Consistency of boundary conditions in crystal optics with spatial dispersion

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Our work deals with the application of the spatial dispersion theory to optical effects in the spectral region of excitonic transitions. The principal problems here are the physics of additional (with respect to the traditional birefringence theory) light waves (ALW's) and formulation of the corresponding additional boundary conditions (ABC's) (with respect to the Maxwell ones). It was substantiation of the ABC choice that aroused an active discussion lasting from the first study of this problem (1957) up to now. As a result of certain theoretical incompleteness, some ABC's have come into conflict with the main physical principles and were rejected. Here we show that this rejection is premature and try to remake them using the experience gained in attempts to reach self-consistency of ABC's and Maxwell boundary conditions within crystal optics with spatial dispersion. The main approach is to put into correspondence a number of formulas for polaritonic reflectance that are used in ALW physics, with the principle of conservation of polaritonic energy.

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Until 1957 all investigations of crystal optics were performed on the basis of the classical birefringence theory that was formed long ago and approved repeatedly. A new stage of the development of the crystal optics with spatial dispersion (SD) started in 1957. It was initiated not least by the physics of additional light waves (ALW's). For the first time an actual possibility for the observation of the corresponding effects was predicted by Pekar in Ref. 1 who made a comprehensive analysis of the spectral region of excitonic resonances.

Dealing with ALW's, a variation of the classical birefringence theory is required. One of its consequences is, in particular, that for a bounded medium the traditional Maxwell-Fresnel boundary conditions (MFBC's) (which relate electromagnetic fields at a boundary between two media) become insufficient to determine unambiguously the amplitudes of all the waves considered. Some additional boundary conditions (ABC's) are required in this case. For the first time such ABC were formulated in the same paper[1] of 1957 in which ALW's were introduced [see Eq. (7) below]. Up to now they are still used by most of researchers, both theorists and experimentalists. Concurrently studies on the refinement of MFBC's were performed. They dealt with taking into account spatial dispersion as a whole and coordination of the whole set of boundary conditions in the region of excitonic transitions.[2]

In spite of a vast number of works that have been published for 45 years, still much controversy remains concerning the correct form of ABC's. Just the same might be said about ALW physics and other problems of spatially dispersive media as a whole. One of the controversies that has also arisen due to the presence of a boundary is related to the problem of fitting generalized MFBC's and ABC's. It is well known[3] that consideration of them separately (as is done in most cases) may lead to violation of fundamental principles. Such a situation often arises at the calculation of the polaritonic reflectance (PR), either on the basis of microscopic consideration or in the continual approximation. The objective of our paper is just in putting into correspondence with the principle of conservation of polaritonic energy a number of formulas for the calculation of PR that are used in ALW physics.

In the early works on ALW's various ABC's were derived on the basis of Pekar's idea. It stated that a correct (i.e., not conflicting with the principal laws of physics) formulation of ABC's is possible on the basis of exciton models only. Such a position remains in a great number of further works in this line (see references to the corresponding works made before the 1990s in Refs. 2–4 as well as the works made during the last decade[5–9]). The model concepts, however, are restricted as compared to actual situations. This natural drawback of such concepts stimulated the attempts of many researchers to build heuristic generalizations of ABC's. These generalizations are based on either the results of model calculations for a set of excitonic states (which is replenished permanently), and/or rather general semiphenomenological guesses, and/or some additional postulates concerning the polarization operator structure or range, etc.[3]

From the very beginning the grounds for such generalizations of ABC's aroused keen discussion. It is revived every time a new group of researchers in the field of SD theory advances fresh results of their investigations. As a consequence of this, many of the results obtained have been rejected (in our opinion, sometimes prematurely) either because they seemed to be in conflict with the principle of conservation of energy or due to some similar reasons. The objective of this work is to advance the way for rehabilitation of such results through self-consistency and inner coordination, as was mentioned above.

The presentation of our paper is closely related to its content. Having in mind the aim to analyze critically (from the viewpoint of their obeying the principle of conservation of energy) and to remake as many of the known formulas for the calculation of PR as possible, we cite all the corresponding formulas known from the literature on ABC's (which are required for achievement of our goal), along with a couple of the pioneer works dealing with each of them. For the sake of simplicity and clearness, we shall restrict ourselves to a simplest model of polarization oscillations in a medium [the exciton effective mass (EEM) approximation], as well as the

simplest geometrical configuration of the "light-medium" system.

Let us consider a case when a monochromatic plane wave (with frequency ω and wave vector $k_0=\omega/c$, where c is the speed of light in vacuum) is incident from vacuum normally to the surface of a semi-infinite optically uniaxial crystal (occupying the half-space $x\geqslant 0$). We assume that this wave is in resonance with some nondegenerate dipole-allowed excitonic state to which a polarization vector $\boldsymbol{P}$ corresponds. This vector (directed along the crystal axis) is parallel to that of the wave electric field $\boldsymbol{E}\{0,0,E\}$ and determines the partial contribution from the state studied into the total crystal polarization. The ideal Maxwell equation in a medium (1) and MFBC (2) now have the standard form[10]

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

where ε_0 is the background permittivity, and

$$E(0^-)=E(0^+),$$

$$\frac{\partial E}{\partial x}(0^-)=\frac{\partial E}{\partial x}(0^+)+4\pi\frac{ik_0}{c}j, \tag{2}$$

Here $0^{\mp}$ designates approaching the crystal boundary from the left and right to the origin of coordinates, respectively. The quantity $\boldsymbol{j}\{0,0,j\}$ (which is at the focus of consideration in this work) is the density of the surface polarization current. The following preliminary assumptions are made here concerning this quantity:

$$j=\sigma(\omega)E(0), \tag{3}$$

where σ is some (unspecified for the moment) surface conductivity of the crystal, and

$$\mathrm{Im}(\sigma)=0. \tag{4}$$

It should be noted that the requirement $\mathrm{Im}(\sigma)\neq 0$ is not contradictory, in principle, to the condition of energy flow continuity at the vacuum-medium boundary (when neglecting spatial dispersion, as well as taking it into account). However, it introduces, without some reasonable grounds, an extra phenomenological parameter (or even a function of frequency) into the theory. By assuming Eq. (4) valid, we believe that this situation holds also in the limiting case of birefringence for those media and spectral regions where the traditional Fresnel formulas for light reflection and transmission are adequate. This is valid, in particular, by ignoring the higher multipole transitions (except the dipole one) in the crystal-field interaction,[11] as it has place in this paper.

Usually, in addition to the above-mentioned conditions, the crystal surface is assumed ideal (due to absence of any mechanisms for intense decay of excitons or their considerable diffuse scattering) to an extent that the requirement $\mathrm{Re}(\sigma)=0$ is assumed also. The experience of model calculations, however, evidences that in SD theory the condition $\mathrm{Re}(\sigma)\neq 0$ may be not only consistent, but necessary as well, even when there are no specific polariton energy sources or drains at the crystal surface. When calculating light wave reflection from crystals in this work, we shall resort to this fact.

In the EEM approximation the material Maxwell equation for a present configuration is determined by the following constitutive equation[1,12]:

$$\frac{\hbar}{2M}\frac{\partial^2 P}{\partial x^2}+(\omega-\omega_{ex}+i\gamma)P=-\frac{\varepsilon_0\Delta}{4\pi}E. \tag{5}$$

Here M is the exciton effective mass (taken positive for the sake of definiteness); ω_{ex} and γ are the resonance frequency and damping, respectively; Δ is equal to the so-called "longitudinal-transverse" splitting of exciton line.

According to the studies of such class of equations in mathematical physics, the only linear boundary conditions for Eq. (5) that are not overdetermined have generally the following form:

$$P(0^+)+\alpha\frac{\partial P}{\partial x}(0^+)+\beta E(0^+)=0. \tag{6}$$

Here α and β are some phenomenological parameters of the theory. Their values may be determined either from experiment and/or by calculation, after having chosen a concrete microscopic model for the excitonic state of a confined crystal. When applying the semiphenomenological approach to the boundary conditions, the parameters introduced above may be treated, naturally, as some functions of frequency to be determined experimentally. We will treat them (as is made traditionally in the studies of ALW's) as constants in the narrow spectral region of the exciton transition considered or as some concrete functions of frequency [e.g., stepwise ones, as is done often for the quantity γ—see Ref. 13—or those of the form (10) in the dielectric approximation] which do not involve other parameters except for those entering Eq. (5) and/or a small number of some additional parameters, if they are phenomenological generalizations of certain model calculations [see text to Eq. (11)].

From the generalized Pekar ABC (6) one may obtain various particular ABC's that are used in when studying ALW's:

(i) The Dirichlet like boundary condition (known in physics of ALW's as the Pekar ABC proper[1]) that corresponds to the case $\alpha=\beta=0$—i.e.,

$$P=0. \tag{7}$$

Historically this is the first and most popular ABC which, in the opinion of many experimentalists, provides the best agreement between the theory and experiment at primary processing of the latter.

(ii) The Neumann boundary condition, with $\alpha^{-1}=0$ in Eq. (6):

$$\frac{\partial P}{\partial x}=0. \tag{8}$$

For the first time this condition has been derived for some specific excitonic models.[14–16] The author of Ref. 14 applied the Frenkel exciton model, subject to the condition that near-surface distortion of the crystal lattice is such that (a) a surface excitonic level may appear and (b) this level coincides

with the excitonic band edge (see also Ref. 2). The authors of Ref. 15 obtained the boundary condition (8) when considering repulsion of the Wannier-Mott exciton from the crystal surface. Due to this repulsion, the exciton is reflected from the crystal surface when its center of mass has not reached the surface yet.

The condition (8) appears also in the method of "auxiliary surface current"; see Ref. 17. However, it corresponds adequately to another problem statement—namely, excitation of polaritons in an infinite medium by a monochromatic light source located in the plane $x=0$ [in this case the current j in Eq. (2) is set by an external field source]. In Ref. 18 an attempt has been made to apply the above method and its results [in particular the form of Eq. (8) for ABC's] to the problem considered here. This attempt has aroused a useful discussion (see Refs. 19–21) that enabled us to refer the work of Ref. 18 to item (ii).

(iii) The mixed uniform boundary conditions, with $\beta=0$ in Eq. (6):

$$P + \alpha\frac{\partial P}{\partial x} = 0. \tag{9}$$

There are a number of studies where α was assumed to be an essentially large and, even more, a complex quantity. Among those studies are such as Ref. 22, where α was considered as one of the main parameters used in fitting experimental data to the results of theoretical calculations. There exist also a series of works, Refs. 23–26, which are well known in the literature concerning ALW's as the "dielectric approximation" (DA). They use the following expression for α:

$$\alpha \equiv -ir = -i\left[\frac{2M}{\hbar}(\omega - \omega_{ex} + i\gamma)\right]^{-1/2}. \tag{10}$$

In the above-mentioned works it was assumed concurrently that $j=0$. In this case some conflicts appear with the law of conservation of polariton energy flow at the crystal boundary. That was the reason why those works were ignored later by the researchers dealing with physics of ALW's. However, in our opinion, it has been done prematurely and the main aim of the paper is to remake and rehabilitate that and analogous approaches to the ABC problem.

The same may be said about a lot of papers on ALW theory, where the parameter α is determined starting from certain semiphenomenological assumptions concerning the structure and/or localization radius r of the polarization operator for a confined or semi-infinite medium. As some generalization of the results of the specific excitonic models (see, e.g., Refs. 14, 27, and 28) for the above-mentioned "light-crystal" configuration it is taken often[4] that $P = \int_0^\infty [\chi(x-x',\omega)+u\chi(x+x')]E(x')dx'$, where $u(|u|\leqslant 1)$ is a generalized phenomenological microscopic parameter setting the exciton interaction with crystal surface, and $\chi(x-x',\omega)$ is the polarization operator for an infinite medium. In the EEM approximation [which corresponds to Eqs. (5) and (9)], $\chi=-i(\Delta\varepsilon_0 Mr/4\pi\hbar)\exp[(i/r)|x-x'|]$ and in compliance with this

$$\alpha = ir(u+1)/(u-1). \tag{11}$$

The limiting cases here are as follows: $u=-1$ corresponds to item (i), while $u=1$ corresponds to item (ii) and $u=0$ results in Eq. (10) of the DA.

(iv) The mixed nonuniform boundary conditions of Dirichlet type, with $\alpha=0$ in Eq. (6):

$$P + \beta E = 0. \tag{12}$$

Reference 5 may serve as a typical example of the application of the boundary condition (12) in the physics of ALW's.[29] Since it was assumed in Ref. 5 that $j=0$, then in accordance with Ref. 30 all the remarks of item (iii), concerning the possible violation of the principle of conservation of energy, refer also to that paper.

Some other limiting versions of the ABC (6) are possible also. It is obvious, however, that in no case is the chosen set of parameters in Eqs. (2)–(6) independent. Their choice must be consistent from the standpoint of the fulfillment of the main physical principles, in particular those of the energy conservation law, the principle of symmetry of the kinetic coefficients, etc.

At this point we would like to draw attention to some previous investigations of this topic. In Refs. 31 and 32 the way of correction and consistence of boundary conditions and the appropriate material equations has been considered for SD effects in natural optically active crystals. The work in Ref. 31 has been done in the framework of birefringence theory but Ref. 32 has used the ALW theory concept [on the base of MFBC's (2) with $j=0$ and ABC's of (9) type with $\mathrm{Im}(\alpha)=0$, in fact]. The latter approach has been used also in Ref. 30 in another context—namely, for correlation of MFBC's and nonuniform ABC's of (12) type. Here we have to stress that in SD theory there exist different expressions for the energy flux vector $\boldsymbol{S}$, which may be called, conditionally, the Poynting-Pekar vector. In Refs. 30 and 31 a particular form of the vector $\boldsymbol{S}$ was used.[3,10] It is quite general with respect to the choice of excitonic model. However, its application is strongly restricted by the following requirements: $\mathrm{Im}(\beta)=0$ and $\mathrm{Im}(\alpha)=0$ [i.e., it does not include the case of DA and many other ones corresponding to expression (11)]; $\gamma=0$ (which seems unlikely in the spectral range of exciton resonance); the "interference" fluxes are to be ignored also. In this paper [see Eq. (13)], as well as is done in Ref. 32, we choose such a form of the above vector which enables us to go beyond those limitations.

If the EEM approximation is applied consistently, then the expressions for the energy dissipation power and for the energy flux density are well defined (see Refs. 33–35). In our case a possible jump of the time-averaged normal component of the energy flux, ${}_\Delta S$, at the vacuum-medium boundary is given by the expression

$${}_\Delta S = \frac{1}{4\pi}\left[-Ej^* + i\frac{2\pi\hbar\omega_{ex}}{M\Delta\varepsilon_0}P\frac{\partial P^*}{\partial x} + \text{c.c.}\right]_{0^+}. \tag{13}$$

It is evident that this jump is related to the presence of surface current and excitonic polarization behavior near the surface. Here an assumption is made that the crystal surface is neither a source nor a drain of polariton energy and the

above-stated quantities are determined mainly by the character of the exciton interaction with the crystal surface, so one may treat them, to a first approximation, as independent of possible dissipation processes at the crystal surface, excitation of surface waves, Raman scattering of polaritons, and so on (see Refs. 2 and 3). If the above conditions are met, then the relation between the boundary parameters must be such as to make $_{\Delta}S$ vanish.

The set of Equations (1)–(6) and (13) supplemented with the requirement for absence of any electromagnetic field sources at infinity $(+\infty)$ and with the condition $_{\Delta}S=0$ makes it possible to get a stable and unique solution of the problem. When speaking about the possibility of obtaining an exact solution, one often means that this solution can be presented in a way that is natural for physicists (the extinction theorem)—namely, as a sum of the incident and reflected waves in vacuum (whose refractive index is unity) and a finite number of plane waves that entered the crystal with the corresponding refractive indices n_j (for the problem studied there are two of them, e.g., $j=1,2$). In this case the final objective of the calculation is the amplitude reflection coefficient R, which can be obtained experimentally. We omit cumbersome but rather apparent intermediate mathematics, bearing in mind to make our paper more compact and help experimentalists to focus their attention on the final results only. The procedure of such calculation is known from numerous works on ALW's.[2,3] As concerning our problem with ABC (6), in the standard representation the coefficient R is of the following form:

$$R=\frac{1-n_{eff}}{1+n_{eff}}, \tag{14}$$

where n_{eff} is the effective refractive index. The latter may be presented as a sum of two terms n'_{eff} and n''_{eff}:

$$n_{eff}=n'_{eff}+n''_{eff}, \tag{15}$$

with

$$n'_{eff}=\frac{n_1+qn_2}{1+q}, \tag{16}$$

$$n''_{eff}=\frac{4\pi}{b}\left[\frac{k_0}{4\pi}\,\mathrm{Im}(\alpha)|D|^2-\mathrm{Re}(\beta * D)\right], \tag{17}$$

$$D=\frac{n_1(n_1^2-\varepsilon_0)+qn_2(n_2^2-\varepsilon_0)}{1+q}. \tag{18}$$

Here q sets the ratio E_2/E_1 between the amplitudes of two waves that propagate in the crystal,

$$q=-\frac{(n_1^2-\varepsilon_0)(1+i\alpha k_0 n_1)+4\pi\beta}{(n_2^2-\varepsilon_0)(1+i\alpha k_0 n_2)+4\pi\beta}, \tag{19}$$

and $n_{1,2}$ are the corresponding refractive indices that are determined by solution of the volume equations (1) and (5). They are the same for all models of excitons considered in the EEM approximation for the considered "light-crystal" configuration:

$$n_{1,2}^2\equiv n_{+,-}^2=\frac{1}{2}(\mu+\varepsilon_0)\pm\sqrt{\frac{1}{4}(\mu-\varepsilon_0)^2+b}, \tag{20}$$

where

$$\mu=(k_0 r)^{-2}\equiv\frac{2M}{\hbar k_0^2}(\omega-\omega_{ex}+i\gamma),$$

$$b=\frac{2M\varepsilon_0\Delta}{\hbar k_0^2}\equiv-(n_1^2-\varepsilon_0)(n_2^2-\varepsilon_0)$$

[here and below all equations are written in the "resonance" approximation identical to that for Eq. (5)].

We would like to note that for the particular microscopic model of Ref. 3 [with $\gamma=0$ in Eq. (5); $\mathrm{Im}(\alpha)=\mathrm{Im}(\beta)=0$ in Eq. (6) and $\mathrm{Re}(\alpha)$, $\mathrm{Re}(\beta)$ being expressed via the definite microparameters] the appropriate expression for R coincides with that given above.

Equations (16)–(18) become considerably simpler for the particular ABC's:

$$\text{(i)}\quad n'_{eff}=\frac{n_1n_2+\varepsilon_0}{n_1+n_2},\quad n''_{eff}=0, \tag{21}$$

$$\text{(ii)}\quad n'_{eff}=\frac{n_1n_2(n_1+n_2)}{l},\quad n''_{eff}=0, \tag{22}$$

where

$$l\equiv n_1^2+n_1n_2+n_2^2-\varepsilon_0, \tag{23}$$

and

$$\text{(iii)}\quad n'_{eff}=\frac{n_1n_2+\varepsilon_0+i\alpha k_0 n_1n_2(n_1+n_2)}{n_1+n_2+i\alpha k_0 l}, \tag{24}$$

$$n''_{eff}=0 \text{ at } \mathrm{Im}(\alpha)=0 \text{ and } n''_{eff}=\mathrm{Im}(\alpha)k_0 b|n_1+n_2+i\alpha k_0 l|^{-2} \text{ at } \mathrm{Im}(\alpha)\neq 0. \tag{25}$$

In the DA, α is set by Eq. (10); for more general case see Eq. (11). One can see from Eqs. (13) and (21)–(25) that, at $j=0$ (i.e., $n''_{eff}=0$), the principle of conservation of energy is fulfilled only at some values of the parameter U in expression (11), in particular at $U=1$ [case (i)] and $U=-1$ [case (ii)]. However, it is not fulfilled for the continuum region of $|U|<1$, in particular for a DA that corresponds to $U=0$. Just this situation has been corrected in our paper.

(iv) In the ordinary situation $\mathrm{Im}(\beta)=0$,

$$n'_{eff}=\frac{n_1n_2+\varepsilon_0-4\pi\beta}{n_1+n_2},\ n''_{eff}=-4\pi\beta\,\mathrm{Re}\left(\frac{1-4\pi\beta l/b}{n_1+n_2}\right). \tag{26}$$

Above [see Eq. (15)], we presented n_{eff} as a sum of two terms so that one could compare easily the results obtained here with those obtained in a number of other theoretical works where it was set $j=0$—i.e., $n''_{eff}\equiv 0$—and fulfillment of the principle of conservation of energy has been lost.

It is traditional practice in physics of ALW's to compare the experimental results with theoretical calculations that use

different ABC's—from (i) to (iv). In most of earlier works only the term n'_{eff} was taken into account in Eq. (15). From our results it follows, however, that to satisfy the principle of conservation of polaritonic energy one should take into account both terms (16) and (17) in the corrected Equation (15) for n_{eff}. Of course, only experiments can give a complete quantitative estimation of the generalizations made in our paper. Tentative estimations made on the basis of the experimental data known from the literature show that modifications of the formulas for PR calculations in the excitonic resonance region are not insignificant. This is not surprising, because it is well known that calculation of PR depends essentially on the boundary conditions chosen.

Some other semiphenomenological parameters, such as the Hopfield dead layer thickness d,[12] are introduced also when performing calculations in the physics of ALW's within the above approximations. The parameter d can be taken into account easily also within the theory considered here. To do this, one should replace n_{eff} in Eq. (14) by $n_{eff,d}$:

$$n_{eff,d} = n\frac{(n_{eff}+n)e^{-2ik_0nd} - n + n_{eff}}{(n_{eff}+n)e^{-2ik_0nd} + n + n_{eff}}, \tag{27}$$

where $n=\sqrt{\varepsilon_0}$.

In conclusion we would like to outline once more some items that are of importance for understanding the paper. Both the formulation and coordination of the Maxwell boundary conditions and those additional to them is one of the crucial problems in SD crystal optics. The program of solving this problem that is stated in our work rests on mandatory observance of the fundamental principles when performing calculations, whatever the values of the expected results. This program regards equally the calculations on the basis of micromodels and those using the continual approximation. In the latter case the above program was realized in our paper when dealing with a certain class of polarization oscillations in a medium.

We have considered here the simplest situation in physics of ALW's and have made only a small number of references to the works where this situation was realized in practice (in addition to those, see the proper references in Refs. 2–4, etc.). Obviously, the results obtained here require further generalization when considering more complicated configurations of the "light-crystal" system for which the parameters α and β in Eq. (6) are tensors, and/or an oblique incidence of light is considered, and/or light polarization rotates at reflection, etc. In other words, the situation becomes more complicated when different limiting ABC's for different polarization vector projections are mixed in a nontrivial way (see, e.g., Ref. 8). Such a generalization is required also when considering close or degenerate excitonic states[32] and excitons of different multiplicity,[36] as well as when choosing an alternative basis for calculation of polariton states.[6] It should be noted once more that all our calculations have been done in the EEM approximation. In those exciton models which are beyond the framework of this approximation, the above equations [including ABC (6) and the expression (13) for the Poynting-Pekar vector] may undergo substantial alterations; see, in particular, Ref. 7 dealing with the N-exponential exciton model.

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(the so-called "*k* method"). In such a case the parameter β in Eq. (12) is determined from Eq. (91) in Ref. 5.

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