Light-induced phase transitions

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A theory of phase transitions occurring under the action of light is developed. It is assumed that the influence of the light on the ordering is exerted through photoelectrons, which interact strongly with the subsystem that becomes ordered (e.g., with the magnetic atoms in a ferromagnetic semiconductor). The steady states of the system can be described with the aid of a nonequilibrium free-energy analog called a synergetic potential, since the rate $\dot{S}$ of entropy production is proportional to its derivative with respect to the light frequncy Ω. The steady-state value of the order parameter is found by minimizing the synergetic potential with respect to the order parameter. Light-induced phase transitions may be continuous or discontinuous, depending on Ω. In the first case the derivatives of $\dot{S}$ with respect to the light intensity Ω, and temperature are discontinuous; in the second, $\dot{S}$ itself is discontinuous. The photoelectron concentrations at which the phase transitions occur are estimated.

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There exist physical mechanisms, besides trivial heating, through which light can affect the state of a crystal that exhibits some kind of order, e.g., magnetic or ferroelectric order. In many cases these mechanisms tend, in contrast to heating, to maintain the order. Then at a given sample temperature (it is determined not only by the temperature of the thermostat, but also by the intensity of the absorbed light) the degree of ordering is found to be higher in light than in darkness, and it is even possible for phase transitions to occur from the disordered to the order state under the action of light. The Curie temperature T_c of the ferromagnetic semiconductor EuS has been experimentally observed to increase under the action of light.[1] In view of this, it has become necessary to construct a theory of light-induced phase transitions, and this is the object of the present paper.

Light-induced phase transitions fundamentally differ from ordinary phase transitions in that they occur under thermodynamic-nonequilibrium conditions, since the absorbed light energy is dissipated. Therefore, the free energy cannot be used to describe such transitions, and we cannot speak of the discontinuities of its derivatives, e.g., the specific heat, at the transition point. In fact, the only clearly defined quantity characterizing nonequilibrium processes is the entropy-production rate $\dot{S}$, and therefore it is precisely its discontinuities that should be investigated in the theory of nonequilibrium phase transitions. The question may naturally be asked: Is is possible to construct for nonequilibrium phase transition a quantity that will play the same role that the free energy plays in equilibrium phase transitions, i.e., one from whose extrema the steady states and the conditions for a nonequilibrium phase transition can be found, and from which the entropy production can be determined?

Below we show that at any rate, for certain systems, it is possible to introduce such a quantity. The production of entropy is proportional to the derivative of this quantity with respect to the light frequency, and therefore this derivative can, by analogy with the thermodynamic potential, be called a synergetic potential. The steady states correspond to the minimum of the synergetic potential of the system.

Apparently, the most effective mechanism through which light exerts its influence on the ordering is the interaction of the photoelectrons with the ordering subsystem. For example, in magnetic semiconductors the exchange interaction between the photoelectrons and the localized magnetic moments of the atoms makes the moments tend to establish a ferromagnetic order. The photoelectrons can affect not only the magnetic, but also the crystallographic order. Thus, they can cause uniaxial distortion of multivalley semiconductors of the Ge or Si type, accumulating in the valleys lying on one of the axes, since such a deformation lowers the bottom of these valleys (a similar phenomenon occurring upon the doping when multivalley semiconductors are doped has been theoretically investigated by Kochelap *et al.*[2]).

Below we consider two models, in the first of which the phase transition does not occur in darkness, while in the second it does occur in darkness, and illumination only shifts the critical point or changes the transition from continuous to discontinuous. In both models the influence of light on the ordering is exerted through photoelectrons. Although the first model is formulated in terms of a magnetoexciton semiconductor, it can easily be reformulated for an antiferromagnetic or a nonmagnetic multivalley semiconductor. The second model, formulated for a ferromagnetic semiconductor, is equally general. In the self-consistent field approximation the derivatives of $\dot{S}$ with respect to the light intensity and frequency and with respect to temperature should be discontinuous at a second-order phase transition point, just as the specific heat is in the Landau-Lifshitz theory. The corresponding photoelectron-concentration derivatives should exhibit similar discontinuities. In the case of a first-order phase transition $\dot{S}$ itself should undergo a jump. This should manifest itself in photoconductivity and Hall-effect anomalies.

We can, by analogy with the theory of equilibrium second-order phase transitions, expect $\dot{S}$ to have discontinuities of a type more complex than jumps in its derivatives when we go outside the limits of the self-consistent field approximation, i.e., when we make a more exact allowance for the fluctuations. It is also natural to assume that the discontinuities in $\dot{S}$ should be observed not only in light-induced, but also in nonequil-

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ibrium phase transitions of other types. Thus, the discontinuity in the thermal coefficient of electrical resistivity $d\rho/dT$ of a ferromagnetic conductor at the Curie point can be regarded as a manifestation of an $\dot{S}\sim d\rho/dT$ discontinuity (strictly speaking, this is a nonequilibrium phase transition even under conditions of weak currents). The $\dot{S}$ discontinuity in these materials is of the same type as the specific-heat discontinuity. This allows us to assume that the specific-heat discontinuity in the case of equilibrium, and the $\dot{S}$ discontinuity in the case of nonequilibrium, phase transitions are affected in qualitatively identical fashion by the fluctuations.

Let us note in conclusion that in principle light-induced phase transitions can occur without the dissipation of the light energy when the light frequency falls within the transmission band of the crystal.[3,4] The effect of light on the magnetization in this case is due to the change occurring under the action of the light in the valence-band states in the magnetic semiconductor, as a result of which the superexchange between the localized spins changes. This light-action mechanism requires much higher light intensities than the mechanism considered in the present paper. The absence of dissipation also allows us to describe such phase transitions as equilibrium transitions, the effect of illumination on which amounts only to the renormalization of the exchange integral.

1. THE INDUCTION OF MAGNETIZATION IN MAGNETOEXCITON CRYSTALS BY LIGHT

In this section we shall consider the simpler case of a phase transition that occurs as $T\to 0$ entirely as a result of the absorption of light. In darkness the order parameter (the mean magnetization) is equal to zero not because the magnetic moments of the atoms are disordered, i.e., not because the statistical averages of the components of the moments are equal to zero, but because their quantum-mechanical averages over the ground state are equal to zero. Therefore, the term "disordered state" has a somewhat arbitrary character here: it does not allow for order-parameter fluctuations. The order appears when the conduction electrons polarize the atom shells and thus make them acquire mutually parallel quantum-mechanical mean moments.

The simplest model of such a magnetoexciton crystal (it is customarily called a singlet magnet in the physics of magnetic phenomena) is as follows.[5] The crystal consists of magnetic atoms whose average moments in the ground and first-excited states $|0\rangle$ and $|1\rangle$ are equal to zero. The moments are quenched, since the opposite directions of the moment J are represented with equal weights in the wave functions, e.g., $|0\rangle\sim\{|J_0^z\rangle+|-J_0^z\rangle\}$, while $|1\rangle\sim\{|J_0^z\rangle-|-J_0^z\rangle\}$, where $|J_0^z\rangle$ is a state with a given $J^z=J_0^z$. But the matrix element of a component of an atom's moment, taken between the ground and first-excited states of the atom, is nonzero (in the cited example it is equal to J_0^z). Therefore, there appears in the crystal as a result of the mixing of these states a moment component proportional to the degree of mixing. This mixing can occur as a result of the interaction with the photoelectrons, and we assume this mechanism below.

The system's Hamiltonian without allowance for the direct exchange between the magnetic moments of the atoms can be written in the form

$$H=\omega\sum_g b_g^*b_g-A\sum_g (b_g^*+b_g)(a_{g\uparrow}^*a_{g\uparrow}-a_{g\downarrow}^*a_{g\downarrow}). \tag{1}$$

Here b_g^* and b_g are the Pauli creation and annihilation operators for the exciton corresponding to the $|0\rangle\to|1\rangle$ transition in the atom g; $a_{g\sigma}^*$ and $a_{g\sigma}$ are the operators for a conduction electron with a spin component σ; A is the electron-atom exchange integral, which is assumed to be positive (it includes the factor J_0^z). The Hamiltonian for the electron-phonon interaction in a multivalley semiconductor of the Ge or Si type has a similar structure, except that the valley number should figure in it instead of the electron spin.

According to (1), in order for the mixing of the states $|0\rangle$ and $|1\rangle$ to occur, the conduction electrons should be spin polarized. When the crystal is illuminated by unpolarized light, the polarization of the photoelectrons can occur only as the result of the magnetization of the crystal. If magnetization fluctuations arise in an originally unmagnetized crystal during its illumination, they entail photoelectron-polarization fluctuations that tend to sustain and intensify the magnetization fluctuations. But the growth of the magnetization fluctuations is impeded by the expenditure of energy on exciton excitation. As a result of the competition between these two factors the fluctuations die down at low light intensities and intensify at high intensities. This means that a phase transition from the unmagnetized into the magnetized state occurs at some critical light intensity.

A quantitative analysis is carried out with the use of phenomenological equations of motion for the occupation numbers of the spin subbands of the conduction band $\nu_\sigma=\langle a_{g\sigma}^*a_{g\sigma}\rangle$ (the symbol $\langle\ldots\rangle$ denotes averaging):

$$\frac{d}{dt}\nu_\sigma=G_\sigma-\frac{\nu_\sigma}{\tau_r}+P_{\sigma,-\sigma}\nu_{-\sigma}-P_{-\sigma,\sigma}\nu_\sigma, \tag{2}$$

where τ_r is the recombination time and $P_{\sigma,-\sigma}$ is the probability for a change to occur in the spin component in unit time.

The effect under consideration is most important in the case of direct band–band transitions. We shall, for simplicity, assume that the conduction electrons interact with the crystal magnetization significantly more strongly than the holes. Then, to first order in the ratio of A to the conduction-band width W, the generation factor for unpolarized or plane-polarized light with frequency Ω can be represented in the form (see, for example, Ref. 6)

$$G_\sigma=R(\Omega-E_g+2A\sigma\eta)^{1/2}\theta(\Omega-E_g+2A\sigma\eta),\qquad \eta=\langle b^*+b\rangle, \tag{3}$$

where E_g is the gap width in the absence of the magnetization η; the constant R, which depends on the energy-band structure, is proportional to the light intensity; $\theta(x)$ is the unit step function; $\hbar=1$.

Since under normal conditions recombination can be assumed to be nonradiative, and it occurs via multistage processes of charge capture by the impurity centers, the recombination process should depend on the

gap width much more weakly than the generation process. Therefore, we can assume that τ_r does not depend on η. Further, in order to simplify the calculation, we shall make the model somewhat cruder by assuming that the coefficients $P_{\uparrow\downarrow}$ and $P_{\downarrow\uparrow}$ in (2) are equal to each other and independent of η at small η ($P_{\sigma,-\sigma}=P$). Thus, in the model being used, the dependence of the numbers ν_σ on the magnetization stems from the dependence of the carrier production on the same quantity.

To investigate the stability of the system against the fluctuations η for $T\Rightarrow 0$, we construct with the aid of (1)–(3) linearized—in η—equations of motion for $\langle b^*\rangle$, $\langle b\rangle$, and $\delta=\nu_\uparrow-\nu_\downarrow$, the time dependence of these quantities being assumed to be exponential ($\sim e^{\lambda t}$). Allowing for the fact that the characteristic variation time λ^{-1} of the fluctuations is long compared to ω^{-1}, we find that for $\Omega>E_g$ the magnetization fluctuations increase in intensity when the light intensity exceeds the critical value

$$V>V_c=U^{-1};$$
$$V=\frac{2R\tau A(\Omega-E_g)^{1/2}}{\omega},\quad U=\frac{A}{\Omega-E_g},\quad \frac{1}{\tau}=\frac{1}{\tau_r}+P. \tag{4}$$

With allowance for (3) the criterion (4) can be expressed in terms of the critical photoelectron concentration ν_c, starting from which the unmagnetized state of the crystal becomes absolutely unstable:

$$\nu>\nu_c=A^{-2}\omega(\Omega-E_g). \tag{5}$$

For $A=0.3$ eV, $\Omega-E_g=0.1$ eV, and $\omega=10^{-3}$ eV, it is $\sim 10^{-4}$ ($\sim 10^{18}$ cm^{-3}).

Let us now investigate the steady state of the system. Instead of explicitly introducing an interaction between the system and the thermostat, it is sufficient to assume on the basis of the condition $T\to 0$ that each atom, whether in darkness or not, is in the ground state, and that the moments of all the atoms are parallel to each other. Allowing for the possibility that the atom will be polarized by the photoelectrons, we write its wave function in the form

$$\Phi=(ub^*+v)\,|0\rangle_{ex}, \tag{6}$$

where $|0\rangle_{ex}$ is the vacuum wave function of the exciton. It follows from (1) and (6) that the magnetization in the steady state is given by the equation

$$\omega\eta-2(1-\eta^2)^{1/2}\alpha(\eta)=0,\quad \alpha=A(\nu_\uparrow-\nu_\downarrow). \tag{7}$$

We can construct an entire family of functions of η with Eq. (7) condition for their extrema. Of these functions we are interested in the one in terms of which the entropy production can be expressed. For this reason, below we shall call it the synergetic potential [see (20), (21)]:

$$K=-\tfrac{1}{2}\omega[1-(1-\eta^2)^{1/2}]-\int\alpha(\eta)\,d\eta. \tag{8}$$

Since as $\eta\to 0$ the quantity $\alpha(\eta)$ also goes to zero, Eq. (7) always has a trivial solution and, furthermore, it can, under certain conditions, also have nontrivial solutions with $\eta\neq 0$. To prove that the nontrivial solution that ensures the minimum of the synergetic potential is the one that should be realized, we need to use the above-obtained results on fluctuation dynamics. Using (3), we represent the synergetic potential (8) in the form of a Landau–Lifshitz-type expansion in powers of η:

$$K=\tfrac{1}{4}\omega\eta^2[1-VU]+\tfrac{1}{16}\omega\eta^4[1-\tfrac{1}{4}VU^3]. \tag{9}$$

Indeed, according to (4) and (9), the nonmagnetic state with $\eta=0$ is stable precisely at the point where $K(0)$ has its minimum value, and loses its stability when the minimum of K corresponds to $\eta\neq 0$. Thus, the physically realizable solutions indeed correspond to the minimum of $K(\eta)$.

It follows from (9) that at frequencies sufficiently far from the absorption edge (under the assumptions made above, when $U<2$), the light-induced phase transition is of second order, and the critical light-intensity value, i.e., the value at which the phase transition occurs, coincides with V_c, (4). If, on the other hand, the light frequency is close to the absorption edge (i.e., if $U>2$), then the light-induced phase transition is of first order, and should occur at some intensity V_1 lower than V_c. The possibility that metals undergo, under the action of light, a discontinuous phase transition from the normal into the superconducting state was pointed out earlier.[8] It was, however, first indicated in a situation similar to the one discussed below.[8]

We consider the case in which for $\eta=0$ the light frequency falls within the transmission band of the crystal, while for $\eta\neq 0$ the light can be absorbed ($E_g-A<\Omega<E_g$). The synergetic potential has in this frequency range a minimum with $\eta\neq 0$, besides the one with $\eta=0$, if the light intensity is sufficiently high. For this to happen in the case in which $E_g-\Omega\ll A$, the intensity should satisfy the following inequality, which follows from (2), (3), and (7) with allowance made for the fact that $P_{\uparrow\downarrow}=\nu_\downarrow=0$:

$$L=(2AR\tau_r/\omega)^2>L_c=4A^{-2}(E_g-\Omega). \tag{10}$$

To the critical intensity value $L=L_c$ corresponds the value $\eta=\eta_c$:

$$\eta_c=2(E_g-\Omega)/A. \tag{11}$$

The solution with $\eta\neq 0$ is stable against relatively weak fluctuations. The question of its stability, however, remains open when the fluctuations are intense. But the magnetic state can in any case be realized, even if as a metastable state, if the crystal is first magnetized by an external field, and then light that will maintain this magnetization is switched on after switching off the field. If we assume that, in the presence of the two $K(\eta)$ minima, the more stable state is the one with the deeper minimum, then the magnetized state becomes more stable than the unmagnetized state beginning with $L>4/3L_c$. The magnetization is then equal to $2\eta_c$.

Light-induced magnetization at low temperatures is possible not only in singlet magnets, but also in antiferromagnets and metamagnets. Especially promising in this connection is the metamagnet EuSe, whose critical field is very low, and which, moreover, possesses a high photoconductivity.[5] The theory, developed in Ref. 8, of the photomagnetization of such materials can

easily be generalized within the framework of the shceme expounded in this section, and the results obtained are qualitatively similar to those obtained for magneto-exciton semiconductors.

2. THE EFFECT OF LIGHT ON THE PHASE TRANSITION IN A FERROMAGNETIC SEMICONDUCTOR

Here we shall investigate the effect of light on a phase transition that occurs in the absence of light. The analysis is carried out, using as an example a ferromagnetic semiconductor in which light causes a Curie-point shift or a change in the order of the phase transition. Naturally, because such a system is not in an equilibrium state, it cannot be characterized by a free energy, and, strictly speaking, the concept of temperature as a thermodynamic parameter of the system does not have any meaning. But these concepts make sense for the thermostat with which the nonequilibrium system in qeustion interacts, and, therefore, temperature figures in the results pertaining to nonequilibrium phenomena.

The analysis will be carried out in the self-consistent field approximation. We shall, for simplicity, assume that the ferromagnet is of the Ising type (i.e., that is has a strong magnetic anisotropy, and that the spin of the magnetic atoms is equal to $\frac{1}{2}$). The Hamiltonian of these spins, which interact with the conduction electrons, can be written in the form

$$H_1=-\frac{1}{2}\sum_{n_{g\sigma}=a_{g\sigma}^{+}a_{g\sigma}} I(\mathbf{g}-\mathbf{f})S_g^{z}S_f^{z}-A\sum S_g^{z}(n_{g\uparrow}-n_{g\downarrow}), \tag{12}$$

Below, instead of the spin operators S_g, we shall use the Pauli operators

$$S_g^{z}=1-2c_g^{+}c_g, \quad S_g^{+}=c_g, \quad S_g^{-}=c_g^{+}. \tag{13}$$

As for the interaction of this system with the thermostat, its details are unimportant, since they do not enter into the final results. Therefore, it is expedient to use that model of the thermostat which will allow the maximum simplification of the calculation. Specifically, we shall assume that the role of a thermostat is played by Fermi quasiparticles, whose Hamiltonian has, when (13) is taken into account, the form

$$H_Q=-Q\sum(c_g^{+}b_{g\mu}+b_{g\mu}^{+}c_g)+\sum\omega_\mu b_{g\mu}^{+}b_{g\mu}. \tag{14}$$

It is assumed that the thermostat operators $b_{g\mu}^{*}$ and $b_{g\mu}$ (g is the atom number and μ stands for the rest of the dynamical variables) anticommute with c_g^{*} and c_g. The spectrum ω_μ of the thermostat quasiparticles is assumed to be continuous (in Ref. 3, where the interaction of light with a ferromagnet in the spin-wave region is considered, it was more convenient to assume the thermostat quasiparticles to be Bose quasiparticles).

The role of the order parameter η in the case under consideration is played by the quantity $\langle S_g^{z}\rangle=1-2m$, where $m=\langle c_g^{*}c_g\rangle$. In order to find m, we construct for the correlators $\langle c^{*}c\rangle$, $\langle c^{*}b\rangle$, and $\langle b^{*}c\rangle$ with the aid of (12)–(14) equations of motion, which can then be solved in the molecular-field approximation with allowance for the properties of the thermostat as a system with a number of degrees of freedom that tends to infinity:

$$\langle b_{g\mu}^{+}b_{g\lambda}\rangle=F_\mu\delta_{\mu\lambda}, \quad F_\mu=\{\exp(\omega_\mu/T)+1\}^{-1}\equiv F(\omega_\mu/T). \tag{15}$$

Assuming that $\omega_\mu \ll J$, and expanding the distribution function F_μ, (15), in powers of ω_μ/T, we obtain from these equations the following expression for the magnetization-fluctuation growth time λ^{-1}:

$$\lambda=\tau^{-1}[\nu/\nu_c(T)-1], \quad \nu_c(T)=8A^{-2}(\Omega-E_g)(T-T_c), \quad T_c=J/2, \quad J=\tfrac{1}{2}\Sigma I(\mathbf{f}). \tag{16}$$

As can be seen from (16), the closer T is to T_c, the lower the critical photoelectron concentration starting from which light-induces magnetization in the crystal. Conversely, when the electron concentration becomes equal to ν_c, the Curie temperature increases by an amount $\Delta T=T-T_c$ connected with ν_c by the relation (16). The foregoing is valid if the light-induced phase transition is of second order. As will be shown below, it can also be of first order. Then the concentration ν_c has the meaning of a threshold concentration, starting from which the paramagnetic state is absolutely unstable.

The steady state of the system can be found from the same equations of motion with the use, as in Ref. 3, of the Laplace transformation. These equations can be linearized by choosing as zero time not the moment at which the light is switched on, but a moment sufficiently distant from it, when the values of the correlators are close to the steady-state values. The asymptotic behavior of $m(t)$ is determined by the singularity of its Laplace transform at the coordinate origin. Then for $m(\infty)$ we obtain the self-consistent expression [see (15)]:

$$m(\infty)=F\left(\frac{J+\alpha}{T}\right), \quad J=J[1-2m(\infty)], \quad \alpha=A(\nu_\uparrow-\nu_\downarrow). \tag{17}$$

For weak magnetizations, it can be obtained from the condition for the extremum of the quantity

$$K=J\eta^2(2-J/T)+\tfrac{2}{3}J\eta^4-\int^{\eta}\alpha(\eta)\,d\eta, \tag{18}$$

which has the structure of the ferromagnet's free energy shifted by the work done by the photoelectrons in establishing the given degree of magnetic order. It will be shown below that the quantity K is the synergetic potential.

As follows from (16) and (18), here also the minimum of the synergetic potential corresponds to the stable state of the system. The formula (18) yeilds for the Curie temperature shift an expression coinciding with (16). For $A=0.6$ eV, $\nu=10^{-4}$, and $\Omega-E_g=0.03$ eV the quantity ΔT attains the value 1.5 K, which is comparable to the light-induced Curie-point shift observed in EuS by Afanas'ev *et al.*[1] Here it is natural to assume that the coefficient of the η^4 term in (18) remains positive when the contribution to it from the photoelectrons is taken into account, so that light does not change the order of the phase transition.

But the phase transition goes over from a continuous into a discontinuous transition (i.e., the term $\sim\eta^4$ changes its sign) when the light frequency falls below some threshold value Ω_1:

$$\Omega_1-E_g=\left[\frac{3\nu_c(T_t)A^4}{32T_c}\right]^{1/3}=\frac{9A}{16}\left(\frac{T_t}{T_c}-1\right)^2, \tag{19}$$

where T_t is the temperature starting from which the paramagnetic state becomes absolutely unstable at the given light intensity. According to (16), it and, hence, Ω_1 increase with the light intensity. If we fix the frequency and raise the light intensity, then the order of the phase transition may be changed: from second to first order.

The situation for $\Omega < E_g$ is similar to the one considered in Sec. 1. If $E_g - \Omega \ll A$, then neglecting the term $\sim\eta^4$ in the potential (18) we obtain a condition for the existence of a K minimum at a finite η. If the photoelectrons can be considered to be totally spin polarized, then this condition is the condition (10) with ω replaced by $2(T - T_c)$, where T is the temperature of the sample. The expression (11) carries over unchanged to the case under consideration.

The main difficulty preventing us from carrying over the results of this section to the case of the Heisenberg magnet is that the direction of the spins of the slow electrons in this magnet is not conserved in the vicinity of T_c, but follows adiabatically the direction of the localized magnetic moment of the crystal, slowly varying in space.[5] But for long ranges of the exchange interaction between the localized spins this effect can be neglected, which enables us to apply the above-obtained results to the Heisenberg magnet as well.

Let us now investigate the behavior of the main kinetic characteristic of nonequilibrium processes (the rate of entropy production) during phase transitions. This quantity is, when we neglect the radiative recombination of the current carriers, and take account of the fact that all the energy of the absorbed light is eventually converted into heat, given by the expression [see (3)]

$$\dot{S} = \frac{\Omega}{2}[G(\Omega - E_g + A\eta) + G(\Omega - E_g - A\eta)], \tag{20}$$

which when allowance is made for the fact that $\partial K/\partial\eta = 0$ in the steady state yields the relation

$$\dot{S} = -\frac{\Omega}{T\tau}\frac{\partial K}{\partial \Omega}, \tag{21}$$

which justifies the term "synergetic potential" for the quantity K, (8), (18).

The entropy generation rate $\dot{S}$ is discontinuous at the light-induced second-order phase-transition point for the same reason as the entropy is in the Landau–Lifshitz theory of equilibrium phase transitions. According to (20) and (3), for small η, when $A\eta \ll \Omega - E_g$, the entropy generation rate is given by the expression

$$\dot{S}(\eta) = \dot{S}(0)[1 - {}^1/_8 U^2\eta^2]; \quad U = A(\Omega - E_g)^{-1}, \qquad \dot{S}(0) = \Omega\nu_R/T\tau, \quad \nu_R = 2R\tau(\Omega - E_g)^{1/2}. \tag{22}$$

It follows from (9) and (18) that, in the vicinity of the phase transition point,

$$\eta^2 = 2(\nu_R/\nu_c - 1)(1 - {}^1/_4 U^2)^{-1} \tag{23}$$

for the magnetoexciton semiconductor and

$$\eta^2 = \nu_c^{-1}\left(\frac{7}{3}T_c\right)[\nu_R - \nu_c(T)]\left[1 - \left(\frac{\Omega_1 - E_g}{\Omega - E_g}\right)^2\right]^{-1} \tag{24}$$

for the ferromagnetic semiconductor, in which the concentrations ν_c and $\nu_c(T)$ are given respectively by the expressions (5) and (16) and Ω_1 is found from (19).

As follows from the formulas (22)–(24), the entropy generation rate is itself continuous at the second-order phase transition point, but the derivative of $\dot{S}$ with respect to the light intensity R exhibits a jump, the derivative $\partial\dot{S}/\partial R$ being smaller in the magnetized state than in the disordered state. If the light frequency does not exceed too much the threshold frequency, beginning with which the second-order phase transition becomes discontinuous, $\partial\dot{S}/\partial R$ may even become negative, i.e., the light may be absorbed at a decreasing rate as its intensity increases. The derivative $\partial\dot{S}/\partial\Omega$ also exhibits a jump at the transition point. This derivative has a higher value in the magnetized state than in the unmagnetized state. Finally, according to (24), a jump of the same sign occurs in the derivative $\partial\dot{S}/\partial T$ (it corresponds at a fixed light intensity to the variation of the ambient temperature).

In those cases in which a first-order phase transition occurs, the transition should be accompanied by a jump in the entropy production rate (in the equilibrium case the entropy itself undergoes a jump). $\dot{S}$ may increase or decrease in the transition from the disordered into the ordered state. For example, it clearly increases when $\Omega < E_g$, since light is not absorbed at all in the disordered state. On the other hand, it may decrease when $\Omega > E_g$. Thus, in the case of a magnetoexciton semiconductor with $U \leq 3$, the inequality $\dot{S}(0) > \dot{S}(\eta)$, which guarantees the decrease of $\dot{S}$ upon the appearance of magnetization, remains valid at all values of $\eta < 1$. At the same time, according to the results of Sec. 1, the phase transition should be discontinuous when $U > 2$.

By analogy with (9), we can expect the above-considered steady state with a homogeneous light-induced magnetization to be unstable under certain conditions. The crystal may break up into magnetized and unmagnetized layers, the former layers having an enhanced and the latter a diminished photocarrier concentration. Since the stratification is due to the flow of photocarriers from one phase into the other, these layers should have a thickness of the order of the diffusion length. The homogeneous state should certainly be stable, in particular, in crystals with high magnetostriction.

In certain crystals, the photoelectrons may, before recombining with the holes, form microregions with a high degree of ferromagnetic order, and localize themselves in them.[5] Thus, besides the collective effect of the photoelectrons on the long-range ferromagnetic order, the electrons can, acting independently of each other, also exert an influence on the short-range order. Consequently, autolocalization of the photoelectrons with appearance of local ferromagnetic order can occur in light of intensity lower than critical, starting from which long-range ferromagnetic order appears.

[1]M. M. Afans'ev, M. E. Kompan, and I. A. Merkulov, Zh. Eksp. Teor. Fiz. 71, 2068 (1976) [Sov. Phys. JETP 44, 1086 (1976)]; Pis'ma Zh. Eksp. Teor. Fiz. 23, 621 (1976) [JETP Lett. 23, 570 (1976)].

[2]V. A. Kochelap, V. I. Pipa, V. N. Piskovoĭ, and V. N. Sokolov, Zh. Eksp. Teor. Fiz. **61**, 2504 (1971) [Sov. Phys. JETP **34**, 1340 (1972)].
[3]V. D. Lakhno and É. L. Nagaev, Zh. Eksp. Teor. Fiz. **74**, 2123 (1978) [Sov. Phys. JETP **47**, 1105 (1978)].
[4]É. L. Nagaev and É. B. Sokolov, Pis'ma Zh. Eksp. Teor. Fiz. **28**, 105 (1978) [JETP Lett. **28**, 97 (1978)].
[5]É. L. Nagaev, Fizika magnitnyk poluprovodnikov (The Physics of Magnetic Semiconductors), Nauka, Moscow, 1979.
[6]J. I. Pankove, Optical Processes in Semiconductors, Prentice-Hall, Englewood Cliffs, N.J., 1971 (Russ. Transl., Mir, Moscow, 1973).
[7]V. F. Elesin, Zh. Eksp. Teor. Fiz. **73**, 355 (1977); **76**, 2218 (1979) [Sov. Phys. JETP **46**, 185 (1977); **49**, 1121 (1979)].
[8]V. M. Matveev and É. L. Nagaev, Fiz. Tverd. Tela (Leningrad) **17**, 2483 (1975) [Sov. Phys. Solid State **17**, 1653 (1975)]; N. S. Lidorenko, V. M. Matveev, and É. L. Nagaev, Dokl. Akad. Nauk SSSR **230**, 1085 (1976) [Sov. Phys. Dokl. **21**, 585 (1976)].
[9]A. I. Larkin and D. E. Khmel'nitskiĭ, Zh. Eksp. Teor. Fiz. **55**, 2345 (1968) [Sov. Phys. JETP **28**, 1245 (1969)].

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