ON THE PHENOMENOLOGICAL THEORY OF NATURAL OPTICAL ACTIVITY

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The interrelationship between different formulations of the constitutive relations is investigated and the form of the boundary conditions is determined for the phenomenological electrodynamics of non-absorbing optically active media.

1. THE proportionality, established in the microscopic theory[1,2] of the parameters responsible for the electric and magnetic parts of optical activity in the phenomenological theory was known only for an isotropic medium.[3] For anisotropic media a term containing the magnetic field gradients was introduced into the constitutive relation for magnetic induction in [4,5], but without assuming that a relation exists between the tensors of magnetic and electric activity. Recent investigations of electromagnetic field energy conservation[6-8] have led to this kind of relationship in the phenomenological constitutive relations for crystals. In these studies it was shown that the electric and magnetic parts of the optical activity are described by a single pseudotensor α of second order and that, analogously to the case of an isotropic medium,[3] the constitutive relations have the form

$$\mathbf{D} = \varepsilon(\mathbf{E} + \alpha \operatorname{rot} \mathbf{E}), \qquad \mathbf{B} = \mu(\mathbf{H} + \tilde{\alpha} \operatorname{rot} \mathbf{H}) \tag{1}$$

(the real tensors ϵ and μ are symmetric: $\tilde{\epsilon} = \epsilon$, $\tilde{\mu} = \mu$); Maxwell's equations are formulated for the four vectors **E**, **D**, **H**, and **B**:

$$\operatorname{rot} \mathbf{E} = -\frac{1}{c}\dot{\mathbf{B}}, \tag{2}$$

$$\operatorname{div} \mathbf{B} = 0, \tag{3}$$

$$\operatorname{rot} \mathbf{H} = \frac{1}{c}\dot{\mathbf{D}}, \tag{4}$$

$$\operatorname{div} \mathbf{D} = 0. \tag{5}$$

From (2) and (4) in conjunction with (1) we obtain the conservation law of electromagnetic energy with the Poynting vector $c[\mathbf{E} \times \mathbf{H}]/4\pi$ and the energy density $(\mathbf{D}\epsilon^{-1}\mathbf{D} + \mathbf{B}\mu^{-1}\mathbf{B})/8\pi$.[8]

Equations (1) differ from the conventional constitutive relations for optically active media (see [9,10], for example). In [9] the form of the conservation law of electromagnetic energy was not clarified, and the energy analysis in [4] has incurred an objection[7] regarding the form of the Poynting vector. It should also be noted that the solution of the boundary problem using the constitutive relations[4] and the customary continuity conditions for the tangential components of the electric and magnetic vectors at the interface between two media has led to difficulties with the conservation of the angular momentum of a free electromagnetic field[11] and with the balance requirement for electromagnetic energy flow when radiation traverses an optically active layer.[12]

The possibility of the new formulation (1) of the constitutive relations, free of the mentioned contradictions, has led to doubt[7] regarding the correctness of the phenomenological treatment of natural optical activity that is found in [9].

In the present communication we establish the interrelationship between different formulations of the constitutive relations and field equations in optically active crystals, analyze the energy relations using the approach of [9] in a way different from [4,10], and show that incorrect boundary conditions are a source of several difficulties in the electrodynamics of the given media.

2. In the phenomenological constitutive relations and field equations of [9], in contrast with (1)–(5), terms that appear when microscopic currents are averaged and that are responsible for the optical activity are included in the definition of electric induction. We shall apply the same procedure directly in the system of phenomenological equations (1)–(5) by redefining the vectors of electric induction and magnetic field strength (see also [13]).

From (1) and (4) we obtain

$$\mathbf{H} = \mu^{-1}\mathbf{B} - \frac{\tilde{\alpha}}{c}\dot{\mathbf{D}}, \tag{6}$$

which we use to write Maxwell's equations (4) and (5):

$$\operatorname{rot} \mathbf{H}' = \frac{1}{c}\dot{\mathbf{D}}', \tag{7}$$

$$\operatorname{div} \mathbf{D}' = 0. \tag{8}$$

Here the redefined vectors of the magnetic field **H'** and electric induction **D'** are

$$\mathbf{H}' = \mu^{-1}\mathbf{B}, \tag{9}$$

$$\mathbf{D}' = \mathbf{D} + \operatorname{rot} \tilde{\alpha}\mathbf{D}. \tag{10}$$

By virtue of (1) and some simple transformations the last equation becomes[1)]

$$\mathbf{D}' = \varepsilon\mathbf{E} + [\gamma\nabla, \mathbf{E}], \tag{11*}$$

where the second-order tensor γ of optical activity is related to the tensors α and ϵ in (1) by

$$\gamma = \operatorname{Sp}(\tilde{\alpha}\varepsilon) - \tilde{\alpha}\varepsilon. \tag{12}$$

*$[\gamma\nabla, \mathbf{E}] \equiv \gamma\nabla \times \mathbf{E}$.

1)Here and subsequently we retain only first-degree terms in the parameters of the activity.

The inverse relation, which expresses the tensor α in terms of γ, will be

$$\alpha = \varepsilon^{-1}(1/2\,\mathrm{Sp}\,\gamma - \tilde{\gamma}). \qquad (13)$$

It follows from (12) and (13) that the tensor γ has the same form as α for different classes of crystal symmetry. In particular, for an isotropic medium (12) and (13) are reduced to the equality $\gamma = 2\epsilon\alpha$.

For a nonmagnetic medium $\mu = 1$ and $\mathbf{B} = \mathbf{H}'$. Consequently, instead of (7) we write

$$\mathrm{rot}\,\mathbf{B} = \frac{1}{c}\dot{\mathbf{D}}'. \qquad (14)$$

The discussion of natural optical activity in [9] was based on Maxwell's equations in the forms (2) and (14) and the constitutive relation (11), which was obtained using the principle of the symmetry of kinetic coefficients. We thus establish the equivalence of the two formulations of the constitutive relations and remove the objections of [7] against the procedure of [9].

3. The energy analysis of the electromagnetic field in an optically active medium with the redefined electric induction and magnetic field vectors proceeds as follows. Using the identity

$$[\gamma\nabla,\ \mathbf{E}] = \mathrm{Sp}\,\gamma\,\mathrm{rot}\,\mathbf{E} - \mathrm{rot}\,\gamma\mathbf{E} - \tilde{\gamma}\,\mathrm{rot}\,\mathbf{E},$$

we can write (11) in the form

$$\mathbf{D}' = \varepsilon\mathbf{E} + (1/2\,\mathrm{Sp}\,\gamma - \tilde{\gamma})\,\mathrm{rot}\,\mathbf{E} + \mathrm{rot}(1/2\,\mathrm{Sp}\,\gamma - \gamma)\mathbf{E}.$$

Equation (7) can now assume the form

$$\mathrm{rot}\left\{\mathbf{H}' - \frac{1}{c}\left(\frac{1}{2}\mathrm{Sp}\,\gamma - \gamma\right)\dot{\mathbf{E}}\right\} = \frac{1}{c}\frac{\partial}{\partial t}\left\{\varepsilon\mathbf{E} - \frac{1}{c}\left(\frac{1}{2}\mathrm{Sp}\,\gamma - \tilde{\gamma}\right)\dot{\mathbf{B}}\right\}. \qquad (15)$$

Scalar multiplication of the left- and right-hand sides of (2) by $\mathbf{H}' - c^{-1}(1/2\,\mathrm{Sp}\,\gamma - \gamma)\mathbf{E}$, similar multiplication of (15) by $\mathbf{E}$, and subtraction of the second result from the first yields

$$\mathrm{div}\frac{c}{4\pi}\left[\mathbf{E}\left\{\mathbf{H}' - \frac{1}{c}\left(\frac{1}{2}\mathrm{Sp}\,\gamma - \gamma\right)\dot{\mathbf{E}}\right\}\right] = -\frac{\partial}{\partial t}\frac{1}{8\pi}\left\{\mathbf{E}\varepsilon\mathbf{E} + \mathbf{B}\mu^{-1}\mathbf{B} - \frac{2}{c}\mathbf{E}\left(\frac{1}{2}\mathrm{Sp}\,\gamma - \tilde{\gamma}\right)\dot{\mathbf{B}}\right\}. \qquad (16)$$

This equation shows that, starting from the traditional form of the constitutive relations for the electromagnetic field in optically active media, we can represent energy conservation with the Poynting vector

$$\mathbf{S} = \frac{c}{4\pi}[\mathbf{E}\mathbf{H}'] - \frac{1}{4\pi}\left[\mathbf{E}, \left(\frac{1}{2}\mathrm{Sp}\,\gamma - \gamma\right)\dot{\mathbf{E}}\right], \qquad (17)$$

which for the redefined fields, in accordance with (6) and (9), assumes the customary form $c[\mathbf{E}\times\mathbf{H}]/4\pi$.

4. We shall now consider the conditions imposed on an electromagnetic field at the interface of optically active media; more exactly, we shall discuss the changes of the conventional boundary conditions that result from redefining the magnetic field and electric induction. For two contiguous media I and II, the boundary conditions for the electric and magnetic vectors of a free electromagnetic field will have the customary forms

$$[(\mathbf{E}_\mathrm{I} - \mathbf{E}_\mathrm{II})\mathbf{q}] = 0, \qquad (18)$$

$$[(\mathbf{H}_\mathrm{I} - \mathbf{H}_\mathrm{II})\mathbf{q}] = 0, \qquad (19)$$

which are based on the continuity requirement for the normal component of energy flow density ($\mathbf{q}$ is the vector of the normal to the interface).

The boundary condition for the redefined magnetic field vector $\mathbf{H}'$ can be obtained from (19). Using (6), (9), and (13), we obtain from (19) the condition

$$[(\mathbf{H}_\mathrm{I}' - \mathbf{H}_\mathrm{II}')\mathbf{q}] = \frac{1}{c}\left[\left\{\left(\frac{1}{2}\mathrm{Sp}\,\gamma_\mathrm{I} - \gamma_\mathrm{I}\right)\dot{\mathbf{E}}_\mathrm{I} - \left(\frac{1}{2}\mathrm{Sp}\,\gamma_\mathrm{II} - \gamma_\mathrm{II}\right)\dot{\mathbf{E}}_\mathrm{II}\right\}\mathbf{q}\right], \qquad (20)$$

which must be obeyed by the redefined vector $\mathbf{H}'$ (the magnetic induction $\mathbf{B}$ for $\mu = 1$) at the boundary between two optically active media. Similarly, from the usual continuity condition for the normal component of electric induction $\mathbf{D}$, by using (10) and (13) we obtain the boundary condition for the redefined induction:

$$(\mathbf{D}_\mathrm{I}' - \mathbf{D}_\mathrm{II}')\mathbf{q} = \mathrm{rot}\{(1/2\,\mathrm{Sp}\,\gamma_\mathrm{I} - \tilde{\gamma}_\mathrm{I})\mathbf{E}_\mathrm{I} - (1/2\,\mathrm{Sp}\,\gamma_\mathrm{II})\mathbf{E}_\mathrm{II}\}\mathbf{q}. \qquad (21)$$

The relations (20) and (21) together with (18) and the usual continuity condition for the normal component of $\mathbf{B}$ comprise the boundary conditions for the electromagnetic field in the traditional description of natural optical activity.

5. When (9) and (11) are used in the theory of the optical activity of crystals it is often overlooked that $\mathbf{H}'$ and $\mathbf{D}'$ are redefined terms different from the usual vectors $\mathbf{H}$ and $\mathbf{D}$ in (1), and that (19) is used as the boundary condition for $\mathbf{H}'$ instead of (20). This last circumstance has important consequences. When the terms with optical activity in (20) are dropped, solutions of the boundary problem are obtained for which balance of the energy flow is not fulfilled at the interface between the optically active and inactive media. This affects the polarization obtained for the reflected wave and the wave transmitted through the optically active medium.

We now refer to the investigation in [14] of the possibility of observing and measuring experimentally the optical activity parameter of transparent uniaxial crystals belonging to planar classes by measuring the ellipticity of the reflected wave. When the optic axis of a crystal is perpendicular to the plane of incidence, from (2), (7), (18), and the boundary condition (20) without its right-hand side we obtain the following expressions for the amplitude factors of the reflected wave:

$$a_1 = \frac{\eta - \eta_e}{\eta + \eta_e}a, \qquad b_1 = \frac{\varepsilon_0\eta - n^2\eta_0}{\varepsilon_0\eta + n^2\eta_0}b + \frac{2ik\alpha_1 n\eta|\mathbf{a}|}{(\eta + \eta_e)(\varepsilon_0\eta + n^2\eta_0)}a, \qquad (22)$$

where a and b are the amplitude factors of the incident wave, $\eta = \mathbf{m}\cdot\mathbf{q}$, $\eta_e = \mathbf{m}_e\cdot\mathbf{q}$, $\eta_0 = \mathbf{m}_0\cdot\mathbf{q}$, $\mathbf{m}_i = n_i\cdot\mathbf{n}$ is the refraction vector, n_i is the refractive index, $\mathbf{n}$ is the wave normal, k is the vacuum wave number, $\mathbf{a} = [\mathbf{m}\times\mathbf{q}]$, and α_1 is the activity parameter. When the complete boundary condition (20) is used the expression for b_1 remains unchanged, but for a_1 we obtain

$$a_1 = \frac{\eta - \eta_e}{\eta + \eta_e}a + \frac{2ik\alpha_1 n\eta|\mathbf{a}|}{(\eta + \eta_e)(\varepsilon_0\eta + n^2\eta_0)}b. \qquad (23)$$

It follows from (22) that for a = 0 (the electric vector of the incident wave oscillates in the incident plane) the reflected wave is polarized linearly, while for b = 0 (the electric vector of the incident wave oscillates in a plane perpendicular to the incident plane) the reflected wave is polarized elliptically with the semiaxes of the ellipse in the ratio

$$\frac{x}{y} = \pm \frac{2\alpha_1 kn\eta |\mathbf{a}|}{(\eta - \eta_e)(\varepsilon_0\eta + n^2\eta_0)}.$$

At the same time, it follows from (23) that for $a = 0$ the reflected wave should be elliptically polarized with the ellipticity

$$\frac{x}{y} = \pm \frac{2\alpha_1 kn\eta |\mathbf{a}|}{(\eta + \eta_e)(\varepsilon_0\eta - n^2\eta_0)}.$$

Consequently the optical activity parameter α_1 can be measured both for $b = 0$ and $a = 0$.

6. The equations for a free electromagnetic field in a medium are invariant under the so-called dual transformations $\mathbf{D} \to \pm\mathbf{B}$, $\mathbf{B} \to \mp\mathbf{D}$, $\mathbf{E} \to \pm\mathbf{H}$, $\mathbf{H} \to \mp\mathbf{E}$. It appears most natural to formulate the constitutive relations consistently with the dual symmetry that is an internal property of Maxwell's equations. Equations (1)–(5) are invariant under the transformations $\mathbf{D} \to \pm\mathbf{B}$, $\mathbf{B} \to \mp\mathbf{D}$, $\mathbf{E} \to \pm\mathbf{H}$, $\mathbf{H} \to \mp\mathbf{E}$ and $\epsilon \rightleftarrows \mu$, $\alpha \to \bar{\alpha}$.

An electromagnetic wave in an optically active medium is described more conveniently by means of (1)–(5) than by (2), (3), (7)–(9), (11), in virtue of the obviously simple boundary conditions and Poynting vector expression, and also because of the symmetric form of the constitutive relations, although in the second version the properties of a nonmagnetic medium ($\mu = 1$) can be described using the single constitutive relation (11).[9]

We note that the transition from (1) to the not dually symmetric forms (9), (11) of the constitutive relations is not unique. A different formulation [the dual of (2), (3), (7)–(9), (11)] of the constitutive relations and field equations can result from redefinition of the electric field strength and the magnetic induction. Expressing $\mathbf{E}$ in terms of $\mathbf{D}$ and $\mathbf{B}$ in accordance with (1) and (2), and substituting into (2), we obtain

$$\operatorname{rot} \varepsilon^{-1}\mathbf{D} = -\frac{1}{c}(\dot{\mathbf{B}} + \operatorname{rot} \alpha\dot{\mathbf{B}}). \tag{24}$$

In virtue of (1) and (4) we can write (24) and (3) as

$$\operatorname{rot} \mathbf{E}' = -\frac{1}{c}\dot{\mathbf{B}}' \tag{25}$$

$$\operatorname{div} \mathbf{B}' = 0, \tag{26}$$

where

$$\mathbf{E}' = \varepsilon^{-1}\mathbf{D}, \tag{27}$$

$$\mathbf{B}' = \mu\mathbf{H} + [\beta\nabla, \mathbf{H}]. \tag{28}$$

Here the tensor β is defined by

$$\beta = \operatorname{Sp}(\alpha\mu) - \alpha\mu = \varepsilon^{-1}\tilde{\gamma}\mu - \operatorname{Sp}(\varepsilon^{-1}\tilde{\gamma}\mu) + {}^1\!/_2[\operatorname{Sp}(\varepsilon^{-1}\mu) - \varepsilon^{-1}\mu]\operatorname{Sp}\gamma. \tag{29}$$

Consequently, a free electromagnetic field in a nonabsorbing anisotropic optically active medium can be described by Maxwell's equations (25), (26) and the constitutive relations (27), (28) together with (4), (5). This version of the phenomenological description of optical activity is possible because the transition to macroscopic quantities can be performed by having the definition of magnetic induction include terms that appear when averaging the microscopic currents that are associated with optical activity. The field equations and constitutive relations in this form will obviously possess the same characteristics as (2), (3), (7)–(9), (11). Specifically and analogously with (20), (21), the boundary conditions for $\mathbf{E}'$ and $\mathbf{B}'$, in accordance with (18), (27), (28), (1), (29), will be

$$\begin{gathered}[(\mathbf{E}_{\mathrm{I}}' - \mathbf{E}_{\mathrm{II}}')\mathbf{q}] = -\frac{1}{c}\Big[\Big\{\Big(\frac{1}{2}\operatorname{Sp}\beta_{\mathrm{I}} - \beta_{\mathrm{I}}\Big)\dot{\mathbf{H}}_{\mathrm{I}} \\ -\Big(\frac{1}{2}\operatorname{Sp}\beta_{\mathrm{II}} - \beta_{\mathrm{II}}\Big)\dot{\mathbf{H}}_{\mathrm{II}}\Big\}\mathbf{q}\Big], \\ (\mathbf{B}_{\mathrm{I}}' - \mathbf{B}_{\mathrm{II}}')\mathbf{q} = \operatorname{rot}\{({}^1\!/_2\operatorname{Sp}\beta_{\mathrm{I}} - \dot{\beta}_{\mathrm{I}})\mathbf{H}_{\mathrm{I}} - ({}^1\!/_2\operatorname{Sp}\beta_{\mathrm{II}} - \dot{\beta}_{\mathrm{II}})\mathbf{H}_{\mathrm{II}}\}\mathbf{q}.\end{gathered} \tag{30}$$

The transformations $\mathbf{B} \to \pm\mathbf{D}$, $\mathbf{H}' \to \pm\mathbf{E}'$, $\mathbf{D}' \to \mp\mathbf{B}'$, $\mathbf{E} \to \mp\mathbf{H}$, $\mu \rightleftarrows \epsilon$, $\gamma \to \beta$ convert (2), (3), (7)–(9), (11), (18), (20), (21) into, respectively, (25), (26), (4), (5), (27), (28), (19), (30), (31). Thus all results obtained for one of these two versions of the phenomenological description of optical activity can be obtained automatically through dual transformations in the corresponding equations of the other version.

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