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Optical orientation and alignment of free excitons in GaSe under resonant excitation. Theory.

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A microscopic theory is constructed for optical orientation and alignment of excitons in semiconductors under resonant excitation; the theory is valid for any ratio of their lifetimes to the momentum relaxation time. The exciton radiation is regarded as a result of resonant scattering of light with multiple elastic scattering of the excited excitons by impurities. It is shown that in the case of n-fold scattering ($n \geq 2$) the probability of backward emission of the light, i.e., at scattering angles close to π, is increased by two times. The theory takes into account the influence of the Faraday rotation, dichroism and birefringence of the light propagating in a crystal in a magnetic field. The influence of inhomogeneous broadening is analyzed. The developed is used to calculate the change in the intensity and in the degree of polarization of exciton radiation in GaSe in longitudinal and transverse magnetic fields following excitation with linearly and circularly polarized light.

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INTRODUCTION

Optical orientation of excitons in semiconductors was first observed by Gross, Ekimov, Razbirin, and Safarov in CdSe crystals.[1] This phenomenon was observed by now in many semiconductors (see the review[2]).

In GaSe crystals, the exciton orientation was first observed by Veshchunov, Zakharchenya, and Leonov.[3] In[3], as well as in a later study[4] the excitons were produced by binding free electrons and holes excited with light. In[5], where resonant excitation was used, both orientation and alignment of the excitons were observed, in accordance with the predictions of the theory.[6] The character of the depolarization of the exciton radiation in the magnetic field (the Hanle effect) differed qualitatively from that customarily observed for excitons and free carriers. Calculation within the framework of the phenomenlogical theory[6] with allowance for the singularities of the band structure of GaSe does not explain the dependences of the polarization on the magnetic field observed in[5]. It was suggested in[5] that the character of these dependences is connected with the fact that in GaSe the exciton lifetime τ_0 is comparable with the momentum relaxation time τ_r, and an analogy was noted there between the orientation of free excitons and the orientation of atoms in gases under cascade excitation. The role of the different energy levels in the atom under resonant excitation of excitons is played in this case by states with different momenta.

If τ_0 and τ_r are comparable in value, the phenomenological theory of[6] does not hold. In this paper we develop a microscopic theory of optical orientation of excitons, which is valid for an arbitrary ratio of τ_0 and τ_r and takes into account the singularities of the optical properties of GaSe.

In the first part of the article we derive a general expression for the density matrix of the secondary radiation following resonant excitation of the excitons. It is shown that this radiation can be regarded as a result of the resonant scattering of light in multiple scattering of the excited excitons by the impurities. In the general case it is necessary to take into account here scattering in all orders, and this can be done in a sufficiently weak interaction of excitons and photons in the crystal The description of the secondary radiation as resonant scattering is a consistent quantum-mechanical description,[1)] that takes into account, in particular, interference phenomena that occur in backward scattering of light. At $\tau_0 \gg \tau_r$, when the predominant contribution is made by high-order scattering, the derived expressions coincide with the formulas of the phenomenological theory that regards secondary radiation as a result of exciton luminescence.

In the second part of the article we calculate the change in the degree polarization of exciton radiation in GaSe under resonant excitation in longitudinal and transverse magnetic fields.

The present paper is the theoretical part of a joint experimental and theoretical investigation of optical orien-

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tation and alignment of excitons in GaSe. A separate article will be devoted to a detailed description of the procedure and results of the experiment and to their comparison with the conclusions of the theory developed here.

I. OPTICAL ORIENTATION IN THE CASE OF NONEQUILIBRIUM EXCITON MOMENTUM DISTRIBUTION

1. General formula for the secondary radiation density matrix

We consider a semi-infinite crystal on the surface $z=0$ of which the exciting light is normally incident. The intensity and polarization of the exciting light in the crystal are determined by the density matrix $d^0_{\alpha\beta}(\omega, z)$ in a basis of polarization vectors $\mathbf{e}_\alpha$. The density matrix of the light radiated by the excitons from the volume V_0, situated at a distance z from the crystal surface, is connected with the exciton Green's function $F_{mm'}(\mathbf{q}, \omega', z)$ by the relation

$$d_{\alpha\beta}(\omega', \mathbf{q}, z)=\sum_{mm'} j_m^{\alpha *} j_{m'}^{\beta} F_{mm'}(\mathbf{q}, \omega', z),$$
$$F_{mm'}(\mathbf{K}, \omega')=\int_{-\infty}^{+\infty} dt'\, e^{i\omega' t'} \langle 0|\tilde{b}_{\mathbf{K}m'}(t)\tilde{b}_{\mathbf{K}m}(t+t')|0\rangle. \quad (1.1)$$

Here $\mathbf{K}$ and m are the wave vector and the index of the spin state of the exciton; $\tilde{b}^+_{\mathbf{K}m}(t)$ and $\tilde{b}_{\mathbf{K}m}(t)$ are the operators of creation and annihilation of the exciton in the state $(\mathbf{K}, m)$ in the Heisenberg representation; ω' and $\mathbf{q}$ are the frequency and wave vector (in the crystal) of the radiated light. $|0\rangle$ is the ground state of the crystal, and $j^\alpha_m = \langle m|\mathbf{j}\mathbf{e}_\alpha|0\rangle$ is the matrix element of the current-density operator following excitation of the exciton into the state m. Formula (1.1) follows directly from the known relation between the vector potential of the radiated light and the extraneous current produced when the crystal is excited (see, e.g., [8]), if we change over to the second-quantization representation in the indicated relation. The value V_0 of the radiating region is chosen in this case such that, on the one hand, the density matrix $d^0_{\alpha\beta}(z)$ hardly changes in the region, and on the other hand, the linear dimensions of the region exceed the wavelength of the light. We note that in (1.1) and subsequently we leave out inessential factors that do not depend on the polarization indices. We consider the case of resonant excitation with light whose average frequency corresponds to the energy of the ground state of the exciton, and whose spectral line width is smaller than the distance between the ground and excited states. Therefore the summation in (1.1) is only over the spin indices of the exciton in the ground state $n=1$.

We calculate the Green's function $F_{mm'}(\mathbf{K}, \omega')$ first for monochromatic excitation. We use for the calculation the diagram technique developed by Keldysh[9] for nonequilibrium systems. To this end we introduce, in analogy with,[9] a contour C with two branches parallel to the time axis, the upper from $-\infty$ to $+\infty$ and the lower from $+\infty$ to $-\infty$. In the approximation linear in the intensity of the exciting light, the single-particle Green's function in (1.1) can be expressed in terms of the two-particle Green's function of the excitons in the absence of exciting light:

$$\langle 0|\tilde{b}^+_{\mathbf{K}m_2}(t_2)\tilde{b}_{\mathbf{K}m_1}(t_1)|0\rangle = \frac{1}{(\hbar c)^2}\sum_{\substack{m_3m_4\\ \alpha\beta}} j_{m_4}^{\alpha} j_{m_3}^{\beta *} d_{\alpha\beta}{}^0(\omega, z)\int_{-\infty}^{+\infty} dt_3\, dt_4\, e^{i\omega(t_3-t_4)} G^{II}_{m_1m_2m_3m_4}(t_1t_2t_3t_4; \mathbf{K}, \mathbf{q}_0), \quad (1.2)$$

$$G^{II}_{m_1m_2m_3m_4}(t_1t_2t_3t_4; \mathbf{K}, \mathbf{K}') = \langle 0|T_C b_{\mathbf{K}m_1}(t_{1+}) b_{\mathbf{K}m_2}(t_{2-}) b_{\mathbf{K}'m_3}(t_{3-}) b_{\mathbf{K}'m_4}(t_{4+}) \exp\left[-\int_C V(\tau)\,d\tau\right]|0\rangle, \quad (1.3)$$

where T_C is the operator of ordering along the contour C, $b^+_{\mathbf{K}m}(t)$ and $b_{\mathbf{K}m}(t)$ are the operators of creation and annihilation in the interaction representation, and $\mathbf{q}_0$ is the wave vector of the exciting light in the crystal. The index $\pm$ indicates the position of the temporal point on the upper (from $-\infty$ to $+\infty$) or lower branch of the contour C, respectively. The operator of the interaction of the excitons with the impurities and with the lattice vibrations V is best represented in the form of two terms, V_1+V_2. The interaction V_1 is responsible for the exciton momentum scattering within the limits of the exciton band. We shall henceforth include in V_1 only scattering by impurities. In this case we have

$$V_1=\sum_{\substack{\mathbf{K}\mathbf{K}'\\ ms}} v_{\mathbf{K}-\mathbf{K}'} \exp\{i(\mathbf{K}-\mathbf{K}')\mathbf{r}_s\}\, b^+_{\mathbf{K}'m} b_{\mathbf{K}m}, \quad (1.4)$$

where $v_{\mathbf{K}-\mathbf{K}'}$ is the amplitude of the scattering of an exciton by an individual impurity center and $\mathbf{r}_s$ is the position of the s-th impurity. The expression (1.3) for the function G^{II} is averaged over the impurity distribution which is assumed, as usual, to be uncorrelated. For simplicity we neglect in (1.4) the scattering with transition from one spin state of the exciton to the other, i.e., we do not take into account the spin relaxation of the excitons. In this case the two-particle Green's function (1.3) is different from zero only at $m_1=m_4$ and $m_2=m_3$.

The operator V_2 describes nonradiative transitions of the exciton into lower states: bound excitons, indirect excitons and electron-hole pairs, etc. At sufficiently low temperatures, when there is no reversed transitions from these states, the contribution of V_2 to the Green's function of the excitons does not depend on the concrete mechanism of the transition and is determined by the "departure" time, i.e., the lifetime of the exciton τ_0, which is assumed henceforth to be independent of the state of the exciton $(\mathbf{K}, m)$.

In our case, when the excitation is produced by monochromatic light and the exciton scattering is elastic, the Green's functions $F_{mm'}(\mathbf{K}, \omega')$ can be written in the form

$$F_{mm'}(\mathbf{K}, \omega')=\delta(\omega-\omega') f_{mm'}(\mathbf{K}, \omega). \quad (1.5)$$

The function $f(\mathbf{K}, \omega)$ determines the probability of the excitation of an exciton with wave vector $\mathbf{K}$ at the frequency of the exciting monochromatic radiation.

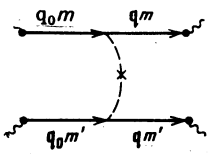

FIG. 1. Simplest diagram for the function $f_{mm'}(\mathbf{q}, \omega)$.

2. Derivation of the equation for the function $f_{mm'}(\mathbf{K}, \omega)$

The simplest diagram that determines $f_{mm'}(\mathbf{K}, \omega)$ is shown in Fig. 1. The upper and lower exciton lines are set in correspondence respectively to the causal and anti-causal Green's functions

$$G_0^c(\omega, \mathbf{K}m) = (\omega - \omega_{\mathbf{K}m} + i\delta)^{-1}, \quad \tilde{G}_0^c(\omega, \mathbf{K}m) = (\omega - \omega_{\mathbf{K}m} - i\delta)^{-1}, \tag{1.6}$$

where $\hbar\omega_{\mathbf{K}m}$ is the exciton energy in the state $(\mathbf{K}, m)$ and $\delta \to +0$. It follows from (1.2) that for each diagram the corresponding expression is multiplied by

$$\sum_{\alpha\beta} j_m^{\alpha} j_{m'}^{\beta *} d_{\alpha\beta}^0(\omega, z).$$

Therefore the diagram of Fig. 1 is set in correspondence with the expression

$$\times \frac{\frac{1}{c^2\hbar^4}\sum_{\alpha\beta} j_m^{\alpha} j_{m'}^{\beta *} d_{\alpha\beta}^0(\omega, z)\, N_s |v_{q-q_0}|^2}{(\omega - \omega_{qm} + i\delta)(\omega - \omega_{qm'} - i\delta)(\omega - \omega_{q_0 m} + i\delta)(\omega - \omega_{q_0 m'} - i\delta)}, \tag{1.7}$$

where N_s is the concentration of the scattering centers.

Substituting (1.7) in (1.5) and (1.5) in (1.1) we obtain the usual expression for the cross section for the scattering of light in single scattering of an exciton by an impurity. Under resonance conditions, however, it is necessary to sum an infinite sequence of diagrams. This summation can be carried out if the average momentum $|\Delta\mathbf{q}|$ are transferred in the exciton scattering exceeds its reciprocal mean free path

$$l_e^{-1} = (v_{q_0}\tau)^{-1}, \quad \frac{1}{\tau} = \frac{1}{\tau_0} + \frac{1}{\tau_r}, \quad v_{q_0} = \frac{\hbar q_0}{m_e},$$

where τ_r is the momentum relaxation time, m_e is the effective mass of the exciton. At $\Delta q \sim q_0$ this is the ordinary condition for the applicability of the kinetic equation

$$q_0 \gg l_e^{-1} \text{ or } \omega_{q_0}\tau \gg 1, \tag{1.8}$$

where $\hbar\omega_{q0} = (2m_e)^{-1}(\hbar q_0)^2$ is the exciton kinetic energy. We also assume the Born approximation to be valid. When these conditions are satisfied, it suffices to take into account in the summation the diagrams without intersection of the impurity lines (Fig. 2a). At $\mathbf{q} + \mathbf{q}_0 \approx 0$ it is necessary to take into account, from among the diagrams with line intersection, those shown in Fig. 2b, which are conveniently represented in the form of Fig. 2c. Each of the diagrams of Fig. 2a or Fig. 2b will be regarded as a sum of diagrams with a specified number of vertical impurity lines and with an arbitrary number of non-intersecting horizontal impurity lines, i.e., lines joining vertices located on one branch of the contour C. This leads to a replacement of the functions G_0^c and $\tilde{G}_0^c$, which are set in correspondence with the exciton lines, by the exact causal and anti-causal Green's functions without allowance for the interaction of the excitons with the light; in this case these functions are defined by the formulas

$$G^c(\omega, \mathbf{K}m) = \tilde{G}^{c*}(\omega, \mathbf{K}m) = \Omega_{\mathbf{K}m}^{-1} = (\omega - \omega_{\mathbf{K}m} + i\Gamma_{\mathbf{K}m})^{-1}, \tag{1.9}$$

where the damping is

$$\Gamma_{\mathbf{K}m}(\omega) = \frac{1}{2}\left[\frac{1}{\tau_0} + \sum_{\mathbf{K}'} Q(\mathbf{K}-\mathbf{K}')\delta(\omega_{\mathbf{K}'m} - \omega)\right], \tag{1.10}$$

$$Q(\mathbf{K}_1 - \mathbf{K}_2) = \frac{2\pi}{\hbar^2} N_s |v_{\mathbf{K}_1 - \mathbf{K}_2}|^2.$$

In formula (1.9), $\omega_{\mathbf{K}m}$ is the exciton frequency renormalized with allowance for the interaction; the quantity $\Gamma_{\mathbf{K}m}$ includes the damping due to the nonradiative departure from the exciton band.

The diagram of Fig. 2a, of order $2n$, describes ordinary scattering of light with n-fold scattering of the excited excitons by the impurities. The diagram in Fig. 2c describes the interference contribution made to the scattering cross section by the backwards scattering. This contribution is of general character, independent of the actual scattering mechanism, and is due to the fact, as seen from Fig. 3, in backward scattering the phase shift of the ray scattered directly by centers 1, 2, ..., n coincides exactly with the phase shift of the ray scattered in succession by the centers $n, n-1, \ldots, 1$. As a result, the intensity of the backwards scattering of n-th order is doubled at $n \geq 2$.[2)]

We now derive an equation for the function $f_{mm'}(\mathbf{K}, \omega)$. It is convenient to write it in the form

$$f = f^{(1)} + f^{(2)} - f_1,$$

where $f^{(1)}$ corresponds to the sum of diagrams of the type 2a, while $f^{(2)}$ corresponds to the sum of diagrams

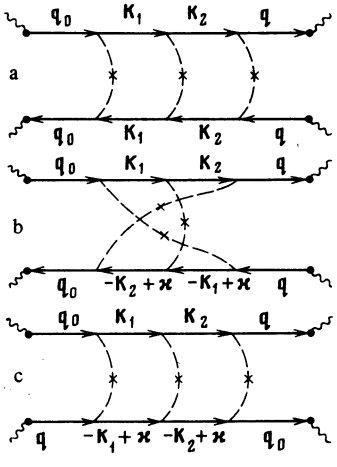

FIG. 2. Normal (a) and anomalous (b, c) diagrams for the function $f_{mm'}(\mathbf{q}, \omega)$.

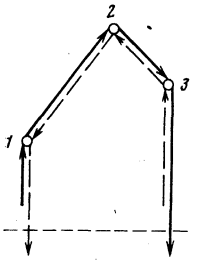

FIG. 3. Interference of backward-scattered waves (in multiple scattering).

2c. The diagram f_1 with one vertical impurity line was included by us not only in $f^{(1)}$ but also in $f^{(2)}$. With this definition, as seen from Figs. 2a and 2c, $f^{(2)} = f^{(1)}$ at $\mathbf{q}+\mathbf{q}_0 = 0$.

Summing diagrams 2a and 2c, which reduces in this case to summation of two "ladders," we obtain for $f^{(1)}$ and $f^{(2)}$ the following equations:

$$\left(\frac{1}{\tau_0} + i\omega_{mm'} + \hat{I}\right) f^{(1)}_{mm'}(\mathbf{K}, \omega) = \Delta_{mm'}(\mathbf{q}_0, \omega) g_{mm'}(\omega) \delta_{\mathbf{K},\mathbf{q}_0}, \tag{1.11a}$$

$$\left[\frac{1}{\tau_0} + i(\omega_{\mathbf{K}m} - \omega_{-\mathbf{K}+\varkappa,m'}) + \hat{I}\right] f^{(2)}_{mm'} = \bar{\Delta}_{mm'}(\mathbf{q}_0, \omega) g_{mm'}(\omega) \delta_{\mathbf{K},\mathbf{q}_0}, \tag{1.11b}$$

where

$$\Delta_{mm'}(\mathbf{K}, \omega) = (2\Gamma_{\mathbf{K}} + i\omega_{mm'})/2\pi\Omega_{\mathbf{K}m}\Omega^{*}_{\mathbf{K}m'}, \tag{1.12a}$$

$$\bar{\Delta}_{mm'}(\mathbf{K}, \omega) = [2\tilde{\Gamma}_{\mathbf{K}} + i(\omega_{\mathbf{K}m} - \omega_{-\mathbf{K}+\varkappa,m'})]/2\pi\Omega_{\mathbf{K}m}\Omega^{*}_{-\mathbf{K}+\varkappa,m'}; \tag{1.12b}$$

$$\varkappa = \mathbf{q}+\mathbf{q}_0, \quad \omega_{mm'} = \omega_{\mathbf{K}m} - \omega_{\mathbf{K}m'}, \quad g_{mm'}(\omega) = \sum_{\alpha\beta} j_m{}^{\alpha} j_{m'}{}^{\beta *} d_{\alpha\beta}{}^{0}(\omega, z),$$

$$2\tilde{\Gamma}_{\mathbf{K}} = \Gamma_{\mathbf{K}} + \Gamma_{-\mathbf{K}+\varkappa},$$

$$\hat{I} f^{(1)}_{mm'} = \sum_{\mathbf{K}'} Q(\mathbf{K}-\mathbf{K}') [\Delta_{mm'}(\mathbf{K}', \omega) f^{(1)}_{mm'}(\mathbf{K}, \omega) - \Delta_{mm'}(\mathbf{K}, \omega) f^{(1)}_{mm'}(\mathbf{K}', \omega)], \tag{1.13}$$

and the expression for $\hat{\tilde{I}} f^{(2)}_{mm'}$ is determined by a formula that differs from (1.13) in that $\Delta_{mm'}(\mathbf{K}, \omega)$ is replaced by $\bar{\Delta}_{mm'}(\mathbf{K}, \omega)$. We have taken into here the fact that at $\omega_{q_0}\tau \gg 1$ and $|\omega_{mm'}| \lesssim \tau^{-1}$ the value of $\Gamma_{\mathbf{K}}$ is independent of m, and at a fixed direction $\mathbf{K}$ does not change in the range of values of K satisfying the condition $|\omega_{\mathbf{K}m} - \omega| \lesssim \Gamma_{\mathbf{K}}$ and greatly exceeds the renormalization of the difference in frequencies $\omega_{mm'}$ as a result of the collisions.

The term f_1 which enters in $f_{mm'}(\mathbf{K}, \omega)$ is equal to

$$f_{1,mm'}(\mathbf{q}, \omega) = \frac{g_{mm'}(\omega) Q(\mathbf{q}-\mathbf{q}_0)}{(2\Gamma_{\mathbf{q}} + i\omega_{mm'})(2\Gamma_{\mathbf{q}_0} + i\omega_{mm'})} \Delta_{mm'}(\mathbf{q}, \omega) \Delta_{mm'}(\mathbf{q}_0, \omega). \tag{1.14}$$

3. Allowance for the change of the polarization of light as it propagates in the crystal

Formulas (1.1), (1.5), and (1.11) determine the density matrix of the radiation from a small volume V_0 in the immediate vicinity of the volume. We are interested in the radiation emitted by the entire crystal and propagating in a narrow solid angle in a direction opposite to the excitation direction. To calculate the density matrix of this radiation it is necessary to integrate expression (1.1) over the entire excited volume. It is necessary to take into account here the fact that when light propagates in a crystal in a magnetic field, its intensity and polarization prior to the absorption point and after the emission can be altered as a result of absorption, dichroism, birefringence, or Faraday rotation. We assume that in the absence of a magnetic field, at the chosen light propagation direction z, a crystal has an optically isotropic behavior. For a uniaxial crystal, such a direction is the principal symmetry axis.

The polarization basis vectors $\mathbf{e}_\alpha$ are conveniently chosen such that the tensor of the transverse permittivity $\varepsilon^{\perp}_{\alpha\beta}$ in this basis is diagonal. We separate here in $\varepsilon^{\perp}_{\alpha\beta}$ the resonance contribution due to excitation of the exciton $n = 1$

$$\varepsilon_{\alpha\beta}{}^{\perp}(\omega, \mathbf{q}) = \delta_{\alpha\beta}\left[n_0{}^2 - \frac{4\pi}{\hbar\omega^2}\sum_{m} \frac{|j_m{}^{\alpha}|^2}{\Omega_{\mathbf{q}m}}\right]. \tag{1.15}$$

Here n_0^2 is the nonresonant (background) isotropic component of the permittivity. The basis $\mathbf{e}_\alpha$ depends on the direction of the magnetic field. In this basis, each of the states m is excited by light of only one polarization, i.e.,

$$j_m{}^{\alpha} j_m{}^{\beta *} = |j_m{}^{\alpha}|^2 \delta_{\alpha\beta}.$$

The density matrix of the exciting light is given in the basis $\mathbf{e}_\alpha$ by

$$d_{\alpha\beta}{}^{0}(\omega, z) = d_{\alpha\beta}{}^{0}(\omega) \exp\{i(\tilde{q}_0{}^{\alpha} - \tilde{q}_0{}^{\beta *})z\}, \tag{1.16}$$

where $d^0_{\alpha\beta}(\omega) = d^0_{\alpha\beta}(\omega, 0)$, and $\tilde{q}^{\alpha}_0$ is the root of the dispersion equation

$$(c\tilde{q}/\omega)^2 = \varepsilon_{\alpha\alpha}{}^{\perp}(\omega, \tilde{\mathbf{q}}).$$

The density matrix of the light radiated backwards, i.e., $d_{\alpha\beta}(\omega', \mathbf{q})$, is determined at $\mathbf{q} = -\mathbf{q}_0$ by the relation

$$d_{\alpha\beta}(\omega', -\mathbf{q}_0) = \int_{z>0} dz \exp\{i(\tilde{q}_0{}^{\alpha} - \tilde{q}_0{}^{\beta *})z\}\, d_{\alpha\beta}(\omega', -\mathbf{q}_0, z). \tag{1.17}$$

Substituting formulas (1.1), (1.5), and (1.16) in this expression, we obtain after integrating with respect to ω and ω' the following expression for the integral density matrix $d_{\alpha\beta}(-\mathbf{q}_0)$ for nonmonochromatic excitation:

$$d_{\alpha\beta}(-\mathbf{q}_0) = \iint d\omega\, d\omega'\, d_{\alpha\beta}(\omega', -\mathbf{q}_0) = \int d\omega \sum_{mm'} \frac{j_m{}^{\alpha *} j_{m'}{}^{\beta} f_{mm'}(-\mathbf{q}_0, \omega)}{-2i(\tilde{q}_0{}^{\alpha} - \tilde{q}_0{}^{\beta *})}. \tag{1.18}$$

The quantity $f_{mm'}(\mathbf{q}, m)$ in (1.18) is the solution of Eqs. (1.11), in which $d^0_{\alpha\beta}(\omega, z)$ is replaced by the value of $d^0_{\alpha\beta}(\omega)$ on the crystal boundary.

4. Criteria for the applicability of the theory

Let us indicate the conditions under which formulas (1.17) and (1.18) are valid. We neglect reradiation, i.e., the secondary production of excitons by the radiated light. This approximation is correct if the nonradiative lifetime τ_0 is small in comparison with the radiative time $\tau_{\text{rad}} = l_{\text{rad}} v_{q_0}^{-1}$, where l_{rad} is the mean free path of the exciton with respect to photon radiation and

is given by

$$l_{\text{rad}}^{-1}=q_0\omega_{LT}/\Gamma. \tag{1.19}$$

Here ω_{LT} is the longitudinal-transverse splitting for the exciton:

$$\omega_{LT}=\frac{4\pi}{\hbar\omega^2 n_0^2}\sum_m |j_m^\alpha|^2. \tag{1.20}$$

Thus, the condition $\tau_{\text{rad}} \gg \tau_0$ indicated above can be expressed in the form

$$\omega_{LT}\tau \ll (\omega_{q_0}\tau_0)^{-1}, \tag{1.21}$$

where $\tau=(2\Gamma)^{-1}$.

Equations (1.11) for $f_{mm'}$ presume the possibility of separating a volume V_0 with linear dimensions that are small in comparison with the quantity $l_{\text{ph}}=|q_0^\alpha-q_0^{\beta*}|^{-1} \approx l_{\text{rad}}$, which determines the length over which a noticeable change takes place in the intensity and polarization of the light, but are large in comparison with the diffusion length of the exciton $l_D=v_{q0}(\tau_0\tau)^{1/2}$, so that we can neglect in (1.11) the diffusion transport of the excitons. This locality condition $l_D \ll l_{\text{rad}}$ can be written in the form

$$\omega_{LT}\tau \ll (\omega_{q_0}\sqrt{\tau_0\tau})^{-1}. \tag{1.22}$$

In addition, we neglect throughout the polariton effect, i.e., we regard the photons and excitons in the crystal as weakly interacting particles. This approach is valid when the exciton mean free path l_e is small in comparison with l_{rad}, i.e.,

$$\omega_{LT}\tau \ll (\omega_{q_0}\tau)^{-1}. \tag{1.23}$$

It is seen that the condition (1.21) is the most stringent and if it is satisfied the inequalities (1.22) and (1.23) are also satisfied. Since Eqs. (1.11) were derived under the condition $\omega_{q0}\tau \gg 1$, the inequality (1.23) leads also to the inequality $\omega_{LT} \ll \tau^{-1}=2\Gamma$. At $\omega_{LT}\tau \ll 1$ the resonant contribution to $\varepsilon_{\alpha\beta}^{\perp}$ is small in comparison with the nonresonant one. Therefore the coefficient of light transmission through the crystal boundary is practically independent of the frequency, and

$$\tilde{q}_0^\alpha-\tilde{q}_0^{\beta*}=-\frac{2\pi}{cn_0\hbar\omega}\sum_m\left(\frac{|j_m^\alpha|^2}{\Omega_{q_0m}}-\frac{|j_m^\beta|^2}{\Omega_{q_0m}^*}\right). \tag{1.24}$$

5. Allowance for inhomogeneous broadening

As shown by experiment,[5] when the excitation is by a broad line $\Delta\omega \gg \Gamma$ the exciton-radiation line width in GaSe is $\delta\omega \gg \Gamma$ and is determined by the inhomogeneous broadening of the exciton spectrum. The influence of the inhomogeneous broadening on the polarization of the radiated light is substantially determined by the correlation length a of the exciton energy fluctuations. At $a \gg l_{\text{ph}} \approx l_{\text{rad}}$, formula (1.18) above remains in force, since no averaging of the quantity $\tilde{q}_0^\alpha-\tilde{q}_0^{\beta*}$ takes place over the light absorption depth l_{ph}, and at $\Delta\omega \gg \Gamma$ averaging over the transverse coordinates does not change the final expression for the integral density matrix of the photons. At $l_{\text{ph}} \gg a \gg q_0^{-1}$, the exponential in (1.16) and (1.17) is replaced by

$$\exp\left\{i\int_0[\tilde{q}_0^\alpha(z_1)-\tilde{q}_0^{\beta*}(z_1)]dz_1\right\}\approx\exp[i(\bar{q}^\alpha-\bar{q}^{\beta*})z],$$

where $\bar{q}^\alpha$ is the value of $\tilde{q}_0^\alpha(\omega)$ averaged over the exciton energy. If the inhomogeneous broadening is $\delta\omega \gg \Gamma$, then we get from (1.24)

$$\bar{q}^\alpha-\bar{q}^{\beta*}=i\pi q_0\omega_{LT}/\delta\omega. \tag{1.25}$$

We took account of the fact that the sum $\sum_m |j_m^\alpha|^2$ in (1.20) does not depend on the polarization α. Thus, at $a \ll l_{\text{ph}}$ there is no birefringence or dichroism of absorption of light at the absorption length, which in this case is equal to $(2\,\text{Im}\bar{q}^\alpha)^{-1}$. If furthermore $a \gg l_D$, then the locality condition is preserved and we have according to (1.18)

$$d_{\alpha\beta}(-\mathbf{q}_0)=\sum_{mm'} j_m^{\alpha*}j_{m'}^\beta f_{mm'}(-\mathbf{q}_0), \tag{1.26}$$

where

$$f_{mm'}(\mathbf{q})=\int d\omega\, f_{mm'}(\mathbf{q},\omega).$$

6. Solution of the equations for $f_{mm'}(\mathbf{K},\omega)$ in the isotropic approximation

It is seen from (1.11) that at $|\omega_{\mathbf{q}0m}-\omega| \lesssim \Gamma$ scattering by impurity centers leads to excitation of excitons that likewise have only $|\omega_{\mathbf{K}m}-\omega| \lesssim \Gamma$, i.e., the scattering is accompanied by a small change of $\omega_{\mathbf{K}m}$. For the sake of simplicity we shall consider henceforth the case of spherical isotropy of $\omega_{\mathbf{K}m}$ and $Q(\mathbf{K}-\mathbf{K}')$. In this case the function $Q(\mathbf{K}-\mathbf{K}')$ at $K' \approx K$ can be expanded in Legendre polynomials $P_l(\cos\theta')$, where θ' is the angle between the vectors K and K':

$$Q(\mathbf{K}-\mathbf{K}')=Q(K,\cos\theta')=\sum_l Q_l P_l(\cos\theta').$$

We substitute this expansion in (1.11a), multiply by $P_{l'}(\cos\theta)$, where θ is the angle between K and q_0, and integrate over the solid angle. As a result we obtain independent expressions for the corresponding components $f_{mm',l}^{(1)}(K,\omega)$ of the expansion of $f_{mm'}^{(1)}(\mathbf{K},\omega)$ in Legendre polynomials. Next, summing these equations over K, we get

$$\int K^2\,dK\, f_{mm',l}^{(1)}(K,\omega),$$

after which the functions $f_{mm',l}^{(1)}(K,\omega)$ are readily determined. At $\mathbf{q}=-\mathbf{q}_0$, when $f=2f^{(1)}-f_1$, we obtain the following expression for $f_{mm'}(-\mathbf{q}_0,\omega)$:

$$f_{mm'}(-\mathbf{q}_0,\omega)=\frac{g_{mm'}(\omega)}{\tau^{-1}+i\omega_{mm'}}\Delta_{mm'}(-\mathbf{q}_0,\omega)\Delta_{mm'}(\mathbf{q}_0,\omega)$$

$$\times\sum_{l=0}^{\infty}(-1)^l Q_l\left[\frac{2}{\tau_0^{-1}+i\omega_{mm'}+2(\Gamma_p^0-\Gamma_p^l)}-\frac{1}{\tau^{-1}+i\omega_{mm'}}\right], \tag{1.27}$$

where $\Gamma_p^0 = \Gamma - 1/2\tau_0$ and $\Gamma_p^l = \Gamma_p^0 Q_l/(2l+1)Q_0$. At $Q(\cos\theta) = Q_0 = \text{const}$, when only the principal term with $l=0$ remains in (1.27), we have

$$f_{mm'}(-\mathbf{q}_0, \omega) = g_{mm'}(\omega)\frac{\Delta_{mm'}(-\mathbf{q}_0, \omega)\Delta_{mm'}(\mathbf{q}_0, \omega)}{(1+i\omega_{mm'}\tau_0)(1+i\omega_{mm'}\tau)} \times Q_0\tau\tau_0\left[2 - \frac{\tau}{\tau_0}\frac{1+i\omega_{mm'}\tau_0}{1+i\omega_{mm'}\tau}\right]. \quad (1.28)$$

According to (1.26), in the case of strong inhomogeneous broadening at a depth l_{ph}, the radiation polarization is determined by the quantity $f_{mm'}(-\mathbf{q}_0)$. In the considered approximation we have in accordance with (1.28)

$$f_{mm'}(-\mathbf{q}_0) = \frac{Q_0\tau\tau_0 g_{mm'}}{\pi(1+i\omega_{mm'}\tau_0)(1+i\omega_{mm'}\tau)^2}\left[2 - \frac{\tau}{\tau_0}\frac{1+i\omega_{mm'}\tau_0}{1+i\omega_{mm'}\tau}\right]. \quad (1.29)$$

When only diagrams of type 2a are considered, the expression in the square brackets in (1.28) and (1.29) is replaced by unity. It is seen that at $\tau_0 \ll \tau_r$, i.e., as $\tau \to \tau_0$, or at $|\omega_{mm'}| \gg \tau^{-1}$, the contribution of the diagrams 2c is immaterial. If we take into account in (1.27) the terms with $l \neq 0$ and the function $Q(\cos\theta)$ is smooth enough, when $Q_l/Q_0 < 2l+1$, the second term in the brackets of (1.28) or (1.29) acquires an additional factor $[2 - Q(-1)/Q_0]$.

Formulas (1.28) and (1.29) are valid for backward scattering. When the direction of the scattered light is inclined to the normal by an angle $\tilde{\theta}$, it can be shown, by solving Eq. (1.11b) by the iteration method, that the contribution of the diagrams 2c decreases: when terms up to second order in $\varkappa$ inclusive are taken into account, the relative change of the contribution of these diagrams is

$$\frac{\Delta f_{mm'}^{(2)}}{f_{mm'}^{(1)} - f_{1,mm'}} = -\frac{8}{3}\frac{(\omega_{q_0}\tilde{\theta})^2\tau_0\tau}{(1+i\omega_{mm'}\tau_0)(1+i\omega_{mm'}\tau)}. \quad (1.30)$$

Thus, the contribution of $f^{(2)} - f_1$ is significant at $\tilde{\theta} \lesssim (q_0 l_D)^{-1}$, i.e., at $\varkappa l_D \lesssim 1$. Therefore at $\tau_0 \sim \tau$ the polarization of the radiation should change substantially when the scattering angle is changed by an amount on the order of $(q_0 l_D)^{-1}$.

7. Transition to the limit of the phenomenological theory

We consider the limiting case $\tau_0^{-1}, |\omega_{mm'}| \ll \tau_r^{-1}$. We note first that at large values of τ_0 the limiting angle $\tilde{\theta}_M = (q_0 l_D)^{-1}$ at which the difference $f^{(2)} - f_1$ is comparable with $f^{(1)}$ is small and when the scattering light is observed in a sufficiently large solid angle the contribution from $f^{(2)} - f_1$ vanishes, and therefore $f = f^{(1)}$. At $|\omega_{mm'}| \ll \tau_r^{-1}$, as seen from (1.24), the birefringence and dichroism are insignificant in a magnetic field. In the zeroth approximation in the parameters τ_r/τ_0 and $\omega_{mm'}\tau_r$, the function $f_{mm'}^0(\mathbf{K}, \omega)$ satisfies the equation $\hat{I} f_{mm'}^0(\mathbf{K}, \omega) = 0$ and is equal to

$$f_{mm'}^0(\mathbf{K}, \omega) = C_{mm'}(\omega)\Delta_{mm'}(\mathbf{K}, \omega). \quad (1.31)$$

Inasmuch as $|\omega_{mm'}| \ll \tau_r^{-1}$ in this case, the quantity $\Delta_{mm'}(\mathbf{K}, \omega) = \Delta(\mathbf{K}, \omega)$ does not depend on the spin indices, i.e., $f_{mm'}(\mathbf{K}, \omega)$ can be written in the form of a product of the function $C_{mm'}(\omega)$, which depends only on the spin indices, and the function $\Delta(\mathbf{K}, \omega)$, which depends only on $\mathbf{K}$. At an arbitrary ratio of τ_r and τ_0, such a representation, as seen from (1.27) and (1.28), is impossible. To determine $C_{mm'}(\omega)$ we sum the right-hand and left-hand sides of (1.11a):

$$\sum_{\mathbf{K}} f_{mm'}(\mathbf{K}, \omega) = \frac{g_{mm'}(\omega)}{\tau_0^{-1} + i\omega_{mm'}}\Delta_{mm'}(\mathbf{q}_0, \omega). \quad (1.32)$$

We substitute in this relation $f_{mm'}^0(\mathbf{K}, \omega)$ in the form (1.31) and integrate with respect to ω. We then obtain for the matrix

$$\rho_{mm'} = \int d\omega \sum_{\mathbf{K}} f_{mm'}^0(\mathbf{K}, \omega) = \int d\omega\, C_{mm'}(\omega) \sum_{\mathbf{K}} \Delta_{mm'}(\mathbf{K}, \omega)$$

the equation

$$(1/\tau_0 + i\omega_{mm'})\rho_{mm'} = g_{mm'}. \quad (1.33)$$

According to (1.18), (1.31), and (1.32), the integral density matrix of the radiated light is in this case

$$d_{\alpha\beta}(\mathbf{q}) \sim \sum_{mm'} j_m^{\alpha *} j_{m'}^{\beta} \int d\omega\, f_{mm'}^0(\mathbf{q}, \omega) \sim \sum_{mm'} j_m^{\alpha *} j_{m'}^{\beta} \rho_{mm'}. \quad (1.34)$$

The expressions (1.33) and (1.34) were used earlier in[6], where a phenomenological theory of optical orientation of excitons in semiconductors was developed. The equation for the matrix $\rho_{mm'}$ in (1.33) was written in the diagonal representation. In an arbitrary basis, with allowance for spin relaxation and also for the possible dependence of the recombination rate on the spin state, this equation can be rewritten in the form[6]

$$\left(\frac{\partial\hat{\rho}}{\partial t}\right)_{\text{rec}} + \left(\frac{\partial\hat{\rho}}{\partial t}\right)_{\text{s.r.}} - \frac{i}{\hbar}[\hat{\mathscr{H}}, \hat{\rho}] + \hat{g} = 0. \quad (1.35)$$

It was indicated in[6] that the phenomenological theory, strictly speaking, is valid for bound excitons, for which the microscopic and phenomenological theories are equivalent. The derivation given above shows that at $\tau_r \ll \tau_0 \ll \tau_{rad}$ the theory can be used also to describe optical orientation of free excitons.[3)] We consider below, using GaSe crystals as an example, the qualitative differences that result at $\tau_0 \sim \tau_r$ from the use of the exact formulas of the microscopic theory in comparison with the results of the phenomenological theory.

II. POLARIZATION OF EXCITON RADIATION OF GaSe IN A MAGNETIC FIELD

1. Exciton spectrum and selection rules

It is known that GaSe is crystallized in three modifications, $\beta(D_{6h})$, $\gamma(C_{3v})$ and $\varepsilon(D_{3h})$. All the modifications have a similar band structure (see, e.g., the review[13] where detailed references to the original papers are given). We shall therefore consider henceforth only the ε modification. The lower conduction band and the upper valence band at the point are simple and correspond to the representations $\Gamma_4(A_2^-)$ and $\Gamma_1(A_1^+)$. When the spin is taken into account, the representation Γ_4 goes over

into Γ_8, and Γ_1 into Γ_7. A noticeable spin-orbit mixing takes place then in the nearest bands Γ_1 and Γ_5, which split into $\Gamma_7+\Gamma_9$. The ground state of the exciton splits as a result of the interaction of the electrons and the holes into three terms: $\Gamma_8\times\Gamma_7=\Gamma_3+\Gamma_4+\Gamma_6(A_1^++A_2^-+E^+)$. The exchange-interaction Hamiltonian $\mathcal{H}_{ex}$ takes the form

$$\mathcal{H}_{ex}=-{}^1/_2({}^1/_2\Delta_1\sigma^e\sigma^h+\Delta\sigma_z^e\sigma_z^h), \tag{2.1}$$

where $\sigma_\alpha^{e,h}$ are Pauli matrices for electrons and holes. In the absence of spin-orbit mixing of the bands, we have $\Delta=0$, the terms Γ_3 and Γ_6 have one and the same energy and correspond to a total exciton $\mathbf{S}=\frac{1}{2}(\sigma^e+\sigma^h)$, equal to 1, and $S_z=0,\pm1$. These states will henceforth be labeled by the indices 0 and ±1. The upper level Γ_4 experiences a splitting Δ_1 that amounts according to the data of[13] to approximately 2 meV, and corresponds to a total spin $S=0$.

In the absence of spin-orbit mixing of the bands, direct optical transitions are allowed only at $e_z\neq0$. When account is taken of the spin-orbit mixing, transitions are allowed both to the state Γ_4, for which only the matrix element $j^z_{\Gamma_4}$ differs from zero and to the state Γ_6, for which the following matrix elements are not equal to zero:

$$j^+_{\Gamma_6,1}=-j^-_{\Gamma_6,-1}=j_0,\quad j^\pm=2^{-1/2}(j_x\pm ij_y). \tag{2.2}$$

The splitting of the exciton state in the magnetic field is described by the Hamiltonian

$$\mathcal{H}_H={}^1/_2\mu_0[H_z(g_\parallel^e\sigma_z^e+g_\parallel^h\sigma_z^h)+\mathbf{H}_\perp(g_\perp^e\boldsymbol{\sigma}_\perp^e+g_\perp^h\boldsymbol{\sigma}_\perp^h)], \tag{2.3a}$$

where $\mathbf{H}$ is the magnetic field intensity vector, $\sigma_\perp\mathbf{H}_\perp=\sigma_xH_x+\sigma_yH_y$, and μ_0 is the Bohr magneton. For the three lowest states of $\Gamma_3+\Gamma_6$ with $S_z=0,\pm1$, the Hamiltonian is

$$\mathcal{H}_H={}^1/_2\hbar(S_z\Omega_\parallel+\mathbf{S}_\perp\boldsymbol{\Omega}_\perp), \tag{2.3b}$$

where

$$\hbar\Omega_\parallel=g_\parallel\mu_0H_z,\quad \hbar\boldsymbol{\Omega}_\perp=g_\perp\mu_0\mathbf{H}_\perp,\quad g_{\parallel,\perp}=g^e_{\parallel,\perp}+g^h_{\parallel,\perp}.$$

According to the data of[13] $g_\parallel=2.7\pm0.2$ and $g_\perp=1.9\pm0.15$. If $\hbar/\tau\ll\Delta_1$ and the splitting of the exciton states $\hbar\omega_{mm'}$ is also smaller than Δ_1, then the state Γ_4 is not excited if the exciton is reasonantly excited by light propagating along the principal symmetry axis C_3. We shall therefore consider only the three lowest states.

2. Longitudinal magnetic field (Faraday geometry)

In this case there are two optically active levels ±1 with energies $\omega_{K,\pm1}=\omega_{K,0}\pm\Omega_\parallel/2$, excited respectively by right- and left-polarized light, and the vectors

$$\mathbf{e}_{\pm1}=2^{-1/2}(\mathbf{e}_x\pm i\mathbf{e}_y)$$

are the eigenvectors of the polarization. Correspondingly we have in this case in accordance with (1.24) and (2.2)

$$-i(\tilde q_0^\alpha-\tilde q_0^{\beta*})=\pi q_0\omega_{LT}\Delta_{\alpha\beta}(q_0,\omega), \tag{2.4}$$

where $\alpha_1\beta=\pm1$. Just as above, we assume that the exciting-light line width $\Delta\omega$ greatly exceeds the line width of the exciton absorption. Substituting (2.4) and (1.28) in (1.18) and integrating with respect to ω from $-\infty$ to $+\infty$, we obtain

$$d_{\alpha\beta}=d_{\alpha\beta}^0\left[1-\frac{1}{2}\frac{\tau}{\tau_0}\frac{1+i\omega_{\alpha\beta}\tau_0}{1+i\omega_{\alpha\beta}\tau}\right]\{(1+i\omega_{\alpha\beta}\tau_0)(1+i\omega_{\alpha\beta}\tau)\}^{-1}. \tag{2.5}$$

Here and below we leave out the common factor of $d_{\alpha\beta}$, since it does not depend on the magnetic field and on the polarization of the light. It is seen from (2.5) that the degree of circular polarization of the radiation, apart from the sign, is equal to the degree of circular polarization of the exciting light and does not vary in a longitudinal magnetic field.

In the case of excitation by linearly polarized light, the degree of linear polarization of the radiation, in the coordinate system that coincides with the polarization plane of the exciting light, takes the form

$$\mathcal{P}'_{\text{lin}}(H_\parallel)=\frac{2\,\mathrm{Re}\,d_{1,-1}}{d_{11}+d_{-1,-1}}=\Phi_1(\Omega_\parallel,\tau_0,\tau)\Big/\left(1-\frac{\tau}{2\tau_0}\right), \tag{2.6}$$

where

$$\Phi_1(\Omega,\tau_0,\tau)=\frac{1}{1+\Omega^2\tau^2}\left[\frac{1-\Omega^2\tau_0\tau}{1+\Omega^2\tau_0^2}-\frac{\tau}{2\tau_0}\frac{1-\Omega^2\tau^2}{1+\Omega^2\tau^2}\right]. \tag{2.7}$$

The degree of polarization of the radiation in a coordinate system rotated around the direction of $\mathbf{q}_0$ by an angle 45° to the plane of polarization of the exciting light is

$$\mathcal{P}''_{\text{lin}}(H_\parallel)=-\frac{2\,\mathrm{Im}\,d_{1,-1}}{d_{11}+d_{-1,-1}}=-\left[\frac{\Omega_\parallel(\tau_0+\tau)}{1+\Omega_\parallel^2\tau_0^2}-\frac{\Omega_\parallel\tau^2}{\tau_0(1+\Omega_\parallel^2\tau^2)}\right]\times\left[\left(1-\frac{\tau}{2\tau_0}\right)(1+\Omega_\parallel^2\tau^2)\right]^{-1}. \tag{2.8}$$

If the inhomogeneous broadening is large and $l_D\ll a\ll l_{ph}$, the expressions for $\mathcal{P}'_{\text{lin}}$ and $\mathcal{P}''_{\text{lin}}$ take according to (1.26) and (1.29) the form

$$\mathcal{P}_{\text{lin}}(H_\parallel)=\Phi_2(\Omega_\parallel,\tau_0,\tau)/(1-\tau/2\tau_0), \tag{2.9}$$

where

$$\Phi_2(\Omega,\tau_0,\tau)=\frac{1}{(1+\Omega^2\tau^2)^2}\left[\frac{1-\Omega^2\tau(2\tau_0+\tau)}{1+\Omega^2\tau_0^2}-\frac{\tau}{2\tau_0}\frac{1-3\Omega^2\tau^2}{1+\Omega^2\tau^2}\right]; \tag{2.10}$$

$$\mathcal{P}''_{\text{lin}}(H_\parallel)=\frac{-1}{(1-\tau/2\tau_0)(1+\Omega_\parallel^2\tau^2)^2}\left\{\frac{2\Omega_\parallel\tau+\Omega_\parallel\tau_0(1-\Omega_\parallel^2\tau^2)}{1+\Omega_\parallel^2\tau_0^2}-\frac{\tau}{2\tau_0}\frac{\Omega_\parallel\tau(3-\Omega_\parallel^2\tau^2)}{1+\Omega_\parallel^2\tau^2}\right\}. \tag{2.11}$$

At $\tau_0^{-1}\ll\tau_r^{-1}$ and $\Omega_\parallel\ll\tau_r^{-1}$ formulas (2.6), (2.8), and (2.9), (2.11) go over into the ordinary Hanle-effect expressions which follow directly from the phenomenological equation (1.35):

$$\mathcal{P}'_{\text{lin}}(H_\parallel)=\frac{1}{1+\Omega_\parallel^2\tau_0^2},\quad \mathcal{P}''_{\text{lin}}(H_\parallel)=-\frac{\Omega_\parallel\tau_0}{1+\Omega_\parallel^2\tau_0^2}. \tag{2.12}$$

In this simplest case $\mathcal{P}'_{\text{lin}}$ decreases monotonically with increasing $H_\parallel$, and the polarization plane is rotated

through an angle $\varphi = -\tan^{-1}(\Omega_\parallel \tau_0)$, i.e., $|\varphi| \leqslant \pi/4$ and $\mathcal{P}^{\prime\prime}_{\mathrm{lin}}$ increases to $\pm\frac{1}{2}$, after which it decreases. According to (2.6)–(2.8), the function $\mathcal{P}^{\prime}_{\mathrm{lin}}(H_\parallel)$ becomes nonmonotonic at $\tau_0 \sim \tau_r$, namely $\mathcal{P}^{\prime}_{\mathrm{lin}}$ reverses sign at a certain value of $H_\parallel$, and then decreases to zero. The angle of rotation of the polarization plane is $|\varphi| \leqslant \pi/2$, and therefore $\mathcal{P}^{\prime\prime}_{\mathrm{lin}}$ does not reverse sign. In the case of strong inhomogeneous broadening, according to (2.9) and (2.11), the rotation angle φ can reach values $\pm 3\pi/4$, so that both $\mathcal{P}^{\prime}_{\mathrm{lin}}$ and $\mathcal{P}^{\prime\prime}_{\mathrm{lin}}$ reverse sign in this case at certain values of $H_\parallel$.

3. Transverse magnetic field (Voigt geometry)

An analysis of the experimental data shows that the exchange-interaction constant Δ of GaSe is small in comparison with $\hbar/\tau_0$ and does not manifest itself in the optical orientation of the excitons. When considering the general case of an arbitrary ratio τ_r/τ_0 we therefore put $\Delta = 0$. For the case $\tau_r \ll \tau_0$ we shall derive formulas with $\Delta \neq 0$.

We choose the x axis parallel to the vector $\mathbf{H}_\perp$. At $\Delta = 0$ the two levels with energies $\omega_{\mathbf{K}1,2} = \omega_{\mathbf{K},0} \pm \Omega_\perp/2$, for which $j^x_{1,2} = j_0/2^{1/2}$, are excited only with light having $e_x \neq 0$, while the third level with energy $\omega_{\mathbf{K},3} = \omega_{\mathbf{K},0}$ and with energy $j^y_3 = j_0$ is excited only at $e_y \neq 0$. In this case the eigenvectors of the polarization of the light are the unit vectors $\mathbf{e}_x$ and $\mathbf{e}_y$ and we have according to (1.24)

$$\begin{aligned} -i(\tilde{q}_0^{\,y} - \tilde{q}_0^{\,y*}) &= \pi q_0 \omega_{LT} \Delta_{33}(q_0, \omega), \\ -i(\tilde{q}_0^{\,x} - \tilde{q}_0^{\,x*}) &= {}^1/_2 \pi q_0 \omega_{LT}[\Delta_{11}(q_0, \omega) + \Delta_{22}(q_0, \omega)], \\ -i(\tilde{q}_0^{\,x} - \tilde{q}_0^{\,y*}) &= {}^1/_2 \pi q_0 \omega_{LT}[\Delta_{13}(q_0, \omega) + \Delta_{23}(q_0, \omega)]. \end{aligned} \tag{2.13}$$

Substituting (1.28) and (2.13) in (1.18) we obtain after integrating with respect to ω

$$d_{yy} = d_{yy}^0\left(1 - \frac{\tau}{2\tau_0}\right), \quad d_{xy} = d_{yx}^* = d_{xy}^0\,\Phi_1\left(\frac{\Omega_\perp}{2}, \tau_0, \tau\right), \tag{2.14}$$

$$d_{xx} = d_{xx}^0\left\{\left(1 - \frac{\tau}{2\tau_0}\right)\left[1 - \frac{1}{2(1+\Omega_\perp^2\tau^2)^{1/2}}\right] + \left[1 - \frac{(1+\Omega_\perp^2\tau^2)^{1/2}}{2}\right]\Phi_1(\Omega_\perp, \tau_0, \tau)\right\},$$

where $\Phi_1(\Omega, \tau_0, \tau)$ is defined by formula (2.7).

The degrees of linear polarization $\mathcal{P}^{(1)}_{\mathrm{lin}}$ in the coordinate frame connected with vector $\mathbf{H}_\perp$, and of $\mathcal{P}^{(2)}_{\mathrm{lin}}$ in the coordinate frame rotated through an angle 45° relative to the vector $\mathbf{H}_\perp$, are given by the relations

$$\mathcal{P}^{(1)}_{\mathrm{lin}} = \frac{d_{xx} - d_{yy}}{d_{xx} + d_{yy}}, \quad \mathcal{P}^{(2)}_{\mathrm{lin}} = \frac{2\,\mathrm{Re}\,d_{xy}}{d_{xx} + d_{yy}}. \tag{2.15a}$$

The degree of circular polarization of the radiation is given by

$$\mathcal{P}_{\mathrm{circ}} = 2\,\mathrm{Im}\,d_{xy}/(d_{xx} + d_{yy}). \tag{2.15b}$$

In the case of strong inhomogeneous broadening we have according to (1.26) and (1.29)

$$\begin{aligned} d_{yy} &= d_{yy}^0(1 - \tau/2\tau_0), \quad d_{xy} = d_{yx}^* = d_{xy}^0\,\Phi_2(\Omega_\perp/2, \tau_0, \tau), \\ d_{xx} &= {}^1/_2 d_{xx}^0[1 - \tau/2\tau_0 + \Phi_2(\Omega_\perp, \tau_0, \tau)]. \end{aligned} \tag{2.16}$$

where the function $\Phi_2(\Omega, \tau_0, \tau)$ is defined by formula (2.10).

In the case $\tau_r \ll \tau_0$, at an arbitrary value of Δ, Eq. (1.35) is best solved in a basis of the state $\pm 1, 0$ which is not diagonal at $\mathbf{H} \perp C_3$ and $\Delta \neq 0$. Solving the system (1.35) for these states at $\mathcal{H}_{ex}$ and $\mathcal{H}_H$ defined by formulas (2.1) and (2.3b), we get

$$\begin{aligned} d_{yy} &= d_{yy}^0, \quad d_{xx} = d_{xx}^0\left[1 - \frac{1}{2}\frac{\Omega_\perp^2\tau_0^2}{1 + (\Omega_\perp^2 + \Delta^2)\tau_0^2}\right] \\ d_{xy} = d_{yx}^* &= d_{xy}^0\left[1 + \tau_0^2\left(\frac{\Omega_\perp^2}{4} + \Delta^2\right) + i\Delta\tau_0\left(\frac{\Omega_\perp\tau_0}{2}\right)^2\right] \\ &\times\left\{\left[1 + \left(\frac{\Omega_\perp\tau_0}{2}\right)^2\right]^2 + \Delta^2\tau_0^2\right\}^{-1} \end{aligned} \tag{2.17}$$

It is seen from (2.15) and (2.17) that the exchange splitting Δ does not alter qualitatively the dependences of the degree of polarization of the radiation on H. However, just as in uniaxial II-VI crystals,[6] if $\Delta \neq 0$ a circular polarization of the radiation appears in a transverse magnetic field if the excitation is by light linearly polarized at an angle 45° to the direction of $\mathbf{H}_\perp$, and conversely, when the excitation is by circularly polarized light, linear polarization appears, $\mathcal{P}^{(2)}_{\mathrm{lin}} \neq 0$. At $\Delta\tau_0 \ll 1$, the maximum degree of $\mathcal{P}_{\mathrm{circ}}$ or $\mathcal{P}^{(2)}_{\mathrm{lin}}$ in the latter amounts to $\frac{1}{4}|\Delta|\tau_0$ and is reached at $\Omega_\perp \sim 2/\tau_0$.

It is seen from (2.14)–(2.17) that in the case of excitation by linearly polarized light with $\mathbf{e} \perp \mathbf{H}_\perp$ the intensity $J = d_{xx} + d_{yy}$ and the degree of polarization of the radiation do not change in a magnetic field. In the case of excitation by linearly polarized light with $\mathbf{e} \parallel \mathbf{H}_\perp$ the degree of polarization of the radiation likewise remains unchanged and stays equal to unity, while the radiation intensity J decreases with increasing $H_\perp$. At $\tau_r \ll \tau_0$ the function $J(H_\perp)$ is monotonic and at $\Omega_\perp\tau_0 \gg 1 \gg \Omega_\perp\tau_r$ we have $J \approx \frac{1}{2}J(0)$. At $\tau_r \sim \tau_0$, $J(H_\perp)$ also first decreases with increasing $H_\perp$, and then in strong fields, owing to the dichroism, in the magnetic field, it again increases and reaches $\Omega_\perp\tau \gg 1$ at the value $J(0)$. If the inhomogeneous broadening is strong, when the dichroism is negligible, the intensity J reaches a minimum value $J_{\min} < \frac{1}{2}J(0)$, and then increases and reaches at $\Omega_\perp\tau \gg 1$ the limiting value $\frac{1}{2}J(0)$. Therefore in a transverse magnetic field, when the excitation is by unpolarized light, just as in the case of excitation by light that is polarized circularly or linearly at an angle 45° to the direction of $\mathbf{H}_\perp$, a polarization $\mathcal{P}^{(1)}_{\mathrm{lin}}$ is produced. At $\tau_r \ll \tau_0$ the variation of $\mathcal{P}^{(1)}_{\mathrm{lin}}$ is monotonic and the maximum value of $|\mathcal{P}^{(1)}_{\mathrm{lin}}(H_\perp)|$ is $\frac{1}{3}$. At $\tau_r \sim \tau_0$ the quantity $|\mathcal{P}^{(1)}_{\mathrm{lin}}(H_\perp)|$ goes through a maximum and then tends to zero at $\Omega_\perp\tau \gg 1$. In the case of strong inhomogeneous broadening $|\mathcal{P}^{(1)}_{\mathrm{lin}}(H_\perp)|$ goes through a maximum value larger than $\frac{1}{3}$, and in strong fields it decreases to $\frac{1}{3}$.

If the excitation is by circularly polarized light, $\mathcal{P}_{\mathrm{circ}}(H_\perp)$ decreases to zero with increasing $H_\perp$. At $\tau_r \ll \tau_0$, the function $\mathcal{P}_{\mathrm{circ}}(H_\perp)$ is monotonic. $\mathcal{P}_{\mathrm{circ}}(H_\perp)$ passes through zero at $\tau_r \sim \tau_0$, reaches a minimum value, and then tends asymptotically to zero. The variation of $\mathcal{P}^{(2)}_{\mathrm{lin}}(H_\perp)$ following excitation by light linearly polarized at an angle 45° to $\mathbf{H}_\perp$ duplicates completely the variation of $\mathcal{P}_{\mathrm{circ}}(H_\perp)$ under circularly polarized pumping.

Thus, by measuring the change of the degree of polarization of the resonant radiation in a magnetic field, we can determine the values of the times τ_0 and τ_r and ascertain whether inhomogeneous broadening takes place over lengths on the order of l_{ph}.

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[1)]A similar method was used by Rebane, Tekhver, and Khizhnyakov to describe secondary resonant radiation of impurity centers in ionic crystals (see the review[7]).

[2)]Diagrams analogous to 2c were taken into account in the calculation of the vertex part in the theory of the Fermi liquid in[10], where they also turned out to be significant at a small summary momentum of the colliding particles. The presence of the singularities that arise when the light is scattered backward upon excitation of hot excitons with emission of optical phonons, was noted in[2]. Analogous singularities of backward scattering of light or radio waves by different inhomogeneities were discussed earlier in a number of papers (see, e.g.,[11]). A more complete bibliography is given in the review[12]. We are grateful to V. I. Tatarskiĭ for calling our attention to these papers.

[3)]In the theory developed above it was assumed that the exciton energy relaxation time is $\tau_\varepsilon \gg \tau_0$. The phenomenological equation (1.35) is valid both at $\tau_r \ll \tau_0 \ll \tau_\varepsilon$, when the energy of the exciton remains practically unchanged during the time τ_0, and in the opposite limiting case $\tau_r \ll \tau_0$ and $\tau_\varepsilon \ll \tau_0$, when the lifetime is long enough for an equilibrium exciton distribution to be established. At $\tau_\varepsilon \approx \tau_0 \approx \tau_{rad}$ it is necessary to take into account in (1.35) also the energy relaxation.

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