

Polariton mechanism of luminescence of crystals; reabsorption, spatial dispersion effects, and role of boundary

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At low temperatures, the spatial dispersion (in particular, the additional light waves) and the polariton effects alter substantially the physical picture of reabsorption of light in crystals. Besides the usual volume reabsorption, which is due to overlap of the light emission and absorption spectra, allowance for the additional light waves leads to the onset of two new reabsorption mechanisms. One is due to exciton collisions with the surface of the crystal, which are accompanied by formation of photons that propagate into the interior of the crystal. Reabsorption corresponds to their absorption in the crystal, so that the intensity of the corresponding process turns out to be proportional to the concentration of the excitons at the boundary. The other reabsorption mechanism comes into being when account is taken of exciton scattering by defects (impurities) in the crystal lattice or of exciton-exciton collisions. Since these processes can be accompanied by interband transitions, a new volume source of reabsorption appears (exciton + impurity → photon + impurity), and can take place even in crystals in which the ordinary reabsorption mechanism is negligible because of the weak overlap of the emission and absorption spectra. We discuss the limits of applicability of the reabsorption equations that can be used in the region of sufficiently low and high temperatures.

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1. INTRODUCTION

Experimental investigations of exciton luminescence of crystals at low temperatures have been going on now for several decades. Most recently, however, spectral and temporal singularities of crystal emission have been observed for many crystals and cannot be interpreted within the framework of the ideas, dating back to Frenkel's paper¹ (see also Ref. 2), on the emission of light by excitons. According to Ref. 1, the absorption of light corresponds to a real process, in which a photon incident on a crystal is transformed into an exciton, luminescence corresponds to the inverse process, etc.

This analysis is valid if both the excitons and the photons are well defined elementary excitations of the medium that interact weakly with one another. However, in crystals in which the excitons have sufficiently large oscillator strengths, the retarded interaction in the region of low values of the wave vector renormalizes the spectrum of the elementary excitations (i.e., of the transverse photons and excitons which appear only when the Coulomb interaction is taken into account). The spectrum of the new elementary excitations, which are superpositions of excitons and photons and usually called polaritons, differ substantially at small values of k from the exciton spectrum (see Fig. 1). The use of polaritons rather than excitons and photons makes it possible in fact to develop in most natural fashion an interpretation of the above-mentioned singularities of exciton luminescence of crystals at low temperatures. The exciton-photon interaction for polaritons has been approximately taken into account even in the harmonic approximation, so that the perturbations are produced only by scattering from phonons and lattice defects. This circumstance no longer makes it possible to speak of luminescence as a process in which an exciton is transformed into a photon, etc. The emission spectrum observed outside the crystal is now the result of elastic and inelastic processes that occur on the surface of the

crystal, and the polariton is completely or partially transformed in these processes into an "external" luminescence photon. At the same time, the polariton scattering inside the crystal (from phonons or from crystal defects) leads only to intraband or interband transitions of the polaritons.

An important factor when it comes to using the foregoing qualitative picture of the so-called polariton mechanism of luminescence^{3,4} (see also Ref. 5) is that in the region of large wavelengths the above-mentioned elementary excitations (polaritons) are in fact photons in the medium and their properties can be completely described within the framework of the macroscopic Maxwell's equations with account taken of the spatial dispersion.⁶ Therefore, for example, an "external" luminescence photon produced when a polariton collides with the surface of a crystal corresponds to a refracted wave. Its intensity, which generally speaking is a function of the frequency and of the time, is directly proportional to the number of polaritons with a given frequency and polarization (see, e.g., Refs. 7 and 8). It follows thus from the foregoing that the determination of the polariton distribution function on the boundary of the crystal is a basic problem that arises within the framework of the polariton mechanism of luminescence.

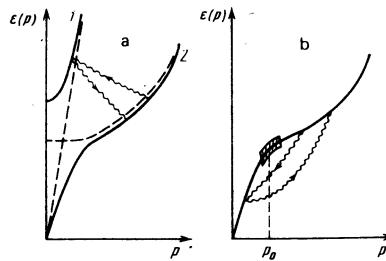


FIG. 1. a) Spectrum of transitions in a crystal. The wavy lines show the interband transitions. b) Lower branch of the polariton spectrum. The wavy lines show the intraband transitions. The "bottleneck" region is shaded.

For a number of problems of exciton physics (energy transfer, capture of excitons by impurities, exciton-exciton collisions, etc.), however, it is necessary to know the polariton distribution function not only on the surface of the crystal but also in its volume. This permits a more general formulation of the problem, and makes it particularly urgent to derive first of all the kinetic equations for the polaritons at different values of the exciton-phonon interaction constant. We shall deal with this question later on.

At this point we emphasize that allowance for polariton effects is particularly important at low temperatures, when the bulk of the polaritons is concentrated in the "bottleneck" region (see Fig. 1). If the temperature of the crystal is not too low, however, and the bottleneck is weakly populated, we can use the kinetic equations not for the polaritons but for the excitons and photons, provided only, as shown below, we introduce into these equations certain correction terms necessitated by the effects of spatial dispersion.

The present article is devoted in fact to a discussion of the evolution of the physical picture of reabsorption of light when the temperature of the crystal is changed. In particular, we shall obtain the aforementioned correction terms for the usual equations of reabsorption, and consider some physical effects corresponding to them.

To proceed to their analysis in the most natural fashion, we shall first consider the polariton effects (i.e., we shall disregard the influence of the retarded interaction on the emission spectrum). In this case one can obtain in quite obvious fashion for the exciton concentration an equation that permits a complete description of both the exciton luminescence and the energy transport by the excitons.

Since the exciton lifetime in a dielectric ($\tau \approx 10^{-5} - 10^{-8}$ sec) is usually much longer than its free path time t_0 with respect to scattering by phonons, at not too low temperatures we have $t_0 \approx 10^{-11} - 10^{-12}$ sec and the exciton manages to collide a tremendous number of times with phonons during its lifetime. Therefore the excitons can be regarded as in a state of thermodynamic equilibrium with the lattice, and the concentration $c(\mathbf{r}, t)$ can be determined from the diffusion equation

$$\frac{\partial c}{\partial t} = D \Delta c - c/\tau + I(\mathbf{r}, t), \quad (1)$$

where D is the exciton diffusion coefficient and $I(\mathbf{r}, t)$ is the source function (see Ref. 5).

In luminescent crystals, the possibility of emission of a luminescence quantum by an exciton from one point of the crystal and its absorption, accompanied by the production of an exciton, at another point can be taken into account by adding to the right-hand side of (1) the following integral term (we disregard the anisotropy of the crystal)

$$\left. \frac{\partial c}{\partial t} \right|_{\text{reabs}} = \frac{1}{4\pi} \int_0^\infty \rho(v) \beta(v) k(v) dv \int_v c(\mathbf{r}, t) \frac{\exp(-k(v)|\mathbf{r}-\mathbf{r}_1|)}{|\mathbf{r}-\mathbf{r}_1|} d\mathbf{r}_1, \quad (2)$$

where $\rho(v)$ is the probability, per unit time, of production of a photon of frequency v in radiative annihilation of the exciton, $k(v)$ is the coefficient of absorption of

light of frequency v in the crystal, and $\beta(v)$ is a quantity equal to the fraction of absorption acts of light of frequency v , as a result of which excitons are produced. The quantity $\beta(v)$ is directly connected with the quantum yield $\eta(v)$ of the exciton luminescence when light of frequency v is excited, namely

$$\eta(v) = \tau \beta(v) \int_0^\infty \rho(v_i) dv_i,$$

so that

$$\beta(v) \rho(v) = \frac{1}{\tau} E(v) \eta(v), \quad (3)$$

where $E(v)$ is the luminescence spectrum normalized to unity:

$$E(v) = \rho(v) / \int_0^\infty \rho(v_i) dv_i, \quad \int E(v) dv = 1.$$

The assumption that the exciton becomes thermalized before its decay that is accompanied by photon emission allows us to regard the quantity $\rho(v)$ in (2) as dependent only on the crystal temperature. The luminescence photon produced at the point \mathbf{r} can also land at the point \mathbf{r}_1 , and experience one or several reflections from the crystal surface. This means that the possibility of reflection of the luminescence light from the crystal boundaries is not taken into account in the kernel of the integral relation (2). It is clear that these reflections increase the role of the reabsorption. Single reflections, the only ones of interest in the case of weak crystals, are taken into account in Ref. 9. Multiple scatterings, which are particularly important for films whose thickness exceeds slightly the photon mean free path, were considered in part in Ref. 10, and we shall return to this question later. The use of relations (1) and (2), supplemented by the usual diffusion boundary conditions, enables us to study not only the distribution of the exciton in luminescent crystals as a function of the character of the source and of the conditions on the crystal boundaries, but also to investigate the time evolution of the spectral composition of the exciton luminescence as a function of its thickness and shape (see Refs. 5 and 11).

We emphasize once more that in the derivation of relations (1) and (2), or of those modifications of these equations which were mentioned above, it is assumed that the excitons and photons are different well defined elementary excitations of the crystal and that the processes corresponding to their transformation into one another (i.e., the processes of absorption of light and luminescence) can be taken into account within the framework of perturbation theory in terms of the operator of the exciton-photon interaction. At sufficiently low temperatures, however, as already mentioned, an appreciable fraction of excitons manages to "slip-off" into those parts of their spectrum (Fig. 1) where the assumption mentioned above is no longer valid. Under these conditions, relations (1) and (2) are strictly speaking not well founded, and the entire problem as a whole calls for a more exact analysis.

2. KINETIC EQUATION FOR POLARITONS AND GENERALIZATION OF THE EQUATIONS OF THE PHENOMENOLOGICAL THEORY OF REABSORPTION

Allowance for the interaction between the Coulomb excitons and the electromagnetic field of the transverse photons in the harmonic approximation can be carried out with the aid of an appropriate $u-V$ transformation (see, e.g., Ref. 11). The Hamiltonian of the new elementary excitations (polaritons) corresponds in the vicinity of an isolated exciton resonance, as shown in Fig. 1a, to two energy branches, $\varepsilon_1(\mathbf{k})$ and $\varepsilon_2(\mathbf{k})$. If $\xi^{(1)}(\xi^{+(1)})$ ($i=1, 2$) are the operators of annihilation (creation) of a polariton with wave vector \mathbf{k} of branch i , then

$$\hat{H}_0 = \sum_i \varepsilon_i(\mathbf{k}) \xi_i^{+(1)} \xi_i^{(1)}. \quad (4)$$

Allowance for the exciton scattering by the lattice phonons leads to the polariton-phonon interaction. In the approximation linear in the displacements of the nuclei, the operator corresponding to this interaction can be written in the form

$$\begin{aligned} \hat{H}_{\text{int}} = & \sum_{\mathbf{p}, \mathbf{q}} [g_{11}(\mathbf{p}, \mathbf{q}) \xi_p^{+(1)} \varphi_q \xi_{p-q}^{(1)} + g_{22}(\mathbf{p}, \mathbf{q}) \xi_p^{+(2)} \varphi_q \xi_{p-q}^{(2)} \\ & + g_{12}(\mathbf{p}, \mathbf{q}) (\xi_p^{+(1)} \varphi_q \xi_{p-q}^{(2)} + \text{H.c.})], \end{aligned} \quad (5)$$

where $\varphi_q = b_q + b_q^*$, b_q (b_q^*) is the operator of annihilation (creation) of a phonon with wave vector \mathbf{q} , the quantities g_{ij} ($i, j=1, 2$)—the bare vertices—correspond to the transition of the polariton from branch i to branch j , accompanied by emission or absorption of a phonon. We have left out from the expression for H_{int} terms of the type $(\xi^* \varphi \xi + \text{H.c.})$; this, as will be shown below, is justified within the framework of the approximation used in the derivation of the Boltzmann kinetic equation.

We assume below that the exciton-phonon interaction is weak enough and that the polariton level width corresponding to this interaction is small at $k \ll \pi/a$ (a is the lattice constant) compared with the longitudinal-transverse splitting Δ of the exciton.¹⁾ If $\tau(p)$ is the free-path time of such a polariton, then the foregoing means that

$$\Delta \gg 1/\tau(p). \quad (6)$$

Only in this case is the renormalization of the exciton spectrum as a result of the retarded interaction of importance. Consequently the use of the polariton mechanism of luminescence is justified only if the inequality (6) is satisfied, and it may become necessary to introduce those generalizations of the transport equations (1) and (2) which were referred to in the introduction.

The equations for the polariton distribution function can be obtained with the aid of the equations for the nonequilibrium polariton Green's function. These equations can in turn be obtained by using the diagram technique developed, for example, in Ref. 12 or 13.

Assuming the exciton-photon interaction to be weak enough, we shall disregard also the effects of exciton autolocalization (see Ref. 14 for the corresponding criteria). In this case the equation for the Green's function can be solved by iteration in the polariton-phonon coupling constant g_{ij} [see (5)]. In second order in

g_{ij} , the equation for the Green's function can be reduced to the Boltzmann equation for the distribution function of the polaritons, in perfect analogy with the procedure used in Ref. 12 for the electron-phonon system. Allowance for the succeeding iteration (see Ref. 15), and also for the terms not accounted for in (5), in the form $(\xi^* \varphi \xi + \text{H.c.})$, does not change the structure of the equation for the polariton distribution function, and merely renormalizes the transition probabilities that enter in this equation. We shall not calculate here these probabilities, and merely show later how they can be connected with the phenomenological quantities $D, \tau, k(\nu), \rho(\nu)$, etc., which enter in Eqs. (1) and (2) and which characterize the excitons when no account is taken of the polariton effects. The Boltzmann equation mentioned above for the distribution functions $f_p^{(i)}(\mathbf{r}, t)$, where i is the number of the branch ($i=2$), takes the following form²⁾

$$\begin{aligned} \frac{\partial f_p^{(i)}(\mathbf{r}, t)}{\partial t} + v_i(\mathbf{p}) \frac{\partial f_p^{(i)}(\mathbf{r}, t)}{\partial \mathbf{r}} = & I_p^{(i)}(\mathbf{r}, t) \\ & + \sum_j \int \frac{d^3 p'}{(2\pi)^3} [W_{pp'}^{ij} f_p^{(j)}(\mathbf{r}, t) - W_{p'p}^{ji} f_{p'}^{(i)}(\mathbf{r}, t)], \end{aligned} \quad (7)$$

where $W_{pp'}^{ij}$ is the probability of polariton scattering from the state (j, \mathbf{p}') into the state (i, \mathbf{p}) , $v_i(\mathbf{p})$ is the group velocity of the polariton (i, \mathbf{p}) , and $I_p^{(i)}(\mathbf{r}, t)$ is the source of the polaritons.

We shall show now how to obtain from the system (7) the transport equations (1) and (2) as well as those generalizations of these equations which are due to the presence of the additional waves, to allowance for the reflection of the luminescence photons from the crystal boundaries, and to scattering of the polaritons by the crystal defects or impurities.

As already emphasized, in Eqs. (1) and (2) we did not take into account the renormalization of the exciton and photon spectrum due to the retarded interaction. This approximation is justified for sufficiently high temperatures, when the average value of the wave vector of the thermalized exciton $k_T = \sqrt{3MT}$ (M is the effective mass of the exciton and T is the temperature in energy units) is much larger than the quantity $k_0 = E(0)/c$ (E is the exciton energy and c is the speed of light). It is precisely in the wave-vector region $k \approx k_0$ that a substantial renormalization of the exciton and photon spectrum takes place (see Fig. 1). At $k_T \gg k_0$ most excitons do not "feel" this renormalization and the excitons can be regarded as in a state close to thermodynamic equilibrium with the lattice at each point of the crystal.

In the approximation indicated above, the branch of the spectrum $i=1$ can be regarded as corresponding to photons, while the branch $i=2$ as corresponding to the excitons; we can then disregard the renormalization of their spectra in the region $k \approx k_0$ (dashed lines). Taking the foregoing into account, it must be recognized, when writing down the equations for the distribution functions the photons $f_p^{(1)}(\mathbf{r}, t) = \varphi_p(\mathbf{r}, t)$ and for the excitons $f_p^{(2)}(\mathbf{r}, t) = f_p(\mathbf{r}, t)$, but for photons the main scattering process in the energy region $\sim E(k_T)$ is a process in which the photon goes over, with participation of the phonons, into an exciton (interband transition, see Fig. 1a). On the other

hand, Raman scattering of photons by phonons (intraband transitions) correspond to the next higher approximation in the exciton-phonon interaction, and can be omitted here. At the same time, the situation for excitons is reversed. The most frequent for them is precisely the intraband process of scattering by phonons. Nonetheless, the exciton-into-photon transformation processes must also be taken into account, since they correspond precisely to luminescence. If the foregoing is taken into account, Eqs. (7) can be rewritten in the form³⁾:

$$\frac{\partial f_p(r, t)}{\partial t} + v(p) \frac{\partial f_p(r, t)}{\partial r} = J(f) + I_p(r, t) \\ + \int \frac{d^3 p'}{(2\pi)^3} [W_{pp'}^{21} \varphi_{p'}(r, t) - W_{p'p}^{12} f_{p'}(r, t)], \quad (8a)$$

$$\frac{\partial \varphi_p(r, t)}{\partial t} + u(p) \frac{\partial \varphi_p(r, t)}{\partial r} = \int \frac{d^3 p'}{(2\pi)^3} [W_{pp'}^{12} f_{p'}(r, t) - W_{p'p}^{21} \varphi_{p'}(r, t)], \quad (8b)$$

where $v(p)$ and $u(p)$ are the group velocities of the excitons and photons, and $J(f)$ is the collision integral that corresponds to the intraband processes of exciton scattering by phonons. Inasmuch as at $p \gg k_0$ the photon velocity u is much higher than the exciton velocity, we can leave out from (8) the time derivative $\partial \varphi / \partial t$, and we have

$$f_p(r, t) = n_p^B c(r, t) + \psi_p(r, t), \quad (9)$$

where $c(r, t)$ is the density of the excitons at the point r and t , n_p^B is the exciton energy Boltzmann distribution normalized to unity, and $\psi_p(r, t)$ is a small "current" increment. Changing over in (8) in the usual manner, for the excitons only, to the diffusion approximation, we find that the functions $c(r, t)$ and $\varphi_p(r, t)$ satisfy the equations

$$\frac{\partial c(r, t)}{\partial t} - D \Delta c(r, t) + \frac{c(r, t)}{\tau} = \int \frac{d^3 p}{(2\pi)^3} \Phi_p \varphi_p(r, t) + I(r, t), \quad (10)$$

$$u(p) \frac{\partial \varphi_p(r, t)}{\partial r} + \Phi_p \varphi_p(r, t) = F_p c(r, t), \quad (11)$$

where

$$\Phi_p = \int \frac{d^3 p'}{(2\pi)^3} W_{p'p}^{21}, \quad I(r, t) = \int \frac{d^3 p}{(2\pi)^3} I_p(r, t), \quad (12)$$

$$F_p = \int \frac{d^3 p'}{(2\pi)^3} W_{pp'}^{12} n_{p'}^B, \quad \frac{1}{\tau} = \int \frac{d^3 p}{(2\pi)^3} F_p.$$

We shall neglect hereafter the anisotropy and assume that the crystal is a plane-parallel plate of thickness d ($0 \leq x \leq d$) and that the source function of the excitons I_p depends only on x . In this case the distribution functions are also functions of x only.

We represent the distribution function $\varphi_p(x, t)$ in the form of a sum

$$\varphi_p(x, t) = \varphi_p^+(x, t) + \varphi_p^-(x, t), \quad (13)$$

where $\varphi_p^+(x, t)$ and $\varphi_p^-(x, t)$ are respectively the distribution functions of the photons moving to the planes $x=d$ and $x=0$. If the vector u of the photon velocity makes an angle θ with the axis x ($0 \leq \theta \leq \pi/2$), then Eq. (11), with allowance for the foregoing, can be written in the form

$$u(p) \cos \theta \frac{\partial \varphi_p^+(x, t)}{\partial x} + \Phi_p \varphi_p^+(x, t) = \frac{1}{2} F_p c(x, t), \\ u(p) \cos \theta \frac{\partial \varphi_p^-(x, t)}{\partial x} - \Phi_p \varphi_p^-(x, t) = -\frac{1}{2} F_p c(x, t).$$

Consequently

$$\varphi_p^+(x, t, \theta) = \frac{\exp\{-k_p x / \cos \theta\}}{2u(p) \cos \theta} \int_0^x F_p c(\xi, t) \exp\left\{-\frac{k_p \xi}{\cos \theta}\right\} d\xi \\ + A_1(p) \exp\left\{-\frac{k_p x}{\cos \theta}\right\}, \quad (14a)$$

$$\varphi_p^-(x, t, \theta) = \frac{\exp\{k_p x / \cos \theta\}}{2u(p) \cos \theta} \int_x^\infty F_p c(\xi, t) \exp\left\{-\frac{k_p \xi}{\cos \theta}\right\} d\xi \\ + A_2(p) \exp\left\{\frac{k_p x}{\cos \theta}\right\}, \quad (14b)$$

where $k_p = \Phi_p/u(p)$ is the extinction coefficient for the photons with momentum p .

The quantities A_1 and A_2 must be obtained with the aid of the boundary conditions. In the general case these conditions and the values of A_1 and A_2 are cumbersome in form and we therefore confine ourselves here to only⁴⁾ a semi-infinite crystal ($d = \infty$). Inasmuch as the function φ_p cannot increase $x \rightarrow \infty$, the value of A_2 is zero in this case. On the other hand the value of A_1 can be obtained from the boundary condition that expresses the flux-balance condition:

$$u(p) \cos \theta \varphi_p^+(0, t) p^2 dp \sin \theta d\theta \\ = r_p(\theta) u(p) \cos \theta \varphi_p^-(0, t) p^2 dp \sin \theta d\theta \\ + \alpha_{p1}(\psi) c(0, t) n_{p1}^B v(p_1) \cos \psi p_1^2 dp_1 \sin \psi d\psi, \quad (15a)$$

where $r_p(\theta)$ is the coefficient of specular reflection of a photon with momentum p when incident on the boundary at an angle θ ; the exciton momentum p_1 and the angle ψ are determined from the condition

$$p \sin \theta = p_1 \sin \psi, \quad \epsilon_1(p) = \epsilon_2(p_1), \quad (15b)$$

and the quantity $\alpha_{p1}(\psi)$ determines the probability of the interband transition induced by the boundary and transforming, upon reflection, an exciton with energy $\epsilon_2(p_1)$ incident on the crystal boundary at an angle ψ into a photon having the same energy. Taking (15b) into account, we get the following condition on the boundary:

$$\varphi_p^+(0, t) = r_p(\theta) \varphi_p^-(0, t) + \alpha_{p1}(\psi) c(0, t) n_{p1}^B.$$

The possibility of this process is due to the allowance for the spatial dispersion in the dielectric tensor, so that $\alpha_{p1}(\psi)$ depends on the form of the additional boundary conditions (ABC) (see Ref. 6).

The values of the function $\varphi_p(x, t)$ in (10) are determined [see (13) and (14)] by the relation

$$\varphi_p(x, t) = \varphi_p^+(x, t, \theta) + \varphi_p^-(x, t, \theta) \\ = \frac{F_p}{u(p) \cos \theta} \int_0^\infty c(\xi, t) \left[\exp\left\{-\frac{k_p(x-\xi)}{\cos \theta}\right\} + r_p(\theta) \exp\left\{-\frac{k_p(x+\xi)}{\cos \theta}\right\} \right] d\xi \\ + \alpha_{p1}(\psi) n_{p1}^B c(0, t) \exp\left\{-\frac{k_p x}{\cos \theta}\right\}. \quad (16)$$

Therefore, if we take into account also (12), the first term in the right-hand side of (10) can be represented as a sum of two contributions, $J_1 + J_2$:

$$J_1 = \int_0^\infty \rho(v) k(v) dv \int_0^\infty d\xi c(\xi, t) [E_1(k(v)|x-\xi|) + R_v(k(v)(x+\xi))], \quad (17a)$$

$$J_2 = c(0, t) \int \frac{p^2 dp}{(2\pi)^2} n_p^B \Phi_p K_p(x) = c(0, t) \mathcal{L}(x), \quad (17b)$$

where

$$E_t(x) = \int_1^x e^{-\mu\eta} \frac{d\eta}{\eta}, \quad R_v(x) = \int_1^x r_p(\eta) e^{-\mu\eta} \frac{d\eta}{\eta},$$

$$K_p(x) = \int_0^{p^2} \exp\left\{-\frac{k_p x}{\cos\theta}\right\} \sin\theta \alpha_p(\theta) d\theta, \quad \rho(v) = \frac{p^2}{(2\pi)^2} \frac{dp}{dv} F_p. \quad (18)$$

The term J_1 is due to reabsorption of the exciton luminescence: the radiation [the polaritons of branch 1 (see Fig. 1) produced in the radiative decay of the exciton at a depth ξ] is absorbed at a depth x with formation of excitons (polaritons of branch 2). Naturally, J_1 is determined by the overlap of the luminescence and absorption spectra [see (17a)].

The presence of the function R_v in the expression for J_1 is due to the reflection of the luminescence from the crystal boundary, so that $R_v=0$ at $r_p=0$. Thus, when reabsorption is taken into account, the term J_1 is the usual term in the kinetic equation (10) (see Refs. 5 and 11).

What is unusual in this equation is the term J_2 . It manifests itself only within the framework of a theory that takes consistent account of the spatial dispersion and the additional waves. The foregoing becomes clearer if it is recognized that to find J_2 it was necessary to take into consideration the boundary condition (15). In this boundary condition, the last term in the right-hand side determines the flux of the photons that go into the interior of the crystal and are produced as a result of the inelastic exciton collisions with the boundary. This is precisely why J_2 is proportional to the electron concentration on the boundary and, in contrast to J_1 , does not vanish even in those crystals where the absorption and luminescence spectra do not overlap. Thus, reabsorption is realized even at $J_1=0$. However, for the existence of the reabsorption connected with the quantity J_2 , the boundary plays an important role: the excitons produce photons only when colliding with the boundary, while the photons are absorbed in the volume. Processes of this kind will be called induced surface reabsorption (ISR).

To estimate the contribution of ISR to transport processes and to formation of the luminescence spectrum, we must know the coefficient $\alpha_p(\psi)$ of the conversion of an exciton into a photon when the exciton is reflected from the surface of the crystal. To estimate α_p we consider the problem of reflection of a polariton of branch 2 with momentum p , $p > p_0$ (the polariton hardly differs from the exciton in this momentum region), incident from the interior of the crystal on its surface, assuming the incidence to be normal. On the surface of the crystal (see Fig. 2) the incident polariton energy flux S breaks up into three fluxes⁵⁾ S_1 , S_2 , and S_3 , where S_1 and S_2 are the reflected fluxes corresponding respectively to the polaritons of the first and second branches, while S_3 is the light energy flux emitted from the crystal. The sought coefficient of conversion of excitons into photons, i.e., the quantity α_p , is equal to the ratio S_1/S . In a cubic crystal, with allowance for spatial dispersion, the dielectric constant for the vicinity of the dipole-allowed exciton resonance can be expressed in the form

$$\epsilon(\omega, k) = \epsilon_\infty + \omega_p^2 / (\omega^2 - \omega^2 + \beta n^2),$$

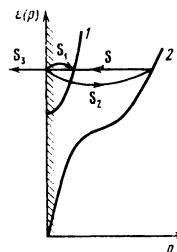


FIG. 2. Polariton fluxes on the boundary: S —flux of polaritons of branch 2 incident on the boundary. S_1 and S_2 —reflected fluxes of polaritons of branches 1 and 2 respectively, S_3 —flux of outgoing photons.

where ω is the frequency, k is the wave vector, $n^2 = k^2 c^2 / \omega^2$, $\beta = \hbar \omega_e \omega^2 / m^* c^2$, ω_e is the frequency of the exciton at $k=0$, $\omega_p = \omega_e (\epsilon_0 - \epsilon_\infty)$, ϵ_0 and ϵ_∞ are respectively the static and high-frequency dielectric constants. In this case (see Ref. 6) the expression for the polariton energy flux of branch i takes the form:

$$S_i = \frac{c}{8\pi} n_i E_i^2 \left(1 + \frac{\beta \omega_p^2}{\phi_i^2} \right),$$

where n_i is the proposed real refractive index, $\phi_i = \omega_e^2 - \omega^2 + \beta n_i^2$, E_i is the amplitude of the electric field in the polariton of branch i . Using this expression for the energy flux and the relation⁶ $\beta \omega_p^2 = -\phi_1 \phi_2$, we can rewrite the coefficient α_p in the form

$$\alpha_p = -\frac{\phi_2}{\phi_1} \frac{n_1 E_1^2}{n_2 E^2}, \quad (19)$$

where E is the amplitude of the electric field intensity of the polaritons of branch 2, which are incident from the inside on the surface of the crystal, whereas E_1 is the amplitude of the electric field intensity of the polaritons of branch 1, which propagate from the boundary into the boundary of the crystal and which are in the considered frequency region in practice photons with refractive index $n_1 \approx \sqrt{\epsilon_\infty}$.

In the presence of additional light waves (polaritons of branch 2), the ordinary boundary conditions that follow from Maxwell's equations are insufficient for the solution of the boundary-value problems, and it is necessary to use also the ABC. We use here the simplest¹⁷ ABC in the form $P_e|_{x=0}=0$, where P_e is the exciton part of the polarization of the medium. The use of ABC and of the ordinary boundary conditions enables use (see, e.g., Ref. 17) to determine expressions for all the electric field intensities. The expression for the intensity E_1 of interest to us and contained in (19) is of the form

$$E_1 = \frac{2n_2(n_2^2 - \epsilon_\infty)}{(n_2 - n_1)(n_1 n_2 + n_1 + n_2 + \epsilon_\infty)} E. \quad (20)$$

From (19) and (20) it follows that the coefficient α_p depends strongly on the parameters of the exciton transition and can vary in a rather wide range. We shall calculate α_p numerically for the crystals CdS and CuCl, which are frequently encountered in experiments. The results of the calculations are shown in Fig. 3. It follows from this figure that α_p depends strongly on the exciton momentum. Excitons in the momentum region $p > p_0$ can be regarded as being in thermodynamic equilibrium with the lattice. To find the mean value $\bar{\alpha}(T)$ it

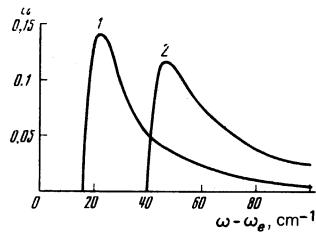


FIG. 3. Plot of $\alpha(\omega)$: 1—for CdS, 2—for CuCl.

is therefore necessary to average α_p over the Boltzmann distribution $n^B(p)$

$$\bar{\alpha}(T) = \int_0^\infty \alpha_p n^B(p) dp.$$

Thus, the effectiveness of conversion of the excitons into photons that move from the boundary into the interior of the crystal turns out to depend on the temperature. However, even from the form of the α_p dependence (see Fig. 3) it follows that the $\bar{\alpha}(T)$ function is not monotonic with increasing T : at small T the value of $\bar{\alpha}(T)$ increases, reaches a maximum in the temperature region $T \approx \omega_{\parallel} - \omega_e$, $\epsilon(\omega_{\parallel}) = 0$, and then decreases rapidly. It follows therefore from the foregoing that the ISR mechanism proposed here can be considered only for the temperature region where $\bar{\alpha}(T)$ is a maximum.

In crystals of the CdS type, for a wide range of temperatures, the term J_1 in the right-hand side of the transport equation for the excitons (10) can be neglected and only the term J_2 need be taken into account. The equation (10) is no longer integral, and this makes its analysis much simpler. We shall return to this question. At present, however, we proceed to an analysis of the influence of the lattice defects (or impurities) on the reabsorption with allowance for the additional waves.

The presence of lattice defects can lead to trapping of the excitons and to luminescence that is sensitized by them. We shall not consider this process here, however. We take into account below only the possibility of elastic and inelastic scattering of polaritons by crystal defects, which leads to their intraband and interband transitions. It was already emphasized in Ref. 4 that the already mentioned interband transitions can lead to a number of spectral singularities of the exciton luminescence. No less interesting, however, is apparently the new reabsorption process, which is discussed below and is controlled by the impurities or lattice defects.

The appearance of additional scattering (i.e., scattering by lattice defects) makes now the polariton scattering probability W_{pp}^{ij} , in Eqs. (7) of a sum of two terms, corresponding to scattering by the phonons and by the impurities (defects). The arguments in Sec. 2 remain valid in this case, too. Now, however, all the kinetic coefficients in (10) and (11) change because of the change of the scattering probability W_{pp}^{ij} . The diffusion coefficient is changed by those polariton-impurity scattering processes which leave the polariton on the same branch of the spectrum (in our case, on the second branch). On the other hand, the processes discussed in Ref. 4, which lead to transfer to another ("optical") branch of the spectrum, alter also the remaining kinetic

coefficients. These coefficients can now be represented in the form

$$\Phi_p = \Phi_p^{\text{ph}} + \Phi_p^{\text{imp}}, \quad F_p = F_p^{\text{ph}} + F_p^{\text{imp}}, \quad \frac{1}{\tau} = \frac{1}{\tau_{\text{ph}}} + \frac{1}{\tau_{\text{imp}}},$$

where the indices ph and imp denote respectively the terms due to scattering by phonons and impurities. In the presence of impurities we obtain also reabsorption terms J_1 and J_2 , in which, however, the meaning of the coefficients $k(\nu)$ and $\rho(\nu)$ is different. Thus for J_1 (volume reabsorption) we obtain from (17a) the expression

$$J_1 = \int_0^\infty d\nu [\rho_{\text{ph}}(\nu)k_{\text{ph}}(\nu) + \rho_{\text{imp}}(\nu)k_{\text{imp}}(\nu) + \rho_{\text{ph}}(\nu)k_{\text{imp}}(\nu) + \rho_{\text{imp}}(\nu)k_{\text{ph}}(\nu)] \\ \times \int_0^\infty d\xi c(\xi, t) [E_1(k(\nu)|x-\xi|) + R_v(k(\nu)(x+\xi))].$$

Thus, in the case discussed here the volume-reabsorption term can be represented as a sum of four terms, three of which are due only to allowance for the polariton scattering by the impurities. As seen from the expression given for J_1 , the exchange-reabsorption term is present even for exciton states with arbitrarily small overlap of the light emission and absorption spectra. In this limiting case we need retain in the expression for J_1 only the terms $\rho_{\text{imp}}(\nu)$ or $k_{\text{imp}}(\nu)$, which are proportional to the impurity or defect concentration.

Thus, allowance for the additional waves leads to new (surface or volume) reabsorption sources. The relative role of these reabsorption sources can vary greatly for different crystals and they must be taken into account first of all in cases when the ordinary reabsorption mechanism does not work. In the next section we discuss the role of ISR in greater detail.

3. LUMINESCENCE DAMPING TIME WITH ALLOWANCE FOR ISR

As already indicated (see Sec. 2), in crystals in which the radiation and emission spectra overlap weakly, we can neglect the volume reabsorption and take into account at sufficiently high temperature only the surface reabsorption. The exciton diffusion coefficient then takes the form

$$\frac{\partial c(x, t)}{\partial t} - D \frac{\partial^2 c(x, t)}{\partial x^2} + \frac{c(x, t)}{\tau} = I_0 \kappa e^{-\kappa x} + \mathcal{L}(x) c(0, t), \quad (21)$$

where I_0 is the intensity of the external source of the excitons, and the expression for $\mathcal{L}(x)$ is given by (17b) and (18).

For simplicity we confine ourselves as before to semi-infinite crystals ($x \geq 0$). We consider the first the solution of Eq. (21) in the stationary case, when the exciton density does not depend on the time. In this case the general solution of the stationary equation, which we denote here by $u(x)$, can be written in the form

$$u(x) = u(0) e^{-\kappa x/l} + \frac{I_0 \kappa \tau}{(1 - \kappa^2 l^2)} (e^{-\kappa x} - e^{-\kappa l}) \\ + \tau u(0) \int_0^\infty \frac{p^2 dp}{(2\pi)^2} \Phi_p n_p B \int_0^{l/\sqrt{\tau}} d\theta \sin \theta \alpha_p(\theta) \frac{\exp\{-k_\nu x / \cos \theta\} - \exp\{-x/l\}}{1 - l^2 k_\nu^2 / \cos^2 \theta} \quad (22)$$

where $l = \sqrt{D\tau}$ is the diffusion displacement length of the exciton. In the diffusion approximation, the boundary

condition on the crystal surface $x=0$ is of the form

$$D \frac{du}{dx} \Big|_{x=0} = \mu u(0), \quad (23)$$

where the surface-annihilation coefficient $\mu \geq 0$ is determined by the possibility of capture of an exciton by traps on the surface and the possibility of its conversion into a luminescence optical quantum. Using the boundary condition (23), we find that the density of the excitons on the crystal surface $u(0)$ is of the form

$$u(0) = \frac{I_0 x \tau}{1 + x l} \left(1 + \frac{\mu l}{D} - \frac{L(1/l)l}{D} \right)^{-1}, \quad (24)$$

where

$$L(k) = \int_0^\infty \mathcal{L}(x) e^{-kx} dx$$

is the Laplace transform of the function $\mathcal{L}(x)$. It is seen from (24) that for the existence of a stationary solution. It is necessary to satisfy the inequality

$$1 + \mu \frac{l}{D} - L(1/l) \frac{l}{D} > 0. \quad (25)$$

Using (17b) and (18) and taking the Laplace transform of the function $\mathcal{L}(x)$, we find that

$$L(1/l) \frac{l}{D} \approx \int_0^\infty \frac{\alpha_r(\theta) \sin \theta d\theta}{1 + k_p l / \cos \theta} = \langle \alpha_r \rangle$$

and consequently the inequality (25) is always satisfied. We consider now the question of the damping time τ_ω of the crystal luminescence. We assume here that during the time of irradiation a stationary exciton distribution has been established in the crystal and is described by formula (22), i.e., $c(x, t)|_{t=0} = u(x)$. According to Refs. 5 and 11, the luminescence damping time is defined as follows:

$$\tau_\omega = \int_0^\infty dt \int c(x, t) e^{-\kappa(\omega)x} dx / \int_0^\infty u(x) e^{-\kappa(\omega)x} dx = \frac{C(k=g(\omega), \omega=0)}{U(k=g(\omega))}, \quad (26)$$

where $g(\omega)$ is the extinction coefficient at the observation frequency, while $C(k, \omega)$ and $U(k)$ are the Laplace transforms of the corresponding functions. Consequently, to determine τ_ω there is no need to solve Eq. (21) with the corresponding initial and boundary conditions, and it suffices to know the Laplace transform $C(k, \omega)$ of the exciton density. Taking the Laplace transform of (21) and putting $\omega=0$, we get

$$C(k, \omega=0) = (L(k) - Dk - \mu) G(k) \frac{1}{2\pi i} \int_r^\infty U(k_i) G(k_i) dk_i \\ \times \left\{ 1 - \frac{1}{2\pi i} \int_r^\infty G(k_i) [L(K_i) - Dk_i - \mu] dk_i \right\}^{-1} + U(k) G(k),$$

where $G(k) = \tau / (1 - D\tau k^2)$, and the integration contour passes to the right of the imaginary axis. The integration is easy, since the functions $U(k)$ and $L(k)$ have no singularities in the right-hand half-plane and consequently only the poles of the function $G(k)$ in the right half-plane contribute to the integrals. We obtain ultimately

$$\tau_\omega = \frac{\tau}{1 - g^2(\omega)l^2} \left\{ 1 - g(\omega)l \frac{U(1/l)}{U(g)} \frac{[1 + \mu/Dg - L(g)/Dg]}{[1 + \mu l/D - L(1/l)l/D]} \right\}. \quad (27)$$

Putting for simplicity $\mu=0$ and assuming that $\langle \alpha_r \rangle \approx 0.1$ (see Fig. 3), we get $\tau_\omega/\tau \approx 1 + \langle \alpha_r \rangle \approx 1.1$.

Allowance for the second boundary increases the role of surface reabsorption. For sufficiently thin films, the contributions from the two boundaries add up, i.e., $\tau_\omega/\tau \approx 1 + \langle \alpha_r \rangle + \langle \beta_r \rangle$ (β is the coefficient of conversion of an exciton into a photon on the second boundary), and at $\alpha=\beta$ we have $\tau_\omega/\tau \approx 1.2$.

Thus, for crystals with weakly overlapping emission and absorption spectra, the contribution of the ISR to the increase of the luminescence damping time turns out to be of the same order as the contribution from the volume reabsorption.^{5,11}

4. CASE OF LOW TEMPERATURES

At sufficiently low temperatures, lower than the longitudinal-transverse splitting Δ of the electron, the greater part of the excitons are in the region of the longitudinal-transverse splitting⁶⁾ and a significant role is assumed by the processes of scattering by photons inside the lower polariton branch, accompanied by the transition of the polariton into the region of low wave vectors. In order not to clutter up the calculations in this section, we shall not consider at all the upper (first) polariton branch, although in principle, of course, it is not difficult to take this branch into account. The distribution function $f_p(r, t)$ of the polaritons in the second branch satisfies an equation that follows from (7) and is of the form

$$\frac{\partial f_p(r, t)}{\partial t} + v(p) \frac{\partial f_p(r, t)}{\partial r} = \int \frac{d^3 p}{(2\pi)^3} [W_{pp'} f_{p'}(r, t) - W_{p'p} f_p(r, t)]. \quad (28)$$

We divide the lower polariton branch into two momentum regions: 1) $p < p_0$, 2) $p > p_0$, where p_0 is the wave vector of the light at the frequency of the exciton resonance (see Fig. 1b). This, as yet formal, division leads us to a system of kinetic equations for the polariton distribution functions in the region 1) $f_p^{(1)}(r, t)$ and in the region 2) $f_p^{(2)}(r, t)$, which coincides completely with the system of equations (7), provided we assume in them that the index i numbers the subdivision regions and not the polariton branches. Just as in the upper polariton branch, the basic scattering process for the "photon-like" polariton in region 1) is scattering by a phonon with a transition into region 2); this enables us to neglect the proper collision integral in this region. In the second region, in turn, as already noted in Sec. 2, the most intense is the scattering by phonons inside a region, and this scattering establishes thermodynamic equilibrium between these particles and the lattice. This enables us to use for these particles the diffusion approximation [we note here that when speaking of the lower-branch polariton everywhere in Sec. 2 we had in mind precisely the "excitonlike" polaritons in region 2)]. This collisionless-hydrodynamic model of the transport phenomena for the lower polariton branch leads, obviously, to the exciton diffusion equation (10) with an integral term in the right-hand side in the form (17a).

The indicated subdivision into only two regions—collisionless and hydrodynamic—is possible (see Refs. 7 and 8) only for crystals with small oscillator strength,

and consequently with a narrow transition region. As already noted, for example, in Ref. 8, the contribution of the polaritons situated inside the bottleneck can in this case be neglected.

For crystals with large oscillator strength (for example, anthracene), the transition region with momentum $p \sim p_0$ is broad, it contains many particles, and can no longer be neglected. In this region the proper collision integral is of the order of the integrals of collisions with transitions into other regions, making it difficult to analyze theoretically the transport phenomena and calling for a numerical solution.

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APPENDIX

We consider a flat crystal of finite thickness d . In this case, besides the boundary condition (15), at $x=0$, it is necessary to have a condition on the second surface $x=d$:

$$\begin{aligned} \varphi_p^-(d, t) u_p \cos \theta \sin \theta d\theta p^2 dp = & R_p(\theta) \varphi_p^+(d, t) u_p \cos \theta \sin \theta d\theta p^2 dp \\ + \beta_p(\psi) c(d) n_{p_1} B v_p \cos \psi \sin \psi d\psi p_1^2 dp_1, \quad (A.1) \\ p \sin \theta = & p_1 \sin \psi, \quad \varepsilon_1(p) = \varepsilon_2(p), \end{aligned}$$

where the quantities R and β are assumed to be different from the corresponding quantities r and α on the surface $x=0$. Substitution of (14a) and (14b) in (15a) and (A.1) leads to a system of algebraic equations for the coefficients A_1 and A_2 , the solutions of which are

$$A_1 = \frac{n_{p_1} B}{\Delta(p, \theta)} \left[\alpha_p(\theta) c(0, t) \exp \left\{ -\frac{k_p d}{\cos \theta} \right\} + r_p(\theta) \beta_p(\theta) c(d, t) \right], \quad (A.2)$$

$$A_2 = \frac{n_{p_1} B}{\Delta(p, \theta)} \left[R_p(\theta) \alpha_p c(0, t) \exp \left\{ -\frac{k_p d}{\cos \theta} \right\} + \beta_p(\theta) c(d, t) \right], \quad (A.3)$$

where

$$\Delta(p, \theta) = \exp \{k_p d / \cos \theta\} - r_p(\theta) R_p(\theta) \exp \{-k_p d / \cos \theta\}.$$

The integral terms ($\sim F_p$) are cumbersome and will not be written out here. Neglect of these terms corresponds to neglect of the volume reabsorption. Leaving out the volume reabsorption terms, we have the following exciton-density diffusion equation

$$\frac{\partial c(x, t)}{\partial t} - D \frac{\partial^2 c(x, t)}{\partial x^2} + \frac{c(x, t)}{\tau} = I_0 \kappa e^{-\kappa x} + c(0, t) \mathcal{L}_1(x) + c(a, t) \mathcal{L}_2(x),$$

where

$$\begin{aligned} \mathcal{L}_1(x) = & \int \frac{d^3 p}{(2\pi)^3} \frac{n_{p_1} B}{\Delta(p, \theta)} \alpha_p(\theta) \left[\exp \left\{ \frac{k_p(d-x)}{\cos \theta} \right\} \right. \\ & \left. + R_p \exp \left\{ -\frac{k_p(d-x)}{\cos \theta} \right\} \right], \end{aligned}$$

$$\mathcal{L}_2(x) = \int \frac{d^3 p}{(2\pi)^3} \frac{n_{p_1} B}{\Delta(p, \theta)} \beta_p(\theta) [\exp \{k_p x / \cos \theta\} + r_p \exp \{-k_p x / \cos \theta\}].$$

¹⁾We use a system of units with $\hbar = 1$.

²⁾If the exciton-autolocalization effects do take place, but the wave vector p at $p \ll \pi/a$ remains for them a "good" quantum number and the inequality (6) also retains its form, then it

seems that the results of the analysis presented below remain in force. All that change are the meaning and method of calculation of the quantities v , W , and I in (7). These quantities can now also be obtained by perturbation theory in which, however, the states of the zeroth approximation should be taken to be the states of the polaritons (excitons) with autolocalization effects taken into account. The problem of autolocalization of electrons in semiconductors is discussed in Ref. 16.

³⁾In the considered approximation, allowance for the polariton effects is nevertheless important when the boundary conditions are considered. It is precisely allowance for these effects which leads to new reabsorption mechanisms (see below).

⁴⁾The results for arbitrary d are given in the Appendix.

⁵⁾This is a manifestation of the polariton nature of the discussed elementary excitations. If we neglect the exciton-photon mixing effect, then the fluxes $\mathbf{S}_1 = \mathbf{S}_2 = 0$ and there is no conversion of excitons into photons on the boundary.

⁶⁾In the same region of the spectrum are located the surface polaritons. See Ref. 18 for their possible role.

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