Transfer of polarized light in crystals at wavelengths corresponding to the exciton part of the spectrum. Influence of reemission

E. L. Ivchenko, G. E. Pikus, and N. Kh. Yuldashev

A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad
(Submitted 7 May 1980)
Zh. Eksp. Teor. Fiz. 79, 1573–1590 (October 1980)

An analysis is made of the influence of reemission and multiple reflection of light from a surface on the intensity and polarization of luminescence in resonant excitation of excitons in a cubic crystal when excitons are scattered elastically by impurities. An integral equation is obtained for a matrix S^R relating the light fluxes incident on and scattered from an internal surface of a crystal. This equation is reduced to a system of simpler integral equations with one variable. Numerical solution of these equations is used to calculate the dependences of the intensity and polarization of light on the refractive index, quantum efficiency of a single scattering event, and directions of propagation of the exciting and scattered light. An analysis is made of the characteristic features which appear in the backscattering as a result of interference effects associated with multiple scattering of excitons. The theory developed is applicable also to the Rayleigh scattering of light by defects in isotropic solid or liquid media.

PACS numbers: 71.35. + z, 72.10.Fk, 61.70.Rj, 78.35. + c

§1. INTRODUCTION

A theory of the optical orientation of excitons by resonant excitation has been developed in earlier investigations[1,2] in which two authors of the present paper participated. It is assumed in these investigations that: a) excitons and photons can be regarded as weakly interacting particles, i.e., that the mean free path of an exciton $l_e = v_{q0}\tau$ is much less than the path $l_{rad} = v_{q0}\tau_{rad}$ of an exciton in the case when a photon is emitted; b) the diffusion length of an exciton $l_{diff} = v_{q0}(\tau_t\tau)^{1/2}$ is short compared with the mean free path of a photon $\alpha^{-1} \approx l_{rad}$; c) the nonradiative exciton lifetime τ_0 is much shorter than the radiative lifetime τ_{rad}. If we bear in mind that $\alpha = 2\tau\omega_{LT}q_0$, we find that the conditions a)–c) can be written in the form

$$\omega_{LT}\tau \ll 1/\omega_{q0}\tau, \quad (1a)$$

$$\omega_{LT}\tau \ll 1/\omega_{q0}(\tau_t\tau)^{1/2}, \quad (1b)$$

$$\omega_{LT}\tau \ll 1/\omega_{q0}\tau_0. \quad (1c)$$

Here, ω_{LT} is the longitudinal–transverse splitting of the exciton branch far from resonance; $\tau = (2\Gamma)^{-1} = (\tau_p^{-1} + \tau_t^{-1})^{-1}$ is the total drift time; τ_p is the exciton momentum relaxation time; $\tau_t = (\tau_0^{-1} + \tau_{rad}^{-1})^{-1}$ is the total exciton lifetime; $\hbar\omega_{q0}$, v_{q0}, and $\mathbf{q}_0$ are the kinetic energy, velocity, and wave vector of an exciton at the point of intersection of the exciton and photon branches.

We can see that the last of the three conditions (1a), (1b), and (1c) is the most stringent. When this condition is satisfied we can ignore the reemission processes. In the case of GaSe crystals investigated earlier,[1,2] the condition (c) is well satisfied, as indicated by a high degree of polarization of the exciton luminescence. Gallium selenide differs from the majority of semiconductors in that it has a small value of ω_{LT} and a short lifetime τ_0, facts associated with the characteristics of its energy band structure. Therefore, one has to justify the application of this theory to other crystals.

We shall consider the influence of reabsorption and reemission on the polarization of the exciton luminescence, i.e., we shall consider the case of an arbitrary relationship between τ_0 and τ_{rad}. We shall assume that the first two conditions (1a) and (1b) are satisfied; this is possible if $\tau_p \ll \tau_t$. In this case the exciton momentum distribution becomes completely isotropic during the total exciton lifetime τ_t. For simplicity, we shall consider a triplet exciton in a cubic crystal. As before,[1] we shall assume that the scattering of excitons is elastic so that the frequencies of the incident and emitted light are identical. Moreover, we shall ignore the spin relaxation of excitons on the assumption that the spin relaxation times τ_s of an electron and a hole in an exciton are longer than τ_t. In the case of spin relaxation of excitons associated with the longitudinal–transverse splitting, the time $\tau_s \sim (\omega_{LT}^2\tau)^{-1}$ is known to be longer than τ_t because of the condition (1b).

When the above conditions are satisfied, the light-scattering matrix differs from the Rayleigh form only by a different frequency dependence of the absorption coefficient α and by a general factor $\tilde{\omega}_0 = \tau_t/\tau_{rad}$, which governs the quantum efficiency of the luminescence for a single scattering of a photon by an exciton. Therefore, the problem considered here is equivalent to that of the Rayleigh scattering of light in a turbid isotropic medium. Problems of this kind have been frequently considered in astrophysics and geophysics, and methods for solving them are given in detail in the monographs of Chandrasekhar[3] and Sobolev.[4] In problems of this kind it is assumed that the density of the scattering medium is low and that its permittivity is practically identical with that of vacuum, so that the reflection and refraction of light at the boundary with vacuum can be ignored. In the case of a solid the reflection of light from an internal surface plays an important role. Agranovich and Konobeev[5] as well as Doronina *et al.*[6] (their work is recounted in the mono-

 0038-5646/80/100793-09$02.40

graph of Agranovich and Galanin[7]) considered the influence of reflection on the luminescence decay time. Gutshabash[8] and Zege with Katsev[9] analyzed the influence of reflection on the intensity of scattered light in the case of specular reflection and also reflection in accordance with the Lambert law. However, in all these cases[4-9] the question of polarization of the luminescence is not considered, the luminescence is considered to be unpolarized, and the equation of propagation for the matrix of the Stokes parameters I is replaced by an approximate equation for the total intensity I.

We shall calculate the change in the polarization of scattered light allowing for the reflection and refraction at a boundary (the results of the present work are reported partly in a short communication[10]). A modification of the proposed method, which will be given in a separate paper, makes it possible to consider the scattering of polarized light also in the opposite limiting case of a strong exciton–photon interaction, when the inequalities opposite to those of (1a)–(1c) are satisfied and one has to allow for the polariton effect.

§2. FORMULATION OF THE PROBLEM

We shall use the S matrix method developed by Chandrasekhar.[3] We shall introduce the Stokes matrix $\mathbf{I}(\Omega)$ which represents the intensity and polarization of the radiation scattered in a crystal when traveling in the direction Ω, defined by the polar angle θ between Ω and the normal to the surface, and by the azimuthal angle φ. The positive values of $\mu=\cos\theta$ represent rays traveling toward the surface and the negative values represent those traveling away from the surface. In Chandrasekhar's book[3] the column matrix I is selected in the form

$$\mathbf{I}=\begin{bmatrix} I_l\\ I_r\\ U\\ V\end{bmatrix};\quad I_l=d_{ll},\quad I_r=d_{rr},\quad U=2\,\mathrm{Re}\,d_{lr},\quad V=2\,\mathrm{Im}\,d_{lr},\qquad (2)$$

$$d_{ij}=(8\pi)^{-1}cn\langle E_iE_j^*\rangle.$$

The unit vector l lies in a meridional plane containing Ω and the normal to the z plane, whereas the unit vector r is perpendicular to this plane; E_l and E_r are the corresponding components of the electric field of the light wave; n is the refractive index.

The degree of linear polarization of the radiation $\mathscr{P}'_{\rm lin}$ in the system of (l,r) axes, the degree of linear polarization $\mathscr{P}''_{\rm lin}$ in the system of (l',r') axes rotated relative to the (l,r) axes by 45°, and the degree of circular polarization $\mathscr{P}_{\rm circ}$ are related to the components of I by

$$\mathscr{P}'_{\rm lin}=(I_l-I_r)/I,\quad \mathscr{P}''_{\rm lin}=U/I,\quad \mathscr{P}_{\rm circ}=V/I,\qquad (3)$$

where $I=I_l+I_r$. The quantity $I(\Omega)/d\Omega$ gives the total intensity of the scattered radiation traveling in the direction Ω in a solid angle $d\Omega$.

We shall assume that a plane light wave is incident on a crystal and after refraction it travels in the direction Ω_0 given by the angles θ_0 and φ_0. Then, the radiation transmitted by the boundary of the crystal ($z=+0$) is described by

$$\mathbf{I}_0(+0,\mu,\varphi)=\pi\mathbf{F}\delta(\mu+\mu_0)\delta(\varphi-\varphi_0),\qquad (4)$$

where $\mu_0=-\cos\theta_0>0$. In this case the equation for the matrix $\mathbf{I}(\tau,\Omega)$ representing the scattered radiation becomes

$$\mu d\mathbf{I}(\tau,\Omega)/d\tau=\mathbf{I}(\tau,\Omega)-\frac{\tilde\omega_0}{4\pi}\int_{-1}^{1}\int_0^{2\pi}d\mu'\,d\varphi'\mathbf{P}(\Omega,\Omega')\mathbf{I}(\tau,\Omega')$$
$$-\frac{\tilde\omega_0}{4}\exp\left(-\frac{\tau}{\mu_0}\right)\mathbf{P}(\Omega,\Omega_0)\mathbf{F},\qquad (5)$$

where $\tau=\alpha z$; z is the distance from the surface; α is the absorption coefficient; $\mathbf{P}(\Omega,\Omega')$ is the Rayleigh scattering matrix.

According to Ref. 3, the matrix $\mathbf{P}(\Omega,\Omega')$ described by the representation (2) is

$$\mathbf{P}(\Omega,\Omega')=\mathbf{Q}\{{}^3/_4\mathbf{P}^{(0)}(\mu,\mu')$$
$$+[(1-\mu^2)(1-\mu'^2)]^{1/2}\mathbf{P}^{(1)}(\Omega,\Omega')+\mathbf{P}^{(2)}(\Omega,\Omega')\}.\qquad (6)$$

Here,

$$\mathbf{Q}=\begin{bmatrix}1&0&0&0\\0&1&0&0\\0&0&2&0\\0&0&0&2\end{bmatrix},$$

$$\mathbf{P}^{(0)}(\mu,\mu')=\begin{bmatrix}2(1-\mu^2)(1-\mu'^2)+\mu^2\mu'^2&\mu^2&0&0\\ \mu'^2&1&0&0\\0&0&0&0\\0&0&0&\mu\mu'\end{bmatrix},$$

$$\mathbf{P}^{(1)}(\Omega,\Omega')=\frac{3}{4}\begin{bmatrix}4\mu\mu'\cos\psi&0&2\mu\sin\psi&0\\0&0&0&0\\-2\mu'\sin\psi&0&\cos\psi&0\\0&0&0&\cos\psi\end{bmatrix},$$

$$\mathbf{P}^{(2)}(\Omega,\Omega')=\frac{3}{4}\begin{bmatrix}\mu^2\mu'^2\cos2\psi&-\mu^2\cos2\psi&\mu^2\mu'\sin2\psi&0\\-\mu'^2\cos2\psi&\cos2\psi&-\mu'\sin2\psi&0\\-\mu\mu'^2\sin2\psi&\mu\sin2\psi&\mu\mu'\cos2\psi&0\\0&0&0&0\end{bmatrix},$$

where $\psi=\varphi'-\varphi$. In Eq. (6), the matrix $\mathbf{P}(\Omega,\Omega')$ is normalized to unity, so that

$$\sum_{i=l,r}\int_{-1}^{1}\int_0^{2\pi}\frac{d\mu\,d\varphi}{4\pi}P_{ij}(\Omega,\Omega')=1\qquad (j=l,r).$$

The boundary condition for Eq. (5) should allow for the reflection by the surface. In the case of a semiinfinite medium and specular reflection, we have

$$\mathbf{I}(+0,\bar\Omega)=\mathbf{R}(\mu)\mathbf{I}(+0,\Omega),\ \mathbf{I}(\infty,\Omega)=0,\qquad (7)$$

where the directions Ω and $\bar\Omega$ are related by the condition for specular reflection $\bar\mu=-\mu<0$, $\bar\varphi=\varphi$. It follows from the Fresnel formulas[11] that the reflection matrix R is

$$\mathbf{R}(\mu)=\begin{bmatrix}R_1&0&0&0\\0&R_2&0&0\\0&0&R_3&-R_4\\0&0&R_4&R_3\end{bmatrix}.\qquad (8)$$

For angles θ smaller than the total internal reflection angle $\theta_0=\sin^{-1}(1/n)$, we have

$$R_1=r_p^2,\ R_2=r_s^2,\ R_3=r_sr_p,\ R_4=0,\qquad (9)$$

where

$$r_p=\frac{\mu-n\mu'}{\mu+n\mu'},\quad r_s=\frac{n\mu-\mu'}{n\mu+\mu'},\quad (1-\mu'^2)=n^2(1-\mu^2).$$

Here, $n=n_{\rm cr}/n_0$, $n_{\rm cr}$, and n_0 are the refractive indices of the crystal and the surrounding medium (in vacuum we have $n_0=1$). If $\theta>\theta_0$, then

$$R_1=R_2=1,\ R_3=\cos\delta,\ R_4=\sin\delta, \tag{10}$$

where δ is found from

$$\mathrm{tg}(\delta/2)=-\mu[(1-\mu^2)-n^{-2}]/(1-\mu^2).$$

The four-component vector representing the energy flux $\mathbf{F}(+0,\Omega_0)$ on the internal surface of the crystal ($z=+0$) is related to the corresponding quantity $\mathbf{F}(-0,\Omega_1)$ on the external surface ($z=-0$) by

$$\mathbf{F}(+0,\Omega_0)=\mu_0^{-1}\mu_1\mathbf{T}(\mu_0,\mu_1)\mathbf{F}(-0,\Omega_1), \tag{11}$$

where the matrix of the transmission coefficients is

$$\mathbf{T}(\mu_0,\mu_1)=\begin{bmatrix} T_1 & 0 & 0 & 0\\ 0 & T_2 & 0 & 0\\ 0 & 0 & T_3 & 0\\ 0 & 0 & 0 & T_3\end{bmatrix},$$

whose components are $T_1(\mu_0)=1-R_1(\mu_0)$, $T_2(\mu_0)=1-R_2(\mu_0)$, $T_3(\mu_0)=(T_1T_2)^{1/2}$, and μ_1 and μ_2 are related by the Snell formula

$$1-\mu_1^2=n^2(1-\mu_0^2). \tag{12}$$

The light scattered out of the crystal is characterized by

$$\mathbf{I}(-0,\Omega_2)=n^{-2}\mathbf{T}(\mu_2,\mu)\mathbf{I}(+0,\Omega), \tag{13}$$

where μ_2 and μ are again related by Eq. (12). In Eq. (13) an allowance is made for the change in the solid angle in which the radiation is scattered out of the crystal because of refraction: it follows from Eq. (12) that $d\Omega_2=n^2\mu/\mu_2 d\Omega$.

§3. EQUATION FOR THE MATRIX S^R

In the absence of reflection the scattered flux $\mathbf{I}(0,\Omega)$ is related to the incident flux at the boundary $\mathbf{I}(0,\overline{\Omega}')$ by the S matrix:[3]

$$\mathbf{I}(0,\Omega)=\frac{\tilde\omega_0}{4\pi\mu_0}\int_0^1 d\mu'\int_0^{2\pi} d\varphi'\mathbf{S}(\Omega,\Omega')\mathbf{I}(0,\overline{\Omega}'). \tag{14}$$

Equation (14) is valid not only at the boundary of a medium but on any plane inside it. This statement represents the principle of invariance, first formulated by Ambartsumyan.[12] This principle is used in Ref. 3 to obtain the integral equation for the matrix S. In the presence of a reflecting boundary we can introduce, by analogy with Eq. (14), a matrix $\mathbf{S}^R$ relating the incident and reflected fluxes on an internal surface in a crystal. For example, when a plane light wave is incident on a crystal [see Eq. (4)], then

$$\mathbf{I}(+0,\Omega)=(4\mu)^{-1}\tilde\omega_0\mathbf{S}^R(\Omega,\Omega_0)\mathbf{F}(+0,\overline{\Omega}_0). \tag{15}$$

The principle of invariance is invalid when the reflection occurs at a boundary. However, Eq. (14) is valid on any other surface inside a crystal. This allows us to derive an integral equation relating the matrices $\mathbf{S}^R$ and S. We shall actually consider a plane parallel to the boundary and located at a distance $\tau\ll 1$. The flux $\mathbf{I}(\tau,\Omega')$ incident on this plane consists of the primary flux (4) and the flux (7) reflected from the boundary. Therefore, it follows from Eqs. (4), (7), and (14) that if $\tau\ll1$, we have

$$\mathbf{I}(\tau,\Omega)=\frac{\tilde\omega_0}{4\mu}\mathbf{S}(\Omega,\Omega_0)\mathbf{F}(\overline{\Omega}_0)$$
$$+\frac{\tilde\omega_0}{4\pi\mu}\int_0^1 d\mu'\int_0^{2\pi} d\varphi'\mathbf{S}(\Omega,\Omega')\mathbf{R}(\Omega')\mathbf{I}(\tau,\Omega'). \tag{16}$$

We shall now assume that $\tau=+0$ and substitute in Eq. (16) the expression (15) for $\mathbf{I}(+0,\Omega)$, which gives the following integral equation for the matrix $\mathbf{S}^R$:

$$\mathbf{S}^R(\Omega,\Omega_0)=\mathbf{S}(\Omega,\Omega_0)+\frac{\tilde\omega_0}{4\pi}\int_0^1\frac{d\mu'}{\mu'}\int_0^{2\pi} d\varphi'\mathbf{S}(\Omega,\Omega')\mathbf{R}(\mu')\mathbf{S}^R(\Omega',\Omega_0). \tag{17}$$

It should be noted that the minimum value of μ_0 for light entering the crystal is $(1-n^{-2})^{1/2}$. However, formally, Eq. (17) describes the matrix $\mathbf{S}^R(\Omega,\Omega_0)$ throughout the full range of μ_0 and μ between 0 and 1.

§4. SYMMETRY PROPERTIES OF THE MATRIX S^R

Before calculating the matrix $\mathbf{S}^R$, we shall consider the general requirements which are imposed on this matrix by the symmetry conditions. In the case of a cubic crystal (and also a uniaxial crystal whose illuminated surface is perpendicular to the principal axis) the matrix $\mathbf{S}^R$ should be invariant under the operations described by the symmetry groups $C_{\infty v}$, i.e., it should not be affected by rotation through any angle about the z axis or reflection in planes passing through this axis. It follows from the first requirement that $\mathbf{S}^R(\Omega,\Omega_0)$ can depend only on the difference between the angles $\varphi_0-\varphi$:

$$\mathbf{S}^R(\Omega,\Omega_0)=\mathbf{S}^R(\mu,\mu_0,\varphi_0-\varphi). \tag{18}$$

Reflection in the xz ($\varphi=0$) plane converts E_l to E_l, E_r to $-E_r$, φ to $2\pi-\varphi$ and φ_0 to $2\pi-\varphi_0$. Therefore, it follows from the second requirement that

$$\mathbf{S}^R(\mu,\mu_0,\varphi_0-\varphi)=\mathbf{D}^{-1}\mathbf{S}^R(\mu,\mu_0,-(\varphi_0-\varphi))\mathbf{D}, \tag{19}$$

where

$$\mathbf{D}=\begin{bmatrix}1 & 0 & 0 & 0\\ 0 & 1 & 0 & 0\\ 0 & 0 & -1 & 0\\ 0 & 0 & 0 & -1\end{bmatrix}.$$

In particular, in the case of a ray scattered in the plane of incidence, i.e., when $\varphi=\varphi_0$, we have

$$\mathbf{S}^R(\mu,\mu_0,0)=\mathbf{D}^{-1}\mathbf{S}^R(\mu,\mu_0,0)\mathbf{D} \tag{20}$$

and consequently, the matrix $\mathbf{S}^R(\mu,\mu_0,0)$ splits into two 2×2 blocks and its nondiagonal components vanish: $S^R_{ik}=S^R_{ki}=0$ ($i=1$ or 2, $k=3$ or 4).

In the case of normal incidence and normal scattering, i.e., when $\mu=\mu_0=1$, the components E_l and E_r transform in accordance with the irreducible representation E_1 of the $C_{\infty v}$ group. Consequently, the components of the Stokes matrix I of Eq. (2) transform in accordance with the representations $E_1\times E_1=A_1+A_2+E_2$, namely $I=(I_l+I_r)$ transforms in accordance with A_1, $Q=(I_l-I_r)$ and U in accordance with E_2, and V in accordance with A_2. Therefore, the matrix $\mathbf{S}^R(1,1,0)$ should be goverened by three linearly independent coefficients S_1, S_2, and S_3 that connect the components I and $\mathbf{I}_0$ and transform in accordance with the same irreducible representation. In the basis of Eq. (2), the matrix $\mathbf{S}^R(1,1,0)$ has the form

$$S^R(1,1,0)=\begin{bmatrix} S & S' & 0 & 0 \\ S' & S & 0 & 0 \\ 0 & 0 & S_2 & 0 \\ 0 & 0 & 0 & S_3 \end{bmatrix},$$

$$S={}^1/_2(S_1+S_2),\quad S'={}^1/_2(S_1-S_2). \tag{21}$$

It should also be noted that in the case of normal incidence the total scattered flux $\pi\mathcal{F}$ [see Eq. (49) below], which also transforms in accordance with the representation A_1, depends only on the component $F=F_l+F_r$ and, therefore, it is independent of the polarization of the radiation.

The scattering matrix satisfies the reciprocity condition

$$\mathbf{Q}^{-1}\mathbf{P}(\Omega,\Omega_0)=\mathbf{K}\tilde{\mathbf{P}}(\bar{\Omega}_0,\bar{\Omega})\mathbf{Q}^{-1}\mathbf{K}^{-1}. \tag{22}$$

Hence, it follows that

$$\mathbf{P}(\Omega,\Omega_0)=\mathbf{K}\mathbf{P}(\Omega,\Omega_0)=\mathbf{G}\tilde{\mathbf{P}}(\bar{\Omega}_0,\bar{\Omega})\mathbf{G}^{-1}. \tag{23}$$

Here,

$$\mathbf{K}=\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix},\quad \mathbf{G}=\mathbf{QK}=\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -2 & 0 \\ 0 & 0 & 0 & 2 \end{bmatrix}.$$

As before, $\bar{\Omega}_0$ corresponds to the case $\mu=-\mu_0$, $\varphi=\varphi_0$ and the tilde represents the matrix transposition operation. As pointed out in Ref. 3, the matrix **S** of Eq. (14) also satisfies a condition similar to Eq. (23):

$$\mathbf{S}(\Omega,\Omega_0)=\mathbf{K}\mathbf{S}(\Omega,\Omega_0)=\mathbf{G}\tilde{\mathbf{S}}(\Omega_0,\Omega)\mathbf{G}^{-1}. \tag{24}$$

This can be proved by applying the operation K to the integral equation for the S matrix[3] or by using the formulas (29)–(32) derived below for this matrix.

We shall show that the matrix $\mathbf{S}^R$ satisfies Eq. (24). We shall do this by writing down, employing the iteration method, the solution of Eq. (17) in the form of an expansion in the powers of $\tilde{\omega}_0$:

$$\mathbf{S}^R(\Omega,\Omega_0)=\sum_{\nu=0}^{\infty}\tilde{\omega}_0^{\nu}\mathbf{S}_\nu^R(\Omega,\Omega_0), \tag{25}$$

where

$$\mathbf{S}_0^R(\Omega,\Omega_0)=\mathbf{S}(\Omega,\Omega_0),$$

$$\mathbf{S}_\nu^R(\Omega,\Omega_0)=\frac{1}{(4\pi)^\nu}\int_0^1\frac{d\mu_1}{\mu_1}\int_0^{2\pi}d\varphi_1\ldots\int_0^1\frac{d\mu_\nu}{\mu_\nu}\int_0^{2\pi}d\varphi_\nu[\mathbf{S}(\Omega,\Omega_1)$$
$$\times\mathbf{R}(\mu_1)\mathbf{S}(\Omega_1,\Omega_2)\ldots\mathbf{S}(\Omega_{\nu-1},\Omega_\nu)\mathbf{R}(\mu_\nu)\mathbf{S}(\Omega_\nu,\Omega_0)]\quad(\nu\geq1). \tag{26}$$

The term $\mathbf{S}_\nu^R$ with $\nu\geq1$ describes the contribution to $\mathbf{S}^R$ associated with the reflection of light ν times from the surface. Application to Eq. (26) for $\mathbf{S}_\nu^R$ of the operation K in accordance with Eq. (23) and the use of Eq. (24) easily demonstrates that $\mathbf{K}\mathbf{S}_\nu{}^R=\mathbf{S}_\nu{}^R$ and, consequently,

$$\mathbf{S}^R(\Omega,\Omega_0)=\mathbf{K}\mathbf{S}^R(\Omega,\Omega_0)=\mathbf{G}\tilde{\mathbf{S}}^R(\Omega_0,\Omega)\mathbf{G}^{-1}. \tag{27}$$

The relationship (27) is the result of invariance under time inversion: when the directions of the incident and scattered rays are reversed, μ becomes $-\mu_0$, μ_0 becomes $-\mu$, φ becomes $\varphi_0+\pi$, and φ_0 becomes $\varphi+\pi$. It follows from Eq. (18) that the change in φ and φ_0 by π does not alter the form of the matrix $\mathbf{S}^R$. Reversal of the sign of the component U is related to the selection of the basis unit vectors: when the direction Ω is reversed, i.e., when μ is replaced with $-\mu$ and φ with $\varphi+\pi$, the value of E_l becomes E_l^*, whereas E_r becomes E_r^*. In general, on the right-hand side of Eq. (27) we should replace $\tilde{\mathbf{S}}^R$ with $\mathbf{S}^{R+}=\tilde{\mathbf{S}}^{R*}$, but since the basis (2) is selected to be real, it follows that $\mathbf{S}^{R+}=\tilde{\mathbf{S}}^R$. The factor Q in Eq. (22) is also associated with the selection of the basis: for the unitary representation $\mathbf{I}'=\mathbf{Q}^{-1/2}\mathbf{I}$ we find that the matrix becomes $\mathbf{G}=\mathbf{K}$. As shown above, it follows from Eq. (20) that in the case of a ray scattered in the plane in incidence, i.e., when $\varphi=\varphi_0$, the matrix $\mathbf{S}^R$ splits into two 2×2 blocks. It follows from Eq. (27) that the components of these blocks are related by

$$S_{ii}{}^R(\mu,\mu_0,0)=S_{ii}{}^R(\mu_0,\mu,0),\quad i=1-4,$$
$$S_{12}{}^R(\mu,\mu_0,0)=S_{21}{}^R(\mu_0,\mu,0),\quad S_{34}{}^R(\mu,\mu_0,0)=-S_{43}{}^R(\mu_0,\mu,0). \tag{28}$$

It should be noted that the matrix **S**, like the matirx $\mathbf{P}(\Omega,\Omega_0)$, has components $S_{i4}=S_{4i}=0$ $(i\neq4)$ which vanish for any value of Ω and Ω_0. The appearance of these components in the case of the matrix $\mathbf{S}^R$ is associated with the reflection of light from a boundary and is explained by the existence of the corresponding components $R_{34}=-R_{43}$ of the matrix **R**.

§5. GENERAL SOLUTION OF THE EQUATION FOR THE MATRIX S^R

The main equation (17) for the matrix $\mathbf{S}^R$ can be reduced to simpler equations. This can be done employing explicit expressions for the matrix **S**. It follows from Refs. 3 and 13 that

$$\mathbf{S}(\Omega,\Omega_0)=\mathbf{Q}\{{}^3/_4\mathbf{S}^{(0)}(\mu,\mu_0)+[(1-\mu^2)(1-\mu_0^2)]^{1/2}\mathbf{S}^{(1)}(\mu,\mu_0)\mathbf{P}^{(1)}(\Omega,\bar{\Omega}_0)$$
$$+S^{(2)}(\mu,\mu_0)\mathbf{P}^{(2)}(\Omega,\bar{\Omega}_0)\}. \tag{29}$$

Here, the matrices $\mathbf{P}^{(1)}$ and $\mathbf{P}^{(2)}$ are given by the expressions in Eq. (6),

$$\mathbf{S}^{(0)}(\mu,\mu_0)=\mathbf{L}(\mu)\tilde{\mathbf{L}}(-\mu_0)\frac{\mu\mu_0}{\mu+\mu_0},$$
$$\mathbf{L}(\mu)=\mathbf{M}(\mu)\mathbf{H}^{(0)}(\mu), \tag{30}$$
$$\mathbf{M}(\mu)=\begin{bmatrix} \mu^2 & \sqrt{2}(1-\mu^2) & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \mu \end{bmatrix},$$

the matrix $\mathbf{H}^{(0)}$ is the solution of the integral equation

$$\mathbf{H}^{(0)}(\mu)=1+\tilde{\omega}_0\mu\mathbf{H}^{(0)}(\mu)\int_0^1\tilde{\mathbf{H}}^{(0)}(\mu')\mathbf{\Psi}^{(0)}(\mu')\frac{d\mu'}{\mu+u'}, \tag{31}$$

where

$$\mathbf{\Psi}^{(0)}(\mu)={}^3/_8\tilde{\mathbf{M}}(\mu)\mathbf{M}(\mu)\mathbf{Q}. \tag{31a}$$

This matrix equation reduces to a system of four equations for the components $H_{ij}^{(0)}$ $(i,j=1$ or $2)$ and one equation for the component $H_{44}^{(0)}$.

The matrix $\mathbf{S}^{(1)}$ in Eq. (29) has the form

$$\mathbf{S}^{(1)}=\begin{bmatrix} S^{(1)}\mathbf{E} & 0 \\ 0 & S_{44}^{(1)} \end{bmatrix},$$

where **E** is a unit 3×3 matrix. These components, like the function $S^{(2)}(\mu,\mu_0)$ in Eq. (29), are given by expressions of the type

$$S_\alpha(\mu,\mu_0)=\frac{\mu\mu_0}{\mu+\mu_0}H_\alpha(\mu)H_\alpha(\mu_0), \tag{32}$$

so that the components $S^{(1)}$, $S_{44}^{(1)}$, and $S^{(2)}$ correspond to the functions $H^{(1)}(\mu)$, $H_r^{(1)}(\mu)$, and $H^{(2)}(\mu)$, which are solutions of the equation analogous with Eq. (31) with the Ψ functions $\Psi^{(1)}(\mu)=\frac{3}{8}(1-\mu^2)(1+2\mu^2)$ for $H^{(1)}$:

$$\Psi_r(\mu)=\tfrac{3}{8}(1-\mu^2) \text{ for } H_r^{(1)} \text{ and } \Psi^{(2)}(\mu)=\tfrac{3}{16}(1+\mu^2)^2 \text{ for } H^{(2)}. \quad (32a)$$

The general equation for the matrix S^R can be written in a form similar to Eq. (29):

$$S^R(\Omega,\Omega_0)=Q\{\tfrac{3}{4}S_R^{(0)}(\mu,\mu_0)+[(1-\mu^2)(1-\mu_0^2)]^{1/2}S_R^{(1)}(\Omega,\Omega_0)+S_R^{(2)}(\Omega,\Omega_0)\}. \quad (33)$$

The matrices $S_R^{(n)}$ contain terms proportional to $\cos n\psi$ or $\sin n\psi$. Since integration with respect to φ' in Eq. (17) causes the products of terms with different values of n to vanish, Eq. (17) splits into three independent equations relating the components $S_R^{(n)}$ and S^n with the same values of $n=0$, 1, and 2. For example, in the case of the matrix $S_R^{(0)}$, we obtain the equation

$$S_R^{(0)}(\mu,\mu_0)=S^{(0)}(\mu,\mu_0)+\frac{3}{8}\tilde\omega_0\int_0^1\frac{d\mu'}{\mu'}S^{(0)}(\mu,\mu')R(\mu')QS_R^{(0)}(\mu',\mu_0). \quad (34)$$

Substituting in Eq. (34) the value of $S^{(0)}$ in the form of (30) and writing down

$$S_R^{(0)}(\mu,\mu_0)=\frac{\mu\mu_0}{\mu+\mu_0}L(\mu)A(\mu,\mu_0)\tilde L(-\mu_0), \quad (35)$$

we obtain the following integral equation for the matrix A:

$$A(\mu,\mu_0)=1+\frac{3}{8}\tilde\omega_0(\mu+\mu_0)\int_0^1\frac{\mu'd\mu'}{(\mu+\mu')(\mu_0+\mu')}\Phi_0(\mu')A(\mu',\mu_0), \quad (36)$$

where

$$\Phi_0(\mu)=\tilde L(-\mu)R(\mu)QL(\mu).$$

The matrix Φ_0 has nonzero components Φ_{11}, Φ_{22}, $\Phi_{12}=\Phi_{21}$, and Φ_{44}. Therefore, the matrix equation (36) splits into two systems of two equations each for the components A_{11}, A_{21} and A_{12}, A_{22} and one equation for the component A_{44}.

The matrices $S_R^{(m)}(\Omega,\Omega_0)$ ($m=1$ or 2) are described by the following equation derived from Eqs. (17) and (33):

$$S_R^{(m)}(\Omega,\Omega_0)=S^{(m)}(\mu,\mu_0)P^{(m)}(\Omega,\overline{\Omega}_0)$$
$$+\frac{\tilde\omega_0}{4\pi}\int_0^1\frac{d\mu'}{\mu'}\int_0^{2\pi}d\varphi'\phi^{(m)}(\mu')S^{(m)}(\mu,\mu')P^{(m)}(\Omega,\overline{\Omega'})R(\mu')QS_R^{(m)}(\Omega',\Omega_0), \quad (37)$$

where $\phi^{(1)}=1-\mu^2$, $\phi^{(2)}=1$. Direct substitution demonstrates that the equation of $S_R^{(1)}$ may be satisfied by assuming that

$$S_R^{(1)}(\Omega,\Omega_0)=\begin{bmatrix}B_1(\mu,\mu_0)E & 0\\ 0 & B_4(\mu,\mu_0)\end{bmatrix}P^{(1)}(\Omega,\overline{\Omega}_0)$$
$$-\begin{bmatrix}B_2(\mu,\mu_0)E & 0\\ 0 & B_3(\mu,\mu_0)\end{bmatrix}P_R^{(1)}(\Omega,\overline{\Omega}_0). \quad (38)$$

Here, E is a unit 3×3 matrix, $P^{(1)}(\Omega,\Omega_0)$ is described by Eq. (6), and

$$P_R^{(1)}(\mu,\varphi;-\mu_0,\varphi_0)=\frac{3}{4}\begin{bmatrix}0&0&0&2\mu\sin\psi\\0&0&0&0\\0&0&0&\cos\psi\\2\mu_0\sin\psi&0&\cos\psi&0\end{bmatrix},\quad \psi=\varphi_0-\varphi. \quad (39)$$

We shall now introduce the matrices

$$B(\mu,\mu_0)=\begin{bmatrix}B_1&B_2\\B_3&B_4\end{bmatrix},\quad b(\mu,\mu_0)=\begin{bmatrix}b_1&b_2\\b_3&b_4\end{bmatrix}. \quad (40)$$

Here, $B_i(\mu,\mu_0)$ are components of the matrix (38), whereas the components $b_i(\mu,\mu_0)$ are given by the relationship

$$B(\mu,\mu_0)=\frac{\mu\mu_0}{\mu+\mu_0}H^{(1)}(\mu)b(\mu,\mu_0)H^{(1)}(\mu_0), \quad (41)$$

where

$$H^{(1)}(\mu)=\begin{bmatrix}H^{(1)}(\mu)&0\\0&H_r^{(1)}(\mu)\end{bmatrix}.$$

Using Eqs. (32), (37), (38), and (41) we can show that the matrix $b(\mu,\mu_0)$ is given by

$$b(\mu,\mu_0)=1+\frac{3}{8}\tilde\omega_0(\mu+\mu_0)\int_0^1 d\mu'\frac{\mu'\Phi_1(\mu')b(\mu',\mu_0)}{(\mu+\mu')(\mu_0+\mu')}, \quad (42)$$

where

$$\Phi_1(\mu)=H^{(1)}(\mu)\chi^{(1)}(\mu)H^{(1)}(\mu),$$
$$\chi^{(1)}(\mu)=(1-\mu^2)\begin{bmatrix}R_3-2\mu^2R_1 & R_4\\ -R_4 & R_3\end{bmatrix}, \quad (43)$$

and $R_i(\mu)$ are the components of the reflection matrix R defined by Eqs. (8)–(10). The matrix equation (42) splits into two pairs of independent equations for the components b_1, b_3 and b_2, b_4.

Equation (37) for the component $S_R^{(2)}$ can be satisfied assuming that

$$S_R^{(2)}(\Omega,\Omega_0)=S_R^{(2)}(\mu,\mu_0)P^{(2)}(\Omega,\Omega_0). \quad (44)$$

If we now introduce the function $a(\mu,\mu_0)$ defined by

$$S_R^{(2)}(\mu,\mu_0)=\frac{\mu\mu_0}{\mu+\mu_0}H^{(2)}(\mu)a(\mu,\mu_0)H^{(2)}(\mu_0), \quad (45)$$

we find that the substitution of Eqs. (44), (45), and (32) in Eq. (37) gives the following equation for $a(\mu,\mu_0)$

$$a(\mu,\mu_0)=1+\frac{3}{16}\tilde\omega_0(\mu+\mu_0)\int_0^1 d\mu'\frac{\mu'\Phi_2(\mu')a(\mu',\mu_0)}{(\mu+\mu')(\mu_0+\mu')}, \quad (46)$$

where

$$\Phi_2(\mu)=\chi_2(\mu)(H^{(2)}(\mu))^2,$$
$$\chi_2(\mu)=R_1(\mu)\mu^4-2R_3(\mu)\mu^2+R_2(\mu). \quad (47)$$

It follows from Eq. (15) that the matrix S^R relates the scattered flux on an inner boundary of a crystal to the incident flux which has entered the crystal.

It follows from Eqs. (11) and (13) that the scattered and incident fluxes on an external boundary are related by

$$I(-0,\mu_2,\varphi)=\frac{\tilde\omega_0}{4\mu_2}S^*(\mu_2,\varphi;\mu_1\varphi_0)F(-0,-\mu_1,\varphi_0). \quad (48)$$

Here,

$$S^*(\mu_2,\varphi;\mu_1,\varphi_0)=\frac{1}{n^2}\frac{\mu_2\mu_1}{\mu\mu_0}T(\mu_2,\mu)S^R(\mu,\varphi;\mu_0,\varphi_0)T(\mu_0,\mu_1),$$

where μ_2 and μ, μ_1 and μ_0 are related by the Snell formula (12).

The total radiation flux emerging from a crystal (per unit surface) is

$$\pi\mathcal{F}=\int_0^1\mu_2 d\mu_2\int_0^{2\pi}d\varphi I(-0,\mu_2,\varphi). \quad (49)$$

The ratio of the emerging to the incident flux crossing the boundary

$$\eta_0=\mathcal{F}/\mu_0F(+0,\mu_0,\varphi_0) \quad (50)$$

represents the total quantum efficiency of the luminescence. It is clear from Eqs. (33), (35), (38), and (44) that in the case of a ray incident normally on the surface and scattered in the reverse direction, i.e., when $\mu_2 = \mu_0 = \mu = \mu_1 = 1$, the degree of polarization of the scattered light in the case when the exciting light is completely (linearly or circularly) polarized is given by the expressions

$$\mathcal{P}_{\text{lin}} = a(1,1)\frac{[H^{(2)}(1)]^2}{\Sigma}, \quad \mathcal{P}_{\text{circ}} = -A_{44}(1,1)\frac{[H_{44}^{(0)}(1)]^2}{\Sigma}, \tag{51}$$

where

$$\Sigma = A_{11}(1,1)[H_{11}^{(0)}(1)]^2 + [A_{12}(1,1) + A_{21}(1,1)]H_{11}^{(0)}(1)H_{22}^{(0)}(1) + A_{22}(1.1)[H_{22}^{(0)}(1)]^2.$$

Here, $H_{ij}^{(0)}$ are the corresponding components of the matrix $\mathrm{H}^{(0)}$ defined in accordance with Eq. (31).

In the case of low values of $\tilde{\omega}_0$, the integral equation for the matrix **S**, like the equation for the matrix $\mathbf{S}^R$, can be solved by iteration in powers of $\tilde{\omega}_0$. In the first approximation, i.e., when allowance is made only for the first- and second-order scattering, we find that if $\mu_2 = \mu_1 = 1$, it follows from Eq. (51) on the assumption that $n^2 = 1$:

$$\mathcal{P}_{\text{lin}}^{(1)} = 1 - 0.160\tilde{\omega}_0, \quad \mathcal{P}_{\text{circ}}^{(1)} = -(1 - 0.315\tilde{\omega}_0); \tag{52}$$

however, if $n^2 = 10$, then

$$\mathcal{P}_{\text{lin}}^{(1)} = 1 - 0.248\tilde{\omega}_0, \quad \mathcal{P}_{\text{circ}}^{(1)} = -(1 - 0.491\tilde{\omega}_0). \tag{53}$$

§6. CHARACTERISTICS OF BACKSCATTERING

As pointed out in Ref. 1, when excitons with a wave vector $\mathbf{q}_0$ are excited, the probability of backscattering of these excitons, i.e., of the scattering in the direction $\mathbf{q} \approx -\mathbf{q}_0$, is doubled for $\tau_t \gg \tau_p$ because of interference. This additional contribution is described by diagrams with intersections (in a certain order) of vertical impurity lines whose contribution is important on deviation of **q** from $-\mathbf{q}_0$ by an angle $\Delta\theta_1 < (q_0 l_{\text{diff}})^{-1}$. Consequently, the intensity of light scattered once backward should be doubled and this intensity is governed by the matrix $\mathbf{S}_0^R(\mu_0, \varphi_0 + \pi, \mu_0, \varphi_0)$, equal—in accordance with Eqs. (53) and (18)—to $\mathbf{P}(\mu_0 - \mu_0, \pi)/2$. At the exit of the crystal the doubling of the radiation scattered singly backward should occur on deviation from the direction of the incident ray by an angle $\varphi < \Delta\theta_2 = \lambda/2\pi l_{\text{diff}}$ in an azimuthal plane and by an angle $\theta < \Delta\theta_2\mu_0/\mu_1$ in a meridional plane. Here, λ is the wavelength of light outside the crystal. Within this angle the scattering matrix is $\mathbf{S}^R + \mathbf{S}_0^R$. Since the polarization of singly scattered light governed by the matrix $\mathbf{S}_0^R$ is stronger than the polarization of multiply scattered light, the degree of polarization of the backscattered radiation increases. Thus, if $\mu_2 = \mu_1 = 1$, then instead of Eq. (53) for the backscattered radiation we find that if $n^2 = 10$, then

$$\mathcal{P}_{\text{lin}}^{(1)} = 1 - 0.124\tilde{\omega}_0, \quad \mathcal{P}_{\text{circ}}^{(1)} = -(1 - 0.246\tilde{\omega}_0). \tag{54}$$

This effect is associated with multiple scattering of excitons in the case when light is scattered once. Multiple scattering of light by excitons should also result, because of interference, in doubling of the backscattered radiation (see the review in Ref. 14). This doubling occurs within an angle $\Delta\theta' \approx \lambda/2\pi l_{\text{rad}}$. Since it follows from the condition (1b) that $l_{\text{rad}} \gg l_{\text{diff}}$, we find that $\Delta\theta' \ll \Delta\theta_2$. As a result of interference associated with multiple scattering of light and with multiple scattering of excitons, the intensity of the radiation is doubled within the angle $\Delta\theta'$ but its polarization is not affected.

§7. RESULTS OF A NUMERICAL CALCULATION

A numerical calculation was carried out in which the integral equations (36), (42), and (46) for **A**, **b**, and a were solved on a computer by the iteration method.[15] The corresponding H functions were determined by the same method from Eq. (31) using the Ψ functions from Eqs. (31a) and (32a), since the tables in Refs. 3 and 13 were insufficient for this purpose. The iteration procedure was continued until the difference between the two consecutive values became less than 0.01% at every point. For $\tilde{\omega}_0$ close to 1, where the convergence of the method was slow, the zeroth approximations were the values of the H functions calculated in Ref. 3 for $\tilde{\omega}_0 = 1$. In all cases the calculated H functions agreed (within the limits of the calculation error) with those given in Ref. 13.

By way of a check, the total scattered flux $\pi\mathcal{F}$ [see Eq. (49)] was calculated for all values of the refractive index and $\tilde{\omega}_0 = 1$, and it was checked whether it was equal to the incident flux πF. The discrepancy between these fluxes did not exceed 0.5%. Figure 1 shows the dependence of the total quantum efficiency of the luminescence $\eta_0 = \mathcal{F}/F$ in the case of normal incidence of the exciting light on the quantum efficiency $\tilde{\omega}_0$ for a single scattering event. As pointed out earlier, if $\mu_0 = 1$, the value of η_0 is independent of the polarization of the exciting light. We can see that on increase in n the value of η_0 decreases steeply even for $\tilde{\omega}_0$ close to unity and this is due to an increase in the number of scattering events N because of the reflection of light. For example, if $\tilde{\omega}_0 = 0.9$, and increase in n^2 from 1 to 10 increases $\bar{N} = \ln\eta_0/\ln\tilde{\omega}_0$ from 8.5 to 28.

At high values of n the radiation leaving the crystal is incident on the surface in a narrow solid angle $\Delta\Omega = \pi/n^2$ and only a small proportion of the radiation escapes within this angle. Therefore, the density of the radiation incident on an internal surface of a crystal,

$$\pi\mathcal{F}_{\text{intern}} = \int_0^1 \mu\, d\mu \int_0^{2\pi} d\varphi\, I(+0, \mu, \varphi),$$

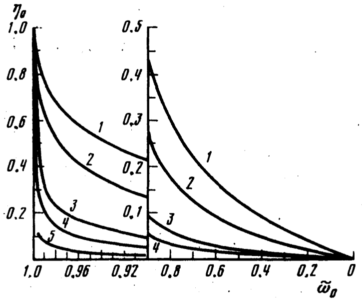

FIG. 1. Dependences of the total quantum efficiency of the luminescence η_0 on the quantum efficiency for a single scattering event $\tilde{\omega}_0$ when $\mu_0 = \mu_1 = 1$. Curves 1–5 correspond to the following values of n^2: 1) 1.0; 2) 2.0; 3) 6.25; 4) 10; 5) 25.

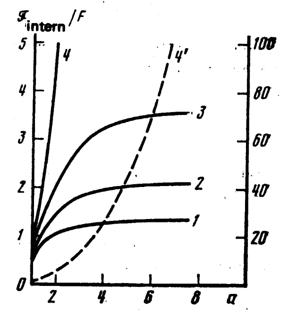

FIG. 2. Dependences of the flux incident on an internal surface of a crystal on n for $\mu_1=\mu_0=1$. Curves 1–4 correspond to the following values of $\tilde{\omega}_0$: 1) 0.95; 2) 0.975; 3) 0.99; 4) 1.0. The right-hand scale applies to the dashed curve 4′; the left-hand scale applies to all the other curves.

may be considerably greater than the flux $\pi\mu_0 F(+0)$ entering the crystal across this surface. Figure 2 shows the dependence of the ratio $\mathcal{F}_{intern}/F(+0)$ on the value of n for a normally incident ray and various values of $\tilde{\omega}_0$. We can see that when $\tilde{\omega}_0=1$, this ratio is a few tens when n is large, but it decreases rapidly when ω_0 is reduced slightly.

Figure 3 shows the angular distribution of the intensity of the scattered radiation in the case when $n^2=10$, calculated for various values of $\tilde{\omega}_0$ when the exciting light is incident normally. This angular distribution depends on the polarization of light and when the exciting light is linearly polarized it depends also on the direction of this light relative to the plane of polarization. Figure 3a corresponds to the propagation of light in the plane of polarization, whereas Fig. 3b corre-

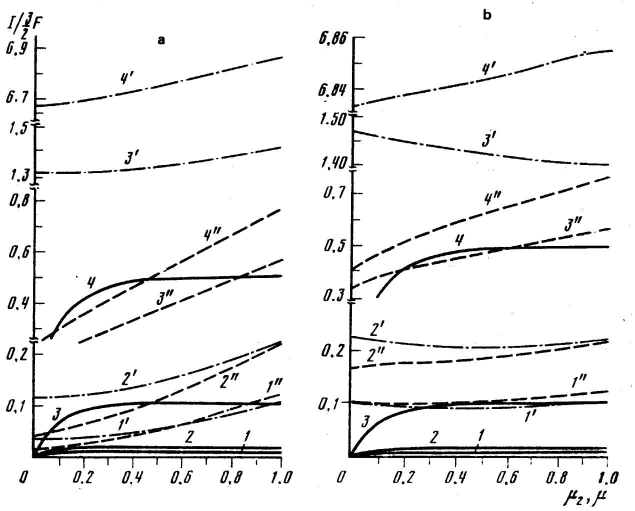

FIG. 3. Angular distributions of the radiation $I(\mu_2)$ emerging from a crystal excited with linearly polarized light when $\mu_1=\mu_0=1$ and the radiation travels in the plane of polarization (a) or when the radiation travels in a plane making an angle of $\pi/4$ with the plane of polarization (b); the latter set of curves applies also to unpolarized and circularly polarized light. The continuous curves are calculated for $n^2=10$ and the dashed curves for $n=1$. The dash-dot curves are the angular distributions of the intensity $I(\mu)$ of light incident on an internal surface of a crystal. Curves 1–4 correspond to the following values of $\tilde{\omega}_0$: 1) 0.6; 2) 0.8; 3) 0.99; 4) 1.0.

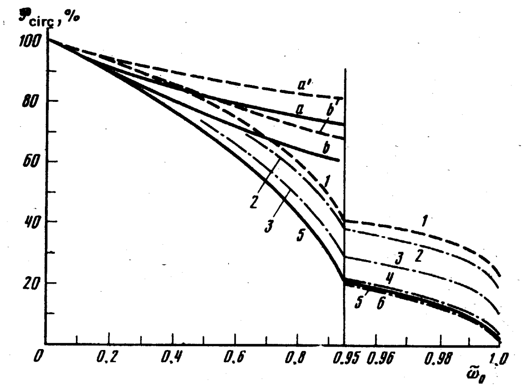

FIG. 4. Dependences of the degree of polarization of the scattered radiation on the value of $\tilde{\omega}_0$ plotted for $\mu_2=\mu=1$ and normal incidence of circularly polarized light. Curves 1–6 correspond to the following values of n^2: 1) 1.0; 2) 1.2; 3) 2.0; 4) 6.25; 5) 10; 6) 25. Curves a and a' correspond to allowance solely for single and double scattering when $n^2=1$ (a') and $n^2=10$ (a) [see Eqs. (52) and (53)]. Curves b and b' again correspond to single and double scattering but an allowance is made also for the contribution of the third-order scattering processes.

sponds to the propagation at an angle of 45° relative to this plane. It follows from Eq. (28) that the same scattering diagram as in Fig. 3b applies also to the excitation by unpolarized or circularly polarized light. For comparison, we are showing the angular distributions in the absence of reflection, i.e., when $n=1$, as well as the dependence on μ of the intensity of light incident on an internal surface of a crystal. Since, in the case of large values of n, a small μ_2 corresponds to μ close to unity, the plot of $I(\mu_2)$ is much flatter for $n^2=10$ than for $n=1$. It follows from the symmetry relationships (28) that the curves in Fig. 3b describe also the dependence of the intensity of light scattered normally to the surface ($\mu_2=1$) on the cosine of the angle of incidence of the radiation μ_0 [apart from a factor μ_0/μ_1 in Eq. (48)]. The curves in Figs. 3a and 3b can also be used to find the intensity of the scattered light in a plane perpendicular to the plane of polarization.

The influence of reemission and reflection from a surface on the polarization is demonstrated in Figs. 4 and 5. These figures give the dependences on $\tilde{\omega}_0$ and

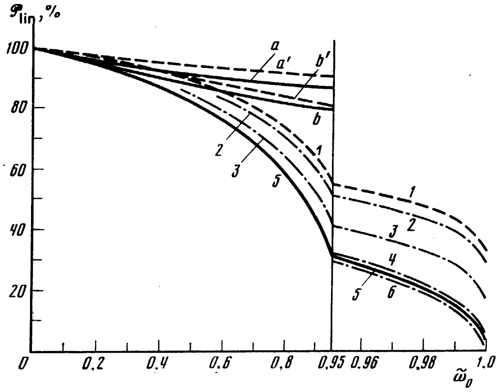

FIG. 5. Dependences of the degree of polarization of the scattered radiation on $\tilde{\omega}_0$ plotted for $\mu_2=\mu=1$ and normal incidence of linearly polarized light. The notation is the same as in Fig. 4.

n of the degree of polarization of normally scattered light in the case of normal incidence of circularly or linearly polarized exciting light. We can see that as $\tilde{\omega}_0$ approaches 1, the degree of polarization falls rapidly, and it decreases on increase in n. For example, when $\tilde{\omega}_0=1$ and n^2 is increased from 1 to 10, $\mathscr{P}_{\rm circ}$ decreases from 23.7 to 1.9%, whereas $\mathscr{P}_{\rm lin}$ decreases from 31.9 to 3.0% (the degrees of polarization for $n=1$ agree with those calculated in Ref. 3). It is clear from Figs. 4 and 5 that an increase in n causes the degree of polarization to fall first rapidly and then reach almost a saturation plateau when the average transmission coefficient $1-\bar{R}$ becomes less than the quantity $[1-\eta_0(\tilde{\omega}_0, n=1)]$ and, therefore, a further increase in $\bar{R}$ does not increase significantly the average number of the scattering events. The dashed curves in Figs. 4 and 5 are calculated by the method of iteration of powers of $\tilde{\omega}_0$ allowing only for single, double, and triple scattering. We can see that if allowance is made for the reflection, these curves are practically coincident with the exact solution for $\tilde{\omega}_0$ below 0.15 and 0.20.

Figure 6 gives the dependences of the degree of circular polarization of light on the cosine of the angle of emergence μ_2. The dash-dot curve shows the corresponding dependence on μ for light incident on an internal surface of a crystal. We can see that at low values of μ_2 the degree of polarization of light for $n^2=10$ is higher than for $n=1$. This is due to the fact that when n is large, small values of μ_2 correspond to μ close to unity and an increase in the degree of polarization because of approach of μ to unity compensates its reduction because of an increase in the number of scattering events $\bar{N}$ on increase in n. This enhances $\mathscr{P}_{\rm circ}$ particularly at low values of $\tilde{\omega}_0$, when—in accordance with Fig. 4—the value of $\mathscr{P}_{\rm circ}$ decreases very smoothly on increase in n. In accordance with the consition (28) the curves in Fig. 6 describe also the dependence of the degree of polarization of light scattered normally to the surface ($\mu=1$) on the angle of incidence μ_0.

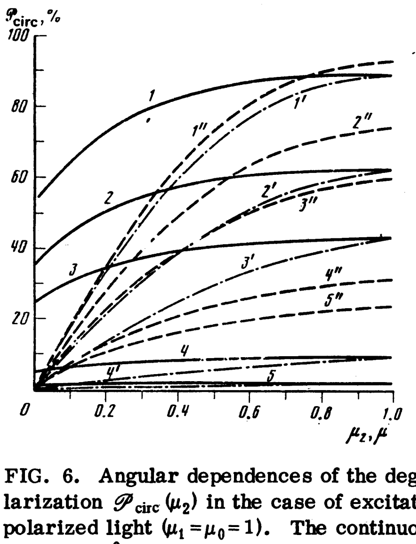

FIG. 6. Angular dependences of the degree of circular polarization $\mathscr{P}_{\rm circ}(\mu_2)$ in the case of excitation with circularly polarized light ($\mu_1=\mu_0=1$). The continuous curves are calculated for $n^2=10$ and the dashed curves for $n=1$. The chain curves show the dependence $\mathscr{P}_{\rm circ}(\mu)$ for the radiation incident on an internal surface of a crystal. Curves 1–5 correspond to the following values of $\tilde{\omega}_0$: 1) 0.2; 2) 0.6; 3) 0.8; 4) 0.99; 5) 1.0.

Figure 7 demonstrates the influence of the interference effects occurring in multiple scattering of excitons by impurities. We can see that the polarization of the back-scattered radiation increases considerably within the selected solid angle. This increase is greatest for $\tilde{\omega}_0=0.6$–0.95. At low values of $\tilde{\omega}_0$, when single scattering of light predominates, the intensity of the backscattered radiation doubles but the polarization does not change when allowance is made for the additional contribution. When $\tilde{\omega}_0$ is close to unity, the contribution of single scattering decreases and a change in the degree of polarization becomes less. Thus, for $\tilde{\omega}_0=1$ and $n^2=10$, the interference effects in backscattering increase $\mathscr{P}_{\rm lin}$ from 3.0 to 4.3% and $\mathscr{P}_{\rm circ}$ from 1.9 to 3.2%. Thus, such interference effects can be detected from a change in the intensity and from a change in the polarization. Precision measurements of the angular distributions make it possible to determine the diffusion length of excitons.

We shall consider briefly the frequency dependence of the intensity and polarization of the scattered light in the case of excitation by a line of considerable width $\Delta\omega \gg \Gamma$. In the Introduction we have given the value of the absorption coefficient $\alpha(\omega_r)=\omega_{LT}q_0/\Gamma$ at a resonance frequency corresponding to the point of intersection of the exciton and photon branches. Away from the resonance frequency ω_r, the absorption coefficient $\alpha(\omega)$ decreases in accordance with the law

$$\alpha(\omega)=\frac{\omega_{LT}q_0\Gamma}{(\omega-\omega_r)^2+\Gamma^2}+\alpha_b, \qquad (55)$$

where for the sake of generality we have included also the background absorption coefficient α_g which in independent of the frequency. Then, the radiative time (when $\tau_p \ll \tau_t$) is given by

$$\frac{1}{\tau_{\rm rad}(\omega)}=\frac{2}{3}\frac{\omega_{LT}q_0v_{g_0}\Gamma}{(\omega-\omega_r)^2+\Gamma^2}. \qquad (56)$$

Therefore, away from the resonance frequency ω_r

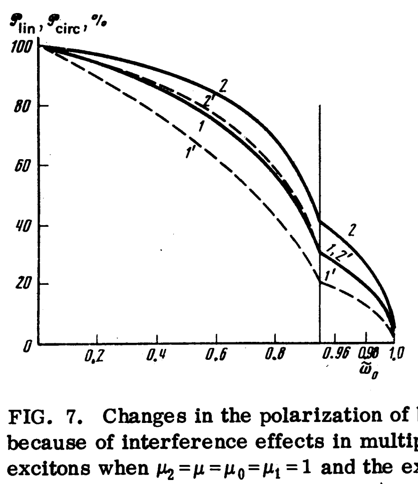

FIG. 7. Changes in the polarization of backscattered radiation because of interference effects in multiple elastic scattering of excitons when $\mu_2=\mu=\mu_0=\mu_1=1$ and the exciting light is circularly polarized (curves denoted by 1) or linearly polarized (curves denoted by 2); it is assumed that $n^2=10$. The continuous curves are calculated allowing for interference and the dashed curves correspond to Figs. 4 and 5.

there is a reduction also in the quantum efficiency for a single scattering of a photon, which—when allowance is made for α_g—is

$$\tilde{\omega}_0 = \frac{\tau_t}{\tau_{rad}} \frac{\alpha - \alpha_b}{\alpha}.$$

When $\tilde{\omega}_0$ is close to unity, the quantum efficiency decreases rapidly on reduction in $\tilde{\omega}_0$. Consequently, for $\tilde{\omega}_0(\omega_r) \approx 1$ the line width of the scattered radiation may be considerably less than Γ. It then follows from Figs. 4 and 5 that the degree of polarization of the radiation increases away from the line center, so that the average degree of polarization of a line may exceed greatly the degree of polarization at the center. In the case of strong inhomogeneous broadening $\Gamma' \gg \Gamma$ with a Lorentzian distribution of the frequency ω_r, Eqs. (55) and (56) should be modified by replacing Γ with Γ'.

We shall conclude by noting that the above theory applies also to the Rayleigh scattering of polarized light by defects in isotropic solid and liquid media, and—under certain conditions—it also describes the resonance scattering of acoustic phonons.[16,17]

The authors are grateful to N. A. Silant'ev for his valuable advice in discussions.

[1]E. L. Ivchenko, G. E. Pikus, B. S. Razbirin, and A. N. Starukhin, Zh. Eksp. Teor. Fiz. **72**, 2230 (1977) [Sov. Phys. JETP **45**, 1172 (1977)].
[2]E. M. Gamarts, E. L. Ivchenko, M. I. Karaman, V. P. Mushinskiĭ, G. E. Pikus, B. S. Razbirin, and A. N. Starukhin, Zh. Eksp. Teor. Fiz. **73**, 1113 (1977) [Sov. Phys. JETP **46**, 590 (1977)].
[3]S. Chandrasekhar, Radiative Transfer, Clarendon Press, Oxford, 1950 (Russ. Transl., IIL, M., 1953).
[4]V. V. Sobolev, Perenos luchistoĭ énergii v atmosferakh zvezd i planet (Transfer of Radiant Energy in the Atmospheres of Stars and Planets), Gostekhizdat, M., 1956.
[5]V. M. Agranovich and Yu. V. Konobeev, Opt. Spektrosk. **6**, 648 (1959); **11**, 369 (1961) [Opt. Spectrosc. (USSR) **6**, 421 (1959); **11**, 199 (1961)].
[6]V. I. Doronina, Yu. V. Konobeev, and Sh. D. Khan-Magometova, Opt. Spektrosk. **28**, 811 (1970) [Opt. Spectrosc. (USSR) **28**, 436 (1970)].
[7]V. M. Agranovich and M. D. Galanin, Perenos élektronnogo vozbuzhdeniya v kondensirovannykh sredakh (Transfer of Electron Excitation in Condensed Media), Nauka, M., 1978.
[8]S. D. Gutshabash, Opt. Spektrosk. **42**, 365 (1977) [Opt. Spectrosc. (USSR) **42**, 204 (1977)].
[9]E. P. Zege and I. L. Katsev, Zh. Prikl. Spektrosk. **31**, 327 (1979).
[10]E. L. Ivchenko and N. Kh. Yuldashev, Fiz. Tverd. Tela (Leningrad) **21**, 2182 (1979) [Sov. Phys. Solid State **21**, 1255 (1979)].
[11]M. Born and E. Wolf, Principles of Optics, 4th ed., Pergamon Press, Oxford, 1970 (Russ. Transl., Nauka, M., 1973).
[12]V. A. Ambartsumyan, Astron. Zh. **19**, 1 (1942); Dokl. Akad. Nauk SSSR **38**, 257 (1943); J. Phys. USSR **8**, 65 (1944).
[13]J. Lenoble, J. Quant. Spectrosc. Radiat. Transfer **10**, 533 (1970).
[14]Yu. N. Barabanenkov, Usp. Fiz. Nauk **117**, 49 (1975) [Sov. Phys. Usp. **18**, 673 (1975)].
[15]A. F. Verlan' and V. S. Sizikov, Metody resheniya integral'nykh uravneniĭ s programmami dlya ÉVM (spravochnoe posobie) [Methods for Solution of Integral Equations with Computer Programs (Handbook)], Naukova Dumka, Kiev, 1978.
[16]I. B. Levinson, Zh. Eksp. Teor. Fiz. **75**, 234 (1978) [Sov. Phys. JETP **48**, 117 (1978)].
[17]V. A. Malyshev and V. L. Shekhtman, Fiz. Tverd. Tela (Leningrad) **20**, 2915 (1978) [Sov. Phys. Solid State **20**, 1684 (1978)].

Translated by A. Tybulewicz

Phase relaxation investigation under conditions of appreciable spin polarization

S. A. Al'tshuler, I. N. Kurkin, and V. I. Shlenkin

Kazan' State University
(Submitted 30 May 1980)
Zh. Eksp. Teor. Fiz. **79**, 1591–1599 (October 1980)

Measurements were made of the phase-memory time T_m of Yb^{3+} ions in $CaWO_4$ single crystals activated simultaneously with Yb^{3+} and Tb^{3+} ions, which have substantially different *g*-factors. Under conditions when T_m is determined by the dipole-dipole interactions between the Yb^{3+} and Tb^{3+} ions, a strong decrease of the relaxation rate T_m^{-1} is observed with decreasing temperature. This is due to the considerable polarization of the Tb^{3+} ions, for which the condition $kT < g\beta H$ is satisfied.

PACS numbers: 61.80.Jh

1. INTRODUCTION

The overwhelming majority of EPR research has been performed under conditions when the high-temperature approximation is valid, i.e., $g\beta H \ll kT$ (the notation here is standard). Yet the courses of the spin-spin and spin-lattice relaxation should change substantially if appreciable spin polarization[1)] takes place, i.e., $g\beta H \gg kT$. Experiments when the low-temperature approximation is valid are of particular interest if the investigations are performed by the electron spin echo (ESE) method, which is highly effective in the study of relaxation processes. No such investigations were performed previously, apparently because of the need for obtaining very low temperatures: thus, at wavelengths ~3 cm we have $g\beta H \sim 0.5$ K so that the hard-

 0038-5646/80/100801-06$02.40