Optical orientation of excitons in cubic crystals

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(Submitted March 15, 1974)
Zh. Eksp. Teor. Fiz. **67**, 788–800 (August 1974)

Optical orientation of bound excitons in cubic crystals in longitudinal and transverse magnetic fields is considered for the case when the main electron spin relaxation mechanism is the interaction between the electron and holes. It is shown that, depending on the ratios of the radiative and nonradiative lifetimes to the hole spin relaxation time, one can observe either an increase or decrease of the luminescence polarization in a longitudinal magnetic field. Orientation damping in a transverse magnetic field turns out to be more complex than in the usual Hanle effect. It is shown that recombinational (nonthermal) exciton polarization may be observed on excitation with nonpolarized light in a longitudinal magnetic field, and exciton alignment occurs in a transverse magnetic field.

1. INTRODUCTION

As we have shown before, the optical orientation of excitons has a number of features which make it different from the orientation of free carriers. The nature of this orientation and its variation under the influence of a magnetic field and strain depend strongly on the fine structure of the exciton levels, e.g., on the exchange interaction. Excitons in uniaxial crystals, in which the optical exciton orientation was first observed[2], were considered in[1]. The effect was later also observed in cubic crystals[3], where the structure of the exciton levels is different from the case of uniaxial crystals due to the different symmetry and, in particular, to the degeneracy of the valence band. In the present paper we consider the optical orientation of cubic-crystal excitons bound to charged centers or to isoelectronic traps in longitudinal and transverse magnetic fields. The spin relaxation time of bound holes in such crystals is usually much shorter than that of electrons.

As D'yakonov and Perel' have pointed out[4,5], the electron-hole exchange interaction may be an effective mechanism of electron spin relaxation under these conditions. It is shown below that electron spin relaxation also occurs as a result of the difference between the exciton lifetimes in its optically active and inactive states, even in the absence of the exchange interaction. We shall treat the case when these two mechanisms of the electron spin relaxation prevail and neglect the other mechanisms associated with the direct electron-lattice interaction.

D'yakonov and Perel' considered the exchange interaction between electrons and holes bound to donor-acceptor pairs. They showed that with the exchange mechanism of electron relaxation, the spin relaxation time increases in a strong longitudinal magnetic field, which leads to an increase in the luminescence polarization. In this paper we consider the case of small exchange splitting and short hole-spin relaxation time, when, due to dynamic averaging, the spin relaxation time of an electron greatly exceeds that of a hole and the degree of hole orientation is modest. We assume an arbitrary exchange splitting and an arbitrary relation between the lifetimes and the spin relaxation times. We also assume, as distinct from[5], that neither the electrons nor the holes return to the band during their lifetime, and neither do they go over to other defects. Under these conditions not only does the electron-hole interaction provide the mechanism for the electron spin relaxation, but it may also cause a redistribution of the angular momentum between an electron and a hole and thereby induce a substantial orientation of the holes. A change of the electron angular momentum can also occur as a result of the hyperfine interaction with nuclei that was considered by D'yakonov and Perel'[6]. As in the case of the interaction with nuclei, the electron-hole interaction leads to a more complex Hanle effect as compared to the case of free electrons.

However, as opposed to the nuclei, the holes are directly involved in recombination, being created and destroyed along with the electrons. Therefore, their momentum cannot accumulate, as distinct from the momentum of the nuclei, and their orientation directly affects the radiation polarization. In addition, the holes possess a considerably larger g factor than the nuclei and their spin relaxation time is much shorter.

All this leads to the fact that in our model the change of orientation in longitudinal and transverse fields in many cases is substantially different from the dependences typical for the models considered by D'yakonov and Perel'[5,6]. For example, it is shown below that the luminescence polarization can not only increase in a longitudinal magnetic field (as was the case in[5,6]), but can also decrease, depending on the relation between the lifetimes and the spin relaxation times.

It is also shown that the simultaneous orientation of the electrons and the holes in an exciton brings about some new effects which distinguish the orientation of excitons from that of free carriers. For instance, recombinational exciton polarization as well as circularly polarized luminescence should be observed on excitation with unpolarized light in a longitudinal magnetic field. However, unlike the thermal orientation also considered by D'yakonov and Perel'[7], this effect does not depend on temperature. In a transverse magnetic field, the exchange interaction leads to alignment of excitons and gives rise to linearly polarized luminescence on excitation with unpolarized light.

The above effects can be observed in the case when the excitons are formed by bound pairs as well as in the case of resonance excitation.

2. BASIC EQUATIONS

Consider a cubic crystal with a simple conduction band and a fourfold-degenerate valence band, both having their extrema at the point Γ. Most A_3B_5 and A_2B_6 cubic crystals have this type of band structure. The exciton ground state in such crystals is eightfold-degenerate. Due to the electron-hole exchange interaction, it is split into two states: threefold-degenerate with $J = 1$, and

fivefold-degenerate with J = 2. The magnitude of the exchange splitting is $\Delta = E_2 - E_1$.

The equation for the exciton density matrix ρ in a magnetic field is of the form:

$$-\left(\frac{\partial\rho}{\partial t}\right)_{rec}-\left(\frac{\partial\rho}{\partial t}\right)_{sr}+i[\mathscr{H}_H\rho]+i[\mathscr{H}_{ex}\rho]=G. \quad (1)$$

(Here and in the following we take $\hbar = 1$.) We assume that both the exchange splitting and the splitting in a magnetic field are small compared to kT. The term $(\partial\rho/\partial t)_{rec}$ describes the radiative and nonradiative exciton recombinations. As the radiative transitions are possible only for the states with J = 1, $(\partial\rho/\partial t)_{rec}$ is determined by two lifetimes: τ_1 for the states with J = 1 and τ_2 for the states with J = 2. The difference $\tau_1^{-1} - \tau_2^{-1} = \tau_{rad}^{-1}$.

As can be shown from symmetry considerations, the recombination term is "diagonal" in the basis Jm_J and is of the form

$$\left(\frac{\partial\rho_{ij}}{\partial t}\right)_{rec}=-\frac{\rho_{ij}}{\tau_{ij}},\quad \frac{2}{\tau_{ij}}=\frac{1}{\tau_{ii}}+\frac{1}{\tau_{jj}}. \quad (2)$$

Here the states 1—3 correspond to J = 1 and m_J = 1, 0, −1; the states 4—8 correspond to J = 2 and m_J = 2, 1, 0, −1, −2. For i, j = 1—3 we have $\tau_{ij} = \tau_1$ and for i, j = 4—8 we have $\tau_{ij} = \tau_2$.

As was pointed out above, we consider only the spin relaxation of holes, assuming that it is defined by a single relaxation time τ_s and is independent of the exciton-level structure, in particular of the exchange and magnetic splittings. The above approximation is valid when the two-phonon processes of hole-spin scattering prevail. In this case the hole-spin relaxation times in bound excitons must be close to the corresponding times for acceptor centers, which have been estimated theoretically[8] and experimentally[9] to be 10^{-9} sec. In this approximation

$$\left(\frac{\partial\rho}{\partial t}\right)^{sr}_{nn',mm'}=-\frac{1}{\tau_s}\rho_{nn',mm'}+\frac{1}{4\tau_s}\sum_{m''}\rho_{nn',m''m''}\delta_{mm'}. \quad (3)$$

Here n, n′ label the electron spins ($n = \pm 1/2$), and m, m′ label the hole spins ($m = \pm 1/2, \pm 3/2$). The operator (3) conserves the total number of excitons $N = \mathrm{Tr}\,\rho$ and the average electron spin $\sigma/2$:

$$\sigma=\mathrm{Sp}\,\rho\hat{\sigma}/\mathrm{Sp}\,\rho. \quad (4)$$

The matrix G describes exciton generation. In the case when the excitons are formed by the binding of free electrons and holes, so that only the electrons are oriented and no appreciable orientation of the holes occurs during the capture[1]1),

$$G=\tfrac{1}{8}G_0(1+p_e\sigma_z). \quad (5)$$

Here p_e is the electron polarization at the moment of capture, G_0 is the total number of excitons generated per unit time, and σ_z is the electron spin operator. In the following we shall also consider certain features of optical orientation that arise in the course of resonance exciton generation.

In (1), it is convenient to combine the depletion term in (3) with the recombination term of (2) by introducing the depletion times (in the basis Jm_J):

$$\frac{1}{T_{ij}}=\frac{1}{\tau_{ij}}+\frac{1}{\tau_s}=\frac{1}{2}\left(\frac{1}{T_{ii}}+\frac{1}{T_{jj}}\right),\quad T_{ij}=T_1 \text{ for } i,j=1-3,$$
$$T_{ij}=T_2 \text{ for } i,j=4-8, \quad (6)$$

and to write the replenishment term in (3) in a form analogous to the generation term (5) by introducing a suitable effective generation G′:

$$G'=\frac{G_0}{8}\frac{\bar\tau}{\tau_s}(I+\bar\sigma\sigma), \quad (7)$$

where $\bar\tau = N/G_0$ is the average lifetime of an exciton and $\bar\sigma$ is defined by (4).

The interaction of electrons and holes with the magnetic field is described in the spherical approximation by the operator $\mathscr{H}_H$[10]:

$$\mathscr{H}_H=\tfrac{1}{2}\mu_0[g_e\sigma\mathbf{H}+2g_h\mathbf{jH}]=\tfrac{1}{2}\mu_0[2g_h\mathbf{JH}+(g_e-g_h)\sigma\mathbf{H})], \quad (8)$$

where g_e and g_h are respectively the g-factors of the electrons and holes, **j** is the hole angular momentum operator, and **J** is the total angular momentum operator.

We shall solve Eq. (1) in the basis (Jm_J), in which σ has matrix elements both diagonal and off-diagonal in **J**. In this case the components diagonal in **J**

$$\sigma^{(1)}=-\tfrac{1}{2}\mathbf{J}^{(1)},\quad \sigma^{(2)}=\tfrac{1}{2}\mathbf{J}^{(2)}, \quad (9)$$

and all the off-diagonal components can be expressed in terms of the reduced matrix element $\sigma_2^1 = -\sqrt{15/2}$ ([11], Sec. 29, 109).

In solving the system of linear equations (1), we shall first regard the quantities $\bar\sigma$ as given and combine (5) and (7) into one generation term $\tilde G = G + G'$. Next, having expressed the components of ρ as functions of σ and substituted them into (4), we define $\bar\sigma$ and $\bar\tau$. Then we evaluate the average total angular momentum $\bar{\mathbf{J}} = \mathrm{Tr}\,\mathbf{J}\rho/\mathrm{Tr}\,\rho$, the average angular momentum of the holes $\bar I_z = \bar J_z - \bar\sigma_z/2$, and the degree of circular luminescence polarization

$$\mathscr{P}_{circ}=(\rho_{11}-\rho_{33})/(\rho_{11}+\rho_{33}). \quad (10)$$

We shall not reproduce the calculations here and shall present only the final formulas. In doing so we shall first consider the case $T_1 = T_2 = T$ which holds for $\tau_1 = \tau_2$ as well as for $\tau_s \ll \tau_1, \tau_2$, and then, in Sections 4 and 5, discuss the peculiarities which arise in the case $T_1 \neq T_2$.

3. OPTICAL ORIENTATION IN A LONGITUDINAL MAGNETIC FIELD ($T_1 = T_2 = T$)

For $T_1 = T_2 = T$, the average exciton lifetime does not depend on the magnetic field and is equal to

$$\frac{1}{\bar\tau}=\frac{1}{8}\left(\frac{3}{\tau_1}+\frac{5}{\tau_2}\right). \quad (11)$$

In the absence of the magnetic field the average electron spin can be expressed in its usual form:

$$\bar\sigma_z=p_e/(1+\bar\tau/\tau_{es}), \quad (12)$$

where τ_{es} is the average electron spin relaxation time and is given by

$$\tau_{es}=T[8+3(\Delta T)^2]/5(\Delta T)^2. \quad (13)$$

The dependence of $\mathscr{P}_{circ}$ on τ_{es} and $\bar\tau$ is more complex:

$$\mathscr{P}_{circ}=-\frac{1}{2}p_e\frac{1+T/\tau_{es}}{1+\bar\tau/\tau_{es}}. \quad (14)$$

In this case the average angular momentum $\bar J_z = -\mathscr{P}_{circ}$.

It is seen from (14) that $\mathscr{P}_{circ}$ approaches its limiting value $-p_e/2$ both at $\tau_s \gg \bar\tau$ (when $T = \bar\tau$) and at $\tau_{es} \gg \bar\tau$ (since $T \leq \bar\tau$). For $\Delta T \ll 1$, when $\tau_{es} \gg T$, expression (14) reduces to the usual form

$$\mathscr{P}_{\text{circ}} = -\frac{1}{2}\frac{p_e}{1+\bar{\tau}/\tau_{es}}. \quad (14a)$$

In this case the holes polarization is small and $\bar{\sigma}_z = -2\mathscr{P}_{\text{circ}}$. Considerable polarization of holes occurs at $\Delta T \gg 1$ and $\tau_s \gg \tau_1, \tau_2$ when

$$\bar{\sigma}_z = {}^3/_8 p_e, \quad \bar{J}_z = {}^5/_{16} p_e. \quad (15)$$

If $\Delta T \gg 1$ and $\tau_s \ll \tau_1, \tau_2$, both the electron polarization and the hole polarization and $\mathscr{P}_{\text{circ}}$ are small:

$$\bar{\sigma}_z = \frac{3}{5}\frac{\tau_s}{\bar{\tau}} p_e, \quad \bar{J}_z = \frac{1}{2}\frac{\tau_s}{\bar{\tau}} p_e, \quad \mathscr{P}_{\text{circ}} = -\frac{4}{5}\frac{\tau}{\bar{\tau}} p_e. \quad (16)$$

In a longitudinal magnetic field, $\bar{\sigma}_z$ is given by (12); in this case τ_{es} depends on the magnetic field and is determined by the quantity K(H):

$$\tau_{es} = TK/(1-K), \quad (16a)$$

$$K = 1 - \frac{1}{8}(\Delta T)^2 \left\{ \frac{3M}{M^2 - (\omega \Delta T^2)^2} + \frac{2}{M} \right\}, \quad (17)$$

$$M = 1 + (\Delta T)^2 + (\omega T)^2, \quad \omega = \mu_0 H_z (g_e - g_h).$$

The expression for $\mathscr{P}_{\text{circ}}(H)$ can be written in the form

$$\mathscr{P}_{\text{circ}} = \mathscr{P}_0(H)\frac{1-\varphi_1(H)}{1+\mathscr{P}_0(H)\varphi_2(H)} \quad (18)$$

where $\mathscr{P}_0(H) = -\bar{J}_z(H)$ is given by (14), in which $\tau_{es}(H)$ is determined by (16a) and (17), and

$$\varphi_1(H) = {}^3/_2(\omega T)^2 R/M, \quad \varphi_2(H) = {}^3/_2 \omega R/\Delta; \quad R = M(\Delta T)^2/[M^2 - (\omega \Delta T^2)^2]. \quad (19)$$

The term $\mathscr{P}_0(H)\varphi_2(H)$ appearing in the denominator of (18) is an odd function in the magnetic field and changes sign as the magnetic field or the orientation sign changes. In other words, $\mathscr{P}_{\text{circ}}$ can vary as the magnetic field changes direction.

As D'yakonov and Perel' pointed out[6], a similar effect can be brought about by the hyperfine interaction between electrons and nuclei if there is a difference in the rates of relaxation of different nuclear polarization momenta. The above calculation shows that with the electron-hole exchange interaction this effect is due to the direct influence of the holes orientation on the radiation polarization. It occurs when all components of the hole density matrix have the same rate of spin relaxation.

Now consider the dependence of τ_{es} and $\varphi_{1,2}(H)$ on the magnetic field. It is seen from (16a) and (17) that for ω exceeding both Δ and T^{-1}, the quantity $K \to 1$ and τ_{es} increases in proportion to H^2. For small exchange ($\Delta T \ll 1$)

$$\tau_{es}(H) = \tau_{es}(0)(1 + (\omega T)^2), \quad (20)$$

where

$$\tau_{es}(0) = 8/5\Delta^2 T. \quad (20a)$$

For $\Delta T \ll 1$ no appreciable hole orientation occurs, $\varphi_1(H)$ and $\varphi_2(H)$ are small, and $\mathscr{P}_{\text{circ}}$ is independent of the direction of H. For large exchange ($\Delta T \gg 1$), τ_{es} depends only on the ratio $\omega/\Delta = z$:

$$\tau_{es} = T\frac{3 + 8z^2 + 11z^4 + 8z^6}{5 + 8z^2 + 5z^4}. \quad (21)$$

It is apparent from (20) and (21) that if $\omega \gg \Delta$, T^{-1} both cases yield

$$\tau_{es} = {}^8/_5 T(\omega/\Delta)^2. \quad (22)$$

For $\Delta T \gg 1$ we have

$$\varphi_1(H) = \frac{3}{2}\frac{z^2}{1+z^2+z^4}, \quad \varphi_2(H) = \frac{3}{2}\frac{z(1+z^2)}{1+z^2+z^4}. \quad (23)$$

As seen from (23), both functions vanish as $H \to 0$ as well as when $H \to \infty$, and at $\omega = \Delta$ they reach the maximum values $\varphi_1 = 1/2$ and $\varphi_2 = 1$.

From the above expressions it follows that, depending on the value of ΔT and the ratio $\tau_s/\tau_{1,2}$, the behavior of $\mathscr{P}_{\text{circ}}$ and $\bar{\sigma}_z$ as functions of H may have different forms.

1. For $\tau_s \gg \tau_1, \tau_2$ and $\Delta\bar{\tau} \ll 1$, the magnetic field does not affect the orientation, and $\mathscr{P}_{\text{circ}}(H) = -\bar{\sigma}_z/2 = -p_e/2$, as occurs in the absence of the field.

2. For $\tau_s \gg \tau_1, \tau_2$ and $\Delta\bar{\tau} \gg 1$, according to (14) and (16a), the total angular momentum $\bar{J}_z = -\mathscr{P}_0 = p_e/2$ and is independent of H, whereas the spin distribution depends on $z = \omega/\Delta$ and with increasing z we have $\bar{\sigma}_z \to p_e$ and $\bar{J}_z \to 0$. In this case, $\mathscr{P}_0 = -p_e/2$ and the $\mathscr{P}_{\text{circ}}(H)$ dependence is defined by the change of $\varphi_1(H)$ and $\varphi_2(H)$: at $z = 0$ and $z \gg 1$ we have $\mathscr{P}_{\text{circ}} = -p_e/2$, whereas for intermediate values of z the function $\mathscr{P}_{\text{circ}}$ passes through a minimum at $z = 1$. In this case the behavior of $\mathscr{P}_{\text{circ}}$ is governed by the sign of $p_e\omega/\Delta$. For $p_e \ll 1$ at $z = 1$ we have $\mathscr{P}_{\text{circ}} = -{}^1/_4 p_e$; for $|p_e| = 1/2$ and $z = 1$ the value of $\mathscr{P}_{\text{circ}} = -{}^1/_3 p_e$ if $zp_e > 0$, and $\mathscr{P}_{\text{circ}} = -{}^1/_5 p_e$ if $zp_e < 0$.

3. For $\tau_s \ll \bar{\tau}$ and $\Delta\tau_s \ll 1$ the dependence of $\mathscr{P}_{\text{circ}}$, $\bar{\sigma}_z$, and $\bar{J}_z$ on H is given by (12) and (14a), and that of τ_{es} by (20). Since, according to (20), τ_{es} increases with the magnetic field, $\mathscr{P}_{\text{circ}}$ in this case increases from $\mathscr{P}_{\text{circ}}(0)$ to $-p_e/2$.

4. For $\tau_s \ll \bar{\tau}$ and $\Delta\tau_s \gg 1$, according to (16), $\mathscr{P}_{\text{circ}}$, $\bar{J}_z$, and $\bar{\sigma}_z$ are small if $H = 0$. In a longitudinal magnetic field, $\mathscr{P}_{\text{circ}}$ increases up to $-p_e/2$ in fields with $\omega \gtrsim (\bar{\tau}/\tau_s)^{1/2}$. Thus, in the case $T_1 = T_2$, a sufficiently strong longitudinal magnetic field always leads to an increase of $\mathscr{P}_{\text{circ}}$ to its limiting value $-p_e/2$.

4. RECOMBINATION ORIENTATION OF EXCITONS

In this section we shall consider some new effects which arise in the case when the lifetimes τ_1 and τ_2 of the optically active and inactive states are different.

According to experiment, radiative recombination prevails over nonradiative for a number of crystals, and in these cases $\tau_1 \ll \tau_2$ (thus, for example, according to[12], $\tau_1 = 3.8 \times 10^{-8}$ sec and $\tau_2 = 4.3 \times 10^{-6}$ sec with $\tau_s < 10^{-9}$ sec for an exciton bound in an isoelectronic trap in GaAs).

If $T_1 \neq T_2$, the average total angular momentum J is not conserved even in the absence of spin-lattice relaxation, and can exceed the average momentum produced by pumping; in this case the holes become polarized even in the absence of the exchange interaction. This gives rise to a new mechanism of electron-spin relaxation, to exciton polarization, and to a circularly polarized luminescence in a longitudinal magnetic field when pumped with a nonpolarized light. In a transverse magnetic field such pumping causes an alignment of excitons if $T_1 \neq T_2$, and a linearly polarized luminescence appears. Moreover, for $T_1 \ll T_2$ the circular polarization of luminescence in a strong longitudinal magnetic field does not increase on excitation with polarized light, but in contrast to the case $T_1 = T_2$ it decreases.

In the case of resonance exciton generation, similar effects occur even if $\tau_1 = \tau_2$ since such excitation itself violates the equivalence of states J = 1 and J = 2. In the absence of the magnetic field at $T_1 \neq T_2$, the average lifetime $\bar{\tau}$ depends on the relation between τ_1 and τ_2, and is given by

$$\bar{\tau}^{-1} = \bar{T}^{-1} - \tau_s^{-1}, \quad \bar{T} = {}^1/_8(3T_1 + 5T_2). \quad (24)$$

For $\tau_s \ll \tau_1, \tau_2$ this expression reduces to (11), and for $\tau_s \gg \tau_1, \tau_2$

$$\bar{\tau} = {}^1/_8(3\tau_1 + 5\tau_2). \quad (25)$$

The expressions (12) and (14) for $\bar{\sigma}_z$ and $\mathscr{P}_{circ}$ remain valid at $\tau_1 \neq \tau_2$; in this case τ_{es} is given by (16a) with

$$K = 1 - \frac{5}{16}\frac{(T_1-T_2)^2}{\bar{T}(T_1+T_2)} - \frac{5}{8}\frac{T_{15}}{\bar{T}}\frac{(\Delta T_{15})^2}{1+(\Delta T_{15})^2}. \quad (26)$$

From (26) it is apparent that K is practically independent of the exchange at $T_1 \ll T_2$ and is equal to $\frac{1}{2}$, i.e., $\tau_{es} = \bar{T}$.

For $T_1 \neq T_2$ the average angular momentum $\bar{J}_z$ is not equal to $-\mathscr{P}_{circ}$ but is related to it by

$$\bar{J}_z = -\mathscr{P}_{circ}\left(1 + \frac{5(T_2-T_1)}{3T_1+5T_2}\right). \quad (27)$$

Let us examine the results obtained. If the spin relaxation time $\tau_s \gg \tau_1, \tau_2$ then always $\mathscr{P}_{circ} = -p_e/2$ and depends on neither the relation between τ_1 and τ_2 nor the strength of the exchange interaction. The total angular momentum $\bar{J}_z$ in this case also does not depend on the exchange splitting, but as the ratio τ_2/τ_1 varies, it increases from $p_e/2$ at $\tau_1 = \tau_2$ to p_e at $\tau_1 \ll \tau_2$. At $\tau_1 \ll \tau_2$, regardless of the magnitude of Δ

$$\bar{\sigma}_z = p_e/2, \quad \bar{j}_z = {}^3/_4 p_e. \quad (27a)$$

If $\tau_1 \ll \tau_s \ll \tau_2$, then again $\bar{\sigma}_z$, $\bar{j}_z$, and $\mathscr{P}_{circ}$ do not depend on the exchange and are equal to

$$\bar{\sigma}_z = \frac{3}{11}p_e, \quad \bar{J}_z = \frac{6}{11}p_e, \quad \bar{j}_z = \frac{9}{22}p_e, \quad \mathscr{P}_{circ} = -\frac{3}{11}p_e. \quad (27b)$$

In this case, practically all the excitons that are in the state J = 1 recombine without changing their spin state whereas those with J = 2 survive until, as a result of spin relaxation, they go into the state J = 1 where the recombination occurs. It is precisely these transitions that are responsible for reducing the degree of orientation of the J = 1 states.

If $\tau_s \ll \tau_1, \tau_2$ and $\Delta\tau_s \ll 1$ then $\mathscr{P}_{circ}$ is given by (14a). In this case τ_{es} involves, besides the exchange-interaction term (20a), an additional term associated with the disparity between τ_1 and τ_2:

$$\frac{1}{\tau_{es}} = \frac{5}{8}\Delta^2\tau_s + \frac{5}{32}\frac{\tau_s}{(\tau_{rad})^2}, \qquad \tau_{rad}^{-1} = \tau_1^{-1} - \tau_2^{-1}. \quad (28)$$

If $\tau_1 \neq \tau_2$, then, in a longitudinal magnetic field, the lifetimes in the states with $m_1 = 1$ and $m_1 = -1$ (split by the magnetic field) turn out to be different due to the mixing of J = 1 and J = 2 states. As a consequence, when the excitons are formed by bound pairs their populations are unequal even on excitation with unpolarized light. This leads to polarization of the excitons and gives rise to circularly polarized luminescence. The effect is smaller if $\tau_s < \tau_1$ since the spin relaxation equalizes the level populations. This feature distinguishes the recombination orientation from the usual thermal orientation associated with a disparity of equilibrium populations in a magnetic field, which arises when the splittings are comparable to kT and $\tau_s \ll \tau_1$.[7]

We shall not list the corresponding relations for $\bar{\sigma}_z$, but give only the final expressions for $\mathscr{P}^{rec}_{circ}$ for two extreme cases: $\tau_s \gg \tau_{1,2}$ and $\tau_s \ll \tau_{1,2}$. For $\tau_s \gg \tau_{1,2}$,

$$\mathscr{P}^{rec}_{circ} = -\frac{6\Delta\tau_{15}}{(1+(\Delta\tau_{15})^2)^{1/2}}\frac{\varepsilon(1-\varepsilon^2)^{1/2}}{(4-\varepsilon^2)^{3/2}}\Phi(\tilde{\omega});$$

$$\Phi(\tilde{\omega}) = \frac{\tilde{\omega}^3}{(1+\tilde{\omega}^2)[1+\tilde{\omega}^2(1+\gamma)]-4\lambda^2\tilde{\omega}^2},$$

$$\tilde{\omega} = \frac{1}{2}\frac{\omega\tau_{15}}{(1+(\Delta\tau_{15})^2)^{1/2}}(1-\varepsilon^2)^{-1/2}(4-\varepsilon^2)^{1/2}, \quad \varepsilon = \frac{\tau_2-\tau_1}{\tau_2+\tau_1}, \quad \gamma = \frac{3\varepsilon}{4-\varepsilon^2}$$
$$\lambda = \frac{\Delta\tau_{15}}{(1+(\Delta\tau_{15})^2)^{1/2}}\left(\frac{1-\varepsilon^2}{4-\varepsilon^2}\right)^{1/2}. \quad (29)$$

It is seen from (29) that $\mathscr{P}_{circ}$ increases at small fields as H^3 and drops at large fields as H^{-1}. The value of $\tilde{\omega}$ that maximizes $\mathscr{P}^{rec}_{circ}$ is close to $\sqrt{3}$ and has little dependence on ϵ and $\Delta\tau_{15}$. The value of $\Phi(\tilde{\omega})$ at $\tilde{\omega} = \tilde{\omega}_{max}$ near 0.3–0.4 $\mathscr{P}^{rec}_{circ}$ is maximal in the case of large exchange, when $\Delta\tau_{15} \gg 1$. In this case, for optimal values of $\tau_2/\tau_1 \approx 5$ at $\tilde{\omega} = \tilde{\omega}_{max}$, the value of $\mathscr{P}^{rec}_{circ}$ comes up to 11%. When $\Delta\tau_{15} \ll 1$ the effect decreases by the amount $\Delta\tau_{15}$.

At small spin relaxation times $\tau_s \ll \tau_1, \tau_2$

$$\mathscr{P}^{rec}_{circ} = -\frac{2\varepsilon}{4-\varepsilon}\frac{\tilde{\omega}}{1+\tilde{\omega}^2}\frac{\Delta\tau_s}{(1+\Delta^2\tau_s\bar{\tau})^{1/2}};$$
$$\tilde{\omega} = \omega\tau_s/(1+\Delta^2\tau_s\bar{\tau})^{1/2}, \quad \bar{\tau}^{-1} = {}^1/_8(3\tau_1^{-1}+5\tau_2^{-1}). \quad (30)$$

Relation (30) suggests that $\mathscr{P}^{rec}_{circ}$ reaches its maximum at $\tilde{\omega} = 1$, i.e., the effect is maximal for $\Delta^2\tau_s\bar{\tau} \gg 1$, when

$$\mathscr{P}^{rec}_{circ} = -\frac{\varepsilon}{4-\varepsilon}\left(\frac{\tau_s}{\bar{\tau}}\right)^{1/2}.$$

It follows from the above expressions that the most favorable conditions for observing the recombination orientation are provided by large exchange splittings and large spin relaxation times. Under these conditions, the limiting values of the degree of circular polarization amount to 5–10% in a wide range of values of τ_2/τ_1. At $\Delta^2\bar{\tau}\tau_s \gg 1$ and $\tau_s \ll \tau_1, \tau_2$ the effect is reduced by the amount $(\tau_s/\bar{\tau})^{1/2}$.

The recombination-orientation effect is strongest in the case of resonance exciton generation with unpolarized light. In this case it occurs at $\tau_1 = \tau_2$ as well, since in resonant excitation of states with J = 1, the rates of pumping into the new states formed by the mixing of states with J = 1 and J = 2 in the magnetic field are different. For resonance (as well as for nonresonance) excitation, the recombination orientation is maximal at $\tau_s \gg \tau_1, \tau_2$. For this case we obtain

$$\mathscr{P}_{circ} = -\frac{3\Delta\tau_{15}}{(1+(\Delta\tau_{15})^2)^{1/2}}(1-\varepsilon)^{1/2}(1+\varepsilon)^{1/2}\Phi_1(\tilde{\omega}),$$
$$\Phi_1(\tilde{\omega}) = \frac{\tilde{\omega}^3}{1+\alpha\tilde{\omega}^2+\beta\tilde{\omega}^4}, \quad \tilde{\omega} = \frac{1}{2}\frac{\omega\tau_{15}}{(1+(\Delta\tau_{15})^2)^{1/2}(1-\varepsilon^2)^{1/2}},$$
$$\alpha = \frac{1}{2}\left[4+3\varepsilon+4\varepsilon^2+\frac{8(1-\varepsilon^2)}{(1+(\Delta\tau_{15})^2)^{1/2}}\right]$$
$$\beta = {}^1/_2(4-\varepsilon^2)(5+3\varepsilon-2\varepsilon^2). \quad (31)$$

As can be seen from (31), at $\Delta\tau \gg 1$ and $\tau_1 = \tau_2$ in the resonance excitation mode, P^{rec}_{circ} reaches a maximum of 24% (at $\tilde{\omega} = 1.3$). For $\Delta\tau \ll 1$

$$\mathscr{P}^{rec\ max}_{circ} = -0.17(\Delta\tau).$$

Relations (29)–(31) suggest that, regardless of the mechanism of excitation, the recombination orientation disappears when $\epsilon \to 1$, i.e. at $\tau_2 \gg \tau_1$. This is explained by the fact that when a crystal emits circularly polarized radiation upon having been excited by unpolarized light, it must acquire an angular momentum to compensate the momentum carried away by the light. In the case under consideration this momentum can only be transferred to the lattice through nonradiative recombination, since the crystal electrons can acquire no angular momentum

after exciton recombination as they return to the initial nondegenerate ground state. The optically inactive states 4 and 8 with M = ±2 gain no momentum on excitation with nonpolarized light in the bound-pair mode of exciton generation. In the resonance-excitation mode these states are not produced at all. Therefore, only the remaining states, which all become optically active in a magnetic field, can transfer momentum to the lattice. This transfer becomes efficient only if the rate of nonradiative recombination is not too small as compared to the radiative rate. In principle, angular momentum can also be transferred to the lattice through spin relaxation. In the nonresonance-excitation mode, this transfer is possible only if there is a disparity in the J = 1 and J = 2 spin relaxation times. It seems that the angular momentum transferred to nuclei in the course of recombination orientation can also be determined directly, by measuring the magnetization of the nuclei. In contrast to the dynamic nuclear polarization due to the Overhouser effect, the above polarization does not depend on temperature.

Due to the effect of recombination orientation, the expression for $\mathscr{P}_{circ}(H)$ in the bound-pair mode of exciton generation on excitation with unpolarized light contains terms that are proportional to the initial electron orientation p_e as well as terms independent of p_e. As is apparent from (29) and (30), for strong magnetic fields the latter terms are small, and the expression for $\mathscr{P}_{circ}(H)$ in this case becomes similar to (18). However, unlike $\varphi_2(H)$, the function $\varphi_1(H)$ does not vanish as $H \to \infty$ for $\tau_2 \neq \tau_1$, and if $\tau_S \gg \tau_1, \tau_2$ it tends to a finite limit equal to $3(\tau_2 - \tau_1)/(3\tau_2 + 5\tau_1)$. Therefore, as $H \to \infty$

$$\mathscr{P}_{\substack{\text{circ}\\ H\to\infty}}(H) = -\frac{1}{2} p_e \frac{8\tau_1}{5\tau_1+3\tau_2}. \tag{32}$$

In this case, one sees that for $\tau_2 \gg \tau_1$, $\mathscr{P}_{circ}(H)$ decreases from $-p_e/2$ to $-\frac{4}{3}p_e\tau_1/\tau_2$ as the magnetic field increases. For $\tau_1 \ll \tau_S \ll \tau_2$ in a strong magnetic field, $\mathscr{P}_{circ}(H)$ also decreases from $-\frac{3}{11}p_e$ to its limiting value $-\frac{10}{9}p_e\tau_1/\tau_S$.

The reduction of the radiation polarization in a strong magnetic field for $\tau_S \gg \tau_2 \gg \tau_1$ is due to the following cause: as seen from (5) and (9), the average angular momentum $\bar{J}_z$ transferred to the system per unit time in the course of exciton formation by bound pairs is equal to $\frac{1}{2}G_0P_e$. It is precisely this value of momentum that is gained by the optically inactive states 4 and 8 with $J_z = \pm 2$, so that for $\tau_S \gg \tau_2$ this entire momentum is transferred to the lattice. All the other states together gain zero net momentum. In a strong magnetic field they all become optically active, and at $\tau_2 \gg \tau_1$, when their entire angular momentum is transferred to the radiation only, the latter can carry away no momentum and remains unpolarized. Polarization becomes possible only when the optically active states can transfer to the lattice an angular momentum compensating the momentum carried away by light, i.e. at $\tau_2 \sim \tau_1$.

At H = 0 the required momentum is transferred to the lattice by the states 5–7, which are optically inactive in the absence of the magnetic field. These states acquire a momentum which is precisely equal to $-\frac{1}{4}G_0p_e$ and compensates the momentum gained by the optically active states 1–3, and in this case $\mathscr{P}_{circ} = -p_e/2$ independently of the time ratio τ_2/τ_1.

In the resonant mode of exciton generation, the optically inactive states are not excited and the entire momentum goes to the optically active states at $\tau_S \gg \tau_1, \tau_2$. The degree of luminescence polarization in a strong magnetic field is then equal to the polarization of the incident light regardless of the ratio τ_1/τ_2, as in the case H = 0.

5. OPTICAL ORIENTATION IN A TRANSVERSE MAGNETIC FIELD

We shall consider the effect that a transverse magnetic field has on optical orientation in two limiting cases: large and small exchange interaction. If the exchange splitting Δ exceeds T_{15}^{-1}, the states with J = 1 and J = 2 can be treated separately, taking into account only those transitions that are due to spin relaxation. In doing so we neglect quantities of order $(\Delta T_{15})^{-2}$ and accordingly consider the magnetic fields for which the Zeeman splitting is less than the exchange splitting. According to (1)–(9), the equation for each of the density matrices $\rho^{(J)}$ can be written in the form:

$$\rho^{(J)} + ig_J T_J [(\mathbf{JH})\rho^{(J)}] = \frac{1}{8} N \frac{T_J}{\bar{T}} \left[1 \mp \frac{\bar{T}}{2\bar{\tau}} \left(p_e J_z + \frac{\bar{\tau}}{\tau_s} (\bar{J}\bar{\sigma}) \right)\right];$$
$$\bar{\sigma} = \tfrac{1}{2}(\bar{\mathbf{J}}^{(2)} - \bar{\mathbf{J}}^{(1)}), \quad g_1 = \tfrac{1}{4}(-g_e + 5g_h),$$
$$g_2 = \tfrac{1}{4}(g_e + 3g_h), \quad \bar{\mathbf{J}}^{(J)} = N^{-1}\,\mathrm{Sp}\,(\rho^{(J)}\mathbf{J}). \tag{33}$$

Multiplying (33) by J_i and taking the traces of both sides of the equation, we obtain a system of equations for $\bar{J}_i^{(J)}$. Evaluating the trace of (33), we find that $\bar{\tau}$ does not depend on the magnetic field in this case and is given by (24).

The total radiation intensity in an arbitrary direction, which in the excitation mode under consideration is determined by $J_i^{2(1)}$, is also independent of H and is equal to

$$J_i^{2(1)} = T_1/4\bar{T}.$$

As a result, the following expression is obtained for the degree of circular polarization of luminescence emitted in the direction of the incident light:

$$\mathscr{P}_{\substack{\text{circ}\\ z}} = \frac{\bar{J}_z^{(1)}}{\bar{J}_z^{2(1)}} = -\frac{1}{2} p_e \frac{\bar{T}}{\bar{\tau}} \frac{1}{1+\omega_1^2 T_1^2} \frac{1-\chi_1-\omega_1 T_1 \chi_2}{(1-\chi_1)^2+\chi_2^2}. \tag{34}$$

As usual, a transverse field gives rise to a circular polarization of the luminescence in the direction perpendicular to both the magnetic field and the incident radiation. At $H = H_x$

$$\mathscr{P}_{\substack{\text{circ}\\ y}} = \frac{\bar{J}_y^{(1)}}{\bar{J}_y^{2(1)}} = \frac{1}{2} \frac{\bar{T}}{\bar{\tau}} \frac{\omega_1 T_1}{1+\omega_1^2 T_1^2} \frac{1-\chi_1+\chi_2/\omega_1 T_1}{(1-\chi_1)^2+\chi_2^2}; \tag{35}$$

$$\chi_1 = \frac{1}{16\tau_s}\left[\frac{T_1}{1+\omega_1^2 T_1^2} + \frac{5T_2}{1+\omega_2^2 T_2^2}\right],$$
$$\chi_2 = \frac{1}{16\tau_s}\left[\frac{\omega_1 T_1^2}{1+\omega_1^2 T_1^2} + \frac{5\omega_2 T_2^2}{1+\omega_2^2 T_2^2}\right]. \tag{36}$$

At $\tau_S \gg \tau_1, \tau_2$, when the replenishment terms in (34), (35) associated with spin relaxation are small, the usual Hanle effect occurs. At $\tau_S \lesssim \tau_2$ and $\omega_2 T_2 \gg \omega_1 T_1$, the $\mathscr{P}_{circ}^{Z}(H)$ curve has two steps: in the field range where $\omega_2 T_2 \gg 1$, $\omega_1 T_1 \ll 1$, the value of $\mathscr{P}_{circ}^{Z}$ drops to

$$\mathscr{P}_{\substack{\text{circ}\\ z}}(0) = \left(1 - \frac{T_1}{16\tau_s}\right) \Big/ \left(1 - \frac{T_1+5T_2}{16\tau_s}\right),$$

and then, for $\omega_1 T_1 \gg 1$, the value of $\mathscr{P}_{circ}^{Z}$ drops to zero in a Lorentzian manner with

$$\omega T = \omega_1 T_1 \Big/ \left(1 - \frac{T_1}{16\tau}\right),$$

Of these two ranges, the second (i.e. when $\omega_1 \sim T_1^{-1}$) involves an appreciable polarization in the direction of the y axis. Such a step-like attenuation of the polarization

$\mathscr{P}_{circ}(H)$ can occur, in particular, at $\tau_1 \ll \tau_S \ll \tau_2$ (and $\omega_1 \approx \omega_2$) when $T_1 = \tau_1$ and $T_2 = \tau_S$. In this case, the polarization decreases from $-{}^3/_{11} p_e$ to $-{}^3/_{16} p_e$ on the first step, i.e. by 30%.

For small exchange splitting, when $\Delta T_{15} \ll 1$, and for $T_2 - T_1 \ll T_2$ (i.e. at $\tau_1 \approx \tau_2$ or $\tau_S \ll \tau_1, \tau_2$) the degree of circular polarization in a transverse magnetic field is given by the same expressions as in the case of free electrons:

$$\mathscr{P}^z_{circ} = -\frac{1}{2} p_e \frac{T_e}{\bar{\tau}} \frac{1}{1+\omega_e^2 \tau_{es}^2}, \quad \mathscr{P}^y_{circ} = \frac{1}{2} p_e \frac{T_e}{\bar{\tau}} \frac{\omega_e \tau_{es}}{1+\omega_e^2 \tau_{es}^2}, \quad \omega_e = g_e \mu_0 H, \quad T_e^{-1} = \bar{\tau}^{-1} + \tau_{es}^{-1}, \tag{37}$$

where τ_{es} is the above-introduced electron spin relaxation time given by (26) and (28). In this case the variation of τ_{es} in the transverse magnetic field need not be taken into account, since at $\omega\tau_{es} \sim 1$ the relative change in τ_{es} is of order $\Delta T_{15} \ll 1$. We note that if the times τ_1 and τ_2 differ substantially and $\tau_S \gtrsim \tau_1$, then at H = 0 a strong polarization of holes occurs, as suggested by (27a) and (27b), and in this case the simple expressions (37) would not describe the polarization attenuation even at $\Delta = 0$.

It has been shown previously [1] that uniaxial crystals in a transverse magnetic field give rise to a linearly polarized luminescence on excitation with circularly polarized light. In the absence of exchange splitting (and at $\tau_1 = \tau_2$), this effect does not occur, as it does not occur in the case of free carriers. However, as will be apparent from (33), for cubic crystals in the bound-pair mode of exciton formation with a large exchange splitting (when $\Delta T_{15} \gg 1$), the linearly polarized luminescence appears entirely as a result of mixing of states with J = 1 and J = 2 by the magnetic field, i.e. at $\omega \sim \Delta$. Since at $\omega \gg T^{-1}$ the circular polarization is attenuated by the Hanle effect as $(\omega T)^{-2}$, its transformation to linear polarization can occur appreciably in cubic crystals only at $\Delta T_{15} \sim 1$.

In the case of a strong magnetic field the excitons may align in the transverse magnetic field upon their excitation with unpolarized light at $\tau_1 \neq \tau_2$. However, the degree of polarization in this case is small: at maximum field strength, when ω_i exceeds both Δ and τ_i^{-1}, it is equal (for $\tau_S \gg \tau_1, \tau_2$) to

$$\mathscr{P}^{lin}_{H\to\infty} = \frac{\tau_1(\tau_2-\tau_1)}{4\tau_1^2+3\tau_2^2+9\tau_1\tau_2}$$

thus not exceeding 3.5%.

The exciton alignment may be sharper in the case of resonant exciton generation with unpolarized light. In this case, for $\Delta\tau_1 \gg 1$ and $\tau_S \gg \tau_1$ in the field domain where $\omega_1 < \Delta$, the polarization

$$\mathscr{P}_{lin} = \frac{(\omega_1\tau_1)^2}{1+3(\omega_1\tau_1)^2},$$

i.e., it reaches 33%. At $\tau_S \ll \tau_1$ this effect is reduced by a factor of τ_S/τ_1. In stronger fields, when $\omega \gg \Delta$, for resonant excitation and $\tau_S \gg \tau_1, \tau_2$ the value of $\mathscr{P}_{lin}$ drops to ${}^3/_{13}$ at $\tau_1 = \tau_2$, and at $\tau_1 \ll \tau_2$ it remains at the level $\mathscr{P}_{lin} = {}^1/_3$.

We note in conclusion that the developed theory is applicable in principle to both electrons and holes localized in donor-acceptor pairs which can be regarded as an exciton localized on a complex defect. It is the electron orientations on such pairs that were treated by D'yakonov and Perel' [5,6]. As was mentioned in the Introduction, our calculations are also valid in the case when the impurity concentration is small and the temperature is low enough so that the time between carrier hops from one impurity to another exceeds the lifetime or the spin relaxation time of electrons and holes. It should be borne in mind, however, that due to the distortion and electric fields produced by a pair the electron localized on it has a symmetry lower than cubic, which leads to a splitting of the degenerate hole levels. For a large exchange interaction, the indicated splitting, like the splitting due to an external transverse deformation, involves a substantial reduction of the degree of orientation (even at a very small deformation splitting comparable to τ_S^{-1} or $\tau_{1,2}^{-1}$). On the other hand, a longitudinal magnetic field reduces the influence of transverse deformation. This effect, as well as an increase in τ_{es}, may also provide an explanation for the increase of the radiation polarization in a longitudinal magnetic field.

The above effects will be considered in more detail in a separate paper.

[1)] If the capture of electrons and holes occurs not directly, but through binding of the generated free electrons, the optically active (in the J = 1 states) free electrons can then rapidly lose their orientation due to the annihilation interaction, and in this case the holes can acquire an appreciable orientation in the course of entrapment. Such an orientation can also be caused by rapid radiative recombination of free excitons with H = 1, which results in a preferential entrapment of the J = 2 excitons. These mechanisms can lead to an orientation of holes (and excitons) on excitation of pairs in an n-material at low temperatures, when all electrons are bound to donors, although under these conditions neither free holes nor bound electrons are oriented.

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Translated by S. Luryi
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