Conservation laws in electrically polarizable spatially dispersive media. I. Dynamic equations

1991 J. Phys.: Condens. Matter 3 2545

(http://iopscience.iop.org/0953-8984/3/15/010)

J. Phys.: Condens. Matter 3 (1991) 2545–2559. Printed in the UK

Conservation laws in electrically polarizable spatially dispersive media: I. Dynamic equations

V G Bar'yakhtar, B I Khudik and I I Obozhin
Institute of Metal Physics, Ukrainian Academy of Sciences, 252680, Kiev-142, USSR

Received 3 January 1989, in final form 16 October 1990

Abstract. The problem of the additional boundary condition in macroscopic crystal optics is considered from the standpoint of conservation laws. The possibility of obtaining this condition in the form of a continuity condition at the interface between two media for macroscopic flux density of the internal angular momentum is substantiated. As is shown, to solve this problem one must describe a medium with excitonic polarization within the framework of a general approach, where mechanical vibrational, polarization and electromagnetic degrees of crystal freedom are considered together. It is shown that, to take the spatial dispersion into account correctly, one must consider the dependence of the crystal energy not only on the first-order space derivatives of polarization but also on those of second order. The consideration of conservation laws and the derivation of the additional boundary condition will be given in part II of this work.

1. Introduction

1.1. Preliminaries

Since the time when Pekar (1957) pointed out the necessity for specifying the additional boundary condition (ABC) in the electrodynamics of spatially dispersive (SD) media, this ABC has been the subject of numerous investigations (see the bibliography in the collective monograph edited by Rashba and Sturge (1982)). Nevertheless, the understanding of the problem seems to be far from perfect. Unquestionable progress has been made in understanding what specific forms the ABC may take. Meanwhile, insight into the physical reasons why the ABC assumes just these forms is still largely illusory.

As far as we can judge from available publications, the most recognized by investigators is the microscopic approach to the ABC problem, where to obtain the ABC they attempt to find a self-consistent solution of Maxwell and Schrödinger equations. This approach is certainly the most precise one, but high accuracy, being an advantage of the microscopic approach, has nevertheless been achieved at the cost of, in our opinion, unnecessary particularity of the ABC problem statement. This in turn has led to a considerable spread in results obtained by different authors but apparently has not revealed the physical idea that lies behind the ABC.

In this sense the possibility of resolving the ABC problem within the framework of a macroscopic phenomenological approach, where the state of the medium is described in terms of macroscopically continuous field variables, gains prime importance. As is known, on treating a problem in this way one makes the problem statement more fine; but in return one makes it more general and profits from the universality of the final results. Unfortunately the macroscopic treatment meets a difficulty that is insurmountable from our point of view without going beyond the scope of the traditional ABC problem formulation.

1.2. The essence of the problem

In standard interpretation the ABC problem is reduced to the question of how to join two macro-asymptotic solutions, polaritons and electromagnetic waves, at the interface of two media, viz. an SD medium and one for which spatial dispersion effects are negligible. As the former, some crystal is considered, provided that the frequency of the waves is close to an exciton resonance line for a given crystal. As the latter, either the vacuum or an exciton-free layer of the same crystal, where the wave frequency does not fall within the resonant region, is chosen.

The main point of the difficulty is that at the interface some transition layer occurs within which polariton solutions go over to those in the form of electromagnetic waves. Since the exciton resonant region as a rule is extremely narrow ($\Delta\omega/\omega_0 = 10^{-3}$), the width of this transition layer may prove to be small enough to become of the same order of magnitude as the effective range parameter in the short-range interaction responsible for the spatial dispersion. If that is the case, the transition layer, unlike the rest of the crystal, may not be described in terms of macroscopic field variables only, so long as there are no grounds for regarding the exciton polarization $p^{\mathrm{ex}}(x)$ to be a *sufficiently* smooth function of coordinates x as Agranovich and Ginzburg (1966) believed or for regarding the wavevector k to be small *everywhere* as assumed by Skettrup (1973).

Thus, the fact that within the transition layer the function $p^{\mathrm{ex}}(x)$ may prove to be insufficiently smooth should be taken into account, thereby bringing into question the validity of reducing the non-local expression for stored energy within the transition layer to the local form by expanding this expression in a power series of weak polarization gradient; though a reduction like this is usual when one treats the ABC problem within the scope of a traditional macroscopic approach interpretation.

If, all the same, on describing the transition layer one employs formally this reduction in the way Agranovich and Ginzburg (1966) did, the series coefficients will be indeterminate quantities; and it is exclusively the ABC that, if it were at hand, could provide the required information on these coefficients.

In order to avoid confusion, it is appropriate to note that our interpretation of the ABC problem is distinct from the one proposed by Birman and Sein (1972), Birman (1982) and Birman and Zeyher (1974) or, say, by Halevi and Fuchs (1984). In their works, to obtain the ABC they proceeded from the fact that the reflectivity for excitons is either assumed to be *given* as a result of a microscopic consideration or thought to be an *arbitrary* phenomenological parameter, which is equivalent to postulating the boundary condition for excitons. An analogous approach has been developed for the case of conducting media in the works by Garcia-Moliner and Flores (1977), Mukhopadhyay (1978) and Forstmann *et al* (1978), where the reflectivity for plasmons was introduced through a parameter. Since according to Pekar (1983) and Zeyher *et al* (1972) the form of the ABC depends in a unique fashion on the reflectivity for excitons, to resolve the ABC problem within the framework of the macroscopic approach must imply, from our point of view, finding this reflectivity from first principles by means of macroscopic methods only.

1.3. Basic principles of our approach

To overcome the difficulty mentioned above without resort to the help of microscopic considerations, we should combine the field variables into a quantity that is macroscopically continuous at the interface. As is known, the flux density of every constant of motion is an example of such a continuous quantity. When the problem of boundary conditions (BC) is considered in mechanics and electrodynamics of continuous media, making use of the conservation laws of linear momentum and of energy is usual. As regards the ABC problem, taking these laws into account is necessary too. Particularly, energy conservation, in accord with Bishop and Maradudin (1976) and Selkin (1977), imposes limitations on the form of the ABC. Unfortunately, making use of only these laws would not resolve the ABC problem for the case of SD media with excitonic polarization.

To be fair we must note that Forstmann (1979) attempted to solve the ABC problem for the case of an excitonic dielectric by proceeding from energy conservation only. Particularly, Forstmann split up the condition of energy flux continuity at the surface into some separate conditions. This approach has already been criticized by Boardman (1982). In our opinion Forstmann's method seems to add nothing new in comparison with the approach proposed by Halevi and Fuchs (1984) as long as exciton reflectivity remains an indeterminate parameter. As a matter of fact, Fortsmann removed this indeterminacy in his work by imposing restrictions on BC for the electromagnetic field, which is certainly equivalent to *postulating* the ABC. We shall return to this question in part II of this work with our ABC being at hand.

Therefore we shall turn our attention to a study of the law of angular momentum (AM) conservation, which, as a rule, was assumed to be beyond the scope of the ABC problem.

More than likely the latter circumstance may be attributed to the fact that, when they considered the ABC problem, the investigators used, consciously or otherwise, the notion originating from the mechanics of *unpolarized* media, where the AM, being generated by the motion of the centres of mass of the crystal unit cells only, contains exclusively an orbital component of the total AM. The point is that in such a case the condition of AM flux density continuity (AM FDC) at the interface may readily be reduced to an analogous condition for linear momentum with the former bringing about no independent BC.

According to de Groot and Suttorp (1972) and also Lax and Nelson (1976a, b), when electrical polarization is the case, an additional contribution to the total AM arises. This contribution is generated by the motions within the unit cells and is called internal angular momentum (IAM).

The main point of the foregoing situation is that the condition of total AM FDC is reducible in the light of the above-stated reasoning to the condition of IAM FDC with the latter not being a simple identity if spatial dispersion occurs.

The presence of spatial dispersion is of paramount importance for the IAM FDC condition to be an independent BC, because otherwise the IAM is constrained to move with the material, and IAM transfer across the transition layer is impossible. That is why, for example, being a corollary of the AM conservation law in the specific case of a non-dispersive medium, the BC (7.6) obtained in Lax and Nelson (1976b) does not contain any additional information in comparison with some other BC derived in the same work.

According to Lam (1977a, b) and Lam and Lax (1978), in the case of an SD medium the IAM may be transferred through the crystal, and consequently we may infer that the condition of the IAM FDC across the transition layer becomes an *independent* BC, which, being expressed in terms of $\boldsymbol{p}^{\mathrm{ex}}$, may be used as the ABC.

The foregoing reasoning is the basis for the novel approach to the ABC problem, and the aim of our work is to obtain the ABC in this way.

1.4. Choosing a formalism

In spite of the fact that the conservation laws of linear momentum and of energy do not immediately bring about the ABC, they are of great importance when one considers the problem in question. Therefore it is necessary to obtain not only the expression for the IAM flux density but also analogous expressions for linear momentum and for energy.

Note that the situation in question is distinct from those considered earlier. First, it is necessary to take into account the polarization excitations of the crystal and the ones in the form of electromagnetic waves. Secondly, to study the conservation law of AM we must use the energy functional, which is invariant under uniform space rotations. And thirdly, as is known from the works of Lax and Nelson (1976a) and Bar'yakhtar and Turov (1988), this rotational invariance implies that the energy of the anisotropic polarized medium depends on the tensor of local rotations of the material $\hat{\boldsymbol{R}}$. Since the latter is expressed in terms of elastic displacement gradients $\partial u_i/\partial x_j$, we must take elastic displacements $\boldsymbol{u}$ into account equally with other degrees of freedom.

Thus, to solve the ABC problem in the way proposed above it is necessary to analyse (for the case of SD media with excitonic polarization) the conservation laws of linear momentum, of energy and of AM within the scope of the general macroscopic approach where mechanical vibrational, polarization and electromagnetic excitations of a crystal are considered together, because they affect each other.

So far as we know, the case of the SD medium with excitonic polarization has not yet been analysed within the scope of such a general approach. In particular, Lax and Nelson (1976a, b) did not take the spatial dispersion into account; while in works by Lam (1977a, b) and Lam and Lax (1978) the electromagnetic field in the SD medium was not considered. Finally, in the works by Agranovich and Ginzburg (1966), Bishop and Maradudin (1976), Selkin (1977), Forstmann (1979) and Tilley (1980), where the energy conservation law was considered, elastic displacements and hence rotational invariance were not taken into account at all.

In the case when dissipative processes are not taken into account, the medium may be described within the scope of the Lagrange theory. In our opinion, the use of such a model approximation in the present work is reasonable. First, according to Pekar (1983) the influence of dissipative processes on the phenomena with which the ABC problem is concerned is not cardinal. On many occasions the dissipation contribution to these phenomena proves to be not a factor at all. On the other hand, the omission of the dissipation will enable us to simplify the presentation and to emphasize the physical ideas behind our approach. And finally, if we use the Lagrange theory, we shall be able on studying the conservation laws to take advantage of applying the standard variational analysis procedure based on the use of the Nöether theorem (see e.g. the monograph by Bogolubov and Shirkov (1980)). One of the disadvantages of this approximation is that we shall not be able to apply our findings freely to the case of conducting (metallic) media. Although plasmon modes might be easily described by introducing some exciton-like polarization vector $\boldsymbol{p}^{\langle c\rangle}$, with charge density $\rho^{\langle c\rangle}$ and current $\boldsymbol{j}^{\langle c\rangle}$ for conduction electrons being expressed as

$$\rho^{\langle c\rangle} = \operatorname{div}(\boldsymbol{p}^{\langle c\rangle}) \qquad \boldsymbol{j}^{\langle c\rangle} = \partial \boldsymbol{p}^{\langle c\rangle}/\partial t$$

nevertheless the realm of application for such a theory would be substantially restricted as long as we confine ourselves to consideration of media with negligible energy dissipation only.

The other important question is concerned with making a choice between the material frame of reference and the spatial one. As for the case when the spatial dispersion is under study with the electromagnetic field being beyond the scope of consideration, the most convenient frame, according to works by Lam (1977a, b) and Lam and Lax (1978), is the material one.

But since to take the spatial dispersion and the electromagnetic field into account simultaneously is one of the specific features of this work, it is reasonable to use here the spatial frame from the very beginning.

At last, we shall not consider surface roughness effects, so translational invariance will be assumed to hold for any direction parallel to the crystal surface.

1.5. Organization of the paper

Briefly, the organization of our work is as follows. In section 2 the Lagrangian for the SD medium with excitonic polarization is built out of the field variables. To make this Lagrangian invariant under displacements and rotations, we apply there the formalism presented by Lax and Nelson (1971a, b, 1976a) and by Bar'yakhtar and Turov (1988). In section 3 by means of the standard variational procedure the equations of motion are derived from the above-mentioned Lagrangian. The obtained equations are analysed in section 4.

The consideration of conservation laws, of the AM conservation law in particular (being, in our opinion, of interest in itself), is postponed by us to part II of the work (Bar'yakhtar *et al* 1991). In the same paper the derivation of the ABC will be given.

2. Invariant Lagrangian

Let us now determine the field variables, keeping in mind that we must take electromagnetic and internal degrees of freedom into account equally with the elastic displacement $\boldsymbol{u}$,

$$\boldsymbol{u} = \boldsymbol{x} - \boldsymbol{X}. \tag{1}$$

Here $\boldsymbol{X}$ is the material coordinate vector of the centre of mass of a unit cell. It rides with and names this unit cell. Here, too, $\boldsymbol{x}$ is the spatial coordinate vector of the same centre of mass. Throughout this work both the material and the spatial frames are supposed to be Cartesian. The electromagnetic excitations will be described by potentials $\boldsymbol{A}$ and Φ. Internal degrees of freedom may be described by introducing a number of internal coordinates in the same way as was done, for example, by Lax and Nelson (1976a, b). However, since in this work we are interested only in the effects that are concerned with the ABC problem, it is reasonable to confine ourselves to taking into consideration those internal variables which are associated with the polarization of the medium. Therefore as internal variables we use the set of polarization excitations in the form of vector field functions $\boldsymbol{p}^{(\mu)}$ ($\mu = 1, \ldots, N$) where $\boldsymbol{p}^{(\mu)}$ are electric dipole moments per unit mass.

Since, below, the stored energy is supposed to be a quadratic polynomial in the quantities $\boldsymbol{p}^{(\mu)}$, this expression may be reduced to a diagonal form; and, consequently, it is convenient to use as internal variables the set of normal polarization modes of the crystal. This set must include the modes associated with the background polarization (i.e. those for which spatial dispersion effects are not a factor at a given frequency) as well as the resonant exciton modes. In other words, we shall describe the SD medium within the scope of the 'N isolated exciton absorption lines' approximation.

However, if one takes into account the fact that normal modes are those whose contributions to the stored energy assume the form of a sum of independent terms, one will come to the conclusion that, if we subjected the Lagrangian to any variational transformation, the contribution of each of these modes to the resultant expression would be formally analogous with the contribution of every other normal mode. That is why, henceforth, for short, we shall consider explicitly only one polarization variable $\boldsymbol{p}$. To generalize this consideration to the case of the N isolated exciton lines approximation, it is sufficient in the resultant expression to sum each term dependent on the polarization vector $\boldsymbol{p}$ over all exciton modes,

$$\ldots + f(\boldsymbol{p}) + \ldots \rightarrow \ldots + \sum_{\mu=1}^{N} f(\boldsymbol{p}^{(\mu)}) + \ldots. \tag{2}$$

Let us now turn to constructing the invariant Lagrangian. To possess the invariant property under space and time displacements, the Lagrangian density function $\mathscr{L}$ must be independent directly of spatial coordinates $\boldsymbol{x}$ and of time t, with these quantities appearing in the expression for $\mathscr{L}$ only indirectly as the arguments of field variables and via the following combination that itself remains unchanged under displacements:

$$\boldsymbol{X}(\boldsymbol{x}, t) = \boldsymbol{x} - \boldsymbol{u}(\boldsymbol{x}, t). \tag{3}$$

Moreover, since this invariance is understood to be under uniform displacements, $\mathscr{L}$ may depend also on the substantive, or material, time derivative $\mathrm{d}\boldsymbol{x}/\mathrm{d}t$ (i.e. one which holds $\boldsymbol{X}$ fixed) and on the derivative of $\boldsymbol{x}$ with respect to the material coordinates $\boldsymbol{X}$.

To possess the invariant property under uniform space rotations, function $\mathscr{L}$ should be built out of the individually rotationally invariant quantities composed of the field variables and of their space and time derivatives. It is conventional to use for these derivatives a compact notation like that given below:

$$\begin{aligned} p_{i,jk} &= \partial^2 p_i/\partial x_j \partial x_k \qquad & x_{i,\mathrm{A}} &= \partial x_i/\partial X_\mathrm{A} \\ \mathrm{d}_t p_i &= \mathrm{d}p_i/\mathrm{d}t \qquad & \partial_t p_i &= (\partial p_i/\partial t)|_{\text{fixed}\,\boldsymbol{x}} \end{aligned} \tag{4}$$

where $\mathrm{d}/\mathrm{d}t$ represents the material time derivative and commas denote space differentiation.

Note that spatial coordinates x_i change under space rotations, but material coordinates X_A, being the 'names' of the crystal unit cells, do not. Therefore properties of some quantity under space rotations depend on which frame, material or spatial, each of the quantity's subscripts is attributed to. In order to facilitate the distinction between the material and the spatial frames of reference we shall use, wherever necessary, upper-case subscripts to denote components in the material coordinate system and lower-case subscripts to denote components in the spatial coordinate system (see, for example, formulae (4)). It should be noted that a distinction like this is necessary only for the cases when the properties of some quantity under space rotations are not obvious.

The total Lagrangian density $\mathscr{L}$ can be decomposed into three terms as

$$\mathscr{L} = \mathscr{L}_{\mathrm{F}} + \mathscr{L}_{\mathrm{I}} + \mathscr{L}_{\mathrm{M}}. \tag{5}$$

(i) The first term is an electromagnetic field Lagrangian density of the form

$$\mathscr{L}_{\mathrm{F}}(\boldsymbol{x}, t) = [\mathscr{E}^2(\boldsymbol{x}, t) - \mathscr{B}^2(\boldsymbol{x}, t)]/8\pi \tag{6}$$

with $\mathscr{E}$ and $\mathscr{B}$ being the total electric field strength and the total magnetic induction, respectively,

$$\mathscr{E} = \boldsymbol{E} + \mathfrak{E} \qquad \mathscr{B} = \boldsymbol{B} + \mathfrak{B}. \tag{7}$$

Here $\boldsymbol{E}$ and $\boldsymbol{B}$ are electric and magnetic fields generated by the material body itself, but $\mathfrak{E}$ and $\mathfrak{B}$ are those generated by the external sources. These sources henceforth will be supposed to be located far beyond the material body. Vector quantities $\boldsymbol{E}$ and $\boldsymbol{B}$ are expressed conventionally in terms of the vector and scalar potentials by

$$\boldsymbol{E} = -\nabla\Phi - (1/c)\partial_t \boldsymbol{A} \tag{8}$$

$$\boldsymbol{B} = \nabla \times \boldsymbol{A} \tag{9}$$

where $\nabla_i\Phi = \partial\Phi/\partial x_i$, and c is the speed of light in vacuum.

(ii) The second term is a field–matter interaction Lagrangian density of the form

$$\mathscr{L}_{\mathrm{I}}(\boldsymbol{x}, t) = \rho(\boldsymbol{x}, t)[\boldsymbol{p}(\boldsymbol{x}, t) \cdot \mathscr{E}(\boldsymbol{x}, t) + \boldsymbol{m}(\boldsymbol{x}, t) \cdot \mathscr{B}(\boldsymbol{x}, t)]. \tag{10}$$

Here $\boldsymbol{p}$ and $\boldsymbol{m}$ are, respectively, the electric and the magnetic dipole moments per unit mass. The mass density ρ for a strained crystal is related to that for an unstrained crystal ρ_0 by

$$\rho(\boldsymbol{x}, t) = \rho_0(\boldsymbol{x} - \boldsymbol{u}(\boldsymbol{x}, t)) \cdot |J| \tag{11}$$

where the Jacobian J of the transformation from $\boldsymbol{x}$ to $\boldsymbol{X}$ is given by

$$J(\boldsymbol{x}, t) = \det X_{\mathrm{A},i}. \tag{12}$$

In the formula (10) the free charge density is set to zero and the corresponding current is dropped since a dielectric is being considered. The necessity for taking the magnetization into consideration may be explained as follows. Despite the fact that in this work the crystal in question is understood to be non-magnetic, the effective magnetic moment arises from the motion of the polarization. Using Minkowski's relations we may approximate $\boldsymbol{m}$ by

$$\boldsymbol{m} = [\boldsymbol{p} \times (\boldsymbol{v}/c)]. \tag{13}$$

Here $\boldsymbol{v} = \mathrm{d}_t\boldsymbol{x}$ is the velocity of the centre of mass of a unit cell.

When one considers the phenomena with which ordinary crystal optics deals, it seems that the magnetization may be ignored in most cases. Nevertheless, it is reasonable in this work to take the vector $\boldsymbol{m}$ of the form (13) into account, because in this case, as will be shown below, the so-called Abraham force term arises in the equation of motion for elastic displacements. This in turn will enable us, when we shall study the conservation law of linear momentum, to avoid certain confusion in making a choice between the expression for linear momentum density in Abraham's form and that in Minkowski's form. (One can find the details of this problem and a bibliography, for example, in the monograph by Møller (1972).)

(iii) And finally, the third term is a matter Lagrangian density $\mathcal{L}_\mathrm{M}$ which may be expressed as follows:

$$\mathcal{L}_\mathrm{M}(x,t) = \rho(x,t)\{\tfrac{1}{2}[v^2(x,t) + (\mathrm{d}_t p)^*_\mathrm{A} \hat{f}^{-1}_\mathrm{AB} (\mathrm{d}_t p)^*_\mathrm{B}] \\ - \Sigma((p)^*; (\nabla p)^{**}; (\nabla(\nabla p))^{***}; \hat{E}; X(x,t))\}. \tag{14}$$

The first and second terms on the right-hand side of (14) correspond, respectively, to the mechanical kinetic energy and to the polarization one. The symbol $\hat{f}$ denotes a certain phenomenological tensor. The quantity Σ corresponds to the stored energy of the crystal per unit mass.

Throughout this work summation over repeated subscripts is implied.

The function $\mathcal{L}_\mathrm{M}$ is expressed in terms of individually rotationally invariant quantities, among which the following ones should be emphasized: (1) The quantity

$$E_\mathrm{AB} = \tfrac{1}{2}(x_{i,\mathrm{A}} x_{i,\mathrm{B}} - \delta_\mathrm{AB}) \tag{15}$$

is the finite strain tensor which describes the changes of the distances in the matter under deformation. In equation (15) and below, $\hat{\delta}$ is the Kronecker delta. (2) A number of invariant quantities related to the polarization vector are

$$(p)^*_\mathrm{A} = p_i R_{i\mathrm{A}} \tag{16a}$$

$$(\mathrm{d}_t p)^*_\mathrm{A} = \mathrm{d}_t p_i R_{i\mathrm{A}} \tag{16b}$$

$$(\nabla p)^{**}_\mathrm{AB} = p_{i,k} R_{i\mathrm{A}} R_{k\mathrm{B}} \tag{16c}$$

$$(\nabla(\nabla p))^{***}_\mathrm{ABC} = p_{i,kl} R_{i\mathrm{A}} R_{k\mathrm{B}} R_{l\mathrm{C}} \tag{16d}$$

where $R_{i\mathrm{A}}$ is the rotation tensor

$$R_{i\mathrm{A}} = x_{i,\mathrm{B}} (\hat{\delta} + 2\hat{E})^{-1/2}_\mathrm{BA}. \tag{17}$$

In relations (16) we use the notation from the works by Bar'yakhtar and Turov (1988) and Bar'yakhtar *et al* (1987).

Note that it is no mere chance that here the quantity (16*d*) dependent on the second-order space derivative of p is involved in the description of the SD medium. Indeed, when one considers the processes of light wave propagation through the medium, one conventionally expresses the energy of the system as a power series in small parameter $k\alpha$ up to the second-power terms, where k and α denote the wavevector magnitude and the effective range parameter in short-range interaction, respectively. Therefore both the term of the form

$$\tfrac{1}{2}(\nabla p)^{**}_\mathrm{AB} B^{(0)}_\mathrm{ACBD}(X(x,t))(\nabla p)^{**}_\mathrm{CD} \tag{18}$$

and that of the form

$$-\tfrac{1}{2}(p)^*_\mathrm{A} B^{(1)}_\mathrm{ACBD}(X(x,t))(\nabla(\nabla p))^{***}_\mathrm{CBD} \tag{19}$$

should be taken into account because they are of the same order of magnitude. In expressions (18) and (19) the symbols $B^{(0)}$ and $B^{(1)}$ denote certain phenomenological tensors.

Nevertheless, within the scope of ordinary crystal optics, where investigators disregard the rotational invariance and set $\hat{R}$ to be the identity matrix, the term (19) is excluded from consideration because of the equivalence between the contributions of the terms (18) and (19) to the equation of motion for $\boldsymbol{p}$. (The term (19) is thought to be reducible to the form (18) by adding to the Lagrangian density function the proper divergence-like term.)

When non-uniform rotations of the matter occur, the above-mentioned transformation of the Lagrangian becomes unsuitable and the situation changes dramatically. Between the terms (18) and (19) there arises an irreducible difference dependent on space derivatives of the rotation tensor. In other words, the terms mentioned above describe the interaction between the polarization and the non-uniform rotations in essentially different ways. This fact will be important later when we shall obtain the expression for the IAM flux density, because the contributions of the terms (18) and (19) to this quantity cannot be reduced to one another.

Note that instead of a number of invariant quantities of the form (16) one may choose the following alternative:

$$(\boldsymbol{p})^{*}_{\mathrm{A}} = p_i x_{i,\mathrm{A}} \tag{20a}$$

$$(\mathrm{d}_t \boldsymbol{p})^{*}_{\mathrm{A}} = \mathrm{d}_t p_i x_{i,\mathrm{A}} \tag{20b}$$

$$(\nabla \boldsymbol{p})^{**}_{\mathrm{AB}} = p_{i,k} x_{i,\mathrm{A}} x_{k,\mathrm{B}} \tag{20c}$$

$$(\nabla(\nabla \boldsymbol{p}))^{***}_{\mathrm{ABC}} = p_{i,kl} x_{i,\mathrm{A}} x_{k,\mathrm{B}} x_{l,\mathrm{C}}. \tag{20d}$$

The notation (20) corresponds to the formalism presented, for example, in the work by Lam and Lax (1978), though it seems that the importance of the term (20*d*) was not recognized by them.

As is seen from equation (17), two methods based on the sets (16) and (20) differ only in the method of description of the interaction between the polarization $\boldsymbol{p}$ and the crystal deformation $\hat{E}$. Since we do not intend to use any specific expression for this interaction, the necessity to distinguish between the above-mentioned sets does not arise.

3. Equations of motion

We shall begin by writing the Lagrangian equation of motion for the generalized coordinates $\boldsymbol{u}, \boldsymbol{p}, \boldsymbol{A}$ and Φ:

$$\partial_t \left(\frac{\partial \mathscr{L}}{\partial v_i} x_{i,\mathrm{A}} \right) + \left(\frac{\partial \mathscr{L}}{\partial u_{\mathrm{A},k}} + \frac{\partial \mathscr{L}}{\partial v_i} x_{i,\mathrm{A}} v_k \right)_{,k} - \frac{\partial \mathscr{L}}{\partial u_{\mathrm{A}}} = 0 \tag{21}$$

$$\rho \mathrm{d}_t \left(\frac{1}{\rho} \frac{\partial \mathscr{L}}{\partial \mathrm{d}_t p_i} \right) = \frac{\partial \mathscr{L}}{\partial p_i} - \left(\frac{\partial \mathscr{L}}{\partial p_{i,k}} \right)_{,k} + \left(\frac{\partial \mathscr{L}}{\partial p_{i,kl}} \right)_{,kl} \tag{22}$$

$$\partial_t \left(\frac{\partial \mathscr{L}}{\partial(\partial_t A_i)} \right) + \left(\frac{\partial}{\partial A_{i,k}} \right)_{,k} = 0 \tag{23}$$

$$\left(\frac{\partial}{\partial \Phi_{,k}} \right)_{,k} = 0. \tag{24}$$

To derive equation (21) we have used the following relation:

$$\bar{\delta} v_i = x_{i,\mathrm{A}} \mathrm{d}_t (\bar{\delta} u_{\mathrm{A}}) \tag{25}$$

where $\bar{\delta}$ denotes the variation that holds $\boldsymbol{x}$ and t fixed.

Although the deformation and rotations of the matter are expressed in equations (15), (16) and (20) via the quantities $x_{i,\mathrm{A}}$, to make connection with the ordinary theory of linear elasticity we shall treat, on using equation (21), the Lagrangian density function as if it had been expanded in powers of the displacement gradients by means of the following iterations:

$$\frac{\partial x_i}{\partial X_k} = \delta_{ik} + \frac{\partial u_i}{\partial x_k} + \frac{\partial u_i}{\partial x_m}\frac{\partial u_m}{\partial x_k} + \dots . \tag{26}$$

The displacement $\boldsymbol{u}$ does not possess a vector property in either the spatial coordinate system or the material one. Nevertheless, the variation $\bar{\delta} u_{\mathrm{A}}$, as is seen from equation (3), differs from the variation $\bar{\delta} X_{\mathrm{A}}$ only in sign,

$$\bar{\delta} u_{\mathrm{A}} = -\bar{\delta} X_{\mathrm{A}}. \tag{27}$$

Therefore $\bar{\delta} u_{\mathrm{A}}$ itself and the derivatives

$$\partial/\partial u_{\mathrm{A}} = -\partial/\partial X_{\mathrm{A}} \tag{28a}$$

$$\partial/\partial u_{\mathrm{A},k} = -\partial/\partial X_{\mathrm{A},k} \tag{28b}$$

are vector quantities in the material frame.

It is not surprising that Lagrangian equations (23) and (24) may be reduced to Maxwell equations of the form

$$\nabla \times \mathscr{H} = (1/c)\partial_t \mathscr{D} \tag{29a}$$

$$\nabla \cdot \mathscr{D} = 0 \tag{29b}$$

where

$$\mathscr{D} = \mathscr{E} + 4\pi \boldsymbol{P} \qquad \text{with } \boldsymbol{P} = \rho \boldsymbol{p} \tag{30}$$

$$\mathscr{H} = \mathscr{B} - 4\pi \boldsymbol{M} \qquad \text{with } \boldsymbol{M} = \rho \boldsymbol{m}. \tag{31}$$

In equations (29) the absence of terms involving external field sources reflects the assumption that these sources are located beyond the space region of our interest.

The equation of motion (21) for $\boldsymbol{u}$ may be reduced to a more tractable form

$$\rho \mathrm{d}_t v_j + t_{jk,k} - f_j = 0 \tag{32a}$$

with

$$f_j = \rho p_i \mathscr{E}_{i,j} + \rho m_i \mathscr{B}_{i,j} + \partial_t((1/c)[\rho \boldsymbol{p} \times \mathscr{B}]_j) \tag{32b}$$

and

$$t_{jk} = \rho \frac{\partial(\mathscr{L}^{(0)}/\rho)}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} - \rho \frac{v_k}{c}[\boldsymbol{p} \times \mathscr{B}]_j + p_{i,j}\pi_{ik} + 2p_{i,lj}\pi_{ikl} - (p_{i,j}\pi_{ikj})_{,l}. \tag{33}$$

Here we used the following notations:

$$\mathscr{L}^{(0)} = \mathscr{L}_{\mathrm{M}} + \mathscr{L}_{\mathrm{I}} \tag{34}$$

and

$$\pi_{mk} = -\partial \mathscr{L}^{(0)}/\partial p_{m,k} \tag{35a}$$

$$\pi_{mkl} = -\partial \mathscr{L}^{(0)}/\partial p_{m,kl}. \tag{35b}$$

The details of the derivation of equations (32) and (33) are given in appendix 1. The last term in equation (32*b*) corresponds to the Abraham force.

4. Analysis of equations

Let us analyse the obtained equations, directing our attention to the difference of the contributions of the terms (18) and (19) to these equations. Just as we expected, the above-mentioned contributions to the equation of motion (32) for $\boldsymbol{u}$, unlike those to the equation of motion (22) for $\boldsymbol{p}$, do not tend to be equivalent even in the limiting case when $u \rightarrow 0$ dealt with by ordinary crystal optics. In order to prove this statement we must express explicitly the dependence of the right-hand side of (33) on space derivatives of $\boldsymbol{p}$ since the first term

$$\rho \frac{\partial(\mathscr{L}^{(0)}/\rho)}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} \tag{36}$$

depends on the aforementioned quantities in an indirect way.

Complete presentation of the calculations is clumsy. But, fortunately, henceforth we shall need an explicit expression only for the antisymmetric portion of the quantity (36), because it is this portion that is involved in the condition of rotational invariance of the Lagrangian and hence in the IAM balance equation. The above-mentioned expression is as follows:

$$\begin{aligned}\varepsilon_{ijk}\rho \frac{\partial(\mathscr{L}^{(0)}/\rho)}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} = &-\varepsilon_{ijk}(\gamma_k \mathrm{d}_t p_j + p_j \pi_k + p_{j,m}\pi_{km} \\ &+ p_{m,j}\pi_{mk} + p_{j,lm}\pi_{klm} + 2p_{m,lj}\pi_{mkl}).\end{aligned} \tag{37}$$

Here we used the notation

$$\gamma_k = -\,\partial\mathscr{L}^{(0)}/\partial(\mathrm{d}_t p_k) \tag{38}$$

$$\pi_k = -\,\partial\mathscr{L}_{\mathrm{M}}/\partial p_k. \tag{39}$$

In appendix 2 we give a rigorous proof of the relation (37) for the case when $\mathscr{L}_{\mathrm{M}}$ is defined by expression (14).

Substituting equation (37) into equation (33) and using the antisymmetrization procedure we obtain

$$\begin{aligned}\varepsilon_{ijk} t_{jk} = &-\varepsilon_{ijk}(\rho(v_k/c)[\boldsymbol{p} \times \mathscr{B}]_j + \gamma_k(\mathrm{d}_t p_j) \\ &+ p_j\pi_k + p_{j,m}\pi_{km} + p_{j,lm}\pi_{klm} + (p_{m,j}\pi_{mkl})_{,l})\end{aligned} \tag{40}$$

where the spatial dispersion contribution (i.e. dependence on the quantities π_{km} and π_{klm}) is seen clearly.

Owing to such an explicit presentation the difference between the contributions of the terms (18) and (19) to the equation of motion for elastic displacements turns out to be an obvious fact. It should be no surprise that this difference remains when $u \rightarrow 0$, for the left-hand side of equation (32*a*) is the functional derivative of $\mathscr{L}$ (with respect to $\boldsymbol{u}$), which is known to be non-vanishing even for the case of infinitesimal amplitudes of mechanical vibrations. The foregoing statement will be important later in derivation of the expression for IAM flux density.

In conclusion, let us compare our results with those obtained earlier. As was to be expected, equations (32), (33) and (40) are the generalization of corresponding relations given by Lax and Nelson (1976a, b) and Lam and Lax (1978), and are consistent with these relations in the limiting cases, respectively, when

$$B^{(0)}_{\mathrm{ABCD}} \equiv 0 \qquad B^{(1)}_{\mathrm{ABCD}} \equiv 0 \qquad f_{\mathrm{AB}} = f\delta_{\mathrm{AB}} \tag{41}$$

and when

$$\mathscr{E} \equiv 0 \qquad \mathscr{B} \equiv 0 \qquad f_{\mathrm{AB}} = f\delta_{\mathrm{AB}}. \tag{42}$$

Appendix 1

Here we derive equations (32) and (33) from equation (21). To begin with, let us note that for the Lagrangian density function $\mathscr{L}$ defined by expressions (5), (6), (10) and (14) the following relation holds:

$$\partial\mathscr{L}/\partial v_i = \rho v_i + (1/c)[\mathscr{B} \times \rho \boldsymbol{p}]_i - \gamma_m p_{m,i}. \tag{A1.1}$$

By using the foregoing relation, the mass conservation law

$$\partial_t \rho + (\rho v_k)_{,k} = 0 \tag{A1.2}$$

and the following identity

$$\mathrm{d}_t(x_{i,\mathrm{A}}) X_{\mathrm{A},j} = v_{i,j} \tag{A1.3}$$

we may rewrite equation (21) as

$$\rho \mathrm{d}_t v_j + \rho(\tfrac{1}{2}v^2)_{,j} - \rho \mathrm{d}_t\left(\frac{1}{\rho}\gamma_m p_{m,l} x_{l,\mathrm{A}}\right) X_{\mathrm{A},j} + \rho \mathrm{d}_t\left(\frac{1}{c}[\mathscr{B} \times \boldsymbol{p}]_l x_{l,\mathrm{A}}\right) X_{\mathrm{A},j}$$
$$+ \left(\frac{\partial \mathscr{L}^{(0)}}{\partial u_{\mathrm{A},k}}\right)_{,k} X_{\mathrm{A},j} - \frac{\partial \mathscr{L}^{(0)}}{\partial u_{\mathrm{A}}} X_{\mathrm{A},j} = 0. \tag{A1.4}$$

The third term on the left-hand side of equation (A1.4) may be rearranged with the use of equations (22) and (A1.3), and also with the use of the expression that the following identity

$$\mathscr{L}^{(0)}_{,j} = \frac{\partial \mathscr{L}^{(0)}}{\partial v_k} v_{k,j} - \gamma_m \mathrm{d}_t(p_{m,j}) - \frac{\partial \mathscr{L}^{(0)}}{\partial u_{\mathrm{A}}} X_{\mathrm{A},j} - \frac{\partial \mathscr{L}^{(0)}}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},kj} + \frac{\partial \mathscr{L}^{(0)}}{\partial p_k} p_{k,j}$$
$$- \pi_{m,k} p_{m,kj} - \pi_{mkl} p_{m,klj} + \rho p_k \mathscr{E}_{k,j} + \rho m_k \mathscr{B}_{k,j} \tag{A1.5}$$

yields for a term

$$\gamma_m \mathrm{d}_t(p_{m,j}).$$

Here equation (28*a*) was used. Substituting the rearranged term in equation (A1.4) and using relation (A1.1) we get, after some algebra, the following equation:

$$\rho \mathrm{d}_t v_j + \mathscr{L}^{(0)}_{,j} + \left(\frac{\partial \mathscr{L}^{(0)}}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j}\right)_{,k} + (\pi_{mk} p_{m,j} + \pi_{mkl} p_{m,lj} - p_{m,j} \pi_{mkl,l})_{,k}$$
$$- \rho p_k \mathscr{E}_{k,j} - \rho m_k \mathscr{B}_{k,j} + \rho \mathrm{d}_t\left(\frac{1}{c}[\mathscr{B} \times \boldsymbol{p}]_j\right) = 0. \tag{A1.6}$$

Keeping in mind that the mass density ρ for a strained crystal is expressed by formulae (11) and (12) and also using the following relation, which is well known in algebra,

$$\partial(\det \hat{\alpha})/\partial\alpha_{ij} = (\det \hat{\alpha})(\hat{\alpha}^{-1})_{ji} \tag{A1.7}$$

we get

$$\partial\rho/\partial u_{\mathrm{A},k} = -\rho x_{k,\mathrm{A}}. \tag{A1.8}$$

Then, from (10) and (14) we find

$$\frac{\partial\mathscr{L}^{(0)}}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} = \rho \frac{\partial(\mathscr{L}^{(0)}/\rho)}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} - \mathscr{L}^{(0)}\delta_{kj}. \tag{A1.9}$$

Finally, substitution of (A1.9) into (A1.6) and some rearrangement gives the equation

$$\begin{aligned} \rho \mathrm{d}_t v_j &+ \left(\rho \frac{\partial(\mathscr{L}^{(0)}/\rho)}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} - \rho\frac{v_k}{c}[p \times \mathscr{B}]_j + p_{m,j}\pi_{mk} + 2p_{m,lj}\pi_{mkl} - (p_{m,j}\pi_{mkl})_{,l}\right)_{,k} \\ &- \rho p_k \mathscr{E}_{k,j} - \rho m_k \mathscr{B}_{k,j} - \partial_t\left(\frac{1}{c}[\rho p \times \mathscr{B}]_j\right) = 0 \end{aligned} \tag{A1.10}$$

which completes the derivation.

Appendix 2

Here we present the proof of the relation (37). To begin with, we shall prove an auxiliary identity:

$$(\partial x_{m,\mathrm{A}}/\partial u_{\mathrm{B},j})X_{\mathrm{B},i} \equiv \delta_{mi} x_{j,\mathrm{A}}. \tag{A2.1}$$

Indeed, keeping in mind that

$$x_{m,\mathrm{A}} X_{\mathrm{A},l} \equiv \delta_{ml} \tag{A2.2}$$

we get

$$\frac{\partial(x_{m,\mathrm{C}}X_{\mathrm{C},l})}{\partial u_{\mathrm{B},j}} = \frac{\partial x_{m,\mathrm{C}}}{\partial u_{\mathrm{B},j}} X_{\mathrm{C},l} - x_{m,\mathrm{C}}\delta_{\mathrm{CB}}\delta_{lj} \equiv 0. \tag{A2.3}$$

Here equation (28*b*) has been used. Multiplying the foregoing equation through by a coefficient $x_{l,\mathrm{A}}X_{\mathrm{B},i}$ we get (A2.1).

Then, by using (A2.1) we can prove that the identity

$$\varepsilon_{ijk}(\partial E_{\mathrm{BC}}/\partial u_{\mathrm{A},k})X_{\mathrm{A},j} \equiv 0 \tag{A2.4}$$

holds for arbitrary B, C and i.

In an analogous way using (A2.1) and (A2.4) and also keeping in mind that the rotation tensor $\hat{\boldsymbol{R}}$ is defined by relation (17) we can prove that the identity

$$\varepsilon_{ijk}\left(\frac{\partial R_{l\mathrm{B}}}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j}\right) = \varepsilon_{ijk}(\delta_{lj}R_{k\mathrm{B}}) \tag{A2.5}$$

holds for arbitrary i, l and B.

Then, using auxiliary equations (A2.1), (A2.4) and (A2.5) we get

$$\varepsilon_{ijk} \frac{\partial(\boldsymbol{p})^{*}_{\mathrm{B}}}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} = \varepsilon_{ijk} p_j R_{k\mathrm{B}} = \varepsilon_{ijk} p_j \frac{\partial(\boldsymbol{p})^{*}_{\mathrm{B}}}{\partial p_k} \tag{A2.6}$$

and

$$\begin{aligned} \varepsilon_{ijk} \frac{\partial(\nabla\boldsymbol{p})^{**}_{\mathrm{BC}}}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} &= \varepsilon_{ijk}(p_{j,m} R_{k\mathrm{B}} R_{m\mathrm{C}} + p_{m,j} R_{m\mathrm{B}} R_{k\mathrm{C}}) \\ &= \varepsilon_{ijk}\left(p_{j,m} \frac{\partial(\nabla\boldsymbol{p})^{**}_{\mathrm{BC}}}{\partial p_{k,m}} + p_{m,j} \frac{\partial(\nabla\boldsymbol{p})^{**}_{\mathrm{BC}}}{\partial p_{m,k}}\right) \end{aligned} \tag{A2.7}$$

where B, C and i are arbitrary.

Arguing by analogy we can prove also that the identities

$$\varepsilon_{ijk} \frac{\partial(\nabla(\nabla\boldsymbol{p}))^{***}_{\mathrm{BCD}}}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} = \varepsilon_{ijk}\left(p_{j,lm} \frac{\partial(\nabla(\nabla\boldsymbol{p}))^{***}_{\mathrm{BCD}}}{\partial p_{k,lm}} + 2p_{m,jl} \frac{\partial(\nabla(\nabla\boldsymbol{p}))^{***}_{\mathrm{BCD}}}{\partial p_{m,kl}}\right) \tag{A2.8}$$

and

$$\varepsilon_{ijk} \frac{\partial(\mathrm{d}_t\boldsymbol{p})^{*}_{\mathrm{B}}}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} = \varepsilon_{ijk} \frac{\partial(\mathrm{d}_t\boldsymbol{p})^{*}_{\mathrm{B}}}{\partial(\mathrm{d}_t p_k)} \mathrm{d}_t p_j \tag{A2.9}$$

hold for arbitrary B, C, D and i.

Using the foregoing relations and keeping in mind that $\mathscr{L}_{\mathrm{M}}$ depends on quantities $u_{\mathrm{A},k}, p_k, p_{k,m}$ and $p_{k,lm}$ only via combinations $\hat{E}$, $(\boldsymbol{p})^*$, $(\nabla\boldsymbol{p})^{**}$, $(\nabla(\nabla\boldsymbol{p}))^{***}$ and $(\mathrm{d}_t\boldsymbol{p})^*$ we get the identity

$$\begin{aligned} \varepsilon_{ijk}\rho \frac{\partial(\mathscr{L}^{(0)}/\rho)}{\partial u_{\mathrm{A},k}} X_{\mathrm{A},j} = -\varepsilon_{ijk}(&\gamma_k \mathrm{d}_t p_j + p_j \pi_k \\ &+ p_{j,m}\pi_{km} + p_{m,j}\pi_{mk} + p_{j,lm}\pi_{klm} + 2p_{m,lj}\pi_{mkl}) \end{aligned} \tag{A2.10}$$

which completes the proof.

References

Agranovich V M and Ginzburg V L 1966 *Spatial Dispersion in Crystal Optics and the Theory of Excitons* (New York: Interscience) ch 3.1, 10.4

Bar'yakhtar V G, Khudik B I and Obozhin I I 1991 *J. Phys.: Condens. Matter* **3** 2561

Bar'yakhtar V G, Obozhin I I and Khudik B I 1987 *Preprint* ITF-87-88R, Institute for Theoretical Physics, Kiev

Bar'yakhtar V G and Turov E A 1988 *Spin Waves and Magnetic Excitations 2* ed A S Borovik-Romanov and S K Sinha (Amsterdam: Elsevier) p 333

Birman J L 1982 *Excitons* ed E I Rashba and M D Sturge (Amsterdam: North-Holland) ch 2

Birman J L and Sein J J 1972 *Phys. Rev.* B **6** 2482

Birman J L and Zeyher R 1974 *Polaritons* ed E Burnstain and F DeMartini (New York: Pergamon)

Bishop M F and Maradudin A A 1976 *Phys. Rev.* B **14** 3384

Boardman A D (ed) 1982 *Electromagnetic Surface Modes* (New York: Wiley)

Bogolubov N N and Shirkov D V 1980 *Introduction to the Theory of Quantized Fields* (New York: Wiley) ch 2

de Groot S R and Suttorp L G 1972 *Foundations of Electrodynamics* (Amsterdam: North-Holland) ch 6

Forstmann F 1979 *Z. Phys.* B **32** 385

Forstmann F, Bagchi A, Rajagopal A K and Stenschke H 1978 *Bull. Am. Phys. Soc.* **23** 278

Garcia-Moliner F and Flores F J 1977 *J. Physique* **38** 851
Halevi P and Fuchs R 1984 *J. Phys. C: Solid State Phys.* **17** 3869
Lam L 1977a *Z. Phys.* B **27** 101
—— 1977b *Z. Phys.* B **27** 349
Lam L and Lax M 1978 *Phys. Fluids* **21** 9
Lax L and Nelson D F 1971a *Phys. Rev.* B **3** 2778
—— 1971b *Phys. Rev.* B **4** 3694
—— 1976a *Phys. Rev.* B **13** 1759
—— 1976b *Phys. Rev.* B **13** 1770
Møller C 1972 *The Theory of Relativity* (Oxford: Clarendon) ch 7.7
Mukhopadhyay G 1978 *Solid State Commun.* **28** 277
Pekar S I 1957 *Zh. Eksp. Teor. Fiz.* **33** 1022
—— 1983 *Crystal Optics and Additional Light Waves* (Menlo Park, CA: Benjamin Cummings) ch 12, 19
Rashba E I and Sturge M D (ed) 1982 *Excitons* (Amsterdam: North-Holland) chs 2 and 4
Selkin A 1977 *Phys. Status Solidi* b **83** 47
Skettrup T 1973 *Phys. Status Solidi* b **60** 695
Tilley D R 1980 *J. Phys. C: Solid State Phys.* **13** 781
Zeyher R, Birman J L and Brening W 1972 *Phys. Rev.* B **6** 4613