On the dielectric approximation in the additional light waves theory: an historical rehabilitation (recovery)

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Abstract. After the basic Pekar papers on crystal optics with spatial dispersion (CSD) among a lot of others there was the "dielectric approximation" (DA) method. But soon it has been fully rejected because, as it was recognized by the main group of authors on the theme, it came in conflict with the law of conservation of energy flux through the ideal vacuum-crystal interface. The objective of this paper is to advance a way for rehabilitation of DA method by using indispensable broadening and essential generalization of the expression just for the energy flux density vector (EFDV) at the vacuum-medium interface (the so-called Poynting-Pekar vector in the additional light waves (ALW) theory).

For the sake of simplicity and clearness, we shall restrict ourselves to a simple model of polarization oscillators in a medium as well as the simplest geometrical configuration of the light-medium system. Also, we shall stand here in the framework of the exciton effective mass (EEM) approximation (the exciton effective mass *M* is taken positive for the sake of definiteness).

Let us consider a case that a monochromatic plane wave (with the frequency ω and wave vector $k_0 = \omega/c$, where *c* is the speed of light in vacuum) is incident from vacuum along the normal to the surface of a semi-infinite optically uniaxial crystal (occupying the half-space $x \geq 0$). We assume that this wave is in resonance with some nondegenerate dipole-allowed excitonic state to which a polarization vector $\mathbf{P}(0,0,P(x))$ corresponds.

The **P** vector, being directed along the crystal axis *C(Z)*, is parallel to that of the wave electric field $\mathbf{E}(0,0,E(x))$ and determines the partial contribution from the state studied into the total crystal polarization.

The ideal Maxwell equation in a medium has the standard form [1]

$$\frac{\partial^2 E}{\partial x^2} + k_0^2 (\varepsilon_0 E + 4\pi P) = 0 , \tag{1}$$

where ε_0 is the background permittivity.

As it was shown in our papers [2], the material equations appropriate for the situation considered may be written in one of the following forms:

1.   In the pioneer Pekar form [3] that arises from supplemented material Maxwell equation in the $(0,\infty)$ half-space to the $(-\infty,\infty)$ space with further reducing to zero the remainder part $(-\infty,0)$ in $x \geq 0$ space (in a more generalized case, it corresponds to $U_p = -1$ or $P(0^+) = 0$ in the point 4, see bellow).

2.   In Tolpygo-Mashkevich form that is based on the simultaneous use of the material and ideal equations in the integral form [4]. We have checked, of course, identicity of this approach to

the problem with that of the points 3, 4, and came to conclusion that there is no reasons to base the paper on the different formulations of the ideal Maxwell equations.

3. One more historical approach is based on the fundamental Hopfield-Thomas paper [5]. In this approach, the Maxwell material equations for the above presented configuration is determined by the following material equation:

$$\frac{\hbar}{2M}\frac{\partial^2 P}{\partial x^2} + (\omega - \omega_{ex} + i\gamma)P = -\frac{\varepsilon_0 \Delta}{4\pi}E . \qquad (2)$$

Here, ω_{ex} and γ are the resonance exciton frequency and damping constant, respectively; the parameter Δ is equal to the constant of the so-called "longitudinal – transverse" splitting of exciton line, which, in its turn, may be written in several possible equivalent forms (see Eq. (9) in *mob*4 of 2004).

According to the studies of such class of equations in mathematical physics, the linear in P boundary condition for Eq. (2) is of the following form

$$P(0^+) + \alpha\frac{\partial P}{\partial x}(0^+) = 0 , \qquad (3)$$

where α, in general, is some rather arbitrary parameter that will be partly determined in the point 4.

4. In the simplest integral form with some free enough parameter $U (|U| \le 1)$ that determine the exciton interaction with crystal surface

$$P(x) = \int_0^\infty [\chi(x - x', \omega) + U\chi(x + x', \omega)]E(x')dx' , \quad (4)$$

where $\chi(x - x', \omega)$ is the polarization operator for an infinite medium:

$$\chi(x - x', \omega) = -i\frac{\varepsilon_0 \Delta M r}{4\pi\hbar}\exp\left(\frac{i}{r}|x - x'|\right),$$

$$\text{with } r = \frac{2M}{\hbar}\left(\omega - \omega_{ex} + i\gamma\right)^{-\frac{1}{2}}. \qquad (5)$$

It is obvious that all the above approaches to the problem must be equivalent, and their parameters should be closely connected to one another. Namely, parameter

$$U_p = -1 \qquad (6)$$

corresponds to $\alpha = 0$ in (3), i.e., is the main and the most often used Pekar (or Dirichlet) condition with $P(0^+) = 0$

$$U_g = 1 \qquad (7)$$

that corresponds to the Ginzburg (or Neumann) case [6] with $\alpha^{-1} = 0$ in Eq. (3), and $\frac{\partial P}{\partial x}(0^+) = 0$

$$U_s = -\left[1 + 2i\frac{\delta d}{r(C + \delta)}\right], \text{ with } C = -\frac{\hbar^2}{2Md^2} , \qquad (8)$$

where d is the lattice constant in the x direction and δ is the energy shift of a molecular level on the surface as compared to that of the crystal bulk. The latter corresponds to the Sugakov case [7] with $\alpha = \delta d/(C + \delta)$ in Eq. (3) – that was the first in ALW theory representation of expression for P in the form of Eq. (4).

All the above relations can be easily checked by using Eq. (4) and its differentiation with respect to x, for arbitrarily x as well as for x approaching to 0^+.

In full compliance with the abovementioned, the DA case was already considered in papers [8] with

$$U_{da} = 0 . \qquad (9)$$

In terminology of the point 3, it determines α in Eq. (3) uniquely:

$$\alpha_{da} = -ir . \qquad (10)$$

At the same time, if the exciton effective mass approximation is applied consistently, then the expression for the energy flux density is also well defined, and the possible (virtual) jump of time-averaged normal components of the energy flux density $_\Delta S_0$ at the vacuum-medium interface is given by the expression known in crystal optics with spatial dispersion:

$$_\Delta S_0 = i\frac{\hbar\omega_{ex}}{2M\Delta\varepsilon_0}\left(P\frac{\partial P^*}{\partial x} - P^*\frac{\partial P}{\partial x}\right)_{0^+} . \qquad (11)$$

For the most cases of Eqs. (6) to (8) type, the necessary turning $_\Delta S$ to zero was easily checked, so from the very beginning this fact does not call any doubt as to its general applicability *ad hoc* and, naturally, has been omitted at the calculation of reflection and passing of EM waves for all above situations, that is for all possible meanings of parameter U, including also the case $U_{da} = 0$.

But soon a big enough group of authors (see, e.g., [3, 6, 9, 10]) who used these to simple calculations for $U = 0$ has noted, that in this case the law of energy conservation violates and $_\Delta S_0$ has a discontinuity at 0^+. Namely, this fact results in total ignoring the DA method in ALW theory as inevitable cost of scientific production.

Now our aim is to reanimate it. To this end, we shall take the generalized expression for $_\Delta S$ (see the papers [2]) including the undetermined for a moment expression for the density of the surface polarization current $\mathbf{j}(0, 0, j_0)$. In this case, $_\Delta S$ takes the form:

$$_\Delta S = -\frac{1}{4\pi}[E_0 j_0^* + c.c] +_\Delta S_0 \qquad (12)$$

with $j_0 = \sigma(\omega)E_0$, and σ as the one more free but not completely independent parameter, because it may be related with the parameter U or α by the relation:

$$(_\Delta S)_{0^+} = 0 . \tag{13}$$

As it has been argued in [2], one can choose σ in such a way that $\mathrm{Im}(\sigma) = 0$ in the absence of real surface conductivity.

So, the boundary conditions for electric field and magnetic induction $B\left(-\dfrac{1}{c}\dfrac{\partial \mathbf{B}}{\partial t} = \mathrm{rot}\mathbf{E} \right)$, in the frame of the taken above space configuration and the time dependence of the fields, take the following form:

$$E(0^-) = E(0^+) , \tag{14}$$

$$\frac{\partial E}{\partial x}(0^-) = \frac{\partial E}{\partial x}(0^+) + 4\pi \frac{ik_0}{c} j_0 . \tag{15}$$

Here, $0^\mp$ designates approaching the crystal boundary on the left and right to the origin of coordinates, respectively.

So, step by step we come to the complete system of consistent equations necessary for solving the assigned problem. As usually, we seek its solution in the framework of the plane waves, namely $P(x), E(x) \to (E_j, P_j)\exp(ik_j x)$ with $k_j = k_0 n_j$ for each possible value of the wave-vector. We obtain two different roots for refraction, n_1 and n_2 in accordance with the second order in x of two joint homogeneous Eqs. (1) and (2) and with the requirement for absence of any electromagnetic field sources at $(+\infty)$ – for half-space configuration.

These two roots for refraction are

$$n_{1,2}^2 \equiv n_{+,-}^2 = \frac{1}{2}(\mu + \varepsilon_0) \pm \sqrt{\frac{1}{4}(\mu - \varepsilon_0)^2 + b} \tag{16}$$

where $\mu \equiv \dfrac{2M}{\hbar k_0^{\,2}}(\omega - \omega_{ex} + i\gamma), b = \dfrac{2M\Delta\varepsilon_0}{\hbar k_0^2}$. And, accordingly, the relations

$$P_j = \frac{1}{4\pi}\left(n_j^2 - \varepsilon_0\right)E_j , \tag{17}$$

$$\frac{\partial P_j}{\partial x}(0^+) = ik_0 n_j P_j(0^+) . \tag{18}$$

As a result, we have the complete system of equations for the problem under consideration. Being written in terminology of E_i, E_R, E_1, E_2 for amplitudes of incident, reflected and two penetrating waves, it has the following form

$$\begin{cases} E_i + E_R = E_1 + E_2 \equiv E_0 \\ E_i - E_R = n_1 E_1 + n_2 E_2 + \dfrac{4\pi}{c}\sigma E_0 \\ \displaystyle\sum_{j=1,2}\left(n_j^2 - \varepsilon_0\right)\left(1 + i\alpha k_0 n_j\right)E_j = 0 \end{cases} \tag{19}$$

where $\sigma = \dfrac{c}{4\pi}k_0 b\,\mathrm{Im}(\alpha)\left|n_1 + n_2 + i\alpha k_0 f\right|^{-2}$ (with $f = n_1^2 + n_2^2 + n_1 n_2 - \varepsilon_0$). It follows from Eq. (13) by a transformation from P_j and $\dfrac{\partial P_j}{\partial x}$ to $E_{1,2}$ (see equations (17), (18)) and then via E_0 from the first and third equations of the system (19).

This fact makes it possible to express all fields arising in crystal and free space via the field E_0 and to construct the reflection amplitude coefficient (R) for the situation considered:

$$R \equiv \frac{E_R}{E_i} = \frac{1 - n_{eff}}{1 + n_{eff}} , \tag{20}$$

where n_{eff} is the effective refractive index:

$$n_{eff} = \frac{n_1 n_2 + \varepsilon_0 + i\alpha k_0 n_1 n_2 (n_1 + n_2)}{n_1 + n_2 + i\alpha k_0 f} + \frac{4\pi}{c}\sigma . \tag{21}$$

It is evident that different parts of the expression for n_{eff} may be combined in many ways, but here we take those of them that can be easily compared with the simplest and well known ones, namely:

the Pekar [3] result ($\alpha = 0, \sigma = 0, P_{ex}(0^+) = 0$) with

$$n_{eff} = \frac{n_1 n_2 + \varepsilon_0}{n_1 + n_2} \tag{22}$$

and the Ginzburg [6] result ($\alpha^{-1} \to 0, \sigma = 0, \dfrac{\partial P_{ex}}{\partial x}(0^+) = 0$) with

$$n_{eff} = n_1 n_2 (n_1 + n_2) f^{-1} . \tag{23}$$

So, we fulfilled the main aim of the paper. First, we have given the way for rehabilitation of DA and a group of other similar cases in the ALW theory, in principle. Second, we have given the correct expressions for the coefficients of reflection and refraction of polariton waves in DA for a specific "light – crystal" orientation (see Eqs. (20) and (21) with $i\alpha = r$, and $\sigma \ne 0$, in principle).

Some additional remarks.

After the first Pekar paper on the ALW problem, a lot of publications on the subject appeared. Many of them deal with different types of generalization (as to the variety of exciton-like states, so for their degeneracy or multiplicity, for oblique incidence with different ABC for different polarization vector projections, etc.) and can be found in the rather complete review [11], in the cited above papers [2] and elsewhere. But here we cited the really pioneer works only (see points 1-4 above) which turned out enough for our aim.

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