamiltonian are those which link the states with the same value of the total spin. In other words, the magnetization of a crystal is not affected by illumination if we ignore its interaction with a thermostat.

The interaction which does not conserve the total spin is, for example, the spin-lattice coupling. Since the lattice is in a state of thermodynamic equilibrium, it acts as the thermostat. We shall use only the most general property of this thermostat: its state does not change because of the interaction with the system under consideration. The properties of the thermostat are

possessed also by a system of large dimensions with a very short relaxation time of its modes. For times much longer than that necessary to establish a thermal equilibrium the detailed structure of the interaction between the crystal and thermostat is unimportant. Therefore, without any loss of generality, we shall use the simplest model of the thermostat.

The Hamiltonian of our system considered on the basis of the s - f model of a magnetic semiconductor can be represented in the form

$$\begin{aligned}
 H &= H_0 + H', \quad H' = H_1 + H_2 + H_V + H_Q, \quad (1) \\
 H_0 &= \sum \omega_q b_q^+ b_q + \sum \varepsilon_{k\sigma} a_{k\sigma}^+ a_{k\sigma} + \sum \hbar_k c_{k\sigma}^+ c_{k\sigma} + \sum \Omega_\mu B_\mu^+ B_\mu, \\
 H_1 &= -A \left(\frac{S}{2N} \right)^{1/2} \sum (a_{k_1}^+ a_{k_2} b_q^+ + a_{k_1}^+ a_{k_2}, b_q), \\
 H_2 &= \frac{A}{2N} \sum (a_{k_1}^+ a_{k_2} - a_{k_1} a_{k_2}^+) b_q^+ b_{q+r}, \\
 H_V &= \sum (V_k e^{-i\omega t} a_{k\sigma}^+ c_{k\sigma} + V_k^* e^{i\omega t} c_{k\sigma}^+ a_{k\sigma}), \quad H_Q = \sum (b_q^+ B_\mu + B_\mu^+ b_q), \\
 \varepsilon_{k\sigma} &= \varepsilon_k - AS\sigma, \quad V_k = \frac{ie}{m_0 \omega} E_0 \langle ck | \hat{p} | vk \rangle.
 \end{aligned}$$

Here, $c_{k\sigma}^+$, $c_{k\sigma}$, and $a_{k\sigma}^+$, $a_{k\sigma}$ are, respectively, the creation and annihilation operators of electrons in the valence and conduction bands; b_q^+ and b_q are the magnon operators; B_μ^+ and B_μ are the thermostat operators. It is assumed that the interaction of the valence-band electrons with the localized f moments can be ignored. The interaction of the conduction-band electrons with the f spins is characterized by the s - f exchange constant A and, to be specific, we shall assume that this constant is positive. The s - f exchange shifts the energy $\varepsilon_{k\sigma}$ of an electron with the spin projection σ compared with the "bare" electron energy ε_k by an amount $AS\sigma$, where S is the value of the f spin. The interaction of electrons with light is characterized by the matrix element $\langle ck | \hat{p} | vk \rangle$ between the valence and conduction band states with the same quasimomentum k ; \hat{p} is the polarization vector of the incident light; E_0 is the amplitude of the electric field.¹⁾

The physical meaning of the thermostat operators B_μ^+ and B_μ and the corresponding dynamic variables μ is not specified. From the calculations given below it is sufficient to note simply that the Hamiltonian H_Q of the interaction between magnons and the thermostat conserves neither the magnetic quasimomentum nor the magnon number. Since the Hamiltonian H_Q does not conserve the number of the thermostat quanta either, the operators B_μ^+ and B_μ should be regarded as of the Bose type. The absorption of magnons is possible if the frequencies Ω_μ are of the same order of magnitude as the magnon frequencies ω_q , i.e., if they are low compared with the electron frequencies.

Since a change in the magnetization under the influence of light is proportional to its intensity, i.e., to E_0^2 , the determination of the magnetization is a nonlinear kinetic problem.

The magnetization is found from the equation of motion for the average magnon numbers $\langle b_q^+ b_q \rangle$ and the higher correlation functions associated with them. The angular brackets mean here the averaging with a nonequi-

librium density matrix $\rho(t)$: $\langle \hat{A} \hat{B} \rangle \equiv \langle \rho(t) \hat{A} \hat{B} \rangle$. The equations of motion for the magnon occupation numbers, obtained by means of the Hamiltonian (1), have the following form for $t \geq 0$:

$$i \frac{d}{dt} \langle b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} \rangle = Q \sum_{\mathbf{k}} (\langle b_{\mathbf{q}}^{\dagger} B_{\mathbf{k}} \rangle - \langle B_{\mathbf{k}}^{\dagger} b_{\mathbf{q}} \rangle) - A \left(\frac{S}{2N} \right)^{1/2} \sum_{\mathbf{k}} (\langle b_{\mathbf{q}+\mathbf{k}}^{\dagger} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}+\mathbf{q}} \rangle - \langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}} b_{\mathbf{q}} \rangle) + \frac{A}{2N} \sum_{\mathbf{k}, \sigma} \sigma (\langle b_{\mathbf{q}}^{\dagger} a_{\mathbf{k}\sigma}^{\dagger} a_{\mathbf{k}\sigma} b_{\mathbf{q}+\mathbf{k}} \rangle - \langle b_{\mathbf{q}+\mathbf{k}}^{\dagger} a_{\mathbf{k}\sigma}^{\dagger} a_{\mathbf{k}\sigma} b_{\mathbf{q}} \rangle). \quad (2)$$

We shall show later that after a sufficiently long time the quantity $m_{\mathbf{q}} \equiv \langle b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} \rangle$ is governed by the first term on the right-hand side of Eq. (2). The equations of motion, accurate to within terms $\sim Q$ inclusive, can also be derived for the mixed magnon-thermostat correlation functions in this equation. It is assumed that because of the translational invariance of a crystal in the spin-wave range, only the diagonal correlation functions $\langle b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} \rangle$ do not vanish. The same is true, by definition of the thermostat, of the correlation functions $\langle B_{\mu}^{\dagger} B_{\mu} \rangle$:

$$\langle B_{\mu}^{\dagger} B_{\mu} \rangle = F(\Omega_{\mu}) \delta_{\mu\mu'} = \{ \exp(\Omega_{\mu}/T) - 1 \}^{-1} \delta_{\mu\mu'}.$$

Bearing these points in mind, we obtain the following equations of motion for $\langle b_{\mathbf{q}}^{\dagger} B_{\mu} \rangle$:

$$i \frac{d}{dt} \langle b_{\mathbf{q}}^{\dagger} B_{\mu} \rangle = (\Omega_{\mu} - \omega_{\mathbf{q}}) \langle b_{\mathbf{q}}^{\dagger} B_{\mu} \rangle + A \left(\frac{S}{2N} \right)^{1/2} \sum_{\mathbf{k}} \langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}} B_{\mu} \rangle - \frac{A}{2N} \sum_{\mathbf{k}, \sigma} \sigma \langle b_{\mathbf{q}+\mathbf{k}}^{\dagger} a_{\mathbf{k}\sigma}^{\dagger} a_{\mathbf{k}\sigma} B_{\mu} \rangle + Q(m_{\mathbf{q}} - F_{\mu}); \quad (3)$$

analogous equations apply also to $\langle B_{\mu}^{\dagger} b_{\mathbf{q}} \rangle$. The calculations given below are based on the use of the small parameters $1/2S$ and $V_{\mathbf{k}}/W$, where $W \sim 10/m^* a^2$ is the width of the conduction band. In the first order in $1/2S$, we can write

$$\sum_{\mathbf{k}\sigma} \sigma \langle b_{\mathbf{q}+\mathbf{k}}^{\dagger} a_{\mathbf{k}\sigma}^{\dagger} a_{\mathbf{k}\sigma} B_{\mu} \rangle = \langle b_{\mathbf{q}}^{\dagger} B_{\mu} \rangle \sum_{\mathbf{k}\sigma} \sigma n_{\mathbf{k}\sigma}, \quad n_{\mathbf{k}\sigma} = \langle a_{\mathbf{k}\sigma}^{\dagger} a_{\mathbf{k}\sigma} \rangle. \quad (4)$$

The result for $\langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}}^{\dagger} B_{\mu} \rangle$ can be obtained in the same order in $1/2S$ by deriving the equations of motion

$$i \frac{d}{dt} \langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}}^{\dagger} B_{\mu} \rangle = (\Omega_{\mu} - \varepsilon_{\mathbf{k}+\mathbf{q}} + \varepsilon_{\mathbf{k}}) \langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}}^{\dagger} B_{\mu} \rangle + Q \sum_{\mathbf{q}'} \langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}}^{\dagger} b_{\mathbf{q}'} \rangle - A \left(\frac{S}{2N} \right)^{1/2} \sum_{\mathbf{q}'} \langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}+\mathbf{q}'}^{\dagger} b_{\mathbf{q}'}^{\dagger} B_{\mu} \rangle + \frac{A}{2N} \sum_{\mathbf{q}', \mathbf{r}} \langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}-\mathbf{r}}^{\dagger} b_{\mathbf{q}'}^{\dagger} b_{\mathbf{r}}^{\dagger} B_{\mu} \rangle + A \left(\frac{S}{2N} \right)^{1/2} \sum_{\mathbf{q}'} \langle a_{\mathbf{k}+\mathbf{q}-\mathbf{q}'}^{\dagger} a_{\mathbf{k}}^{\dagger} B_{\mu} b_{\mathbf{q}'}^{\dagger} \rangle + \frac{A}{2N} \sum_{\mathbf{q}', \mathbf{r}} \langle a_{\mathbf{k}+\mathbf{q}-\mathbf{r}}^{\dagger} a_{\mathbf{k}}^{\dagger} B_{\mu} b_{\mathbf{q}'}^{\dagger} b_{\mathbf{r}}^{\dagger} \rangle + V_{\mathbf{k}} e^{-i\omega t} \langle B_{\mu} a_{\mathbf{k}+\mathbf{q}}^{\dagger} c_{\mathbf{k}} \rangle - V_{\mathbf{k}+\mathbf{q}} e^{i\omega t} \langle B_{\mu} a_{\mathbf{k}}^{\dagger} c_{\mathbf{k}+\mathbf{q}} \rangle. \quad (5)$$

The higher magnon correlation functions, which include the operators $b^{\dagger} b$, can be omitted in the spin-wave range. The terms $\sim V$ are also omitted because they give a correction to the correlation function which is $\langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}}^{\dagger} B_{\mu} \rangle \sim V_{\mathbf{k}}^2$, which can easily be demonstrated by writing down the equation of motion for correlation functions of the $\langle B^{\dagger} a^{\dagger} c \rangle$ type. The term $\sim Q$ may be ignored because such correlation functions are already included in Eq. (2) but without the small parameter Q . If the other terms in Eq. (5) are subjected to decoupling of the type (4), we obtain

$$\langle a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}}^{\dagger} B_{\mu} \rangle = \left(i \frac{d}{dt} - \Omega_{\mu} + \varepsilon_{\mathbf{k}+\mathbf{q}} - \varepsilon_{\mathbf{k}} \right)^{-1} A \left(\frac{S}{2N} \right)^{1/2} (n_{\mathbf{k}+\mathbf{q}} - n_{\mathbf{k}}) \langle b_{\mathbf{q}}^{\dagger} B_{\mu} \rangle. \quad (6)$$

Thus, if the average number of the conduction electrons $n_{\mathbf{k}\sigma}(t)$ are known, the system (3)–(6) is closed. It is sufficient to find the numbers $n_{\mathbf{k}\sigma}(t)$ in the principal approximation in respect of $1/S$. Ignoring the influence of magnons on electrons and bearing in mind that for $t \leq 0$ all the electrons are in the valence band and there are none of them in the conduction band, we obtain—in agreement with the well-known results on the state of a two-level system in the presence of a periodic perturbation^[4]

$$n_{\mathbf{k}\sigma}(t) = n_{\mathbf{k}\sigma}^0 [1 - \cos(\Delta_{\mathbf{k}\sigma} t + 4|V_{\mathbf{k}}|^2)^{1/2} t], \quad (7)$$

$$n_{\mathbf{k}\sigma}^0 = 2|V_{\mathbf{k}}|^2 / (\Delta_{\mathbf{k}\sigma}^2 + 4|V_{\mathbf{k}}|^2), \quad \Delta_{\mathbf{k}\sigma} = \varepsilon_{\mathbf{k}\sigma} - \omega - h_{\mathbf{k}}.$$

We shall later find it important that, because of the difference between the phases of the oscillations corresponding to different values of \mathbf{k} , the contribution of the oscillatory parts of $n_{\mathbf{k}\sigma}(t)$ to $\langle b_{\mathbf{q}}^{\dagger} B_{\mu} \rangle$ is negligible for $t \gg t_A = \max[\Delta_{\mathbf{k}\sigma}^2 + 4|V_{\mathbf{k}}|^2]^{-1/2}$. In fact, after substitution of Eqs. (4), (6), and (7) into Eq. (3) and summation over \mathbf{k} by the steepest-descent (saddle-point) method, we find that it is proportional to $1/\sqrt{t}$ [in the expression for the correlation function (6) it is convenient, before summation over \mathbf{k} , to transform the differential operator by means of the identity

$$\mathcal{L}^{-1} = \int_0^{\infty} dp e^{p\hat{t}}, \quad \mathcal{L} = i \frac{d}{dt} + c$$

and then carry out the summation].

The time t_A is of the order of the atomic value, whereas we are interested in the behavior of the system at times much longer than not only the atomic times but also longer than the time needed to establish a thermal equilibrium. Therefore, we may assume that a steady-state electron distribution is established suddenly at $t=0$. This allows us to carry out the Laplace transformation of the correlation functions with respect to time. As a result, Eq. (3) and the conjugate equation assume the form (allowing for the smallness of Ω_{μ} compared with W)

$$(ip + \bar{\omega}_{\mathbf{q}}(p) - \Omega_{\mu}) \langle b_{\mathbf{q}}^{\dagger} B_{\mu} \rangle_p = Q(m_{\mathbf{q}}(p) - F_{\mu}/p) + \langle b_{\mathbf{q}}^{\dagger} B_{\mu} \rangle_{t=0}, \quad (8)$$

$$(ip - \bar{\omega}_{\mathbf{q}}(-p) + \Omega_{\mu}) \langle B_{\mu}^{\dagger} b_{\mathbf{q}} \rangle_p = -Q(m_{\mathbf{q}}(p) - F_{\mu}/p) + \langle B_{\mu}^{\dagger} b_{\mathbf{q}} \rangle_{t=0}.$$

Here,

$$\bar{\omega}_{\mathbf{q}}(p) = \omega_{\mathbf{q}} + v(p),$$

$$v(p) \approx \frac{A}{2N} \sum_{\mathbf{k}, \sigma} \sigma n_{\mathbf{k}\sigma}^0 - A^2 \frac{S}{2N} \sum_{\mathbf{k}} \frac{n_{\mathbf{k}}^0 - n_{\mathbf{k}+\mathbf{q}}^0}{ip + \varepsilon_{\mathbf{k}+\mathbf{q}} - \varepsilon_{\mathbf{k}} + AS}. \quad (9)$$

In the equation system (8) allowance is made for the vanishing of the correlation functions of the $\langle B_{\mu}^{\dagger} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}+\mathbf{q}} \rangle$ type at the initial moment (at $t=0$ there are no electrons in the conduction band). After application of the Laplace transformation of Eq. (2) and substitution there of the Laplace transforms of the correlation functions from the system (8), we obtain the following equation for the determination of the Laplace transform $m_{\mathbf{q}}(p)$ of the magnon distribution function

$$m_{\mathbf{q}}(p) \left\{ ip - Q^2 \sum_{\mu} \left[\frac{1}{\bar{\omega}_{\mathbf{q}}(p) - \Omega_{\mu} + ip} - \frac{1}{\bar{\omega}_{\mathbf{q}}(-p) - \Omega_{\mu} - ip} \right] \right\} = -Q^2 \sum_{\mu} \frac{F_{\mu}}{p} \left[\frac{1}{\bar{\omega}_{\mathbf{q}}(p) - \Omega_{\mu} + ip} - \frac{1}{\bar{\omega}_{\mathbf{q}}(-p) - \Omega_{\mu} - ip} \right] + G(p) + \bar{m}_{\mathbf{q}}(T). \quad (10)$$

Here, $G(p)$ represents the terms which have no singularities at $p=0$. They appear because of the terms $\sim (B_\mu^* b_\mu)_{t=0}$ and the two last sums on the right-hand side of Eq. (2), which also give rise to terms with singularities of the $1/p$ type of higher order in $1/2S$ than those in Eq. (9). Here, $\bar{m}_q(T)$ denotes the thermodynamic-equilibrium magnon distribution function which is equal to m_q at $t=0$.

The asymptotic behavior of the Laplace original is governed by that singularity of its Laplace transform which is furthest to the right. It is clear from Eq. (10) that such a point for m_q is $p=0$. In the vicinity of this point a considerable contribution to the sums over μ on the left- and right-hand sides of Eq. (10) is made by terms with $\Omega_\mu \approx \bar{\omega}_q(0)$. If $\bar{\omega}_q(p)$ is real in the limit $p \rightarrow 0$, the singular part of the Laplace transform in the same limit $p \rightarrow 0$ is $F(\bar{\omega}_q)/p$, i.e., in the limit $t \rightarrow \infty$ the function $m_q(t)$ becomes identical with $F(\bar{\omega}_q)$.

The quantity $\bar{\omega}_q(0)$ can be regarded as real in the case when we can ignore electron transitions to the conduction subband with the down spins. According to Eq. (7) this occurs if $\min \Delta_{k\downarrow} \gg \min \Delta_{k\uparrow}$, $\max V_k$. Then, the only important electron transitions to the states in the up-spin subband are those located near its minimum so that the numbers $n_{k\downarrow}$ can be regarded as equal to zero and among the occupation numbers $n_{k\uparrow}$ practically the only ones which differ from zero are those with $|\mathbf{k} - \mathbf{K}| < q_0 = (2m^*AS)^{1/2}$, where \mathbf{K} is the quasimomentum corresponding to the minimum of the conduction band and m^* is the effective mass. However, for these values of \mathbf{k} the energy denominator in the second term of Eq. (9) does not vanish at $p=0$. In this case, in spite of the absence of a thermodynamic equilibrium in respect of electrons, magnons in an illuminated crystal are in a thermodynamic equilibrium but their frequencies are a function of the illumination intensity.

If the above conditions are not satisfied, the energy denominator in the second term of Eq. (9) does vanish at $p=0$, whereas the numerator remains finite. Thus, magnons are found to be damped. [The expression (9) is similar in structure with the well-known result of the Ruderman-Kittel theory of indirect exchange, except that in the latter case the numbers $n_{k\sigma}^0$ of Eq. (7) are replaced with the Fermi electron distribution functions. Since these depend only on the electron energy, the numerator vanishes at the same time as the denominator and, therefore, there is no magnon damping in the indirect exchange case.] Then, the expression for the photomagnetization includes the density of the energy levels of the thermostat $\rho_T(E)$:

$$m_q(\infty) \approx \int \frac{F(E) \bar{\omega}_q'' \rho_T(E) dE}{(\bar{\omega}_q' - E)^2 + (\bar{\omega}_q'')^2} \bigg/ \int \frac{\rho_T(E) \bar{\omega}_q'' dE}{(\bar{\omega}_q' - E)^2 + (\bar{\omega}_q'')^2}, \quad (11)$$

$$\rho_T(E) = \sum_\mu \delta(E - \Omega_\mu), \quad \bar{\omega}_q(p) = \bar{\omega}_q' + i\bar{\omega}_q''$$

($p \rightarrow 0$). At low temperatures allowance for the finite damping makes the temperature dependence of $m_q(\infty)$ different from the Bose type. However, at high temperature $\bar{\omega}_q'$, $\bar{\omega}_q'' \ll T$, when $F(E)$ is proportional to T , allowance for the finite damping does not alter the nature of the temperature dependence of $m_q(\infty)$ and affects

only the renormalized frequency. Under typical conditions when illumination shifts the magnon frequencies only slightly, we have the inequality $\bar{\omega}_q' \gg \bar{\omega}_q''$. If we assume that $\rho_T(E)$ varies with E more slowly does $F(E)$, we find from Eq. (11) that truncation of integration with respect to E at a distance $\sim \bar{\omega}_q''$ from $\bar{\omega}_q'$ gives

$$m_q(\infty) \approx T \bar{\omega}_q' / [(\bar{\omega}_q')^2 + (\bar{\omega}_q'')^2]. \quad (12)$$

It follows from Eq. (12) that even if the real ν' and imaginary ν'' parts of the magnon frequency shift $\nu_q = \bar{\omega}_q - \omega_q$ are of the same order of magnitude, the above assumption of the slowness of the change in $\rho_T(E)$ allows us to consider only ν' since the correction to $m_q(\infty)$ is linear in respect of ν_q' and quadratic in respect of $\nu_q'' = \omega_q''$.

3. In estimating the magnitude of the photoferrromagnetic effect it is assumed that the valence band maximum and the conduction band minimum lie at the center of the Brillouin zone and the effective electron and hole masses m^* are similar. We shall assume that the frequency of light ω is "almost sufficient" to cause electron transitions from the valence to the conduction band, i.e., that the inequality $p_\dagger^2 \ll q_0^2$ is satisfied, where $q_0^2 = 2m^*AS$, $p_\dagger^2 = p_0^2 - q_0^2/4$, $p_0^2 = m^*(E_g - \omega)$ and E_g is the band gap in the absence of magnetic order. Under these conditions the average number of electrons in the down-spin subband is negligible and the magnon dispersion law (7), (9) can be approximated by

$$\nu_q = \frac{\mathcal{L}q^2}{q_0^2 + q^2}, \quad \mathcal{L} = \frac{Aa^2 u^4}{16\pi^2} \int_0^\infty \frac{dk k^2}{(k^2 + p_\dagger^2)^2 + u^4} = \frac{Aa^2}{32\pi} u^4 \left(\frac{2}{(p_\dagger^2 + u^2)^{3/2} + p_\dagger^2} \right)^{1/2}$$

$$u^2 = 4|V_0|^2 (m^*)^2. \quad (13)$$

For unpolarized light, the quantity $|V_0|^2$ is

$$V_0^2 = \frac{e^2 E_0^2}{m_0^2 \omega^2} |\langle c0 | p_x | v0 \rangle|^2. \quad (14)$$

The square of the matrix element occurring in the above expression can be estimated from the expression for the effective mass m^* in a band $\gamma^{(5)}$:

$$\frac{m_c}{m_\dagger^*} = 1 + \frac{2}{m_0} \sum_\delta \frac{|\langle \gamma 0 | p_x | 0 \delta \rangle|^2}{\epsilon_{\gamma\delta} - \epsilon_{00}}, \quad (15)$$

where ϵ_{00} is the energy of the state of an electron with $k=0$ in a band δ . If we ignore the contributions of the more distant bands and allow for the negative nature of the electron effective mass in the valence band ($m_\dagger^* = -m^*$), we find from Eq. (15) that

$$|\langle c0 | p_x | v0 \rangle|^2 \approx \frac{E_g m_0}{2} \left(1 + \frac{m_0}{m^*} \right). \quad (16)$$

Assuming that $E_g \approx 1$ eV, $m^* = m_0$, $E_0 = 3 \times 10^6$ V/cm, $p_\dagger \sim u$, and $a = 3$ Å, we obtain the following estimate for the parameter \mathcal{L} : $\mathcal{L} \approx 1/2A \cdot 10^{-3}$

According to the results of Ref. 6, the magnon frequency shift (13) shifts the Curie point by

$$\Delta T_c \sim \mathcal{L} S / a q_0, \quad (17)$$

which corresponds to $\sim 3^\circ\text{K}$ for $AS = 0.5$ eV. This esti-

mate is obtained for the most powerful lasers. The effect is still noticeable for lasers whose power is an order of magnitude lower. The fact that the laser frequency corresponds to the transparency range of a crystal ensures the stability of the effect, at least during short pulses.

If the valence band originates from the f -type states, then not only virtual photoelectrons but virtual photoholes interact strongly with the magnetization of a crystal and, therefore, both types of carrier contribute to the photoferrromagnetic effect.

Moreover, the participation of the f electrons in virtual optical transitions should give rise to a dependence of the effect on the nature of polarization of the incident light. In the case of real optical transitions such a dependence is simply a consequence of the spontaneous dichroism of a ferromagnet, caused by the spin-orbit interaction. In the case of virtual transitions, we can speak of virtual dichroism caused by the same interaction. This virtual dichroism is possible also because of the spin-orbit interaction in the conduction band if this band originates from hybridization of s -type atom-

ic states with other states, for example, with d -type states in EuO and EuS.

¹The terms with the opposite sign of the frequency are omitted because of the smallness of their contribution to the effect in H_V .

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Differential thermoelectric power of excitonic insulator

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The thermoelectric coefficient η_{ex} of an excitonic insulator was calculated. The case when the electron and hole scattering amplitudes by the impurities are different is considered. It is shown that in this case the temperature dependence of η_{ex} goes through a maximum. It is also shown that allowance for the terms of higher order in the impurity interaction compared with the Born approximation leads to an additional contribution to η_{ex} . This contribution is of zeroth order in T/μ (μ is the chemical potential of the system) and is characteristic of only the exciton phase. As a result, the thermoelectric power of an excitonic insulator is higher than that of semimetals at temperatures close to the transition point.

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Keldysh and Kopaev^[1] have shown that semimetals with electron and hole Fermi surfaces of like shape are unstable to formation of electron-hole pairs at arbitrarily weak electron-hole attraction. As a result a transition to the state of an excitonic insulator, with a gap in the energy spectrum, takes place at a certain critical temperature T_d .^[2] Allowance for the interband transitions^[3] fixes the phase of the order parameter of the excitonic insulator and makes states with homogeneous particle flow impossible. Realignment takes place, but there is no superfluidity. When the electron-hole attraction greatly exceeds the interaction terms responsible for the interband transitions, allowance for the latter leads to small corrections to T_d and to other quantities that describe the realignment.

Although an analogy exists between the excitation spectra of a superconductor and excitonic insulator, the kinetic properties are different in a number of

cases. Thus, absorption of ultrasound of frequency $\omega < 2\Delta$ has, in contrast to a superconductor, a maximum below T_d .^[4,5] This is due to the fact that the perturbation upsets the electron-hole symmetry, the coherence factors enter with a plus sign and have the same form as for the nuclear-spin relaxation rate in superconductors.

The electric conductivity and thermal conductivity in an excitonic insulator decrease monotonically with decreasing temperature,^[6,7] inasmuch as in this case the coherence factors enter with a minus sign. The expression for the thermal conductivity is then analogous to the corresponding expression for superconductors.^[8]

Zittartz^[7] has shown that in the calculation of the thermoelectric coefficient the coherence factors take the same form as for the thermal conductivity and the electric conductivity. It may seem therefore that the