PHENOMENOLOGICAL ELECTRODYNAMICS OF GYROTROPIC MEDIA

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Submitted April 12, 1972

Zh. Eksp. Teor. Fiz. 63, 838–843 (September, 1972)

Some comments on the phenomenological electrodynamics of gyrotropic media are made with regard to the problems discussed in the literature. An important problem is that of obtaining boundary conditions with an accuracy to terms of the order of a/λ (a is the atomic size and λ the wavelength). The corresponding boundary conditions cannot be derived from the field equations and material equations for space and contain unknown functions of frequency which can be calculated only within the framework of the microscopic theory.

ALTHOUGH the phenomenological electrodynamics of gyrotropic media has been under development for more than a decade, questions concerning this field continue to be under discussion (see the article of Bokut' and Serdyukov[1], where references to a number of other articles are also given). Concretely, in[1] they deal with the form of the "material equations" in a gyrotropic medium, expressions for the density and for the energy flux, and finally, the boundary conditions. An important question in this case is that of the boundary conditions with allowance for terms of order a/λ (a is the atomic dimension and λ is the wavelength), i.e., terms of the same order as the gyrotropic effect itself. Since it is precisely here that we disagree with the statements made in[1] and a few other articles, it is appropriate to present here the corresponding remarks, and to elucidate the question in a sufficiently systematic form.

1. Gyrotropy is a spatial-dispersion effect, and should therefore naturally be considered within the framework of general electrodynamics of media with spatial dispersion. The corresponding field equations can be written in the form

$$\operatorname{rot}\mathbf{B}=\frac{1}{c}\frac{\partial\mathbf{D}}{\partial t}+\frac{4\pi}{c}\mathbf{j}_{ext},\tag{1}$$

$$\operatorname{div}\mathbf{D}=4\pi\rho_{ext},\tag{2}$$

$$\operatorname{rot}\mathbf{E}=-\frac{1}{c}\frac{\partial\mathbf{B}}{\partial t},\tag{3}$$

$$\operatorname{div}\mathbf{B}=0,\tag{4}$$

where $\mathbf{j}_{ext}$ and ρ_{ext} are respectively the current and charge densities corresponding to the external sources, and to the remaining notation is quite obvious and agrees, for example, with that of[2]. It must be emphasized only that we do not distinguish between the magnetic induction **B** and the magnetic field intensity **H**, since these quantities coincide in general in the case of the optical frequencies considered here (see[3]; ferromagnets are a possible exception[4]). All the properties of the medium are reflected in the "material equation," the connection between **D** and **E**. Within the framework of the linear theory, this connection is in general of the form

$$D_i(\mathbf{r},t)=\int_{-\infty}^{t}dt'\int d\mathbf{r}'\hat{\varepsilon}_{ij}(t,t',\mathbf{r},\mathbf{r}')E_j(\mathbf{r}',t').\tag{5}$$

This form takes into account the possible (and actually existing) dependence of the electric induction **D** on **B**, since the field **B** can be expressed in terms of **E** with the aid of (3), and by the same token its influence is reflected on the form of the kernel $\hat{\epsilon}_{ij}$ in the integral equation (5).

If the medium is homogeneous (invariant) in time and in space, then the kernel $\hat{\epsilon}_{ij}$ depends only on the differences $\tau = t - t'$ and $\mathbf{R} = \mathbf{R} - \mathbf{r}'$. Under these conditions, it is possible to introduce the permittivity tensor $\epsilon_{ij}(\omega, \mathbf{k})$ so that

$$D_i(\omega,\mathbf{k})=\varepsilon_{ij}(\omega,\mathbf{k})E_j(\omega,\mathbf{k}),\tag{6}$$

$$\varepsilon_{ij}(\omega,\mathbf{k})=\int_0^\infty d\tau\int d\mathbf{R}\,e^{-i(\mathbf{kR}-\omega\tau)}\,\hat{\varepsilon}_{ij}(\tau,\mathbf{R}),\tag{7}$$

with

$$E_j(\mathbf{r},t)=\int E_j(\omega,\mathbf{k})\,e^{i(\mathbf{kr}-\omega t)}\,d\omega\,d\mathbf{k}$$

and analogously for other quantities.

From the principle of the symmetry of the kinetic coefficients it follows that in the general case

$$\varepsilon_{ij}(\omega,\mathbf{k},\mathbf{B}_{ext})=\varepsilon_{ij}(\omega,-\mathbf{k},-\mathbf{B}_{ext}),\tag{8}$$

$\mathbf{B}_{ext}$ is either the external magnetic induction or a parameter characterizing the magnetization of the lattice or a sublattice of the magnetic medium, i.e., of a ferro- or antiferromagnet (strictly speaking, we have in mind here only antiferromagnets, since we have put **B** = **H**).

In the optical region, the effects of spatial dispersion are in general small, since they are characterized by the parameter a/λ (gyrotropic medium) or even $(a/\lambda)^2$ (nongyrotropic medium). In a gyrotropic medium it is therefore usually sufficient to use the expansion

$$\varepsilon_{ij}(\omega,\mathbf{k})=\varepsilon_{ij}(\omega)+i\gamma_{ijl}(\omega)k_l\tag{9}$$

or the analogous expansion for the reciprocal tensor $\epsilon^{-1}_{ij}(\omega, \mathbf{k})$. By virtue of (8) we have

$$\varepsilon_{ij}(\omega,\mathbf{B}_{ext})=\varepsilon_{ji}(\omega,-\mathbf{B}_{ext}),\quad\gamma_{ijl}(\omega;\mathbf{B}_{ext})=-\gamma_{jil}(\omega,-\mathbf{B}_{ext}).\tag{10}$$

In the absence of absorption and for real ω and **k**, the tensor $\epsilon_{ij}(\omega, \mathbf{k})$ is Hermitian, i.e.,

$$\varepsilon_{ij}(\omega,\mathbf{k})=\varepsilon_{ji}^*(\omega,\mathbf{k}),$$
$$\varepsilon_{ij}(\omega)=\varepsilon_{ji}^*(\omega),\quad\gamma_{ijl}(\omega)=-\gamma_{jil}^*(\omega),\tag{11}$$

where the variable $\mathbf{B}_{ext}$, as above, has been omitted when it does not change sign.

If we write the tensor γ_{ijl} in the form $\gamma_{ijl} = \gamma_{ijl} + i\gamma''_{ijl}$, $\gamma'_{ijl} = \mathrm{Re}\,\gamma_{ijl}$, $\gamma''_{ijl} = \mathrm{Im}\,\gamma_{ijl}$, then we have in a non-absorbing medium as a result of (11)

$$\gamma'_{ijl} = -\gamma'_{jil}, \quad \gamma''_{ijl} = \gamma''_{jil}. \tag{12}$$

For a nonmagnetic medium (at $\mathbf{B}_{ext} = 0$), by virtue of (10), we have $\gamma_{ijl} = -\gamma_{ijl}$, and obviously $\gamma''_{ijl} = 0$. In a transparent magnetic medium, generally speaking, $\gamma''_{ijl} \neq 0$, a fact with which the relatively recently noted interesting effect of gyrotropic birefringence in antiferromagnetic crystals is connected[5-7].

The foregoing scheme for constructing the electrodynamics of gyrotropic media is in our opinion of maximum simplicity, completeness, and economy. In particular, we see no advantages whatever in writing down the material equation in a more symmetrical form, such as (see[1,6])

$$D_i = \varepsilon_{ij}(\omega, \mathbf{k})E_j + \alpha_{ij}(\omega, \mathbf{k})H_j, \quad B_i = \beta_{ij}(\omega, \mathbf{k})E_j + \mu_{ij}(\omega, \mathbf{k})H_j. \tag{13}$$

Both forms, (6) and (13), are uniquely related with each other[1,6], and the greater symmetry of (13) with respect to the electric and magnetic quantities is illusory (particularly within the framework of the considered three-dimensional formulation of electrodynamics) because of the absence of magnetic charges and currents, and also because the vectors **E** and **D** behave differently from **B** and **H** upon inversion of the spatial coordinates and the time. What is principal, however, by virtue of the considerations advanced in[3], is that in the optical frequency region one sees no way of physically distinguishing the magnetization $\mathbf{M} = (\mathbf{B} - \mathbf{H})/4\pi$ from a certain polarization current $\partial \mathbf{P}/\partial t = c\,\mathrm{curl}\,\mathbf{M}$ (the arguments presented in[3] are generally speaking incorrect in the case of ferromagnets[4], but remain in force for gyrotropic bodies, where the coefficients $\boldsymbol{\gamma}_{ijl}$ or $\boldsymbol{\alpha}_{ij}$ and $\boldsymbol{\beta}_{ij}$ are all of the order of a/λ). There are therefore even less grounds for introducing the indistinguishable parts of the induction **D**, defined separately by the fields **E** and **B**.

2. The articles[1,6] can, however, give the impression that the use of quantities such as (13) offer us advantages from the point of view of simplicity of the boundary conditions (we have in mind here the boundary separating a gyrotropic medium 2 from vacuum 1). Actually, however, in[1,6] the boundary conditions for the constraints in the form (13) are simpler, and, concretely, the equality of the components of the fields **E** and **H** tangential to the interface are simply postulated. Then the boundary conditions obtained for the field **B** and **D** from (1) and (4) are somewhat more complicated.

Equations (1)—(4), as is well known, lead in the general case to the boundary conditions (**n** is the normal to the sharp separation boundary between media 1 and 2; allowance for a transition layer will be discussed later on)

$$[\mathbf{n}(\mathbf{B}_2 - \mathbf{B}_1)] = 4\pi c^{-1}\mathbf{i}, \quad D_{2n} - D_{1n} = 4\pi\sigma, \tag{14}$$

$$[\mathbf{n}(\mathbf{E}_2 - \mathbf{E}_1)] = 0, \quad B_{2n} - B_{1n} = 0, \tag{15}$$

where **i** and σ are the densities of the surface currents and charges (it is assumed that there are no external surface currents and charges $\mathbf{i}_{ext}$ and σ_{ext}). We note furthermore that the additional terms that can appear in the presence of higher derivatives are arbitrarily referred to **i** and σ, and consequently do not enter explicitly in (14) (this will also be discussed below). The connection of the quantities **i** and σ with the field vectors is determined by (5), which can be resolved fully only within the framework of the microscopic theory[1)]. Neglecting spatial dispersion and absorption, we can simply put $i = \sigma = 0$, but when spatial dispersion is taken into account there are no longer general grounds for this procedure. To avoid cumbersome expressions, we confine ourselves below to the case of an isotropic but gyrotropic medium, neglecting the second derivatives with respect to the coordinates. In this case the quite general connection between **D** and **E** is

$$\begin{aligned}\mathbf{D} &= \varepsilon\mathbf{E} + \delta_{\mathrm{I}}\,\mathrm{rot}\,\mathbf{E} + \mathrm{rot}\,(\delta_{\mathrm{II}}\mathbf{E}) \\ &= \varepsilon\mathbf{E} + (\delta_{\mathrm{I}} + \delta_{\mathrm{II}})\,\mathrm{rot}\,\mathbf{E} + [\nabla\delta_{\mathrm{II}}\mathbf{E}],\end{aligned} \tag{16}$$

where ϵ, δ_{I}, and δ_{II} are functions of the coordinates **r** and of the frequency ω, where δ_{I} and δ_{II} are pseudoscalars that differ from zero only if the medium has no symmetry center. Of course, in a homogeneous medium we have

$$\mathbf{D} = \varepsilon\mathbf{E} + \delta\,\mathrm{rot}\,\mathbf{E}, \quad \delta = \delta_{\mathrm{I}} + \delta_{\mathrm{II}}. \tag{17}$$

In an inhomogeneous medium, which must be considered on going to the limiting case of a separation boundary, we see, however, no grounds of general (phenomenological) character that would allow us to put $\delta_{\mathrm{I}} = 0$ or $\delta_{\mathrm{II}} = 0$. From (1), (2), and (16) we arrive in the usual manner[2)] at the following boundary conditions (we have put $j_{ext} = \sigma_{ext} = 0$; the conditions (15) remain in force in this case):

$$\begin{gathered}[\mathbf{n}(\mathbf{B}_2 - \mathbf{B}_1)] = \frac{\delta_{\mathrm{II},2}}{c}\left[\mathbf{n}\left(\frac{\partial E}{\partial t}\right)_2\right], \\ \varepsilon E_{2n} - E_{1n} + \delta_{\mathrm{I},2}\,\mathrm{rot}_n\,\mathbf{E}_2 = 0,\end{gathered} \tag{18}$$

Since in vacuum we have $\delta_{\mathrm{I},1} = \delta_{\mathrm{II},1} = 0$ and $\epsilon_1 = 1$.

Since the coefficients δ_{I} and δ_{II} are not known, we cannot find in general form fields of the type (13) for which the simplest boundary conditions (14) with $i = \sigma = 0$, used in[1,6], hold.

From (1) and (3) we get Poynting's theorem

$$\frac{1}{4\pi}\mathbf{E}\frac{\partial\mathbf{D}}{\partial t} + \frac{1}{4\pi}\mathbf{B}\frac{\partial\mathbf{B}}{\partial t} = -\frac{c}{4\pi}\mathrm{div}[\mathbf{EB}] - \mathbf{j}_{ext}\mathbf{E}. \tag{19}$$

substituting (16) in (19) and neglecting, for simplicity, the frequency dispersion and the absorption (i.e., the dependence of ϵ and $\delta_{\mathrm{I,II}}$ on ω), we obtain

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*$\nabla\delta_{\mathrm{II}}\mathbf{E}] = \nabla\delta_{\mathrm{II}} \times \mathbf{E}$.

1)Conditions (14) and the relation (5) are sufficient to solve any boundary-value problem and, for example, the introduction of additional boundary conditions in media with spatial dispersion is only a device of limited significance (for details see [2], Sec. 10; solutions of boundary-value problems with allowance for spatial dispersions is discussed also in [8]).

2)To obtain the boundary conditions on a sharp boundary it is necessary to integrate the corresponding equations over the thickness of the layer and this thickness tend to zero. This can give rise to additional terms in the presence of higher derivatives. In the case of (16), for example, div D = 0 leads to $D_{2n} - D_{1n} = \delta_{\mathrm{II},2}\,\mathrm{curl}_n\mathbf{E}_2$.

$$\frac{\partial}{\partial t}\left\{\frac{\varepsilon E^2+B^2+\delta(\mathbf{E}\,\mathrm{rot}\,\mathbf{E})}{8\pi}\right\} = -\frac{c}{4\pi}\,\mathrm{div}\left\{[\mathbf{EB}]-\frac{\delta}{2c}\left[\mathbf{E}\frac{\partial\mathbf{E}}{\partial t}\right]\right\} + \frac{\nabla(2\delta_{II}-\delta)}{8\pi}\left[\mathbf{E}\frac{\partial \mathbf{E}}{\partial t}\right] - \mathbf{j}_{ext}\mathbf{E}. \qquad (20)$$

The conditions (15) and (18) obviously ensure conservation of the energy balance for any value of the coefficient $\delta_{II,2}$. Thus, as expected, the energy conservation law (the Poynting theorem) imposes no limitations whatever on the choice of the boundary conditions[3)]. We note that in[2], Sec. 10.9, we applied to a gyrotropic medium the boundary condition (18) with $\delta_{II} = 0$. Since in this case the incident wave was assumed to be linearly polarized, allowance for terms of order of δ_{II} in the boundary conditions would affect only the terms of order δ_{II}^2 in the expression for the amplitudes of the reflection and transmission coefficients (for linearly polarized waves, the terms with $\mathbf{E} \times d\mathbf{E}/dt$ in (20) and (21) vanish). In the general case of arbitrarily polarized waves, the choice of the boundary conditions (i.e., for example, the use of a fixed value $\delta_{II,2}$, which is important only in the case of normal incidence of the waves on the medium) affects the intensity of the reflected wave already in the first order in δ_{II}. An investigation of the gyrotropy in a volume (we have in mind primarily measurement of the rotation of the plane of polarization) makes it possible to determine the coefficient $\delta = \delta_I + \delta_{II}$. In normal incidence of waves on a gyrotropic medium, the boundary conditions, as already mentioned (see also (18)), depends only on δ_{II}.

Thus, it is seemingly possible to determine experimentally both coefficients δ_I and δ_{II}, and also other coefficients that appear in the more general case of an anisotropic gyrotropic medium. Unfortunately, the situation is physically much more complicated and less favorable, and calls for a concrete analysis with allowance for the physical properties of the interface. The point is that in the case of reflection from even a non-
The presence of a term proportional to $\nabla(2\delta_{II} - \delta)$ is very important when account is taken of the interface at which this term becomes discontinuous. The fact that such a term appears for an inhomogeneous gyrotropic medium even in the absence of absorption is, of course, quite remarkable; however, in light of the remarks made in[2], Sec. 3, this circumstance is not specially surprising.

Within the limits of a homogeneous medium, the energy flux is

$$\mathbf{S} = \frac{c}{4\pi}[\mathbf{EB}] - \frac{\delta}{8\pi}\left[\mathbf{E}\frac{\partial\mathbf{E}}{\partial t}\right]$$

and it is clear at the same time from (20) that in the stationary case the energy balance is not reduced to equality of the fluxes $S_{2n} = S_{1n}$ on the two sides of the boundary. There follows instead from (20) the energy conservation law (the medium 1 is assumed to be vacuum):

$$S_{2n}-\frac{(2\delta_{II,2}-\delta_2)}{8\pi}\left[\mathbf{E}\frac{\partial\mathbf{E}}{\partial t}\right]_{2n} = \frac{c}{4\pi}[\mathbf{EB}]_{2n} - \frac{\delta_{II,2}}{4\pi}\left[\mathbf{E}\frac{\partial \mathbf{E}}{\partial t}\right]_{2n} = S_{1n} = \frac{c}{4\pi}[\mathbf{EB}]_{1n}. \qquad (21)$$

gyrotropic medium, the Fresnel formulas are approximate and require corrections of the order of l/λ, where l is the characteristic length, for example the effective thickness of the transition layer or, one might say, the physical thickness of the interface (for details see[9-11]). When account is taken of the transition layer, additional terms of order l/λ appear in the boundary conditions (14) and (15) (these terms were not taken into account at all in[1]). The atomic dimension $a \sim 3 \times 10^{-8}-10^{-7}$ is, generally speaking, the minimal value of l. At the same time, the coefficients δ, δ_I and δ_{II} in a gyrotropic medium are also just of the order of a. It is clear therefore that in the general case one can assume that different effects leading to deviation from the Fresnel formulas (see, in particular,[11]) exceed the gyrotropic effects (in particular, corrections of order δ_{II} in the boundary conditions) on the interface. An investigation of the gyrotropy by determining the ellipticity of the reflected waves (see[12,1]) should therefore become much more complicated when the aforementioned factors are taken into account[9-11].

This conclusion does not mean, however, that an investigation of gyrotropy in reflected light or in general in the presence of boundaries is not worthwhile. To the contrary, we assume that the corresponding analysis is essential and can lead to interesting results, but it should be carried out both with allowance of the "transition" layer[9-11] and with allowance for the most general boundary conditions (for example, conditions of the type (18) with arbitrary δ_I and δ_{II}; for an isotropic gyrotropic medium, the corresponding analysis is given in[13]). It is also necessary to take into account here the frequency dependence of all the quantities, which is particularly strong near the frequencies of surface excitons of different types (see[2], Sec. 10.10). For estimating purposes, it is also of interest to calculate the values of δ_I and δ_{II} (see above) for different albeit crude microscopic models of the medium.

3)The term

$$A = \frac{\nabla(2\delta_{II}-\delta)}{8\pi}\left[\mathbf{E}\frac{\partial\mathbf{E}}{\partial t}\right]$$

vanishes for linearly polarized waves, and also in the general case if $\delta_I = \delta_{II} = \delta/2$. If $A \neq 0$, then part of the energy of the incident wave should go over on the separation boundary, for example, into some surface waves (see [14]) in this connection. The requirement $\delta_I = \delta_{II}$ has thus a definite physical meaning. It seems to us, however, that such a requirement is not obligatory.

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Translated by J. G. Adashko
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