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ATTENUATION OF THE ELECTROMAGNETIC FIELD IN THE TRANSPARENCY REGION OF INSULATING CRYSTALS

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Abstract. Proceeding from general Maxwell equations, a straightforward method for the calculation of the light absorption coefficient in the region of an isolated exciton absorption band has been worked out. The treatment is based on a simple expansion of the real and the imaginary part of the transverse dielectric constant, specified as the *weak absorption approximation*. The method has been applied to the calculation of the long-wavelength edge of the exciton absorption band $\Gamma(3/2)$ of solid xenon.

Key words: excitons, polaritons, light absorption.

1. INTRODUCTION

Since the pioneering studies by Anselm and Firsov [1], and Toyozawa [2, 3], a quite thorough theory of lineshapes of optical absorption bands in the region of exciton resonances has been worked out. Apart from the papers dealing with the overall lineshape, a vast number of publications concern absorption near band edges, especially the origin of the exponential behaviour of absorption in the low-energy side, known as the Urbach tail (see, e.g., [4] and [5] for review). Probably the most fruitful starting point for the explanation of this phenomenon has been proposed by Schreiber and Toyozawa [6]. In a remarkable series of numerical experiments, performed for a large number of finite Monte-Carlo samples, they ascribed the Urbach tail to the discrete localized states below the renormalized exciton band edge. According to their model, momentarily trapped excitons can be formed below the free exciton states due to fluctuations in the random potential of lattice vibrations. The existence of localized states as well as the validity of the Urbach rule has been

demonstrated simply by numerically solving the standard model of linear on-site exciton–phonon interaction, neglecting intersite correlation, and including the adiabatic approximation for the harmonic lattice vibrations.

The present report also touches upon the low-energy side of the exciton absorption bands. However, it does not pretend to offer any new interpretation of the Urbach tail. This would probably be excessively speculative in the framework of the approach specified below. Exponential tailing is usually observed in the near-edge region of the absorption bands [5]. Here, on the contrary, the presumable transparency region of insulating crystals is examined. This study has partly been stimulated by some unexpected experimental results concerning absorption in the off-resonance region of condensed xenon, which could be a very promising medium for gamma-scintillators [7]. Unfortunately, registration of scintillations happens to be restricted due to abnormally strong self-absorption (0.5–1 cm^{-1}) of the emission from quasi-molecular scintillation centres Xe_2^* [8, 9]. Consequently, quite remarkable absorption has been detected in the region more than 1 eV below the bottom of the exciton zone, which is in conflict with the predictions of the common polariton theory where the dielectric constant tensor is expressed in terms of renormalized damping polaritons [10, 11].

Recently the author has proposed a different approach [12] where the dielectric constant depends on the spectrum of renormalized damping excitons, not directly on polaritons. Nonstationarity of polariton states in this model, i.e. their frequency shift and damping, are also determined by the corresponding parameters of excitons. The absorption coefficient, when calculated according to [12], tails off much slower as compared to the polariton theory (illustrations are given in Section 4). In addition to intraband scattering, [12] takes account of the interband exciton–phonon processes. This may be important with regard to the interpretation of the mentioned anomalous absorption in the far off-resonance region in condensed xenon where the energetically lowest dipole "allowed" exciton zone $\Gamma(3/2)$ overlaps the "forbidden" zone $\Gamma'(3/2)$ [13]. This essential peculiarity of rare gas crystals does not find any reflection in the polariton picture. Indeed, from the point of view of the polariton theory the absorption in the off-resonance region is practically independent of the presence of "forbidden" exciton zones, whereas according to [12] the interband scattering to the "forbidden" zone $\Gamma'(3/2)$ can contribute to the damping of $\Gamma(3/2)$ excitons in Xe. In the region of small wave vectors this contribution may gradually become predominant. As a result, the absorption coefficient in the far off-resonance region would tail off slower than expected. The above-given qualitative considerations are supported by the corresponding model calculations in Section 4.

Therefore, the starting point for the theoretical discussion in this paper is the general expression for the dielectric constant tensor. The crystals under study are considered to be isotropic and nongyrotropic. Provided that free charges are absent, the optical properties of such crystals in the

region of an isolated exciton zone $\Omega(\mathbf{k})$, characterized by the oscillator strength F, can be described in terms of the transverse dielectric constant scalar

$$\varepsilon_{\perp}(\mathbf{k}, \omega) = \varepsilon_0 + \frac{\Omega_p^2 F}{[\Omega(\mathbf{k}) + d_{\mathbf{k}}]^2 - (\omega + i g_{\mathbf{k}})^2}, \qquad (1)$$

where Ω_p^2 is the quadrate of plasma frequency, and the physical background of the parameters $d_{\mathbf{k}}$ and $g_{\mathbf{k}}$ depends on the model used for their specification. According to [12], they should be interpreted as the frequency shift and damping of excitons with the wave vector $\mathbf{k}$, whereas in the polariton theory $g_{\mathbf{k}}$ would correspond to the damping of polaritons [10, 11].

The goal of this paper is to elaborate an easily applicable method for the calculation of the absorption coefficient in the region energetically far below the exciton resonance, independent of the specification of the damping parameter $g_{\mathbf{k}}$. As will be demonstrated, a special *weak absorption approximation* suits well for this purpose.

2. ENERGY FLUX DENSITY

In a simplified version of the polariton theory, presuming weakness of the damping parameter $g_{\mathbf{k}}$, the recommended way for the calculation of the absorption coefficient can be expressed by the formula [14, 15]

$$\mu(\omega_{\mathbf{k}}) = \frac{g_{\mathbf{k}}(\omega_{\mathbf{k}})}{v_{\mathbf{k}}}, \qquad (2)$$

where $v_{\mathbf{k}}$ is the group velocity of free polaritons. In other words, the absorption coefficient would simply be inversely proportional to the mean free path length of polaritons.

The viewpoint expressed by Eq. (2) is not very rigorous, because it is based on several simplifying suppositions. First, it is assumed that a monochromatic electromagnetic wave can create in the crystal *free* polaritons of the same frequency, i.e. the solutions of the dispersion relation

$$\frac{c^2 k^2}{\omega^2} = \varepsilon_{\perp}(\mathbf{k}, \omega), \qquad (1')$$

neglecting damping. Second, the *complex* frequency shift $\delta\omega_{\mathbf{k}}$ of polaritons is supposed to be determined by the *complex* increment of the wave vector:

$$\delta\omega_{\mathbf{k}} = v_{\mathbf{k}} \delta\mathbf{k}, \quad v_{\mathbf{k}} \equiv \frac{d\omega_{\mathbf{k}}}{d\mathbf{k}}.$$

Third (probably the most arbitrary presumption), damping affects only the *imaginary* part of the polariton frequency and wave vector, whereas their real component remains unchanged:

$$\delta \mathbf{k} = i \cdot \mathrm{Im}\, \mathbf{k},$$

$$\delta \omega_{\mathbf{k}} = i g_{\mathbf{k}} (\omega_{\mathbf{k}}).$$

Making use of those assumptions, one readily comes to Eq. (2) indeed. However, apart from the conceptual differences concerning the physical meaning of the damping parameter, none of the mentioned approximations enter into the model specified below.

According to the general (inhomogeneous) Maxwell equations, the electromagnetic field in the crystal can be expressed through the so-called foreign currents. For an optically isotropic crystal the general expression is as follows [16]:

$$\left[k^2 - \frac{\omega^2}{c^2} \varepsilon_{\perp} (\mathbf{k}, \omega) \right] \mathbf{A} (\mathbf{k}, \omega) = \frac{4\pi}{c} \mathbf{j} (\mathbf{k}, \omega), \tag{3}$$

where $\mathbf{k}$ is the wave vector ($k = |\mathbf{k}|$), c is the speed of light, $\mathbf{A}(\mathbf{k}, \omega)$ and $\mathbf{j}(\mathbf{k}, \omega)$ are the Fourier transforms of the vector potential and the current vector, respectively. Therefore, the vector potential

$$\mathbf{A} (\mathbf{r}, t) = 4\pi c \int d\omega \int d^3 k \frac{\mathbf{j} (\mathbf{k}, \omega)\, e^{i(\mathbf{kr} - \omega t)}}{k^2 c^2 - \omega^2 \varepsilon_{\perp} (\mathbf{k}, \omega)}. \tag{4}$$

To retain the rigorousness of the further treatment, we make use of the approximation of a semi-infinite crystal. Let us assume that a monochromatic electromagnetic wave of the frequency ω falls onto the surface of the crystal perpendicular to the plane $z = 0$. The system of coordinates should be chosen such that the direction of the falling wave would coincide with that of the x-axis. Under these conditions the components of the foreign currents induced on the surface become [17]

$$j_x (z, t) = j_0 e^{-i\omega t} \delta (z), \quad j_y = j_z = 0, \tag{5}$$

whose only component of the corresponding Fourier transform different from zero is

$$j_x (\omega', k) = \frac{j_0}{2\pi} \delta (\omega' - \omega)\, \delta (k_x)\, \delta (k_y), \tag{6}$$

where $k \equiv k_z$.

Substituting Eq. (6) into (4), one obtains

$$A_x(z,t) = \frac{2e^{-i\omega t} j_0}{\omega} \int_{-\infty}^{\infty} dN \frac{e^{i\frac{\omega}{c}Nz}}{N^2 - \varepsilon_\perp(N,\omega)}, \quad N \equiv \frac{ck}{\omega} . \tag{7}$$

The methods commonly used for the evaluation of integrals similar to Eq. (7) are based on the theory of complex variables, i.e. they require the calculation of the poles of the integrand. In the present case, however, to make such an approach really possible, one should specify the parameters $d_{\mathbf{k}}$ and $g_{\mathbf{k}}$ in Eq. (1) for complex wave vectors, which need not be an easy task at all. This gives a reason for introducing the weak absorption approximation mentioned above and performing integration in Eq. (7) directly along the real axis.

As is known, the electric and magnetic field strengths can be expressed via the vector potential:

$$\mathbf{E} = -\frac{1}{c}\frac{\partial \mathbf{A}}{\partial t}, \quad \mathbf{H} = \mathrm{rot}\,\mathbf{A} . \tag{8}$$

Therefore, in accordance with Eq. (7), one can write

$$E_x(z,t) = \frac{4ie^{-i\omega t} j_0}{c} \int_0^{\infty} dN \frac{\cos\left(\frac{\omega}{c}Nz\right)}{N^2 - \varepsilon_\perp(N,\omega)} , \tag{9}$$

$$H_y(z,t) = -\frac{4e^{-i\omega t} j_0}{c} \int_0^{\infty} dN \frac{\sin\left(\frac{\omega}{c}Nz\right)}{N^2 - \varepsilon_\perp(N,\omega)} \tag{10}$$

(only different from zero components of vectors are written).

Equations (9) and (10) lead to the following expression for the energy flux density:

$$S_z \equiv \frac{c}{16\pi}(E_x H_y^* + E_x^* H_y) = \frac{2|j_0|^2}{c} \times$$

$$\times \left\{ \left[\int_0^{\infty} dN \cdot \cos\left(\frac{\omega}{c}Nz\right) F_2 \right] \left[\int_0^{\infty} dN \cdot N \sin\left(\frac{\omega}{c}Nz\right) F_1 \right] - \right. \tag{11}$$

$$\left. - \left[\int_0^{\infty} dN \cdot \cos\left(\frac{\omega}{c}Nz\right) F_1 \right] \left[\int_0^{\infty} dN \cdot N \sin\left(\frac{\omega}{c}Nz\right) F_2 \right] \right\},$$

where

$$F_1 \equiv \frac{N^2 - \varepsilon_1}{(N^2 - \varepsilon_1)^2 + \varepsilon_2^2}, \quad F_2 \equiv \frac{\varepsilon_2}{(N^2 - \varepsilon_1)^2 + \varepsilon_2^2}, \tag{12}$$

$$\varepsilon_1 \equiv \mathrm{Re}\ \varepsilon_\perp (N, \omega), \quad \varepsilon_2 \equiv \mathrm{Im}\ \varepsilon_\perp (N, \omega).$$

3. WEAK ABSORPTION APPROXIMATION

As we are examining the off-resonance region, the quantity $g_{\mathbf{k}}$, which is responsible for the imaginary part of the dielectric constant, has to be a small parameter, independent of its more specific physical background. It then follows that the poles of the integrands on the right side of Eq. (11) must be very close to some resonant value, which can be determined from the equation

$$N_r^2 \equiv \frac{c^2 k_r^2}{\omega^2} = \varepsilon_1 (N_r, \omega), \tag{13}$$

where ε_1 is defined by Eq. (12). Note that k_r cannot be identified with the wave vector of the free polaritons corresponding to the frequency ω, although it does not differ very much from the latter.

Direct integration along the real axis can be performed easily, if one takes into account that integrals in Eq. (11) are determined by a shallow range of wave vectors in the immediate vicinity of k_r. Consequently, it is reasonable to approximate

$$\begin{aligned} \varepsilon_1 &= \varepsilon_{1r} + \varepsilon'_{1r}(k - k_r), \quad \varepsilon_{1r} \equiv \varepsilon_1 (k_r, \omega), \\ \varepsilon'_{1r} &\equiv \varepsilon'_1 (k_r, \omega), \quad \varepsilon_2 = \varepsilon_{2r} \equiv \varepsilon_2 (k_r, \omega). \end{aligned} \tag{14}$$

Equation (14) can be treated as the mathematical formulation of the so-called *weak absorption approximation* used in this study. Within this approximation

$$F_1 = \frac{F_m (k_0 - k_r)\ (k - k_r)}{(k - k_r)^2 + (k_0 - k_r)^2} \tag{15}$$

and

$$F_2 = \frac{F_m (k_0 - k_r)^2}{(k - k_r)^2 + (k_0 - k_r)^2}, \tag{16}$$

where

$$F_m \equiv F_2(k_r) = \frac{1}{\varepsilon_{2r}},$$

$$k_0 \equiv k_r \left(1 + \frac{\varepsilon_{2r}}{2\varepsilon_{1r} - \varepsilon'_{1r} k_r} \right),$$

i.e.

$$F_1(k_0) = F_2(k_0) = \frac{1}{2} F_m .$$

Using Eqs. (14)–(16), one can rewrite Eq. (11), simultaneously replacing the bounds of integration with $N_r - \Delta$ and $N_r + \Delta$ (the quantity Δ will be specified below):

$$S_z = \frac{2|j_0|^2 F_m^2 x_0^2}{c} (I_1 I_2 - I_3 I_4) . \tag{17}$$

Here

$$I_1 = x_0 \int_{-\Delta}^{\Delta} dx \, \frac{\cos\left(\frac{\omega}{c} xz + k_r z \right)}{x^2 + x_0^2}, \quad x \equiv \frac{ck_0}{\omega} - N_r , \tag{18}$$

$$I_2 = N_r \int_{-\Delta}^{\Delta} dx \, \frac{x \sin\left(\frac{\omega}{c} xz + k_r z \right)}{x^2 + x_0^2}, \tag{19}$$

$$I_3 = \int_{-\Delta}^{\Delta} dx \, \frac{x \cos\left(\frac{\omega}{c} xz + k_r z \right)}{x^2 + x_0^2} , \tag{20}$$

$$I_4 = N_r x_0 \int_{-\Delta}^{\Delta} dx \, \frac{\sin\left(\frac{\omega}{c} xz + k_r z \right)}{x^2 + x_0^2}, \tag{21}$$

and the value of the parameter Δ is limited by the inequality

$$N_r > \Delta \gg x_0 . \tag{22}$$

Inequality (22) represents an additional condition related to the weak absorption approximation, which allows us to replace the bounds of

integration $\pm\Delta$ in Eqs. (18)–(21) with $\pm\infty$ (the inequality is fulfilled when $\varepsilon_{2r} \ll 2\varepsilon_{1r}$). Thereafter, the evaluation of the integrals is elementary:

$$I_1 = \pi \cos(k_r z)\, e^{-(k_0 - k_r)z},$$

$$I_2 = \pi N_r \cos(k_r z)\, e^{-(k_0 - k_r)z},$$

$$I_3 = -\pi \sin(k_r z)\, e^{-(k_0 - k_r)z},$$

$$I_4 = \pi N_r \sin(k_r z)\, e^{-(k_0 - k_r)z},$$

i.e.

$$I_1 I_2 - I_3 I_4 = \pi^2 N_r e^{-2(k_0 - k_r)z},$$

and, consequently,

$$S_z = S_0 e^{-(k_2 - k_1)z},$$

$$S_0 \equiv \frac{2|j_0|^2 k_r^3 \pi^2 c^2}{\omega^3 (2\varepsilon_{1r} - \varepsilon'_{1r} k_r)^2}, \tag{23}$$

$$k_1 \equiv k_r\left(1 - \frac{\varepsilon_{2r}}{2\varepsilon_{1r} - \varepsilon'_{1r} k_r}\right), \quad k_2 \equiv k_r\left(1 + \frac{\varepsilon_{2r}}{2\varepsilon_{1r} - \varepsilon'_{1r} k_r}\right).$$

As can be inferred from Eq. (15), k_1 and k_2 in Eq. (23) are just the extremum points of the function F_1 at a given frequency ω. In accordance with the commonly used definition of the absorption coefficient μ

$$\frac{dS_z}{dz} = -\mu S_z, \tag{24}$$

one therefore comes to a formula of an especially simple form

$$\mu = k_2 - k_1. \tag{25}$$

According to Eqs. (13) and (23), when $\varepsilon'_{1r} k_r \ll 2\varepsilon_{1r}$, the latter formula can be rewritten as

$$\mu = \frac{k_r \varepsilon_{2r}}{\varepsilon_{1r}} = \frac{\omega \varepsilon_{2r}}{c\sqrt{\varepsilon_{1r}}}. \tag{26}$$

4. MODEL CALCULATIONS

In the framework of the *weak absorption approximation*, specified by Eq. (14) and complemented with Eq. (22), the calculation of the absorption coefficient at any given photon energy in the off-resonance region reduces to a simple problem of finding extremum points of the function F_1 defined by Eqs. (12) and (15). Such a problem can be readily solved with the help of even a simple computer as may be inferred from Fig. 1. However, for the actual employment of the method one should specify the damping parameter $g_{\mathbf{k}}$, which is a much more complicated task. The adjustment of the frequency shift $d_{\mathbf{k}}$ is less important because one may assume that only renormalized frequencies are observable.

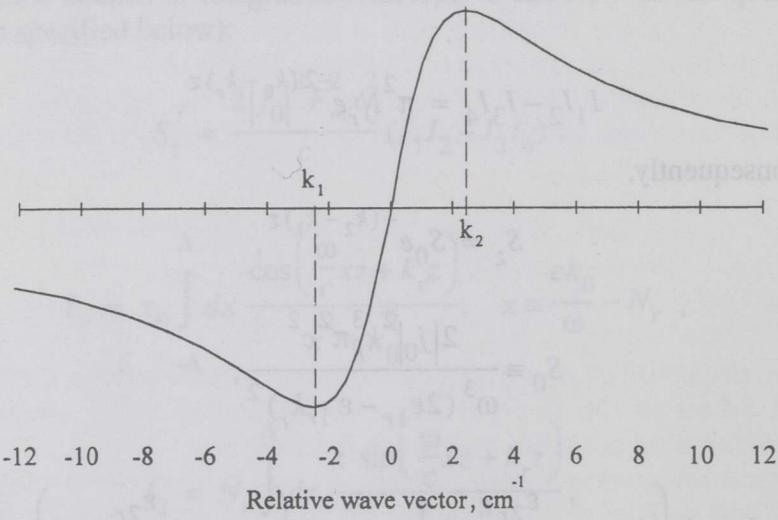

Fig. 1. A graphical illustration of the method used for the calculation of the absorption coefficient $(k_2 - k_1)$ within the weak absorption approximation. The curve corresponds to the function F_1 defined by Eq. (12), depending on the wave vector at a given photon energy $E = 8$ eV. The intraband scattering mechanism of excitons according to Eq. (33) is presumed. The zero-point corresponds to the resonant value k_r that can be determined from Eq. (13) ($k_r = 704\,922.81$ cm^{-1} for the curve presented).

In this section the method is applied to the low-energy side of the lowest-energy absorption band of crystalline xenon. The band is related to the exciton zone $\Gamma(3/2)$ characterized by the effective mass $M = 2.45\, m_e$ [18]. Only longitudinal acoustical phonons are of importance for the exciton–phonon coupling [2]. Their dispersion can be well approximated by the formula [19]

$$E_q = E_L \sin\left(\frac{\pi}{2} \cdot \frac{q}{q_m}\right), \tag{27}$$

where q is the modulus of the phonon wave vector, q_m is its maximum value, and $E_L = 5.4$ meV [19] is the corresponding maximum energy.

Now let us try to specify the damping parameter. In the framework of the polariton theory [10, 11], for the idealized class of crystals under study,

$$g_{\mathbf{k}} = \pi \sum_{\mathbf{q}} \left| F_{11}(\mathbf{k}, \mathbf{q}) \right|^2 \alpha(\mathbf{k})\, \alpha(\mathbf{k}+\mathbf{q}) \times$$

$$\times \{ \bar{n}_q \delta [E(\mathbf{k}) - E(\mathbf{k}+\mathbf{q}) + \omega_q] +$$

$$+ (\bar{n}_q + 1)\, \delta [E(\mathbf{k}) - E(\mathbf{k}+\mathbf{q}) - \omega_q] \}, \qquad (28)$$

while according to another model discussed here [12]

$$g_{\mathbf{k}} = \pi \sum_{\mathbf{q}, n} \left| F_{1n}(\mathbf{k}, \mathbf{q}) \right|^2 \times$$

$$\times \{ \bar{n}_q \delta [\Omega_1(\mathbf{k}) - \Omega_n(\mathbf{k}+\mathbf{q}) + \omega_q] +$$

$$+ (\bar{n}_q + 1)\, \delta [\Omega_1(\mathbf{k}) - \Omega_n(\mathbf{k}+\mathbf{q}) - \omega_q] \}, \qquad (29)$$

$E(\mathbf{k})$ in Eq. (28) denotes the polariton frequency, whereas the symbol $\Omega_1(\mathbf{k})$ in Eq. (29) has been used for the main dipole allowed exciton zone ($\hbar = 1$). The values of the subscript $n > 1$ in the last formula take account of the possible interband exciton–phonon processes. At this point, as [12] may not be yet available to the readers, some comments are probably needed in connection with the physical meaning of Eq. (29). This formula, which is an essential constituent of earlier theories of the lineshape of the exciton absorption bands [2], seems to be incompatible with the polariton theory. Indeed, in those preliminary versions of the theory aiming at direct calculation of the absorption coefficient, the exciton–photon coupling has been treated applying the perturbation methods. In [12], two independent approaches for the calculation of the transverse dielectric constant are proposed, whereas the problem of light absorption is discussed only in passing. Both derivations presented in [12], proceeding from the Green functions of excitons and transversal photons, respectively, lead to Eq. (1) of the present report. It is essential that the corresponding damping term is expressed by Eq. (29). Therefore, [12] proposes a new viewpoint on this well-known formula. This viewpoint does not actually contradict to the polariton theory, while polaritons can be still treated as the solutions of the dispersion relation (1') where, however, damping cannot be neglected any more.

The quantity

$$\alpha(\mathbf{k}) = \frac{E(\mathbf{k})\, \Omega_1(\mathbf{k})}{\Omega_1^2(\mathbf{k}) + \dfrac{\varepsilon_0 \left[\Omega_1^2(\mathbf{k}) - E^2(\mathbf{k}) \right]^2}{\Omega_p^2 F}} \qquad (30)$$

in Eq. (28) is the so-called exciton component of polaritons [20]. Only the case $T = 0$ is examined here, i.e. the mean occupation number of phonons $n_q = 0$.

In fact, the most difficult problem is the adjustment of the exciton–phonon coupling parameters F_{1n} entering into Eqs. (28) and (29). However, in the far off-resonance region, i.e. in the case of small exciton wave vectors, we can make use of the approximation of a quasi one-dimensional crystal. To a first approximation, assuming $\mathbf{k} = 0$, one therefore obtains [21]

$$F_{1n} = A_{1n} \sin^{1/2}\left(\frac{\pi}{2} \cdot \frac{q}{q_m}\right) \cos\left(\frac{\pi}{2} \cdot \frac{q}{q_m}\right), \tag{31}$$

where the constants A_{1n} can be treated as some variational parameters within this approach.

Now intraband polariton–phonon scattering should be nearly elastic [22]. It means that $q = 2k \cdot \sin(\theta/2)$, where θ is the angle of scattering, i.e. q should also be a small parameter and according to Eq. (31) the quadrate of F_{11} is nearly proportional to q. Therefore, in the framework of the polariton theory [10, 11] one comes to the following formula for the damping parameter in the off-resonance region:

$$g_{\mathbf{k}} = A_{11}^2 \frac{2\pi^2}{q_m} \frac{\alpha^2(\mathbf{k})}{v_k} \left(\frac{k}{q_m}\right)^3, \tag{32}$$

where v_k is the group velocity of polaritons.

Analogous formulas can be derived for the model of damping excitons [12]. In this case, in accordance with the mentioned peculiarities of solid xenon, the contributions from two different exciton–phonon coupling channels should be summed up. Here, the effective mass approximation with $M = 2.45\ m_e$ for both exciton zones, $\Gamma(3/2)$ and $\Gamma'(3/2)$, has been used. The intraband scattering of excitons with small wave vectors can be assisted by low-energy phonons only. The corresponding damping term therefore becomes

$$g_{11}(\mathbf{k}) = A_{11}^2 \frac{\pi^2}{B} \left(1 - \frac{k_0}{k}\right)^3 \left(\frac{k}{q_m}\right)^2, \tag{33}$$

where $B \approx 0.8$ eV [13, 23] is the width of the exciton zone in the effective mass approximation and

$$k_0 = \frac{\pi}{4} \cdot \frac{E_L}{B} q_m.$$

Note that the intraband scattering channel switches off when $k \leq k_0$. However, as results from the last formula, k_0 is actually a very small parameter.

Comparing Eqs. (32) and (33), one can easily conclude that the polariton theory predicts the off-resonance absorption to tail off much quicker as compared to the model proposed in [12] (see also Figs. 2, 3 and the Table). As already mentioned, the interband scattering in the case of solid xenon does not enter into the polariton picture. According to [12], on the contrary, the interband scattering of excitons is an important constituent of their damping. These processes can be taken into account through the terms with $n = 2$ in Eq. (29). The energy separation between the $\Gamma(3/2)$ and $\Gamma'(3/2)$ exciton zones $\Delta E \approx 0.1$ eV [13]. Therefore, the interband scattering is assisted by phonons whose wave vectors should be close to some characteristic value q_0 that can be determined from the relation

$$\Delta E = \frac{q_0^2}{2M}.$$

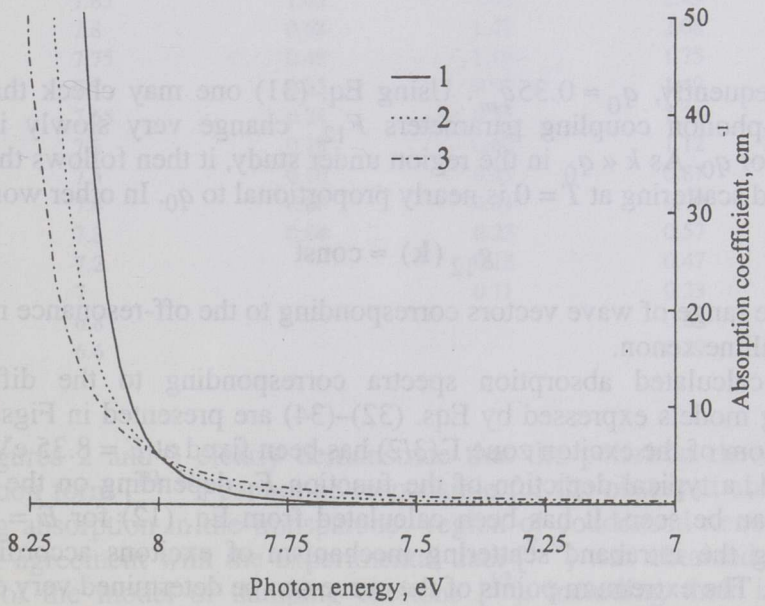

Fig. 2. Calculated low-energy tails of the lowest-energy exciton absorption band of crystalline xenon at $T = 0$ K for various theoretical models. Curve 1 corresponds to the polariton theory [10, 11] where the parameter g_k in Eq. (1) is directly related to the damping of polaritons according to Eq. (32). Two more curves correspond to the model [12] assuming intraband (curve 2) and interband (curve 3) scattering of excitons on phonons, expressed by Eqs. (33) and (34), respectively. The bottom of the exciton zone is fixed at $E = 8.35$ eV.

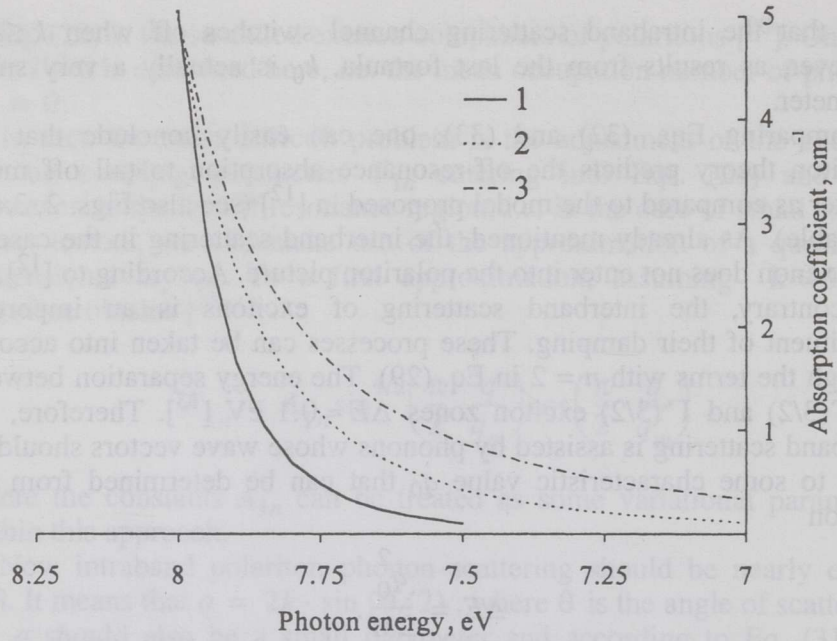

Fig. 3. The same calculated absorption curves (see explanations to Fig. 2) depicted on a different scale.

Consequently, $q_0 \approx 0.35 q_m$. Using Eq. (31) one may check that the exciton–phonon coupling parameters F_{12} change very slowly in the vicinity of q_0. As $k \ll q_0$ in the region under study, it then follows that the interband scattering at $T = 0$ is nearly proportional to q_0. In other words,

$$g_{12}(\mathbf{k}) \approx \text{const} \tag{34}$$

in a wide range of wave vectors corresponding to the off-resonance region of crystalline xenon.

The calculated absorption spectra corresponding to the different damping models expressed by Eqs. (32)–(34) are presented in Figs. 1–3. The bottom of the exciton zone $\Gamma(3/2)$ has been fixed at $E = 8.35$ eV [23]. In Fig. 1, a typical depiction of the function F_1 depending on the wave vector can be seen. It has been calculated from Eq. (12) for $E = 8$ eV, assuming the intraband scattering mechanism of excitons according to Eq. (33). The extremum points of the curve can be determined very easily. This, in turn, immediately fixes the value of the absorption coefficient according to Eq. (25). The curves, quite similar to the presented one, can be calculated for a wide range of wave vectors. The procedure does not include any peculiarities related to the damping mechanism of elementary excitations in the crystal. On the other hand, different damping models expressed by Eqs. (32)–(34) are easy to compare. The proportionality factors in those formulas have been arbitrarily chosen to give $\mu = 5$ cm^{-1} at $E = 8$ eV, independent of the model. The parameters needed for the calculation of polariton dispersion curves have been taken from [23].

Absorption coefficient (cm^{-1}) of solid Xe at $T = 0$ K for various theoretical models. The value $\mu = 5\ cm^{-1}$ at $E = 8$ eV, independent of the model, has been fixed by an appropriate choice of the proportionality factors of the exciton–phonon coupling. The bottom of the exciton zone is located at $E = 8.35$ eV

Photon energy (eV)	Polariton–phonon coupling [10, 11]	Exciton–phonon coupling [12]	
		intraband scattering	interband scattering
8.3	23080	935.5	144.2
8.25	1134	143.5	46.1
8.2	195.7	48.0	23.0
8.175	100.3	31.7	17.5
8.15	56.3	22.2	13.8
8.125	34.0	16.1	11.2
8.1	21.4	12.1	9.22
8.075	14.2	9.43	7.74
8.05	9.76	7.47	6.59
8.025	6.91	6.04	5.67
8	5.00	5.00	5.00
7.95	2.79	3.43	3.83
7.9	1.68	2.56	3.06
7.85	1.05	1.90	2.49
7.8	0.68	1.47	2.08
7.75	0.48	1.16	1.75
7.7	0.33	0.92	1.49
7.65	0.24	0.75	1.28
7.6	0.18	0.62	1.12
7.5	0.10	0.43	0.87
7.4	0.06	0.31	0.70
7.3	0.04	0.23	0.57
7.2		0.18	0.47
7		0.11	0.33
6.8			0.25
6.6			0.19

Figures 2 and 3 clearly demonstrate that the polariton theory in its common form [10, 11] cannot be responsible for the observed abnormally strong absorption in the transparency region of condensed xenon. Much better agreement with the experimental data [8, 9] was obtained with the help of the model of damping excitons [12], including their interband scattering mechanism, which becomes predominant in the far off-resonance region of crystalline xenon.

5. CONCLUSION

In this paper, the validity of the weak absorption approximation for the calculations in the low-energy off-resonance region of exciton absorption bands is demonstrated. The applicability of the approach does not depend

on the physical meaning of the damping parameter $g_{\mathbf{k}}$ entering into the general expression for the dielectric constant. Nevertheless, to make use of this method, one should specify the mechanism of damping in the wave vector representation. The approximation is usable in a wide range of frequencies below the exciton resonance and even relatively close to it, as can be seen from the Table. However, at the frequencies approaching resonance, i.e. $\omega \approx \Omega(\mathbf{k})$, the calculations abruptly become impossible, as the absorption cannot be considered weak any more.

The validity of the method has been verified for the examination of the origin of unexpectedly strong absorption in the presumable transparency region of solid xenon. Three possible damping mechanisms of the elementary excitations of the crystal have been compared. In spite of a somewhat simplified character of those calculations (e.g. only $T = 0$ case was examined) it seems reasonable that the effect is essentially related to the interband scattering of excitons from the dipole allowed zone $\Gamma(3/2)$ into the forbidden zone $\Gamma'(3/2)$, according to the model proposed in [12]. In fact, this model does not contradict to the polariton theory. It only ascribes the nonstationariness of polariton states directly to the damping of excitons.

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ELEKTROMAGNETVÄLJA NÕRGENEMINE MITTEJUHTIVATE KRISTALLIDE LÄBIPAISTVUSE PIIRKONNAS

Matti SELG

Üldistest Maxwelli võrranditest lähtudes on välja töötatud otsene meetod valguse neeldumiskoefitsiendi arvutamiseks isoleeritud eksitontsooni piirkonnas. Käsitlus baseerub transversaalse dielektrilise konstandi reaal- ja imaginaarosa lihtsatel rittaarendustel, mida on nimetatud *nõrga neeldumise lähenduseks*. Meetodit on rakendatud tahke ksenooni eksitonneeldumisriba $\Gamma(3/2)$ pikalainelise serva arvutamiseks.