Physics – Uspekhi **38** (3) 325 – 332 (1995)

PACS numbers: 41.20–q; 41.90.+e; 78.20.Wc

Boundary conditions for electromagnetic field on the surface of media with weak spatial dispersion

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Contents

Abstract. On the basis of general phenomenological consideration the contradictions which often arise between different approaches to the electrodynamics of bounded media with spatial dispersion are shown to be a result of incorrect assumptions which have been made by its supporters. The important problems in the field of optics of bounded media with spatial dispersion are pointed out. The different methods of solution are discussed.

1. Introduction

The question of boundary conditions on the surface of media with spatial dispersion (SD) has a long history (see, for example, [1 – 5]) and includes two main problems. One of them arises if the incident wave frequency ω is close to an electron resonance. In this case the dispersion equation can have three different roots [1] and for unique solution of the problem the so-called additional boundary conditions are required [1, 4].

In our opinion, a sufficiently logical presentation of the reasons underlying this problem and of principal methods of its solution is given in [1]. We only note that the problem arises in the region of a strong SD. However, even in the case of media with a weak SD, that is when the wavelength λ of the incident light is far longer than the spatial scale of nonlocality d of the optical response of the crystal, very serious and often fundamental disagreements about the form of the constitutive relations and boundary conditions to be used in the electrodynamics of bounded optically active media remain among the investigators [6 – 12].

While in some cases, despite essentially different initial assumptions, the results obtained do not generally contradict each other [7 – 11], in other cases they are completely different [6, 7, 12]. This primarily relates to the question as to whether [6, 12] or not [7 – 11] the polarisation characteristics of light should be changed during its reflection (under the conditions of normal incidence onto the surface) from linear gyrotropic nonmagnetic (i.e. not having spontaneous magnetic moment) media.

We also note that in the majority of the experiments performed 'on reflection' polarisation effects have not been observed under the conditions given above [13, 14]. At the same time, optical activity measurements 'on reflection' (for example, under the condition of total internal reflection [15] or in case of nonlinear media [9, 10]) can provide an effective alternative method of spectroscopy of chiral materials which in a number of cases (for thin layers or strongly absorbing media) is much more convenient than the methods currently used which are based on measuring the polarisation characteristics of transmitted light.

Attempts to discover polarisation effects during light reflection from the surface of high-temperature superconductors [16 – 18] have been amongst the most important topics of recent studies. The presence or absence of polarisation effects during these measurements is directly dependent on the anion superconductivity theory being true or false [18 – 20].

It is easy to understand that the degree of justification of such a point of view largely depends on the answer to the critical question: which boundary conditions and constitutive relations should be used to interpret the contradictory

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Received 17 October 1994
Uspekhi Fizicheskikh Nauk **165** (3) 339–346 (1995)
Translated by K A Postnov; edited by H Milligan

experimental results [13, 14, 16–18, 21, 22] on light reflection from media with nonlocal optical response?

Using the general phenomenological consideration, in the present paper we show that all contradictions which arise between different approaches to the electrodynamics of bounded media with SD are, in fact, a result of certain insufficiently correct assumptions and statements which have been made by its supporters.

In our opinion, the main fallacy is the assumption that the boundary conditions and constitutive relations for electric and magnetic fields can be considered independently of each other [7, 14, 23] and, hence, the 'correct' boundary conditions and 'correct' constitutive relations exist separately. Another widespread error is in the treatment of the influence of the surface polarisation current on the reflection [1, 9, 10, 24–26]. It has been incorrectly taken into account [12] or completely ignored [6].

Below we also present a short review of positive results obtained by different authors, formulate current problems in the optics of bounded (mostly linear) media with SD, and discuss possible methods of their solution.

2. Two basic approaches to electrodynamics of unbounded media with spatial dispersion

2.1 General statements

For the analysis of the interaction of radiation with matter we (in common with all authors) start from the Maxwell system of equations for electromagnetic field in a medium, which directly follows from the traditional averaging of the microscopic equations [24, 27]:

$$\operatorname{curl}\boldsymbol{E} = -\frac{1}{c}\frac{\partial \boldsymbol{B}}{\partial t}\,, \tag{1a}$$

$$\operatorname{curl}\boldsymbol{B} = \frac{1}{c}\frac{\partial \boldsymbol{E}}{\partial t} + \frac{4\pi}{c}\boldsymbol{j}\,, \quad \operatorname{div}\boldsymbol{E} = 4\pi\rho\,, \quad \operatorname{div}\boldsymbol{B} = 0\,, \tag{1b}$$

where $\boldsymbol{j} = \overline{\rho v}$ is the density of current induced in the medium (polarisation current), ρ is the density of bound charge, v is velocity of its motion ($\partial\rho/\partial t + \operatorname{div}\boldsymbol{j} = 0$). Here and below we assume no charge density or density of current from external sources.

We emphasise that a physical sense of the electric field strength $\boldsymbol{E}$ and of magnetic induction $\boldsymbol{B}$ entering into Eqns (1) is uniquely determined by the expression for Lorentz force acting on a test point charge q moving with a velocity $\boldsymbol{u}$ [24]:

$$\boldsymbol{F} = q\left[\boldsymbol{E} + \frac{1}{c}\,(\boldsymbol{u}\times\boldsymbol{B})\right]\,. \tag{2}$$

Obviously, the system of Eqns (1) is not a closed one, as one assumes the ρ and $\boldsymbol{j}$ to be dependent on $\boldsymbol{E}$ and $\boldsymbol{B}$, but the form of this dependence (the so-called constitutive relations) is not used and is not specified here.

2.2 'Symmetric' constitutive relations for electric and magnetic field induction

In the subsequent analysis of the system of Eqns (1) one often represents the current $\boldsymbol{j}$ by a sum of two components:

$$\boldsymbol{j} = \frac{\partial \boldsymbol{P}'}{\partial t} + c\operatorname{curl}\boldsymbol{M}\,, \tag{3}$$

and introduces vectors of electric field induction $\boldsymbol{D}'$ and magnetic field strength $\boldsymbol{H}$:

$$\boldsymbol{D}' = \boldsymbol{E} + 4\pi\boldsymbol{P}'\,, \quad \boldsymbol{H} = \boldsymbol{B} - 4\pi\boldsymbol{M}\,, \tag{4}$$

where the primes are used for clarity in the further presentation†. In that case the Maxwell Eqns (1) take a symmetric form

$$\operatorname{curl}\boldsymbol{E} = -\frac{1}{c}\frac{\partial \boldsymbol{B}}{\partial t}\,, \tag{5a}$$

$$\operatorname{curl}\boldsymbol{H} = \frac{1}{c}\frac{\partial \boldsymbol{D}'}{\partial t}\,, \quad \operatorname{div}\boldsymbol{D}' = 0\,, \quad \operatorname{div}\boldsymbol{B} = 0\,. \tag{5b}$$

The constitutive equations can also be transformed to a symmetric form by using temporal and spatial Fourier components ($k = 2\pi/\lambda$) [1, 28]:

$$\begin{aligned} D'_i(\omega,\boldsymbol{k}) &= \varepsilon'_{ij}(\omega,\boldsymbol{k})\,E_j(\omega,\boldsymbol{k}) + \alpha_{ij}(\omega,\boldsymbol{k})\,H_j(\omega,\boldsymbol{k})\,,\\ B_i(\omega,\boldsymbol{k}) &= \beta_{ij}(\omega,\boldsymbol{k})\,E_j(\omega,\boldsymbol{k}) + \mu_{ij}(\omega,\boldsymbol{k})\,H_j(\omega,\boldsymbol{k})\,. \end{aligned} \tag{6}$$

We note that one often writes down the relations (6) in a somewhat different way: by expressing $\boldsymbol{D}'$ and $\boldsymbol{H}$ in terms of $\boldsymbol{E}$ and $\boldsymbol{B}$ (and then using (5a) only in terms of $\boldsymbol{E}$ [12]) or by expressing $\boldsymbol{D}'$ in terms of $\boldsymbol{E}$ and $\boldsymbol{B}$ in terms of $\boldsymbol{H}$ [8].

Sometimes the approach depicted above is called symmetric [7]. In our opinion, it has a number of significant shortcomings. First, the relations (3) and (4), and, hence, (6) are not unique since they introduce four new quantities ($\boldsymbol{P}'$, $\boldsymbol{M}$, $\boldsymbol{D}'$ and $\boldsymbol{H}$) using only three relationships.

Sometimes by writing (3) one says that $\boldsymbol{P}'$ and $\boldsymbol{M}$ are connected with the electric and magnetic moments of the medium, respectively. This, however, is not quite correct, as it remains unclear to what degree these quantitative definitions of $\boldsymbol{P}'$ and $\boldsymbol{M}$ determine them qualitatively, because in the optical frequency range the notion of the magnetic moment of the medium loses its physical meaning [4, 27].

Frequently encountered references to the possibility of quantum mechanical calculations of $\boldsymbol{P}'$ and $\boldsymbol{M}$ are also poorly justified, as unique quantum mechanical calculation is possible only for the total density of the polarisation current $\boldsymbol{j}$ [29, 30] and not for its separate parts. Therefore, the question will arise: how can the expressions obtained be separated into two parts?

This problem is practically unsolved because of the artificiality of the representation in Eqn (3) which is not based upon deep physical considerations. Moreover, as follows from Eqns (3)–(5) and is even more obvious from Eqns (1), the wave equation for $\boldsymbol{E}$ in a homogeneous medium has the form

$$\Big\{\boldsymbol{k}\,[\boldsymbol{k}\cdot\boldsymbol{E}(\omega,\boldsymbol{k})]\Big\} + \frac{\omega^2}{c^2}\,\boldsymbol{E}(\omega,\boldsymbol{k}) = \frac{4\pi\mathrm{i}\omega}{c}\,\boldsymbol{j}(\omega,\boldsymbol{k})\,. \tag{7}$$

Knowing $\boldsymbol{E}$, it is easy to find $\boldsymbol{B}$.

Thus, from the point of view of the electrodynamics of unbounded media, we are ultimately interested only in the dependence of $\boldsymbol{j}(\omega,\boldsymbol{k})$ on $\boldsymbol{E}(\omega,\boldsymbol{k})$. When using Eqns (3), (4) and (6), it is often quite difficult to find this dependence in case of linear homogeneous media. If the medium is inhomogeneous [12, 31] or nonlinear [32], the computational procedure becomes very complicated although, of

†We will denote the medium polarisation and electric field induction used in Landau–Lifshitz approach by $\boldsymbol{P}$ and $\boldsymbol{D}$ without primes, respectively, to avoid ambiguity.

course, it can be done, especially if one introduces various simplifying assumptions [32].

In our opinion, it is this complexity of consistent generalisation to the case of inhomogeneous and nonlinear media that is the most significant defect of the 'symmetric' approach to electrodynamics. Moreover, at the phenomenological level of consideration, this problem is quite artificial: first, the expression for $\boldsymbol{j}$ is split into a few terms in an arbitrary way, then one writes down the constitutive relation for each term, and finally one finds the dependence of the total polarisation current $\boldsymbol{j}$ on the electric field strength $\boldsymbol{E}$.

Clearly, it is much simpler to proceed from system (1) directly and to write down the constitutive relation immediately for the total polarisation current $\boldsymbol{j}$, or (which is practically the same thing), to consider that $\boldsymbol{M} = 0$, and $\boldsymbol{j} = \partial \boldsymbol{P}/\partial t$.

2.3 Landau – Lifshitz constitutive relation for the electric field induction

As we pointed out above, in electrodynamics of unbounded media it is most natural to introduce the total polarisation of the medium $\boldsymbol{P}$ (and the corresponding electric field induction $\boldsymbol{D}$) which includes the total polarisation current $\boldsymbol{j}$ [27]:

$$\frac{\partial \boldsymbol{P}}{\partial t} = \boldsymbol{j}\,, \qquad \boldsymbol{D} = \boldsymbol{E} + 4\pi \boldsymbol{P}\,. \tag{8}$$

Note that $\boldsymbol{j}(\boldsymbol{r}_0, t)$ and hence $\boldsymbol{P}(\boldsymbol{r}_0, t)$, $\boldsymbol{D}(\boldsymbol{r}_0, t)$ can, in general, depend on the fields $\boldsymbol{E}$ and $\boldsymbol{B}$ not only at the point $\boldsymbol{r}_0$, but at the adjacent points as well. On the other hand, the field $\boldsymbol{B}(\boldsymbol{r}, t)$ is related to the field $\boldsymbol{E}(\boldsymbol{r}, t)$ by Eqn (1a). This allows one to consider, in phenomenological treatment, that $\boldsymbol{P}(\boldsymbol{r}_0, t)$ and $\boldsymbol{D}(\boldsymbol{r}_0, t)$ depend only on the field $\boldsymbol{E}(\boldsymbol{r}, t)$ in the entire space.

In the approach presented, the Maxwell system of equations and the constitutive relation (the latter is written for linear media for brevity) take the form, respectively [1, 27]

$$\operatorname{curl} \boldsymbol{E} = -\frac{1}{c}\frac{\partial \boldsymbol{B}}{\partial t}\,, \qquad \operatorname{div} \boldsymbol{D} = 0, \qquad \operatorname{div} \boldsymbol{B} = 0\,, \tag{9a}$$

$$\operatorname{curl} \boldsymbol{B} = \frac{1}{c}\frac{\partial \boldsymbol{D}}{\partial t}\,, \tag{9b}$$

$$\boldsymbol{D}(t, \boldsymbol{r}) = \int_{-\infty}^{t} \mathrm{d}t' \int \hat{\varepsilon}(t, t'; \boldsymbol{r}, \boldsymbol{r}')\, \boldsymbol{E}(t', \boldsymbol{r}')\, \mathrm{d}\boldsymbol{r}'\,. \tag{10}$$

Eqn (10) for homogeneous unbounded media can be written as follows†:

$$D_i(\omega, \boldsymbol{k}) = \varepsilon_{ij}(\omega, \boldsymbol{k})\, E_j(\omega, \boldsymbol{k})\,. \tag{11}$$

We stress once more that by assuming $\boldsymbol{B} = \boldsymbol{H}$ in Eqn (9b), one does not neglect by any magnetic effects at all. All are taken into account by the constitutive relation (10), which, in particular, can be reflected in symmetry properties of the tensor $\varepsilon_{ij}(\omega, \boldsymbol{k})$. For example, use of the symmetry principle for kinetic coefficients [33] leads to the relationship [1, 24, 27]

$$\varepsilon_{ij}(\omega, \boldsymbol{k}, \boldsymbol{B}_{\text{ext}}) = \varepsilon_{ji}(\omega, -\boldsymbol{k}, -\boldsymbol{B}_{\text{ext}})\,, \tag{12}$$

†All questions arising from the introduction of the tensor $\varepsilon_{ij}(\omega, \boldsymbol{k})$ are discussed in detail in Ref. [1].

where $\boldsymbol{B}_{\text{ext}}$ is the magnetic field induction which is a constant in time and is not zero under the influence of an external magnetic field or a magnetic structure (ferromagnetics and antiferromagnetics).

3. Boundary conditions on the surface of media with nonlocal optical response

3.1 Procedure for obtaining boundary conditions

It should be stressed that all the analyses given above indicate only a higher or lower degree of formalism, as well as the convenience or inconvenience, of phenomenological introduction of various characteristics of the medium, and cannot be in any way taken as pro or contra arguments for one or another approach to the electrodynamics of unbounded media. It is also clear that both approaches considered will give similar (within the accuracy of the notation) results [1].

Differences usually arise when considering the interaction of electromagnetic waves with the surface (cf. for example [6, 7, 12]). Apparently, habit plays a major role here. While working mainly with media without SD, many people became used to assuming the tangent components of the fields $\boldsymbol{E}$ and $\boldsymbol{H}$ (or $\boldsymbol{E}$ and $\boldsymbol{B}$ in the Landau – Lifshitz approach) to be continuous at the interface between two media, without considering the applicability of this statement.

Meanwhile, it is well known and even quoted in textbooks in general physics [34] that small violations of the Fresnel formula exist during reflection through angles close to the Brewster angle [34, 35]. In particular, the reflection coefficient does not vanish for any incident angle, although for the Brewster angle it is very small.

The deviations from the Fresnel formula are explained by the presence of a thin transition layer near the surface of the reflecting medium (including the medium not showing natural optical activity) of a size $d_0 \ll \lambda$. The properties of the layer (including optical ones) differ from those of the bulk medium itself.

The transition layers can arise due to external reasons (impurity, processing, gas adsorption etc.) or peculiarities in the molecular structure of the reflecting medium itself near the surface [34, 35]. The latter, in fact, is a manifestation of a 'hidden', extremely weak nonlocality of the optical response of the medium, which is unavoidably present (at least due to discreteness) in any medium.

Existence of a more noticeable SD can lead to a very much increased influence of the transition layers on the reflection of light than in media with a practically local response. This is due, in particular, to the space scale of the transition layers d_0 being not less than the nonlocality scale optical response d of the medium.

In this connection it comes into question how one should phenomenologically take into account the influence of the transition layers on light reflection. As a matter of fact, the boundary conditions are traditionally obtained from the Maxwell equations as a result of extrapolation to the limit, that is by assuming that a sharp interface exists between homogeneous media.

One can solve this problem by assuming that by passing from one medium to another, all properties of matter and, hence, electromagnetic field characteristics vary continuously although sufficiently fast. Then, by approximately solving the Maxwell equations in the narrow transition region, one can obtain a relationship between the fields at its opposite boundaries to the required approximation in terms of the small parameter kd_0.

By comparing this relationship with that which would pertain between the fields at the same points in case of a sharp interface between homogeneous media, one can easily find the form of the altered boundary conditions by using conditions which in the sharp interface model would allow one, nevertheless, to take into account the influence of the transition layer on the reflection and refraction of light.

Everything indicates that the appearance of additional terms in the boundary conditions is a result of the existence of a surface (that is, existing only on the interface boundary) polarisation current $\boldsymbol{i}$ and bounded charge σ at the sharp interface between two media.

In the case of an interface between two homogeneous media which is smooth, plane and homogeneous in the transverse direction, the constitutive relation for temporal Fourier components of the electromagnetic field in the Landau–Lifshitz approach ($\boldsymbol{E}$, $\boldsymbol{B}$, $\boldsymbol{D} \propto \exp(-\mathrm{i}\omega t)$) can be written as

$$\boldsymbol{D}(z, \boldsymbol{E}) = \begin{cases} \boldsymbol{D}_{(1)}(\boldsymbol{E}), & z \leqslant z_1, \\ \boldsymbol{D}_{(\mathrm{s})}(z, \boldsymbol{E}), & z_1 \leqslant z \leqslant z_2, \\ \boldsymbol{D}_{(2)}(\boldsymbol{E}), & z \geqslant z_2. \end{cases} \tag{13}$$

Here and in what follows the z axis is directed perpendicular to the interface from the first medium to the second one; $z_{1,2}$ are the boundaries of the inhomogeneity region ($z_2 - z_1 \approx d_0$).

A particular form of the functionals $\boldsymbol{D}_{(1)}, \boldsymbol{D}_{(2)}$ and $\boldsymbol{D}_{(\mathrm{s})}$ is defined by optical and symmetry properties of the homogeneous media 1, 2 (far away from their boundaries) and, accordingly, of the inhomogeneous layer between them. For media with SD they contain differentiation operators. For example, for homogeneous nonlinear media with SD [9, 10, 36, 37]

$$\boldsymbol{D}_{(m)}(\boldsymbol{E}) = \hat{\varepsilon}_m \boldsymbol{E} + 4\pi \Big\{ \hat{\gamma}_m^{(1)} \vec{\nabla} \boldsymbol{E} + \hat{\chi}_m^{(2)} \boldsymbol{E}\boldsymbol{E} + \hat{\gamma}_m^{(2)} \boldsymbol{E} \vec{\nabla} \boldsymbol{E} + \hat{\chi}_m^{(3)} \boldsymbol{E}\boldsymbol{E}\boldsymbol{E} + \hat{\gamma}_m^{(3)} \boldsymbol{E}\boldsymbol{E} \vec{\nabla} \boldsymbol{E} + \ldots \Big\}. \tag{14}$$

In the smooth interface model an explicit dependence of $\boldsymbol{D}$ on z given by expression (13) is continuous

$$\boldsymbol{D}_{(\mathrm{s})}(z_m, \boldsymbol{E}) = \boldsymbol{D}_{(m)}(\boldsymbol{E}), \quad m = 1, 2,$$

and sufficiently differentiable. It is clear from here that the fields $\boldsymbol{E}$ and $\boldsymbol{B}$ satisfying the Maxwell equations (9) and the constitutive relation (13) are continuous and differentiable, and, hence, no boundary conditions (excluding, of course, conditions at infinity) are required to solve the problem of radiation passage from the first into the second medium.

In the sharp interface boundary model the functional relation (13) takes the form

$$\boldsymbol{D}_{(0)}(z, \boldsymbol{E}) = \begin{cases} \boldsymbol{D}_{(1)}(\boldsymbol{E}), & z < z_0 \\ \boldsymbol{D}_{(2)}(\boldsymbol{E}), & z > z_0, \end{cases} \tag{15}$$

where z_0 is an arbitrarily chosen location of the interface boundary ($z_1 \leqslant z_0 \leqslant z_2$) of the model. It is clear that $\boldsymbol{D}_{(0)}$ has a discontinuity at $z = z_0$, and hence, the fields $\boldsymbol{E}^{(0)}(z)$ and $\boldsymbol{B}^{(0)}(z)$, which are the solutions to the equations (9) and (15) in the entire space, can have a discontinuity at the point $z = z_0$.

Our task is to find boundary conditions for the fields at the point $z = z_0$, such that in the approximation given by the parameter kd_0 the fields $\boldsymbol{E}^{(0)}$ and $\boldsymbol{E}$, as well as $\boldsymbol{B}^{(0)}$ and $\boldsymbol{B}$, are equal at the point $z = z_m$ provided that they are equal at $z = z_l$, where $m = 1, 2$ and $l = 1 + \delta_{1m}$.

In the framework of the 'symmetric' approach relationships similar to Eqns (13) and (15) must be constructed for $\boldsymbol{D}'(z, \boldsymbol{E})$ and $\boldsymbol{D}'_{(0)}(z, \boldsymbol{E})$ (with the help of the corresponding function $\boldsymbol{D}'_{(1)}$, $\boldsymbol{D}'_{(2)}$ and $\boldsymbol{D}'_{(\mathrm{s})}$), and for $\boldsymbol{B}(z, \boldsymbol{H})$ and $\boldsymbol{B}_{(0)}(z, \boldsymbol{H})$ (through $\boldsymbol{B}_{(1)}$, $\boldsymbol{B}_{(2)}$, and $\boldsymbol{B}_{(\mathrm{s})}$) as well.

Naturally, the procedure described can be performed with equivalent (within the accuracy of the notation) results for any incident angles in the framework of any of the approaches to the electrodynamics of unbounded media considered in Section 2. However, for simplicity below we will restrict ourselves to the consideration of normal light falling onto the interface in the first approximation in terms of the parameter kd_0.

3.2 Boundary conditions corresponding to 'symmetric' constitutive relations

One of the significant differences of the 'symmetric' approach is that in fact it explicitly assumes the possible existence of a surface current of bounded charges at the sharp interface between the homogeneous layer (the second term in Eqn (3) becomes a δ-function in this case). This, in fact, allows one to obtain from Eqns (5), without any difficulty, sufficiently correct boundary conditions which, as far as we know, are used by all supporters of the 'symmetric' approach. In the notation of Section 3.1 these conditions have the form

$$\boldsymbol{E}_{1\mathrm{t}}^{(0)} = \boldsymbol{E}_{2\mathrm{t}}^{(0)}, \quad D'_{(1)\mathrm{n}}(\boldsymbol{E}_1^{(0)}) = D'_{(2)\mathrm{n}}(\boldsymbol{E}_2^{(0)}),$$

$$B_{(1)\mathrm{n}}(\boldsymbol{H}_1^{(0)}) = B_{(2)\mathrm{n}}(\boldsymbol{H}_2^{(0)}), \tag{16}$$

$$\boldsymbol{n} \times (\boldsymbol{H}_2^{(0)} - \boldsymbol{H}_1^{(0)}) = 0. \tag{17}$$

Here and in what follows $\boldsymbol{n}$ is normal to the interface boundary directed from the medium 1 to the medium 2, indices 'n' and 't' correspond to the normal and tangent vector components, respectively, and the notation $\boldsymbol{E}_1^{(0)} = \boldsymbol{E}^{(0)}(z_{0-})$, $\boldsymbol{E}_2^{(0)} = \boldsymbol{E}^{(0)}(z_{0+})$ etc. is used.

However, as we pointed out above, introducing quantities $\boldsymbol{M}$ and $\boldsymbol{P}'$, strictly speaking, is not fully unique and hence the boundary condition (17) is not fully correct (the vector $\boldsymbol{H}_\mathrm{t} = \boldsymbol{B}_\mathrm{t} - 4\pi\boldsymbol{M}_\mathrm{t}$ cannot be continuous for arbitrary choice of $\boldsymbol{M}$†). The cause of this problem is not only that the 'symmetric' approach has defects. It is primarily connected with an error made in deriving Eqn (17).

It is the case that in obtaining Eqn (17), we take into account, in fact, only a near-surface inhomogeneity of the 'magnetic moment of the medium' $\boldsymbol{M}$ (the arbitrary nature of this notion has already been pointed out in Section 2.2), and a possible inhomogeneity of its 'electric moment' $\boldsymbol{P}'$ is ignored completely.

Taking account of the latter (in accordance with Section 3.1) changes the right-hand side of Eqn (17):

$$\boldsymbol{n} \times \left(\boldsymbol{H}_2^{(0)} - \boldsymbol{H}_1^{(0)}\right) = \frac{4\pi}{c}\,(\boldsymbol{i}'_{1,2})_\mathrm{t}, \tag{18}$$

†Let us recall that unlike $\boldsymbol{M}$ and $\boldsymbol{P}'$, the electric field strength $\boldsymbol{E}$ and magnetic induction $\boldsymbol{B}$ are uniquely defined [see Eqn (2)].

where

$$\boldsymbol{i}'_{1,\,2} = -\frac{\mathrm{i}\omega}{4\pi}\int_{z_1}^{z_{0-}} \Delta\boldsymbol{D}'\,\mathrm{d}l - \frac{\mathrm{i}\omega}{4\pi}\int_{z_{0+}}^{z_2} \Delta\boldsymbol{D}'\,\mathrm{d}l\,, \tag{19}$$

and $\Delta\boldsymbol{D}' = \boldsymbol{D}'(z, \boldsymbol{E}) - \boldsymbol{D}'_{(0)}(z, \boldsymbol{E}^{(0)})$ characterises the difference of the real electric field induction $\boldsymbol{D}'$ near the surface from the induction $\boldsymbol{D}'_{(0)}$ from the sharp interface boundary model.

Integration of Eqn (19) is carried out over the surface (transition) layer depth (excluding the point $z = z_0$ where the vector $\boldsymbol{D}'_{(0)}$ is undetermined), and z_0 is defined by the chosen model (sharp) interface between two media. Use of Eqn (18) instead of Eqn (17) for the normal incidence of light onto a linear isotropic gyrotropic medium (i.e. such media as are considered in the majority of papers) for the traditional [2, 8, 13, 31, 38] choice of constitutive relations (6):

$$\boldsymbol{D}' = \varepsilon'\boldsymbol{E} - \mathrm{i}g\boldsymbol{H}\,, \quad \boldsymbol{B} = \mu\boldsymbol{H} + \mathrm{i}g\boldsymbol{E}\,, \tag{20}$$

satisfying the symmetry principle for kinetic coefficients [33] and molecular models [3], will only lead to a phase change of the reflected and transmitted waves.

In this simplest case taking account of $\boldsymbol{i}'_{1,\,2}$ is equivalent, in fact, to a small shift (by an amount smaller than d_0) of the model interface boundary between the media. That is, it is equivalent to a small correction to the optical depth of the medium, which takes into account the inhomogeneity of its optical properties near the surface [see the end of Section 4 after Eqns (35) and (36)].

Nevertheless, use of Eqn (17) instead of Eqn (18) is potentially dangerous, as this can lead, in particular, to recurrent new 'contradictions' between different approaches to the electrodynamics of anisotropic media with SD.

Now we turn to the analysis of boundary conditions arising in the Landau–Lifshitz approach.

3.3 Boundary conditions in Landau–Lifshitz electrodynamics. Constitutive relation for the surface polarisation current

If one uses the Maxwell equations in the form (1) or (9) and carries out the procedure described in Section 3.1, then in case of the normal incident of light the corresponding boundary conditions take the form [1, 24]

$$\boldsymbol{E}^{(0)}_{1\mathrm{t}} = \boldsymbol{E}^{(0)}_{2\mathrm{t}}\,,\; B^{(0)}_{1\mathrm{n}} = B^{(0)}_{2\mathrm{n}}\,,\; D_{(1)\mathrm{n}}(\boldsymbol{E}^{(0)}_1) = D_{(2)\mathrm{n}}(\boldsymbol{E}^{(0)}_2)\,, \tag{21a}$$

$$\boldsymbol{n}\times\left(\boldsymbol{B}^{(0)}_2 - \boldsymbol{B}^{(0)}_1\right) = \frac{4\pi}{c}\,(\boldsymbol{i}_{1,\,2})_{\mathrm{t}}\,, \tag{21b}$$

where the surface current of bound charge $\boldsymbol{i}_{1,\,2}$ is expressed in terms of $\Delta\boldsymbol{D}$ in just the same manner as $\boldsymbol{i}'_{1,\,2}$ was expressed by $\Delta\boldsymbol{D}'$ in Eqn (19), and $\Delta\boldsymbol{D} = \boldsymbol{D}(z, \boldsymbol{E}) - \boldsymbol{D}_{(0)}(z, \boldsymbol{E}^{(0)})$ characterises the difference between the real electric field induction $\boldsymbol{D}$ near the surface and the induction $\boldsymbol{D}_{(0)}$ occurring in the sharp interface boundary model.

It should be noted that by taking into account the relationship of $\boldsymbol{D}$ with $\boldsymbol{j}$ [see Eqn (8)], as well as the fact that in the zero approximation with the parameter kd_0 the field $\boldsymbol{E}^{(0)}_{\mathrm{t}}(z) = \boldsymbol{E}_{\mathrm{t}}(z)$, $\boldsymbol{i}_{1,\,2}$ can be expressed as

$$\boldsymbol{i}_{1,\,2} = \int_{z_1}^{z_{0-}} \Delta\boldsymbol{j}(\zeta)\,\mathrm{d}\zeta + \int_{z_{0+}}^{z_2} \Delta\boldsymbol{j}(\zeta)\,\mathrm{d}\zeta\,, \tag{21c}$$

where $\Delta\boldsymbol{j}(z) = \boldsymbol{j}(z, \boldsymbol{E}(z)) - \boldsymbol{j}_{(0)}(z, \boldsymbol{E}^{(0)}(z))$ characterises the difference of the real density of polarisation current near the surface $\boldsymbol{j} = -(\mathrm{i}\omega/4\pi)(\boldsymbol{D} - \boldsymbol{E})$ from the density of polarisation current $\boldsymbol{j}_{(0)} = -(\mathrm{i}\omega/4\pi)(\boldsymbol{D}_{(0)} - \boldsymbol{E}^{(0)})$ occurring in the sharp interface boundary model.

The expressions (21b) and (21c) in particular demonstrate that the quantity $\boldsymbol{i}_{1,\,2}$ has indeed the sense of a 'surface' polarisation current which should be assumed present if we want to consider the interface between the media 1 and 2 as sharp.

In order to convince ourselves of the full equivalence of the boundary conditions (18) and (21b), we note that due to Eqns (3), (4), (8) and (19)

$$\boldsymbol{i}_{1,\,2} = c\Big\{\boldsymbol{n}\times\left[\boldsymbol{M}_{(2)}(\boldsymbol{H}^{(0)}_2) - \boldsymbol{M}_{(1)}(\boldsymbol{H}^{(0)}_1)\right]\Big\} + \boldsymbol{i}'_{1,\,2}\,, \tag{22}$$

where $\boldsymbol{M}_{(m)}(\boldsymbol{H}) = \left(\boldsymbol{B}_{(m)}(\boldsymbol{H}) - \boldsymbol{H}\right)/4\pi$, $m = 1, 2$. Thus, the boundary conditions (18) and (21b) for $\boldsymbol{H}^{(0)}$ and $\boldsymbol{B}^{(0)}$ express the same thing with different notation.

As a result, the question naturally arises as to why the 'symmetric' and 'asymmetric' approaches often yield essentially different results. It is easy to show that either the surface current has been completely ignored [6], or it has been wrongly calculated [12].

In connection with the above, we stress that although from the formal point of view in the Landau–Lifshitz approach it is sufficient to use only one constitutive relation such as Eqn (10) relating vectors $\boldsymbol{D}$ and $\boldsymbol{E}$, it turns out to be very inconvenient in practice.

In the case above, two physically different phenomena, the polarisation current inside the homogeneous medium and that arising close to the boundary, are combined into one constitutive relation. They differ not only in 'place of appearance', but also in having different causes and symmetric properties.

In particular, as we pointed out above, an inhomogeneity intrinsic to interfaces between media with SD [9, 10, 25] has a significant influence on the polarisation current near the surface. The symmetric properties inside and on the surface of the media can differ even for ideal surfaces [9, 10, 25, 26, 39], let alone for surfaces with possible crystal lattice defects, oxides, coatings and so on.

This emphasises the need for two constitutive relations†: one for $\boldsymbol{D}$ inside the medium (assuming homogeneity or, if necessary, a weak inhomogeneity of the properties of the medium with a scale much greater than that of the nonlocality of the optical response of the medium), and another for the surface polarisation current $\boldsymbol{i}_{1,\,2}$ [9, 10, 25]:

$$\boldsymbol{i}_{1,\,2} = \hat{\kappa}^{(1)}\boldsymbol{S} + \hat{\kappa}^{(2)}\boldsymbol{SS} + \hat{\kappa}^{(3)}\boldsymbol{SSS} + \ldots\,, \tag{23}$$

where vector $\boldsymbol{S} = \boldsymbol{E} + 4\pi(\boldsymbol{P}\cdot\boldsymbol{n})\boldsymbol{n}$, in contrast to vectors $\boldsymbol{E}$ and $\boldsymbol{P}$, changes only gradually within the transition layer and thus is more convenient for writing down the constitutive relation for the surface polarisation current. In the expression (23) and below index '0', the notation for the sharp interface model, is omitted for brevity.

It is then convenient to separate the contributions into $\boldsymbol{i}_{1,\,2}$ caused by specific surface mechanisms (primarily inhomogeneity near the surface and symmetric features

†The situation often arises in the case of bounded media with a strong SD, when changing from (10) to (11) introduces the need for additional boundary conditions [1].

of the surface), and the contributions caused by the properties of the adjacent media as a whole†. For the latter, the corresponding constitutive tensors can be related to those characterising the optical properties of the homogeneous medium.

Establishing relationships between these tensors and their forms for different media is one of the most important tasks in electrodynamics of bounded media. Presently, such relationships can be considered partially established only for linear (not necessarily isotropic) nonmagnetic media which the symmetry principle for kinetic coefficients can be applied to:

$$\kappa_{jp}^{(1)}(\omega) = -i\omega\left[\chi_{jp}^{(s)} - \frac{1}{2}\gamma_{1,\,jpz}^{(1)} + \frac{1}{2}\gamma_{2,\,jpz}^{(1)}\right], \quad j, p = x, y\,. \tag{24}$$

The symmetric tensor $\hat{\chi}^{(s)}$ in expression (24) characterises near-surface inhomogeneity of the dielectric permittivity of the medium $\hat{\varepsilon}$, and antisymmetric (by permutation of the first two indices) tensors $\hat{\gamma}_1^{(1)}$ and $\hat{\gamma}_2^{(1)}$ describe gyrotropic bulk properties of the homogeneous media 1 and 2, respectively [see Eqn (14)]. Note that the relationship $\boldsymbol{E} = \boldsymbol{E}(\omega)\exp(-i\omega t)$ was used in Eqn (24).

The relationship (24) can be obtained by substituting the expression for electric field induction in a homogeneous linear medium, which satisfies the symmetry principle for kinetic coefficients, into the formula for the surface polarisation current (see, e.g., [40])‡.

A result similar to relationship (24) was obtained for the first time in Refs [9, 10] for the interface between gyrotropic and nongyrotropic media ($\gamma_1^{(1)} = 0$). The symmetric approach was used and subsequently (with the help of the symmetry principle for kinetic coefficients) a relationship between the constitutive tensors arising with those known from the Landau–Lifshitz approach was established.

Using the notation from [9, 10] we have

$$\chi_{jp}^{(s)} = \tilde{L}\chi_{s,\,jp}^{(1)} + \left(f_{qj}\,e_{qzp} + f_{qp}\,e_{qzj}\right)\,,$$

where $\tilde{L}$ is the effective size of the near-surface inhomogeneity ($\tilde{L} \sim d_0$), $\hat{\chi}_s^{(1)}$ is a symmetric tensor characterising the near-surface inhomogeneity of the tensor $\hat{\varepsilon}'$ arising in the symmetric approach by writing down the constitutive relation for $\boldsymbol{D}'$ [see Eqn (6)], $\hat{f}$ is a gyration pseudotensor ($\gamma_{2,\,jpr}^{(1)} = e_{jpq}f_{qr}$), and e_{jpr} is the Levi–Civita symbol [41].

We note that in Refs [9, 10] nonlinear media were also considered. According to Eqn (22), tensors $\hat{\kappa}^{(n)}$ that are contained in expression (23) were separated into two parts:

$$\kappa_{jp_1\ldots p_n}^{(n)} = -\mathrm{i}\omega\left(e_{jzq}\,\beta_{qp_1\ldots p_n}^{(n)} + \tilde{L}\chi_{s,\,jp_1\ldots p_n}^{(n)}\right)\,, \tag{25}$$

which are connected with a 'magnetic moment' of the medium $\boldsymbol{M}$ and a near-surface inhomogeneity $\boldsymbol{P}'$, respectively.

One should, however, bear in mind that, as we have repeatedly pointed out, such partition is not unique (due to the ambiguity of introducing $\boldsymbol{P}'$ and $\boldsymbol{M}$ in the symmetric approach), and hence, tensors $\hat{\beta}^{(n)}$ and $\hat{\chi}_s^{(n)}$ taken separately have no certain physical meaning. Tensor $\hat{\kappa}^{(n)}$ alone is uniquely defined.

It should be emphasised that it is due to the relationship (24) that polarisation effects during reflection from linear nonmagnetic media are prohibited (in the first approximation by the SD parameter kd in the case of normal incidence of light) [9, 10].

4. Reflection of light from a linear isotropic gyrotropic medium: comparison of different approaches

As an example of our point of view we consider a particular problem of reflection (under normal incidence) of a plane wave from an optical system consisting of an isotropic gyrotropic nonmagnetic medium without absorption and a mirror behind it with a reflection coefficient $R = 1$.

The electric field in vacuum in front of the medium ($z < 0$) is

$$\boldsymbol{E}^{(1)} = \boldsymbol{E}_{\mathrm{i}}\exp(-\mathrm{i}\omega t + \mathrm{i}kz) + \boldsymbol{E}_{\mathrm{r}}\exp(-\mathrm{i}\omega t - \mathrm{i}kz)\,, \tag{26}$$

where $\boldsymbol{E}_{\mathrm{i}}$ and $\boldsymbol{E}_{\mathrm{r}}$ are amplitudes of the incident and reflected waves, respectively, and $k = \omega/c$.

The constitutive relations for isotropic gyrotropic media in the 'symmetric' and Landau–Lifshitz approaches are written, respectively, as [2, 8, 11, 27]

$$\boldsymbol{D}' = \varepsilon'\boldsymbol{E} - \mathrm{i}g\boldsymbol{H}\,, \quad \boldsymbol{B} = \mu\boldsymbol{H} + \mathrm{i}g\boldsymbol{E}\,, \tag{27}$$

$$\boldsymbol{D} = \varepsilon\boldsymbol{E} - 4\pi f_0\,\mathrm{curl}\,\boldsymbol{E}\,, \tag{28}$$

with $\varepsilon = \varepsilon'\mu - g^2$, $g = 2\pi\omega f_0/c$, and $\mu = 1$ (we consider a nonmagnetic medium in the present case).

The eigenwaves in isotropic gyrotropic media are known to be circularly polarised and have different wave vectors [10, 12]:

$$q_\pm^{(1)} = kn_\pm^{(1)}\,, \quad q_\pm^{(-1)} = kn_\pm^{(-1)}\,, \quad n_\pm^{(\ )} = \pm g + \quad \varepsilon^{1/2}\,,$$

where $\quad = -1,\ 1$.

The electric field can be written as follows:

$$\boldsymbol{E}^{(2)} = \boldsymbol{e}_+E_{\mathrm{t}+}\exp[-\mathrm{i}\omega t + \mathrm{i}q_+^{(1)}z] + \boldsymbol{e}_-E_{\mathrm{t}-}\exp[-\mathrm{i}\omega t + \mathrm{i}q_-^{(1)}z]$$
$$+\boldsymbol{e}_+\tilde{E}_+\exp[-\mathrm{i}\omega t + \mathrm{i}q_+^{(-1)}z] + \boldsymbol{e}_-\tilde{E}_-\exp[-\mathrm{i}\omega t + \mathrm{i}q_-^{(-1)}z]\,. \tag{29}$$

In Eqn (29) $\boldsymbol{e}_\pm = (\boldsymbol{i} \mp i\boldsymbol{j})/\sqrt{2}$ are unit vectors with different polarisation ($\boldsymbol{i}$ and $\boldsymbol{j}$ are unit vectors aligned with axes x and y, respectively, and axis z is normal to the interface boundary); $\boldsymbol{E}_{\mathrm{t}}$ and $\tilde{\boldsymbol{E}}$ are amplitudes of the waves expanding in the medium toward and away from the mirror, respectively.

Naturally, the boundary conditions at the front surface of the medium ($z = 0$) for different angles of approach will be different. However, for brevity we write them down in a unified form:

†The relationships (22), in particular, clearly demonstrate that introducing 'magnetic moment of the medium' in the 'symmetric' approach framework selects, in fact, the surface current fraction that is essentially caused by the bulk properties of the medium. This is not too bad on its own, but it is important to bear in mind the possibility of the existence of surface currents $\boldsymbol{i}'_{1,2}$ of a purely near-surface origin (which are not directly and uniquely connected with the bulk properties of the medium).

‡One can show that the series of integrals arising will converge, although the series for $\boldsymbol{D}$ obtained in [40] can diverge under our conditions (inhomogeneity scale $d_0 \sim d$). Physically, this is because the surface polarisation current is always limited.

$$E_{i\pm} + E_{r\pm} = E_{t\pm} + \tilde{E}_{\pm}\,, \quad E_{i\pm} - E_{r\pm} = n_0(E_{t\pm} - \tilde{E}_{\pm})$$
$$- \frac{4\pi i\omega}{c}\,\beta\chi_0^{(s)}(E_{t\pm} + \tilde{E}_{\pm}) \pm \alpha g(E_{t\pm} + \tilde{E}_{\pm})\,. \quad (30)$$

Here $n_0 = \varepsilon^{1/2}$ and $E_{i,\,r\pm} = (\boldsymbol{e}_{\pm}^{*}\boldsymbol{E}_{i,\,r})$ are amplitudes of proper waves. The values of parameters α and β in expression (30), $\alpha = 0$ and $\beta = 0$, correspond to the boundary conditions obtained in the 'symmetric' approach framework from Eqns (16) and (17) using Eqns (26), (27) and (29) [8].

If one does use Landau – Lifshitz approach and Eqns (21) and (23), then taking into account Eqns (26), (28), (29) and equality (24), which for the interface between the isotropic and isotropic gyrotropic media (where $\hat{\gamma}_1^{(1)} = 0$, $\gamma_{2,\,jpr}^{(1)} = f_0\, e_{jpr}$, and $\chi_{jp}^{(s)} = \chi_0^{(s)}\delta_{jp} + \Delta\chi^{(s)}\delta_{jz}\delta_{zp}$) takes the form

$$\kappa_{jp}^{(1)}(\omega) = -i\omega\left[\chi_0^{(s)}\delta_{jp} + \Delta\chi^{(s)}\delta_{jz}\delta_{zp} + \frac{1}{2}f_0\, e_{jpz}\right],$$

we get the boundary conditions (30) with $\alpha = 0$ and $\beta = 1$ [9, 10]. In this case they differ from the previous ones in having an additional term describing the influence of the near-surface inhomogeneity of the electric permittivity of the medium, ε.

The case of $\beta = 0$ and $\alpha = 1$ corresponds to completely ignoring the surface polarisation current effect [6], and that of $\beta = 0$ and $\alpha = -1$ to the boundary conditions proposed in Ref. [12].

We will assume that the mirror is attached tightly to the medium ($z = L$, where L is the length of the medium). Then the boundary conditions at the rear surface of the medium for all four cases will have the form:

$$E_{t\pm}\exp[iq_{\pm}^{(1)}L] + \tilde{E}_{\pm}\exp[iq_{\pm}^{(-1)}L] = 0\,. \quad (31)$$

From Eqns (30) and (31) one can easily find the coefficients of reflection for circular polarised waves $R_{\pm} \equiv E_{r\pm}/E_{i\pm}$ from the system under consideration:

$$R_{\pm} = \frac{(1 - n_0 - \gamma_{\pm}) - (1 + n_0 - \gamma_{\pm})\exp(i\psi)}{(1 + n_0 + \gamma_{\pm}) - (1 - n_0 + \gamma_{\pm})\exp(i\psi)}\,, \quad (32)$$

where $\psi = 2kL\sqrt{\varepsilon}$, and $\gamma_{\pm} = -4\pi i\omega\beta\chi_0^{(s)}/c \pm \alpha g$.

For a medium with no absorption ($\mathrm{Im}\,\chi_0^{(s)} = \mathrm{Im}\,g = \mathrm{Im}\,\varepsilon = 0$, $\gamma_{\pm}^{*} = -\gamma_{\mp}$, where the asterisk means complex conjugation) we obtain in the first approximation by the small parameter of spatial dispersion kd ($g \sim k\chi_0^{(s)} \sim kd$)

$$|R_{\pm}|^2 = 1 \pm \alpha\delta\,, \quad \delta = \frac{4g(\cos\psi - 1)}{(1 + \varepsilon) - (1 - \varepsilon)\cos\psi}\,. \quad (33)$$

The intensity of the light reflected by the system is

$$W_r = (1 + \alpha\delta b_0)W_0\,, \quad (34)$$

where $b_0 = (|E_{i+}|^2 - |E_{i-}|^2)/2W_0$ and $W_0 = (|E_{i+}|^2 + |E_{i-}|^2)/2$ are the incident light ellipticity and intensity, respectively. Obviously, b_0 can be either positive or negative. Moreover, the signs of α, δ and b_0 are fully independent.

Therefore, if one uses the boundary conditions (30) with $\alpha \neq 0$ (i.e. neglects the surface polarisation current [6] or uses an insufficiently accurate expression for the latter [12]), in a steady-state regime the intensity W_r of the reflected light can be higher than that of the incident radiation W_0, which evidently contradicts the energy conservation law.

We compare now the results obtained by using the 'symmetric' approach with boundary conditions (16) and (17) ($\alpha = 0$ and $\beta = 0$) and the correct Landau – Lifshitz approach ($\alpha = 0$ and $\beta \neq 0$). As is seen from Eqn (33), in these two cases $|R_{\pm}| = 1$ and hence Eqn (32) can be rewritten as

$$R_{\pm} = \exp(i\Phi)\,. \quad (35)$$

Here $\Phi = \Phi_0 + 2\beta k\Delta$ characterises phase difference between the incident and reflected light at the point $z = 0$, with $\Delta = 4\pi\chi_0^{(s)}/(1 - \varepsilon)$ and

$$\tan\Phi_0 = \frac{2n_0\sin\tilde{\psi}}{(n_0^2 - 1) + (n_0^2 + 1)\cos\tilde{\psi}}\,, \quad (36)$$

where $\tilde{\psi} = 2k\sqrt{\varepsilon}\tilde{L}$ with $\tilde{L} = L - \beta\Delta$.

It is seen from Eqns (35) and (36) that the influence of the inhomogeneity in the near-surface electric permittivity $\chi_0^{(s)}$, which is taken into account by the correct Landau – Lifshitz approach, is reduced for an isotropic medium to a change (easily explained in physical terms) in the optical length of the medium. (This is equivalent to a small shift of its front surface by an amount Δ).

The change in the optical length of the medium leads to a phase change for the reflected and transmitted waves compared with the phases calculated by using the traditional 'symmetric' boundary conditions (16) and (17) [see Eqns (35) and (36) for $\beta = 0$]. This is caused by the fact that, as was pointed out in Section 3.2, the boundary conditions (17) do not fully take into account inhomogeneity of the optical properties of the medium inside the layer near the surface.

The shortcomings of the 'symmetric' approach can however, be removed if instead of boundary condition (17), one uses the precise boundary condition (18). In that case the results obtained by using the correct 'symmetric' and Landau – Lifshitz approaches will be identical.

5. Conclusion

It is clear that the boundary conditions (16) and (18) and conditions (21) (each in conjunction with the corresponding constitutive relations and Maxwell equations) reflect the existence of one and the same thing, the surface polarisation current, and, hence, are essentially equivalent.

A frequently encountered mistake, which is repeated to one or another extent by many people discussing electrodynamics of bounded media with spatial dispersion [2, 7, 14, 23], is reduction of the problem to the question: which constitutive relations, symmetric or asymmetric, are correct or 'the most' correct? Both are correct, but each of them requires its own boundary conditions.

The reason is that by the intrinsic logic of electrodynamics, first come the Maxwell equations, then the constitutive relations, and only after that boundary conditions, expressions for energy, Umov – Poynting vector etc., which are consequences of the Maxwell equations and the constitutive relations and thus vary with the latter. Moreover, it proves that when considering the boundary problems it is convenient and often necessary to introduce a new constitutive relation — the equation for the surface polarisation current!

Each of the approaches considered has certain advantages. The 'symmetric' constitutive relations appear to be convenient for considering electrodynamics of moving or magnetic media [2, 42]. For solving problems on radiation interaction with nonlinear crystals without magnetic structures, the most effective is the Landau – Lifshitz approach [27, 36, 37, 43] complemented, if necessary, by the corresponding constitutive relation (23) for the surface polarisation current (see also [9, 10]).

Acknowledgements. The authors acknowledge Professor N I Koroteev, Dr. K N Drabovich and S N Volkov for useful discussions.

The work has been carried out with partial support from the Russian Foundation for Basic Research (project 93-02-15026).

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