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The reflection of electromagnetic waves from uniaxial optically active media

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Abstract. This paper investigates the reflection of electromagnetic waves at the boundary between an isotropic non-active medium and a uniaxial optically active medium. The anisotropic gyrotropic medium is described by constitutive relations, invariant to duality transformations. A Fresnel-type boundary problem is solved in two geometries: (i) optical axis perpendicular to the reflecting boundary and (ii) optical axis perpendicular to the plane of incidence. The components of the electric field and the intensity of the reflected and refracted waves are determined. Differential reflection methods for studying the optical activity are discussed in the cases of: ordinary reflection of left and right circularly polarized waves; total reflection of circularly polarized waves; and total reflection of linearly polarized waves.

1. Introduction

Natural optical activity has been known since 1811 and measurements of optical rotatory power and circular dichroism have become traditional techniques for the study of optically active media. The reflection of electromagnetic waves from optically active media, however, has not been thoroughly investigated so far. Its theoretical study is of conceptual and practical interest since reflection measurements in some cases are the only possible method for the study of gyrotropy (in point groups C_{3v}, C_{4v} and C_{6v}) and in other cases are an alternative to the transmission techniques. The coherent process of specular reflection generates a sufficiently strong signal to be detected by standard voltage and current measurements (unlike Raman scattering [1], which requires photon-counting detection). The theoretical understanding of the effect of optical activity on the reflection would make possible the correct interpretation of experimental results from precise optical measurements where the presence of gyrotropy is not known in advance or where gyrotropy appears as a residual effect of mechanical and optical processing.

Considerable progress has been made in the theoretical treatment of the reflection from isotropic optically active media [2–9]: Fresnel amplitudes have been derived for transparent and absorbing isotropic media and differential reflection methods have been proposed [4, 6, 7, 10, 11]. Furthermore, the multibeam reflection from an isotropic chiral slab has been investigated [4, 6, 12, 13].

Many optically active materials, however, are intrinsically anisotropic or become anisotropic as a result of mechanical processing. Both the theoretical and the experimental studies of the optical activity of anisotropic media are much more difficult than the analogous studies of isotropic media. Although the gyrotropy of some anisotropic media is stronger than that of cubic crystals and isotropic materials, its contribution to the refractive indices is masked by the strong linear birefringence and dichroism (in directions different from the optical axis). This makes the standard transmission methods of gyrotropy investigation inapplicable and raises the question of the use of reflection studies. Moreover, there is a group of crystals possessing a 'weak' gyrotropy (crystals from the point groups C_{3v}, C_{4v}, C_{6v}) for which reflection spectroscopy is a unique method of gyrotropy study (owing to their antisymmetric gyrotropic tensor, these crystals exhibit neither optical rotation nor circular dichroism).

We were the first to investigate theoretically the effect of gyrotropy on the reflection of electromagnetic waves (EMWs) from a uniaxial optically active medium [6, 14] in two different geometries of experimental interest [15, 16]. The approach to the solution of the boundary problem, presented in [6, 14] is correct, but the analytical results for the Fresnel coefficients are not precise owing to the application of incorrect boundary conditions. In the paper of Silverman and Badoz [17] the problem of reflection of EMWs from a birefringent optically active medium is precisely studied but in a rather unrealistic simplifying approximation. The authors take into account the influence of the anisotropy only on the permittivity tensor and not on the gyrotropy tensor of the medium. We find the claims of the authors that this approximation is appropriate for intrinsically isotropic optically active materials with stress-induced birefringence not fully convincing, since the stress would surely induce anisotropy of the gyrotropic properties of the material too. Moreover this assumption in [17] automatically excludes from consideration the most interesting case of 'weak' gyrotropy.

The present paper treats the reflection of EMWs from a uniaxial, transparent, optically active medium taking into consideration the anisotropy both of the permittivity and of the gyrotropy tensors. A Fresnel-type boundary problem is solved where the form of the boundary conditions is determined by the form of the constitutive relations used for the description of the optical activity of the medium. This research is a natural continuation of our study of the reflection of electromagnetic waves from isotropic optically active media, presented in [7], which we shall denote as paper I. It proves that the strong birefringence in uniaxial media is not an obstacle for the manifestation of the much weaker gyrotropic effect. Differential reflection signals turn out to be linear in the components of the gyrotropic tensor and are within the sensitivity of the contemporary detection techniques. Reflection studies of anisotropic gyrotropic materials provide much richer spectroscopic information than analogous studies of isotropic gyrotropic materials [18].

Experimental investigations of anisotropic optically active materials by reflection spectroscopy are not numerous owing to the relatively weak gyrotropic effects and the extremely high requirements for precision of the crystal cut orientation and clean surface preparation in order to avoid artefacts. The 'weak' gyrotropy of CdS crystals (point group C_{6v}) in the excitonic region was investigated by reflection spectroscopy [15]. A successful study of the gyrotropy of an induced cholesteric solution was done by differential total reflection of linearly polarized EMWs [16]. The experiment in this case does not face the complications connected with surface preparation. Reflection studies of TeO_2 (point group D_4) illustrated the difficulties in the application of reflection methods [19]. A misalignment of only 1° of the optical axis and the normal to the reflecting boundary resulted in the complete masking of the gyrotropy effects by the linear birefringence.

The organization of this paper is as follows. In section 2 we discuss the correlation of the boundary conditions to the constitutive relations, a problem correctly solved by Fedorov [20]. Section 3 treats the characteristic waves in a uniaxial optically active medium. In section 4 we derive the Fresnel amplitudes for light reflection and refraction in two geometries: (i) optical axis perpendicular to the reflecting boundary (figure 1); (ii) optical axis perpendicular to the plane of incidence (figure 2). In section 5 differential reflection methods are considered for the case of ordinary and total reflection. Section 6 contains some concluding remarks.

2. Constitutive relations and boundary conditions

Different sets of constitutive relations could be used to describe the electromagnetic properties of optically active media [20, 21]. The following constitutive equations [20]:

$$\begin{aligned} \boldsymbol{D} &= \epsilon \cdot (\boldsymbol{E} + \boldsymbol{\Gamma} \cdot \nabla \times \boldsymbol{E}) \\ \boldsymbol{B} &= \mu \cdot (\boldsymbol{H} + \boldsymbol{\Gamma}^{\mathrm{T}} \cdot \nabla \times \boldsymbol{H}) \end{aligned} \tag{1}$$

are invariant under the duality transformation: $\boldsymbol{D} \to \pm\boldsymbol{B}$, $\boldsymbol{B} \to \pm\boldsymbol{D}$, $\boldsymbol{E} \to \pm\boldsymbol{H}$, $\boldsymbol{H} \to \pm\boldsymbol{E}$, $\epsilon \rightleftarrows \mu$, $\boldsymbol{\Gamma} \rightleftarrows \boldsymbol{\Gamma}^{\mathrm{T}}$. As usual $\boldsymbol{E}$ and $\boldsymbol{H}$ are the electric and magnetic vectors of the EMW, $\boldsymbol{D}$ and $\boldsymbol{B}$ are the electric and magnetic inductions, ϵ and μ are the real, symmetric permittivity and permeability tensors, and $\boldsymbol{\Gamma}$ is the axial gyrotropic tensor (with transpose $\boldsymbol{\Gamma}^{\mathrm{T}}$). For the case of an isotropic optically active medium, equations (1) reduce to Condon's constitutive relations [22]. The equation of conservation of energy takes the usual form $\nabla \cdot \boldsymbol{S} + \partial u/\partial t = 0$ with standard Poynting vector $\boldsymbol{S} = (c/4\pi)(\boldsymbol{E} \times \boldsymbol{H})$ and energy density $u = (1/8\pi)(\boldsymbol{D} \cdot \epsilon^{-1} \cdot \boldsymbol{D} + \boldsymbol{B} \cdot \mu^{-1} \cdot \boldsymbol{B})$ [20]. Standard boundary conditions for the continuity of the tangential components of $\boldsymbol{E}$ and $\boldsymbol{H}$ and continuity of the normal components of $\boldsymbol{D}$ and $\boldsymbol{B}$ are valid when the constitutive relations (1) are used [20]:

$$(\boldsymbol{E}^{(1)} - \boldsymbol{E}^{(2)}) \times \boldsymbol{q} = 0 \qquad (\boldsymbol{H}^{(1)} - \boldsymbol{H}^{(2)}) \times \boldsymbol{q} = 0 \tag{2a}$$

$$(\boldsymbol{D}^{(1)} - \boldsymbol{D}^{(2)}) \cdot \boldsymbol{q} = 0 \qquad (\boldsymbol{B}^{(1)} - \boldsymbol{B}^{(2)}) \cdot \boldsymbol{q} = 0. \tag{2b}$$

Here $\boldsymbol{q}$ is the normal to the boundary; the indices (1) and (2) refer to the media on the two sides of the interface.

It is well known that Maxwell's equations are invariant under the transformation

$$\begin{aligned} \boldsymbol{E}' &= \boldsymbol{E} + (1/c)\partial \boldsymbol{P}/\partial t \qquad & \boldsymbol{B}' &= \boldsymbol{B} - \operatorname{curl} \boldsymbol{P} \\ \boldsymbol{H}' &= \boldsymbol{H} + (1/c)\partial \boldsymbol{Q}/\partial t \qquad & \boldsymbol{D}' &= \boldsymbol{D} + \operatorname{curl} \boldsymbol{Q} \end{aligned}$$

where $\boldsymbol{P}$ and $\boldsymbol{Q}$ are arbitrary vectors. Taking $\boldsymbol{P}$ and $\boldsymbol{Q}$ in the form $\boldsymbol{P} = -\theta\boldsymbol{\Gamma} \cdot \boldsymbol{B}$ and $\boldsymbol{Q} = (1-\theta)\boldsymbol{\Gamma}^{\mathrm{T}} \cdot \boldsymbol{D}$ and varying the constant θ, a number of sets of constitutive relations could be obtained [20, 23]. Usually the natural optical activity is weak—the quantity $\gamma_{ij} = \Gamma_{ij}\omega/c \simeq (10^{-3}\text{–}10^{-6})$. In the first-order approximation when we can neglect terms $\mathrm{O}(\Gamma_{ij}^2)$ and higher, all these sets of constitutive relations are equivalent to the relations (1). However, different modified boundary conditions and expressions for Poynting's vector $\boldsymbol{S}$ and energy density u correspond to each of these sets of material equations. A widely used set of constitutive relations (proposed by Born and Wolf [24]) is that obtained for $\theta = 0$:

$$\boldsymbol{D}' = \epsilon \cdot \boldsymbol{E} + (\rho \cdot \nabla) \times \boldsymbol{E} \qquad \boldsymbol{B} = \mu \cdot \boldsymbol{H} \tag{3}$$

where

$$\rho = \mathrm{Sp}(\epsilon \cdot \boldsymbol{\Gamma}) - (\epsilon \cdot \boldsymbol{\Gamma})^{\mathrm{T}}. \tag{4}$$

All optical properties of the medium are then contained in the permittivity tensor $\epsilon_{ij}(\omega, k) = \epsilon_{ij}(\omega) - \mathrm{i}e_{ijl_1}\rho_{l_1l}k_l$ (k = wavevector of the plane wave, ω = frequency, e_{ijl} = Levi-Chivita symbol). For a non-magnetic medium the permeability μ is a scalar of value close to unity ($\mu = 1$). In the case of Born's constitutive relations (3), the boundary conditions, S and u take the form [20]:

$$(E^{(1)} - E^{(2)}) \times q = 0 \tag{5a}$$

$$(H^{(1)} - H^{(2)}) \times q = \frac{1}{c}\left((\Gamma^{\mathrm{T}} \cdot \epsilon)^{(1)} \cdot \frac{\partial E^{(1)}}{\partial t} - (\Gamma^{\mathrm{T}} \cdot \epsilon)^{(2)} \cdot \frac{\partial E^{(2)}}{\partial t}\right) \times q \tag{5b}$$

$$S = \frac{c}{4\pi}\left[E \times \left(H - \frac{1}{c}(\Gamma^{\mathrm{T}} \cdot \epsilon) \cdot \frac{\partial E}{\partial t}\right)\right] \tag{5c}$$

$$u = \frac{1}{8\pi}\left[E \cdot \left(\epsilon \cdot E - \frac{2}{c}(\epsilon \cdot \Gamma) \cdot \frac{\partial B}{\partial t}\right) + B \cdot \mu^{-1} \cdot B\right]. \tag{5d}$$

(Surely it is not correct to apply the standard boundary conditions (2) when Born's constitutive relations (3) are used. This was the subtle but important mistake in [14].)

3. Characteristic waves

Using the constitutive relations (1) and Maxwell's equations in the absence of free charges and currents the electric and magnetic vectors of a plane wave $E = E_0 \mathrm{e}^{-\mathrm{i}(\omega t - k \cdot r)}$ and $H = H_0 \mathrm{e}^{-\mathrm{i}(\omega t - k \cdot r)}$ are to $\mathrm{O}(\Gamma)$ given by:

$$\epsilon \cdot E = -(k \times H)/k_c + \mathrm{i}k_c(\epsilon \cdot \Gamma \cdot \mu) \cdot H \tag{6a}$$

$$\mu \cdot H = (k \times E)/k_c + \mathrm{i}k_c(\epsilon \cdot \Gamma \cdot \mu)^{\mathrm{T}} \cdot E \tag{6b}$$

where $k_c = \omega/c$.

In a non-magnetic medium ($\mu = 1$) equations (6) lead to the wave equation:

$$\epsilon \cdot E = -[k' \times (k' \times E) + \mathrm{i}k' \times (\epsilon \cdot \gamma)^{\mathrm{T}} \cdot E + \mathrm{i}(\epsilon \cdot \gamma) \cdot (k' \times E)] \tag{7}$$

where $k' = (k'_1, k'_2, k'_3)$ is the normalized wavevector $k' = k/k_c$ and γ is the normalized gyration tensor $\gamma = k_c\Gamma$. In a uniaxial medium (when $z \equiv$ optical axis) the permittivity and gyrotropy tensors take the form:

$$\begin{pmatrix} \epsilon_o & 0 & 0 \\ 0 & \epsilon_o & 0 \\ 0 & 0 & \epsilon_e \end{pmatrix} \qquad \begin{pmatrix} \gamma_1 & \gamma_{12} & 0 \\ -\gamma_{12} & \gamma_1 & 0 \\ 0 & 0 & \gamma_3 \end{pmatrix}. \tag{8}$$

(In [17] $\gamma_{12} = 0$, $\gamma_1 = \gamma_3 = \gamma$, which considerably simplifies the problem but is an unrealistic assumption, especially for the crystal classes $\mathrm{C_{3v}}$, $\mathrm{C_{4v}}$ and $\mathrm{C_{6v}}$ where only $\gamma_{12} \neq 0$.)

Then (6) may be expressed as:

$$\begin{pmatrix} \epsilon_o - (k_2'^2 + k_3'^2) & k'_1k'_2 - \mathrm{i}\rho_3k'_3 & k'_1k'_3 + \mathrm{i}(\rho_1k'_2 - \rho_{12}k'_1) \\ k'_1k'_2 + \mathrm{i}\rho_3k'_3 & \epsilon_o - (k_1'^2 + k_3'^2) & k'_2k'_3 - \mathrm{i}(\rho_1k'_1 + \rho_{12}k'_2) \\ k'_1k'_3 - \mathrm{i}(\rho_1k'_2 - \rho_{12}k'_1) & k'_2k'_3 + \mathrm{i}(\rho_1k'_1 + \rho_{12}k'_2) & \epsilon_e - (k_1'^2 + k_2'^2) \end{pmatrix} \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix} = 0 \tag{9}$$

where $\rho_1, \rho_3, \rho_{12}$ are the non-zero components of the tensor ρ (4):

$$\rho_1 = \epsilon_o\gamma_1 + \epsilon_e\gamma_3 \qquad \rho_3 = 2\epsilon_o\gamma_1 \qquad \rho_{12} = \epsilon_o\gamma_{12}. \tag{10}$$

The gyrotropic parameters γ_{ij} are small ($\gamma_{ij} \simeq 10^{-4}$–$10^{-6}$) and in all our considerations we neglect terms quadratic to γ_{ij}. Non-zero solutions $\boldsymbol{E} \neq 0$ of the system (9) exist if the determinant of the coefficients is equal to zero. Two characteristic waves exist in a uniaxial optically active medium in directions not parallel to the optical axis, when $k_1' \neq 0$ and/or $k_2' \neq 0$ (we are using the principal axis coordinate system of the crystal): (i) 'ordinary' EMW (o-wave) for which

$$k_o'^2 = \epsilon_o \tag{11a}$$

and (ii) 'extraordinary' wave (e-wave) for which, writing $\boldsymbol{k}_e' = (k_{e1}', k_{e2}', k_{e3}')$,

$$k_e'^2 = \epsilon_e - k_{e3}'^2(\epsilon_e - \epsilon_o)/\epsilon_o. \tag{11b}$$

The contributions of the gyrotropic parameters to the refractive indices of the o- and e-waves are quadratic in γ_{ij} and are neglected. (Linear gyrotropic contributions to the refractive indices appear only for the characteristic waves along the optical axis (for $\boldsymbol{k} = (0, 0, k)$) [21]. They are left and right circularly polarized waves with refractive indices $k'^{l,r} = \epsilon_o^{1/2} \pm \rho_3/2$.)

By substitution of (11a) or (11b) in (9) the polarization of the ordinary or of the extraordinary wave can be determined. For a wavevector $\boldsymbol{k} = k(\sin\varphi, 0, \cos\varphi)$, forming an angle $\varphi \neq 0$ with the optical axis, the following results are obtained:

(i) The 'ordinary' wave (see (11a)) is $\boldsymbol{E}^o = (E_1^o, E_2^o, E_3^o)$, where

$$E_1^o = -\mathrm{i}k_{o3}'\frac{[\rho_1 k_{o1}'^2 + \rho_3(\epsilon_e - k_{o1}'^2)]}{k_{o1}'^2(\epsilon_o - \epsilon_e)}E_2^o = \mathrm{i}\frac{\epsilon_o^{1/2}\cos\varphi}{\epsilon_o - \epsilon_e}\left[\left(1 - \frac{\epsilon_e}{\epsilon_o\sin^2\varphi}\right)\rho_3 - \rho_1\right]E_2^o \tag{12a}$$

$$E_3^o = \mathrm{i}\frac{(\rho_1 k_{o1}'^2 + \rho_3 k_{o3}'^2)}{k_{o1}'^2(\epsilon_o - \epsilon_e)}E_2^o = \mathrm{i}\frac{\epsilon_o^{1/2}}{\epsilon_o - \epsilon_e}\left(\rho_1\sin\varphi + \rho_3\frac{\cos^2\varphi}{\sin\varphi}\right)E_2^o. \tag{12b}$$

Hence, owing to the gyrotropy, a component parallel to the optical axis (E_3^o) appears in the o-wave. A longitudinal component ($E_\parallel^o = \mathrm{i}\rho_3(\cot\varphi/\epsilon_o^{1/2})E_2^o$) arises in directions $\boldsymbol{k}$ non-orthogonal to the optical axis (when $\varphi \neq \pi/2$). For directions $\boldsymbol{k} = (k, 0, 0)$ perpendicular to the optical axis ($\varphi = \pi/2$) the ordinary wave is transverse

$$E_1^o = 0 \tag{12a$'$}$$

$$E_3^o = \mathrm{i}[\epsilon_o^{1/2}/(\epsilon_o - \epsilon_e)]\rho_1 E_2^o. \tag{12b$'$}$$

(ii) The extraordinary EMW ($k_e' = \{\epsilon_e/[1 + \cos^2\varphi(\epsilon_e - \epsilon_o)/\epsilon_o]\}^{1/2}$, see (11$b$)) is $\boldsymbol{E}^e = (E_1^e, E_2^e, E_3^e)$, where

$$E_1^e = \frac{\epsilon_e}{\epsilon_o k_{e1}'}(-k_{e3}' + \mathrm{i}\rho_{12})E_3^e = \frac{\epsilon_e}{\epsilon_o\sin\varphi}(-\cos\varphi + \mathrm{i}\rho_{12}/k_e')E_3^e \tag{13a}$$

$$E_2^e = \mathrm{i}\frac{\epsilon_e\{\rho_1 + \rho_3[(\epsilon_e/k_{e1}'^2) - 1]\}}{(\epsilon_o - \epsilon_e)k_{e1}'}E_3^e = \mathrm{i}\frac{\epsilon_e[\rho_1 + \rho_3(\epsilon_e/k_e'^2\sin^2\varphi - 1)]}{(\epsilon_o - \epsilon_e)k_e'\sin\varphi}E_3^e. \tag{13b}$$

In directions $\boldsymbol{k} = (k, 0, 0)$ perpendicular to the optical axis ($\varphi = \pi/2$) $k'_{\mathrm{e}} = \epsilon_{\mathrm{e}}^{1/2}$ and the polarization of $\boldsymbol{E}^{\mathrm{e}}$ is

$$E_1^{\mathrm{e}} = \mathrm{i}(\epsilon_{\mathrm{e}}^{1/2}/\epsilon_{\mathrm{o}})\rho_{12}E_3^{\mathrm{e}} \tag{13a'}$$

$$E_2^{\mathrm{e}} = \mathrm{i}[\epsilon_{\mathrm{e}}^{1/2}/(\epsilon_{\mathrm{o}} - \epsilon_{\mathrm{e}})]\rho_1 E_3^{\mathrm{e}}. \tag{13b'}$$

Thus for crystal classes with $\rho_{12} = 0$ the extraordinary wave is transverse.

In the crystal classes C_{3v}, C_{4v}, C_{6v} the only non-zero component of the gyration tensor is ρ_{12}. The ordinary wave in these crystals is not affected by the gyrotropy, while the extraordinary wave possesses a longitudinal 'gyrotropic' component.

The components of $\boldsymbol{H}$ are obtained from equations (6*b*).

The present results for the polarization of the characteristic waves $\boldsymbol{E}^{\mathrm{o}}$ and $\boldsymbol{E}^{\mathrm{e}}$ coincide with the results obtained in [6, 14] using Born's constitutive relations (3). The gyrotropy tensor ρ'_{ij} in [6, 14] is related to the tensor ρ_{ij} (4) by $\rho'_{ij} = -\rho_{ij}$.

4. Reflection of EMWs at the boundary: isotropic inactive medium–uniaxial optically active medium

4.1. Reflection of EMWs: optical axis perpendicular to the reflecting boundary

A plane wave $\boldsymbol{E} = (E_{\mathrm{s}}, E_{\mathrm{p}})$ of frequency ω and wavevector $\boldsymbol{k} = (k_1, k_2, k_3) = k(\sin\theta, 0, \cos\theta)$ propagates in an isotropic inactive medium with refractive index n and is reflected from a uniaxial non-absorbing optically active medium with permittivity and gyration tensors (8) (figure 1). The optical axis of this medium is assumed to be perpendicular to the plane boundary. Two waves are refracted in the gyrotropic medium at angles φ^{o} and φ^{e} respectively. Their wavevectors and polarization are determined from equations (11*a*), (12*a,b*) and equations (11*b*), (13*a,b*) respectively. The reflected wave $\boldsymbol{E}''(E''_{\mathrm{s}}, E''_{\mathrm{p}})$ has both s- and p-polarized components. The usual boundary conditions (2*a*) hold for continuity of the tangential components of $\boldsymbol{E}$ and $\boldsymbol{H}$.

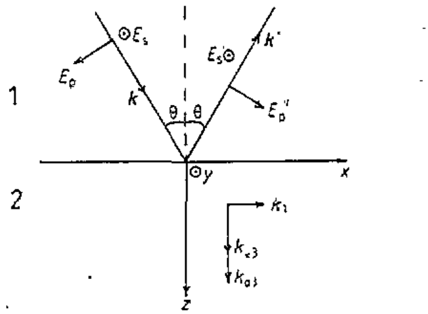

Figure 1. Reflection of an EMW from the boundary: isotropic inactive medium (1)–uniaxial optically active medium (2). The optical axis z is perpendicular to the boundary; θ = angle of incidence; $\boldsymbol{k}$ and $\boldsymbol{k}''$ = wavevectors of the incident and of the reflected wave; k_l = *tangential component of the wavevector;* $k_{\mathrm{o}3}, k_{\mathrm{e}3}$ = normal (to the boundary) components of the wavevectors of the two refracted waves; E_{p}, E_{s}, E''_{p}, E''_{s} = components of the electric field of the incident and reflected waves.

Snell's law takes the form:

$$n\sin\theta = k'^{\mathrm{o}}\sin\varphi^{\mathrm{o}} = k'^{\mathrm{e}}\sin\varphi^{\mathrm{e}} = k'_1. \tag{14}$$

The solution of the boundary problem gives the following results for the components E''_{s}, E''_{p} of the reflected wave:

$$E''_{\mathrm{s}} = \tilde{A}E_{\mathrm{s}} + \mathrm{i}CE_{\mathrm{p}} \tag{15a}$$

$$E''_{\mathrm{p}} = \tilde{B}E_{\mathrm{p}} - \mathrm{i}CE_{\mathrm{s}} \tag{15b}$$

where

$$\tilde{A} = [n\cos\theta - (k'_{o3} + \mathrm{i}\rho_{12})]/[n\cos\theta + (k'_{o3} + \mathrm{i}\rho_{12})] \tag{16a}$$

$$\tilde{B} = [\epsilon_o\cos\theta - n(k'_{e3} - \mathrm{i}\rho_{12})]/[\epsilon_o\cos\theta + n(k'_{e3} - \mathrm{i}\rho_{12})] \tag{16b}$$

$$C = \frac{2n\cos\theta\epsilon_o[\![\rho_1 k_1^2(k'_{o3} - k'_{e3}) + \rho_3\{\epsilon_e(k'_{o3} - k'_{e3}) + k_1^2[k'_{e3}(\epsilon_o + \epsilon_e)/2\epsilon_o - k'_{o3}]\}]\!]}{k_1'^2(\epsilon_o - \epsilon_e)(n\cos\theta + k'_{o3})(\epsilon_o\cos\theta + nk'_{e3})}. \tag{16c}$$

In a non-gyrotropic medium $C = 0$ and the coefficients $\tilde{A}$ and $\tilde{B}$ are reduced to the well known Fresnel amplitudes. In the case of reflection from an opticallly inactive medium ($\rho = 0$) the incident s- (p-) polarized component E_s (or E_p) generates a reflected component only of the same type of polarization E''_s (or E''_p). As a result of the gyrotropy of the reflecting medium, weak contributions of polarization, perpendicular to the polarization of the incident wave, appear in the reflected wave (the terms with coefficients C in equations (15*a*,*b*)). This result is fully analogous to the result for the isotropic case.

The components E_2^o and E_3^e of the two refracted waves are

$$E_2^o = ME_s - \mathrm{i}mE_p \tag{17a}$$

$$E_3^e = NE_p - \mathrm{i}\eta E_s \tag{17b}$$

where

$$M = 2n\cos\theta/[n\cos\theta + k'_{o3} + \mathrm{i}\rho_{12}] \tag{18a}$$

$$N = 2\epsilon_o k'_1 n\cos\theta/\{\epsilon_e[\epsilon_o\cos\theta + n(k'_{e3} - \mathrm{i}\rho_{12})]\} \tag{18b}$$

$$m = 2n\cos\theta\left[\frac{\epsilon_o\rho_1(n\cos\theta + k'_{e3})}{\epsilon_o - \epsilon_e} + \frac{\rho_3}{k_1'^2}\left(\frac{k'_{e3}}{2} + \frac{\epsilon_o(\epsilon_e - k_1'^2)(n\cos\theta + k'_{e3})}{k_1'^2(\epsilon_o - \epsilon_e)}\right)\right]$$
$$\times [(n\cos\theta - k_{o3})(\epsilon_o\cos\theta + nk'_{e3})]^{-1} \tag{18c}$$

$$\eta = 2n\cos\theta\epsilon_o k'_1\left[\frac{\rho_1(\epsilon_o\cos\theta + nk'_{o3})}{\epsilon_o - \epsilon_e} + \rho_3\left(\frac{k'_{o3}[n(\epsilon_e - k_1'^2)\epsilon_e k'_{o3}\cos\theta]}{k_1'^2(\epsilon_o - \epsilon_e)} - \frac{\cos\theta}{2}\right)\right]$$
$$\times [\epsilon_e(n\cos\theta + k'_{o3})(\epsilon_o\cos\theta + nk'_{e3})]^{-1}. \tag{18d}$$

In the geometry of reflection considered here, the 'weak' optical activity of crystals C_{3v}, C_{4v}, C_{6v} ($\rho_1 = \rho_3 = 0$, $\rho_{12} = -\rho_{21} \neq 0$) does not cause the appearance of a reflected component with polarization perpendicular to the polarization of the incident wave ($C = 0$, see equation (16*c*)).

4.2. Reflection of EMWs: optical axis perpendicular to the plane of incidence

In this section we consider the reflection of an EMW with a wavevector $\boldsymbol{k} = k(\sin\theta, \cos\theta, 0)$ from a uniaxial optically active medium when the optical axis ($\equiv \hat{z}$) is perpendicular to the plane of incidence (figure 2). According to equations (12′), (13′) the following two EMWs are refracted on the birefringent medium:

(i) ordinary EMW of wavevector $k_o'^2 = \epsilon_o$ and polarization $E^o = (-E_2^o k_{o2}'/k_1', E_2^o, E_3^o)$, where

$$E_3^o = \mathrm{i}\frac{\epsilon_o \rho_1}{k_1'(\epsilon_o - \epsilon_e)} E_2^o \tag{19}$$

(ii) extraordinary EMW of wavevector $k_e'^2 = \epsilon_e$ (see equation (11b) for $k_{e3}' = 0$) and polarization $E^e = (E_1^e, E_2^e, E_3^e)$, where

$$E_1^e = \mathrm{i}\left(\frac{k_1'\rho_{12}}{\epsilon_o} - \frac{k_{e2}'\rho_1}{\epsilon_o - \epsilon_e}\right) E_3^e \tag{20a}$$

$$E_2^e = \mathrm{i}\left(\frac{k_{e2}'\rho_{12}}{\epsilon_o} + \frac{k_1'\rho_1}{\epsilon_o - \epsilon_e}\right) E_3^e. \tag{20b}$$

(Equations (19), (20) correspond to equations (12′), (13′) in a coordinate system rotated by an angle θ around the z axis.)

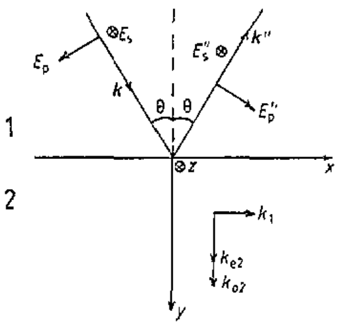

Figure 2. Reflection of an EMW from the boundary: isotropic inactive medium (1)–uniaxial optically active medium (2). The optical axis z is perpendicular to the plane of incidence; k_{o2}, k_{e2} = normal (to the boundary) components of the wavevectors of the two refracted waves. For the other notations see figure 1.

The solution of the boundary problem gives the following results for the components E_s'' and E_p'' of the reflected wave:

$$E_s'' = AE_s + \mathrm{i}(C + D)E_p \tag{21a}$$

$$E_p'' = BE_p - \mathrm{i}(C - D)E_s \tag{21b}$$

where

$$A = (n\cos\theta - k_{e2}')/(n\cos\theta + k_{e2}') \tag{22a}$$

$$B = (\epsilon_o\cos\theta - nk_{o2}')/(\epsilon_o\cos\theta + nk_{o2}') \tag{22b}$$

$$C = \frac{2n\cos\theta[\epsilon_o\rho_1(k_{e2}' - k_{o2}') + \rho_3 k_{o2}'(\epsilon_o - \epsilon_e)/2]}{(\epsilon_o - \epsilon_e)(n\cos\theta + k_{e2}')(\epsilon_o\cos\theta + nk_{o3}')} \tag{22c}$$

$$D = n^2\rho_{12}\sin 2\theta/[(n\cos\theta + k_{e2}')(\epsilon_o\cos\theta + nk_{o3}')]. \tag{22d}$$

The components E_2^o and E_3^e of the refracted waves are given by

$$E_2^o = ME_p - \mathrm{i}mE_s \tag{23a}$$

$$E_3^e = NE_s - \mathrm{i}\eta E_p \tag{23b}$$

where

$$M = 2n\cos\theta\, k_1'/(\epsilon_o\cos\theta + nk_{o2}') \tag{24a}$$

$$N = 2n\cos\theta/(n\cos\theta + k_{e2}') \tag{24b}$$

$$m = \frac{2n\cos\theta\, k_1'}{(n\cos\theta + k_{e2}')(\epsilon_o\cos\theta + nk_{o2}')}\left(\frac{\rho_1}{\epsilon_o - \epsilon_e}(\epsilon_o\cos\theta + nk_{e2}') - \frac{\rho_3}{2}\cos\theta - \frac{nk_1'}{\epsilon_o}\rho_{12}\right) \tag{24c}$$

$$\eta = \frac{2n\cos\theta}{(n\cos\theta + k_{e2}')(\epsilon_o\cos\theta + nk_{o2}')}\left(\frac{\epsilon_o\rho_1}{\epsilon_o - \epsilon_e}(n\cos\theta + k_{o2}') - \frac{\rho_3}{2}k_{o2}' - k_1'\rho_{12}\right). \tag{24d}$$

5. Differential reflection methods

Conventional polarization measurements of the intensity of the reflected component of polarization, perpendicular to the polarization of the incident wave, would give a signal of too weak intensity ($I_s''/I_p^o \sim \rho_{ij}^2 \simeq 10^{-8}$ to 10^{-12})—beyond the sensitivity of detectors. By use of differential reflection methods, a signal of considerably stronger intensity (linear in $\rho_{ij} \sim 10^{-4}$ to 10^{-6}) can be obtained. Differential spectroscopy could be used both in the case of ordinary reflection (no critical angles) and in the case of total reflection.

5.1. Differential circular ordinary reflection

We consider the ordinary reflection of left and right circularly polarized waves (LCP and RCP waves) in the geometry shown in figure 2 (the optical axis is perpendicular to the plane of incidence). The LCP and RCP incident waves of equal intensity $E^{l,r}(E_s^{l,r}, E_p^{l,r}) = (E/\sqrt{2})(1, \pm\mathrm{i})$ generate reflected waves of the following polarization (see (21)):

$$E_s''^{l,r} = [A \mp (C + D)]E/\sqrt{2} \tag{25a}$$

$$E_p''^{l,r} = \mathrm{i}[\pm B - (C - D)]E/\sqrt{2}. \tag{25b}$$

The differential circular reflection δ is

$$\delta = (I''^{l} - I''^{r})/(I''^{l} + I''^{r}) = 2[(A+B)C + (A-B)D]/(A^2 + B^2) \tag{26}$$

where $I'' = E_s''^2 + E_p''^2$. In the case of normal incidence ($\theta = 0$) δ takes the form

$$\delta = \frac{4n^2(\epsilon_o - \epsilon_o^{1/2}\epsilon_e^{1/2})[\rho_1\epsilon_o^{1/2}/(\epsilon_o^{1/2} + \epsilon_e^{1/2})^{1/2} + \rho_3]}{(n^2 + \epsilon_e)(n^2 + \epsilon_o) - 4n^2\epsilon_o^{1/2}\epsilon_e^{1/2}}. \tag{27}$$

In crystal classes C_{3v}, C_{4v}, C_{6v} ($\rho_1 = \rho_3 = 0$, $\rho_{12} \not\equiv 0$) δ takes the form

$$\delta = \frac{4n^2\cos^2\theta\sin\theta(n^2k_{o2}' - \epsilon_e k_{e2}')\rho_{12}}{(n^2\cos^2\theta + k_{e2}'^2)(\epsilon_o^2\cos^2\theta + n^2k_{o2}'^2) - 2n^2\cos^2\theta\,\epsilon_o k_{e2}' k_{o2}'}. \tag{28}$$

For media with $\rho_1 \neq 0$ and $\rho_3 \neq 0$ reflection measurements of the gyrotropy can be done at normal incidence (experimentally easy to perform), while for crystals with $\rho_{12} \neq 0$ measurements at oblique incidence are necessary.

It is necessary to point out that Born's constitutive relations (3) and the respective modified boundary conditions (5*a*,*b*) give the same results as the results presented above. So the proposal of Silverman and Badoz [17] for an experimental check of the adequacy of the different sets of constitutive relations by the differential circular reflection curves could not be realized. All sets of constitutive equations give equivalent results when applied with the appropriate boundary conditions.

5.2. Differential total reflection methods

We consider the total reflection of EMWs in the geometry shown in figure 2 (optical axis perpendicular to the plane of incidence). If $n^2 > \epsilon_o$, $n^2 > \epsilon_e$, total reflection (TR) occurs for angles of incidence larger than the critical angles, $\theta > \theta^o_{cr}$ and $\theta > \theta^e_{cr}$, where $\sin\theta^o_{cr} = \epsilon_o^{1/2}/n$, $\sin\theta^e_{cr} = \epsilon_e^{1/2}/n$. The TR is described by treating in equations (22), (24) k'_{o2} and k'_{e2} as imaginary quantities: $k'_{o2} = \mathrm{i}k''_{o2}$, $k'_{e2} = \mathrm{i}k''_{e2}$ (k''_{o2}, k''_{e2} are real quantities). Thus all Fresnel amplitudes become complex quantities.

The calculations give the following results for the intensities I''_s and I''_p of the s and p components of the reflected light when the incident wave is linearly polarized ($E_s = E\sin\gamma$, $E_p = E\cos\gamma$, γ = azimuth of $\boldsymbol{E}$):

$$I''_s = I_s + \chi E_s E_p \tag{29a}$$

$$I''_p = I_p - \chi E_s E_p \tag{29b}$$

where $I_s = I_0\sin^2\gamma$, $I_p = I_0\cos^2\gamma$ are intensities of the s and p components of the incident EMW of intensity I_0, and χ is a gyrotropic quantity:

$$\chi = \frac{4n\cos\theta\left[-n\left(\frac{\rho_1(k''_{e2}-k''_{o2})\epsilon_o}{\epsilon_o-\epsilon_e}+\frac{\rho_3k''_{o2}}{2}\right)(\epsilon_o\cos^2\theta+k''_{o2}k''_{e2})+\rho_{12}n\sin\theta\cos\theta(n^2k''_{o2}-\epsilon_ok''_{e2})\right]}{(n^2\cos^2\theta+k''^2_{e2})(\epsilon_o^2\cos^2\theta+n^2k''^2_{o2})}. \tag{29c}$$

The non-absorbing birefringent optically active medium does not attenuate the TR ($I''_s + I''_p = I_s + I_p = I_0$) but it acts like a transformer of the polarization (of small efficiency)—see the terms proportional to χ in (29*a*,*b*). In the case of TR in order to obtain a reflected signal, related to the gyrotropy, an analyser should be placed on the path of the reflected wave to absorb the s- or p-polarized reflected component.

We propose two modifications of differential spectroscopy in the case of TR:

(i) Differential TR of linearly polarized EMWs of equal intensities I_0 and azimuths γ and $(-\gamma)$:

$$I''_s(\gamma) - I''_s(-\gamma) = I_0\chi\sin 2\gamma. \tag{30}$$

This signal is maximum when $\gamma = \pi/4$. The method of differential TR of linearly polarized waves was used for the investigation of the gyrotropy of liquid crystals in the frequency ranges of the intramolecular vibrations [16]. However in this case the gyrotropic medium is absorbing and the TR is attenuated. This situation needs a special treatment.

(ii) Differential TR of left and right circularly polarized EMWs of equal intensities I_0. For an incident LCP (RCP) wave the s- and p-polarized reflected components in the case of total reflection take the form:

$$I''^{l(r)}_s = \tfrac{1}{2}I_0(1 \pm \chi') \tag{31a}$$

$$I''^{l(r)}_p = \tfrac{1}{2}I_0(1 \mp \chi') \tag{31b}$$

where

$$\chi' = 4n\cos\theta\left[\cos\theta\left(\frac{\rho_1(k''_{e2}-k''_{o2})\epsilon_o}{\epsilon_o-\epsilon_e}+\frac{\rho_3k''_{o2}}{2}\right)(n^2k''_{o2}-\epsilon_ok''_{e2}) + \rho_{12}n^2\sin\theta(\epsilon_o\cos\theta+k''_{o2}k''_{e2})\right][(n^2\cos^2\theta+k''^2_{e2})(\epsilon_o^2\cos^2\theta+n^2k''^2_{o2})]^{-1}. \tag{31c}$$

The differential circular reflection signal in the case of TR is

$$(I_s^{\prime\prime l} - I_s^{\prime\prime r})/(I_s^{\prime\prime l} + I_s^{\prime\prime r}) = \chi'. \tag{32}$$

The modifications of differential spectroscopy in TR, mentioned above, give signals that are linear in the components ρ_1, ρ_3 and ρ_{12} of the gyration tensor (see (30), (32)). Thus successful experimental studies of the gyrotropy of anisotropic materials through reflection spectroscopy are possible [15, 16].

6. Conclusion

This paper completes the investigations of paper I [7] for the case of reflection of electromagnetic waves from uniaxial optically active materials. It solves the reflection problem using duality-invariant constitutive relations and the standard boundary conditions consistent with them. The anisotropy complicates the theoretical treatment in two ways: (i) by the strong birefringence and (ii) by the existence of several different non-zero components of the gyration tensor. Nevertheless, reflection studies of the gyrotropy of anisotropic media are possible and in some cases they are the only method available (in crystals of point groups C_{3v}, C_{4v} and C_{6v}).

The problem treated is of theoretical and experimental interest. Reflection studies of anisotropic gyrotropic materials can give spectroscopic data for the components of the gyration tensor. They may also be used for the evaluation of the quality of optical elements. Materials that are expected to be isotropic may become anisotropic as a result of mechanical processing and may considerably change their optical behaviour. Knowledge of the process of reflection from aniostropic gyrotropic materials would avoid misinterpretations of the results from measurements done with such optical elements.

The potential for the experimental realization of reflection spectroscopy, especially in the case of total reflection, will be revealed by further investigations.

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