Deriving the transmission and reflection coefficients of an optically active medium without using boundary conditions

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A new wave vector space method that can solve wave propagation problems in bounded media without the use of boundary conditions is applied to a half space of an optically active medium to obtain the transmission and reflection coefficients for oblique incidence. The calculation is aided by a fundamental constitutive derivation that produces the mechanisms, the interchange symmetry, and the frequency and wave vector dispersion for both the bulk and surface interactions. It is also aided by a recent fundamental derivation of the energy propagation vector. This derivation resolves a number of long-standing controversies concerning this problem: it shows that no surface wave need be generated as has been proposed and its solution shows that neither $\mathbf{H}_{\text{tang}}$ nor $\mathbf{D}_{\text{norm}}$ are continuous at the surface.

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INTRODUCTION

In spite of the fact that optical activity (natural circular birefringence or gyrotropy) has been known since Biot's discovery of it in 1812, the correct forms of the transmission and reflection coefficients for a plane wave are still not agreed upon in the literature. This situation is caused by optical activity being a nonlocal or wave vector dispersive interaction[1], that is, its polarization at a particular point in the medium depends not only on the electric field at that point but also on the first spatial derivative of the electric field there. Thus the bulk response of the medium is modified as its surface is approached and the form that the boundary conditions must take is then far from obvious. The debate on them has been at the heart of the difficulties of this problem.

The boundary conditions, however, are not the only source of confusion. Another source is the constitutive relation or relations responsible for the phenomenon. It is known that a third-rank gyration tensor must enter the wave equation to account for the circular polarization modes, but several combinations of phenomenological constitutive relations for $\mathbf{D}$ and $\mathbf{H}$ have been proposed which satisfy this need [2–9]. Some [2–4] have assumed that electric and magnetic fields enter these relations symmetrically. We regard this as a misuse of symmetry because, though there is a certain symmetry between electric and magnetic fields in Maxwell's equations, the sources of material response have no symmetry in nature. We have electric monopoles (charge) but no magnetic monopoles and we have magnetic dipoles (intrinsic spin) but no electric dipoles and it is these sources that produce constitutive relations. Another constitutive relation, derived by Born and Huang [5] from long-wavelength lattice dynamics, contains the entire optical activity effect in the constitutive relation for $\mathbf{D}$. However, a recent long-wavelength lattice-dynamical derivation by the author [6] showed that Born missed the two largest sources of crystalline optical activity and found only a tertiary contribution. The larger two are a combination of electric dipole and magnetic dipole interactions and a combination of electric dipole and electric quadrupole interactions and are completely analogous to the results of quantum mechanics. These mechanisms lead to constitutive relations for $\mathbf{D}$ and $\mathbf{H}$ that evidence no symmetry but still produce a third-rank gyration tensor of the correct interchange symmetry in the wave equation. Agranovich and Ginzburg [7,8] and Bokut' and collaborators [9] used phenomenological constitutive relations for $\mathbf{D}$ only.

The third source of confusion is the proper form of the energy flow vector in the medium. An examination of the Poynting theorem readily shows that the derivation of $\mathbf{E} \times \mathbf{H}$ for the energy flow vector does *not* apply to any of the constitutive relations assumed or derived for optical activity. Bokut' and others [4,7–12] found several forms of the energy flow vector depending on the constitutive assumptions chosen. They also found that the boundary conditions were modified in each case. The approach was phenomenological, no frequency dispersion was considered, and in the end [12] no demanding basis for a particular choice could be found. The work, however, did clearly show the interaction between the constitutive relations, the boundary conditions, and the energy flow vector.

Agranovich and Ginzburg [7,8] and Bokut' *et al.* [9] use a constitutive equation for $\mathbf{D}$ that involves two macroscopic tensors involved in optical activity, one of which has a gradient that contributes. The sum of them determines the bulk crystal response while the second produces a surface layer response and so should affect the transmission and reflection coefficients. Agranovich and Ginzburg [7,8] find that such a model does not usually produce an energy conservation law between the incident, reflected, and transmitted waves but instead appears to require under many circumstances energy deposition in the surface layer which, they suggest, would take the form of surface waves. In a recent series of papers Silverman and collaborators [13–16] calculated and attempted to

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measure a dependence of the transmission and reflection coefficients on the constitutive relation (either Condon [3] or Born and Huang [5]) using, however, the conventional Maxwell boundary conditions and the traditional Poynting vector. We do not believe use of either of these is justified.

We now have available the three techniques necessary to derive definitive results for the transmission and reflection coefficients of an optically active medium and, in so doing, answer the vexing questions about boundary conditions, constitutive relations, and the energy flow vector. First, we have already published a fundamental long-wavelength lattice-dynamical derivation of the constitutive relations of optical activity [6]. That derivation is based on a Lagrangian theory of interactions in dielectrics [17] which is formulated in microscopic physics before a long-wavelength limit is taken. It contains all long-wavelength modes of motion, electromagnetic, optic (both ionic and electronic), acoustic, and, just recently, intrinsic spin [18], interacting at all orders of nonlinearity in a crystal of any symmetry and structural complexity, limited only by known microscopic physics and the conservation laws. That derivation considered only the bulk optical activity interaction. Here we generalize it to include the alteration of the interaction near a surface.

Second, we recently developed a method of studying wave propagation in bounded media *without the use of any boundary conditions* [19,20]. We have demonstrated this mathematical method [19] by calculating the well-known Fresnel reflection and transmission coefficients for a local dielectric medium. The traditional method of doing this uses the Maxwell boundary conditions to match fields found from the ordinary, real-space form of the Maxwell equations. The new method, instead, operates in wave vector space. The real advantage of the new method becomes evident in treating nonlocal or wave vector dispersive interactions and was successfully applied to the exciton-polariton problem [20]. The new method can calculate the transmitted and reflected waves resulting from an incident wave on a material boundary without having to calculate the rapid field variation in a surface layer where the bulk nonlocal interaction is being modified by the presence of the surface. Nevertheless, the surface layer is accounted for and its effect on the observed waves is included. Thus the failure of certain Maxwell boundary conditions (those on normal $\mathbf{D}$ and tangential $\mathbf{H}$) is sidestepped but can be deduced from the solution. The failure of these two boundary conditions is expected [20] because the divergence of the quadrupolarization (important to optical activity) enters $\mathbf{D}$. Since that derivative acts on a material property, the quadrupole charge density (as well as the electric field), a term in each of these boundary conditions becomes indeterminate (infinity times zero) as the pillbox volume or loop area is shrunk onto the abrupt material surface. Thus the limit cannot be taken and the boundary conditions in ordinary space can only relate a field on two sides of a thin but finite and unknown layer [9,21]. The wave vector space method avoids this problem entirely.

Third, in our recent generalization of the Lagrangian formulation to include intrinsic spin [18] we carried the multipole expansions of the bound charge and current densities of a dielectric to include magnetic dipole and electric quadrupole terms (which are at the same level) in order to compare the entry of magnetization terms from the motion of bound charge and from intrinsic spin into the theory. Dependence of the stored energy of the matter on one higher-order derivative with respect to the optic mode coordinate was also included. Thus the energy conservation law there can be applied directly to an optically active medium. We emphasize that this too results from a fundamental, not phenomenological, derivation. We have not, however, included in this treatment the intrinsic spin contribution to optical activity [6].

The result of the present calculation is the transmission and reflection coefficients for a plane wave incident from a vacuum on a half space of an optically active crystal. Since the presence of linear birefringence acts only to complicate and obscure the smaller component of circular birefringence, we consider here only isotropic media (crystal classes 432 and 23 and noncrystalline media lacking a center of symmetry). We find that the most natural expression of the theory is for a circularly polarized (either of two senses) incident wave. For an arbitrary angle of incidence this produces transmitted and reflected waves of both senses of circular polarization. Our results support the presence of a surface layer represented in this long-wavelength theory as surface distributions of the various fields. It is evident, however, that these fields do not represent a surface wave. Furthermore, there is no need for an additional surface wave since we demonstrate energy conservation at the surface between the incident, reflected, and transmitted waves.

Because of the unfamiliarity of the mathematical method used here, it is worthwhile to describe its physical concept before beginning. We believe its lack of any need to use boundary conditions results from two causes. One is that in wave vector space where it operates there are no physical boundaries where solutions need to be matched. The second is that the usual boundary conditions of Maxwell theory are derived from the differential equations. Thus, when the equations are transformed to wave vector space, the physical content of the boundary conditions is transformed with them. The philosophy of the method is quite akin to a quantum mechanical calculation of a scattering problem though the geometry is much different. In such a calculation the incident beam is represented as a plane wave and the outgoing scattered spherical wave is evaluated in the asymptotic region (the far-field region in optics terminology). The rapid variations of the wave function near the scattering center correspond to large wave vectors compared to that in the asymptotic region. They need not be evaluated, since they are not observed, but their existence is inherent to the process and they thus affect the scattered wave. In the optics problem we consider an incident plane wave on a half space of matter that has a nonlocal, or wave vector dispersive, interaction with the light wave. This interaction varies rapidly in a surface layer whose width is comparable to the range of the nonlocal interaction and so possesses large wave vector components. These components are not observed but do have an effect on the amplitudes of the (far-field) transmitted and reflected waves. This new wave vector space method avoids evaluating the rapid variations of the interaction in the surface layer while still accounting for its effect on the observed long-wavelength waves of the solution.

FORMULATION

We previously published a derivation of the constitutive relations of optical activity in a bulk crystal from a fundamental Lagrangian theory of interactions in dielectrics [6]. The only change needed here is allowing the various material parameters to be functions of position and allowing the space derivative in the Euler-Lagrange equation to act on the material parameters. Thus we can write down the equations of motion of the electromagnetic field and matter directly from that work [6]. The two Maxwell equations needed to form the electric field wave equation are

$$\nabla \times \mathbf{H} - \frac{\partial \mathbf{D}}{\partial t} = \mathbf{0}, \tag{1}$$

$$\nabla \times \mathbf{E} + \frac{\partial \mathbf{B}}{\partial t} = \mathbf{0}, \tag{2}$$

where

$$\mathbf{D} \equiv \epsilon_0 \mathbf{E} + \mathbf{P} - \nabla \cdot \overleftrightarrow{\mathbf{Q}}, \tag{3}$$

$$\mathbf{H} \equiv \frac{1}{\mu_0}\mathbf{B} - \mathbf{M}, \tag{4}$$

and the polarization $\mathbf{P}$, magnetization $\mathbf{M}$, and the quadrupolarization $\overleftrightarrow{\mathbf{Q}}$ are expressed in terms of the optic mode normal coordinates, $\eta^{Tr} = N^r + \eta^r$, by

$$P_i = \sum_r c_i^r \eta^{Tr}, \tag{5}$$

$$M_i = \frac{1}{2}\epsilon_{ijk} \sum_{rs} q_{jk}^{rs} \eta^{Tr} \dot{\eta}^s, \tag{6}$$

$$Q_{ij} = \frac{1}{2} \sum_{rs} q_{ij}^{rs} \eta^{Tr} \eta^{Ts}. \tag{7}$$

Here the summations run over the number of optic modes of the primitive unit cell, $\mathbf{c}^{\mathbf{r}}$ is a measure of the electric dipole charge density of the optic mode [6], q_{ij}^{rs} is a measure of the quadrupolar charge density [6], and ϵ_{ijk} is the permutation tensor. The optic mode coordinate in general possesses a spontaneous value N^r in the absence of any perturbations of the medium. Its varying part is η^r. Since optical activity is a linear phenomenon, only the parts of $\mathbf{M}$ and $\overleftrightarrow{\mathbf{Q}}$ linear in η^r are retained and N^r can be dropped from $\mathbf{P}$ entirely.

It should be remarked that we have defined $\mathbf{D}$ and $\mathbf{H}$ in the conventional way, that is, with $\nabla \cdot \overleftrightarrow{\mathbf{Q}}$ contributing to $\mathbf{D}$ and with $\mathbf{M}$ contributing to $\mathbf{H}$. Others [8,22] prefer to make the interaction look entirely electric by deleting $\mathbf{M}$ from $\mathbf{H}$ and by adding $\int \nabla \times \mathbf{M} dt$ to $\mathbf{D}$. While this definitional change is permissible, it does not make the interaction entirely electric because the magnetic dipole interaction still enters the optic mode dynamical equation where there is no $\mathbf{D}$ to absorb it. While the magnetization resulting from the motion of bound charge combines in the optic mode equation with the quadrupolarization term, any magnetization from intrinsic spin [18] (which has not been included here for simplicity) cannot combine in such a way. That dynamical equation takes the form for an inhomogeneous medium (of which a bounded medium is an example)

$$\frac{\partial^2 \eta^r}{\partial t^2} = c_j^r E_j - \Omega_r^2 \eta^r + \sum_s N^s q_{jk}^{sr} E_{j,k} + \sum_s (L_k^{sr} - L_k^{rs}) \eta_{,k}^s + \sum_s L_{k,k}^{rs} \eta^r, \tag{8}$$

where Ω_r is the transverse optic frequency of the mode, $\mathbf{L}_k^{sr}$ is a stored energy coefficient, and the subscript notation ",k" means $\partial/\partial x_r$. The term involving q_{kj}^{rs} arises half from magnetization coupling and half from quadrupolarization coupling [6].

The other dynamical equation needed besides Eq. (8) is the electric field wave equation found by combining Eqs. (1) and (2) into

$$\nabla \times (\nabla \times \mathbf{E}) + \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = -\mu_0 \frac{\partial}{\partial t}\left(\frac{\partial \mathbf{P}}{\partial t} - \frac{\partial \nabla \cdot \overleftrightarrow{\mathbf{Q}}}{\partial t} + \nabla \times \mathbf{M} \right), \tag{9}$$

where the right side can be linearized in η^r with the use of Eqs. (5)–(7).

TRANSFORMATION TO $(\mathbf{k}, \omega)$ SPACE

We use the following notation for the Fourier transform and the inverse transform for space and time dependent fields:

$$F(\mathbf{k}, \omega) = \frac{1}{(2\pi)^4} \int F(\mathbf{x}, t) e^{-i(\mathbf{k}\cdot\mathbf{x} - \omega t)} d\mathbf{x} dt, \tag{10}$$

$$F(\mathbf{x}, t) = \int F(\mathbf{k}, \omega) e^{i(\mathbf{k}\cdot\mathbf{x} - \omega t)} d\mathbf{k} d\omega. \tag{11}$$

Material properties such as c_i^r, q_{ij}^{rs}, and L_k^{rs} are taken as only space dependent. For a half space of matter (and the other half free space) we take the space dependence of each of these three material quantities (represented generically as m) to be

$$m(\mathbf{x}) = m\theta(z), \tag{12}$$

where z is the coordinate measured perpendicular to the surface and positively inward and the step function $\theta(z)$ is defined by

$$\theta(z) = \begin{cases} 1, & z > 0 \\ 0, & z < 0. \end{cases} \tag{13}$$

Note that $\theta(z)$ is not defined at $z = 0$. The $\mathbf{k}$ space transform of Eq. (12) is

$$m\mathbf{k} = m\theta(\mathbf{k}) = \frac{m\delta(k_x)\delta(k_y)}{2\pi i(k_z - i\epsilon)}, \quad (14)$$

where δ is the Dirac delta function and ϵ is an infinitesimal positive quantity needed to obtain a convergent transform.

Inverse transforms of the form of Eq. (11) are substituted into the wave equation (9) for $\mathbf{E}$ and η^r with the expressions for $\mathbf{P}$, $\mathbf{M}$, and $\overleftrightarrow{\mathbf{Q}}$, Eqs. (5) – (7), linearized in η^r. The transforms of the material quantities c_i^r and q_{ij}^{rs} are taken as a function of $\mathbf{k}''$ while $\mathbf{k}'$ is used for η^r. The substitution of $\mathbf{k} = \mathbf{k}'' + \mathbf{k}'$ then leads to

$$\int \left\{ -k_j \epsilon_{ijk} \epsilon_{klm} k_l E_m(\mathbf{k},\omega) - \frac{\omega^2}{c^2} E_i(\mathbf{k},\omega) - \frac{\mu_0 \omega^2}{2\pi i} \sum_r \int \frac{c_i^r \eta^r(k_x,k_y,k_z',\omega) dk_z'}{k_z - k_z' - i\epsilon} + \frac{\mu_0\omega^2}{2\pi} k_j \sum_{rs} \int \frac{N^r q_{ji}^{rs} \eta^s(k_x,k_y,k_z',\omega) dk_z'}{k_z - k_z' - i\epsilon} \right\} e^{i(\mathbf{k}\cdot\mathbf{x} - \omega t)} d\mathbf{k} d\omega = 0. \quad (15)$$

In general this integral can vanish only if the integrand vanishes. Using the $\epsilon - \delta$ identity on the first term and introducing $k_0 = \omega/c$, we obtain

$$k_i k_j E_j(\mathbf{k},\omega) - k^2 E_i(\mathbf{k},\omega) + k_0^2 E_i(\mathbf{k},\omega) + \frac{k_0^2}{2\pi i \epsilon_0} \sum_r c_i^r \int \frac{\eta^r(k_x,k_y,k_z',\omega)}{k_z - k_z' - i\epsilon} dk_z' - \frac{k_0^2}{2\pi\epsilon_0} k_j \sum_{rs} N^r q_{ji}^{rs} \int \frac{\eta^s(k_x,k_y,k_z',\omega) dk_z'}{k_z - k_z' - i\epsilon} = 0. \quad (16)$$

The differential equation has now been converted to an integral equation.

Reduction to an algebraic equation can be accomplished with use of a theorem [19]. Divide the transform $F(k_z)$ into two parts,

$$F(k_z) = F^{(+)}(k_z) + F^{(-)}(k_z), \quad (17)$$

where $F^{(+)}(k_z)$ contains poles only in the upper half k_z plane $[\mathrm{Im}(k_z) > 0]$ while $F^{(-)}(k_z)$ contains poles only in the lower half k_z plane. Poles that might be thought to fall on the real axis $[\mathrm{Im}(k_z) = 0]$ are pushed off the axis by a necessary $\pm i\epsilon$ in the transform [19]. The theorem states

$$\frac{1}{2\pi i} \int \frac{F(k_z') dk_z'}{k_z - k_z' - i\epsilon} = F^+(k_z). \quad (18)$$

Using it converts the integral equation (16) to an algebraic equation

$$\{(k_0^2 - k^2)\delta_{ij} + k_i k_j\}[E_j^{(+)}(\mathbf{k},\omega) + E_j^{(-)}(\mathbf{k},\omega)] + \frac{k_0^2}{\epsilon_0} \left\{ \sum_r c_i^r - ik_\ell \sum_{rs} N^s q_{\ell i}^{sr} \right\} \eta^{r(+)}(\mathbf{k},\omega) = 0. \quad (19)$$

The optic mode equation (8) can be converted to an algebraic equation by a similar procedure except that surface distribution fields $\eta^{s(0)}$ and $\mathbf{E}^{(0)}$ arise as in the following term:

$$-\frac{1}{2\pi} \sum_s L_\ell^{rs} \int \frac{k_\ell' \eta^s(\mathbf{k}',\omega)\delta(k_x - k_x')\delta(k_y - k_y') d\mathbf{k}'}{k_z - k_z' - i\epsilon} = -ik_\ell \sum_s L_\ell^{rs} \eta^{s(+)}(\mathbf{k},\omega) + \sum_s L_3^{rs} \eta^{s(0)}(k_x,k_y,\omega), \quad (20)$$

where

$$\eta^{s(0)}(k_x,k_y,\omega) \equiv \frac{1}{2\pi} \int \eta^s(k_x,k_y,k_z',\omega) dk_z'. \quad (21)$$

Surface fields such as this always arise when the interaction is wave vector dispersive (nonlocal) and play a crucial role in determining the form of the solution. In a long-wavelength theory such as this, integrals like Eq. (21) can be shown [20] to be convergent. The optic mode equation then becomes

$$\omega^2 \eta^{r(+)} - \Omega_r^2 \eta^{r(+)} + \mathbf{c}^r \cdot \mathbf{E}^{(+)} + ik_\ell \sum_s N^s q_{\ell j}^{rs} E_j^{(+)} + ik_\ell \sum_s (L_\ell^{sr} - L_\ell^{sr}) \eta^{s(+)} + \sum_s L_3^{rs} \eta^{s(0)} - \sum_s N^s q_{3j}^{sr} E_j^{(0)} = 0. \quad (22)$$

The first three terms are by far the largest or zero-order terms while the remaining four terms are small optical activity terms which are first-order corrections. Thus an iterative method of solution is justified and $\eta^{s(+)}$ and $\eta^{s(0)}$ can be eliminated from the first-order terms by substitution of zero-order solutions

$$\eta^{s(+)} = \frac{\mathbf{c}^s \cdot \mathbf{E}^{(+)}}{\Omega_s^2 - \omega^2}, \tag{23}$$

$$\eta^{s(0)} = \frac{\mathbf{c}^s \cdot \mathbf{E}^{(0)}}{\Omega_s^2 - \omega^2}. \tag{24}$$

The solution of Eq. (22) is thus

$$\begin{aligned}\eta^{r(+)}(\mathbf{k},\omega) = &\left\{ \frac{c_j^r}{\Omega_r^2 - \omega^2} + ik_\ell \sum_s \left[\frac{N^s q_{\ell j}^{sr}}{\Omega_r^2 - \omega^2} + \frac{(L_\ell^{sr} - L_\ell^{rs}) c_j^s}{(\Omega_r^2 - \omega^2)(\Omega_s^2 - \omega^2)} \right] \right\} E_j^{(+)}(\mathbf{k},\omega) \\ &+ \sum_s \left[\frac{L_3^{rs} c_j^s}{(\Omega_r^2 - \omega^2)(\Omega_s^2 - \omega^2)} - \frac{N^s q_{3j}^{sr}}{\Omega_r^2 - \omega^2} \right] E_j^{(0)}(k_x, k_y, \omega). \end{aligned} \tag{25}$$

Substitution of this into the wave equation (19) leads to

$$\left\{ k_0^2 \left[\kappa_{ij}(\omega) + ik_\ell g_{ij\ell}(\omega) \right] - k^2 \delta_{ij} + k_i k_j \right\} E_j^{(+)}(\mathbf{k},\omega) + \left\{ (k_0^2 - k^2)\delta_{ij} + k_i k_j \right\} E_j^{(-)}(\mathbf{k},\omega) - k_0^2 g_{ij3}^S(\omega) E_j^{(0)}(\mathbf{k},\omega) = 0, \tag{26}$$

where the dielectric tensor, the bulk optical activity tensor, and the surface optical activity tensor are defined, respectively, by

$$\kappa_{ij}(\omega) \equiv \delta_{ij} + \frac{1}{\epsilon_0} \sum_r \frac{c_i^r c_j^r}{\Omega_r^2 - \omega^2}, \tag{27}$$

$$g_{ij\ell}(\omega) \equiv g_{ij\ell}^S - g_{ji\ell}^S = -g_{jil}(\omega), \tag{28}$$

$$g_{ij\ell}^S(\omega) \equiv \frac{1}{\epsilon_0} \sum_{rs} \left[\frac{N^s q_{\ell j}^{sr} c_i^r}{\Omega_r^2 - \omega^2} - \frac{c_i^r L_\ell^{rs} c_j^s}{(\Omega_r^2 - \omega^2)(\Omega_s^2 - \omega^2)} \right]. \tag{29}$$

The bulk constitutive relation (28) agrees with the previous derivation [6]. It is worth repeating a few remarks about it. This constitutive derivation has found the frequency dispersion of g_{ijk}, its interchange symmetry, and its contributing mechanisms. Tracing the origin of the terms containing the factors $q_{\ell j}^{sr}$ and c_i^r reveals that they arise from equal contributions from two combinations of mechanisms: a combination of magnetic dipole and electric dipole interactions and a combination of electric quadrupole and electric dipole interactions. These terms are exactly the terms found from a quantum mechanical derivation from precisely these combinations of interactions. Both of these combinations were missed by Born and Huang [5]. The other terms that involve L_ℓ^{rs} represent the first-order wave vector dispersive contributions from the shorter range bonding forces and thus can be expected to be smaller. Their denominator indicates they would arise from one-higher-order perturbation theory in quantum mechanics. These are the only terms found by Born and Huang [5]. The present derivation has also produced the surface interaction of Eq. (29).

Since the circular birefringence arising from optical activity is always small compared to linear birefringence when it exists, the latter acts only to complicate and obscure optical activity. It is thus helpful to restrict what follows to cubic crystals (classes 432 or 23) or amorphous media lacking a center of symmetry (solutions of optically active molecules) which lack linear birefringence. Thus we assume for the remainder of the derivation that

$$\kappa_{ij}(\omega) = \kappa(\omega)\delta_{ij}, \tag{30}$$

$$g_{ij\ell}(\omega) = g(\omega)\epsilon_{ij\ell}. \tag{31}$$

Thus the algebraic equations we must solve are the three components of the transformed (and processed) electric field wave equation,

$$\left\{ k_0^2(\kappa\delta_{ij} + ig\epsilon_{ij\ell}k_\ell) - k^2\delta_{ij} + k_i k_j \right\} E_j^{(+)}(\mathbf{k}) + \left\{ (k_0^2 - k^2)\delta_{ij} + k_i k_j \right\} E_j^{(-)}(\mathbf{k}) - k_0^2 g_{ij3}^S E_j^{(0)}(k_x, k_y) = 0, \tag{32}$$

which now contains two unknown vector transforms, $\mathbf{E}^{(+)}(\mathbf{k})$ and $\mathbf{E}^{(-)}(\mathbf{k})$, and a surface field distribution that represents the altered material response in a surface layer whose thickness is finite but small compared to the wavelength of the light in the medium.

DETERMINING THE TRANSFORM

If the Fourier transform of the electric field of a propagating wave is taken, it is seen to have a first-order pole in $\mathbf{k}$ space at its propagating wave vector [19]. Clearly the transformed electric field wave equation (32) should remain meaningful for such important values. This is possible only if the coefficient of the particular component of the electric field that diverges vanishes. When the propagating eigenmode involves more than a single electric field component, it is the coefficient of the eigenmode that must vanish at the poles. Since the second and fourth terms in the coefficient of $\mathbf{E}^{(+)}(\mathbf{k})$ mix the components appearing in the equation, the eigenmodes of $\mathbf{E}^{(+)}(\mathbf{k})$ must be found by diagonalizing the coefficient, that is,

$$\mathbf{K}^{(+)}\mathbf{E}^{(+)} = \mathbf{\Lambda}^{(+)}\mathbf{E}^{(+)}, \tag{33}$$

where

$$K_{ij}^{(+)} \equiv k_0^2(\kappa\delta_{ij} + ig\epsilon_{ij\ell}k_\ell) - k^2\delta_{ij} + k_ik_j \tag{34}$$

and $\mathbf{\Lambda}^{(+)}$ is a diagonal matrix. Since $\mathbf{K}^{(+)}$ is Hermitian, its eigenvalues are real. To simplify the algebra without restricting the problem we choose a coordinate system in which the propagation vector is in the xz plane,

$$\mathbf{k} = k_x\hat{\mathbf{i}} + k_z\hat{\mathbf{k}}, \tag{35}$$

where $\hat{\mathbf{i}}$, $\hat{\mathbf{j}}$, and $\hat{\mathbf{k}}$ are unit vectors in the x, y, and z directions. The eigenvalues of $\mathbf{E}^{(+)}$ are found to be

$$\mathbf{\Lambda}^{(+)} = \begin{bmatrix} \kappa k_0^2 - k^2 + gk_0^2k & 0 & 0 \\ 0 & \kappa k_0^2 - k^2 - gk_0^2k & 0 \\ 0 & 0 & \kappa k_0^2 \end{bmatrix}, \tag{36}$$

where k is the magnitude of $\mathbf{k}$ and we denote the rows and columns in order by $+, -$, and L. The corresponding normalized eigenvectors are found to be

$$\hat{\mathcal{E}}_+ = \frac{1}{\sqrt{2}k}(-k_z\hat{\mathbf{i}} + ik\hat{\mathbf{j}} + k_x\hat{\mathbf{k}}), \tag{37a}$$

$$\hat{\mathcal{E}}_- = \frac{1}{\sqrt{2}k}(-k_z\hat{\mathbf{i}} - ik\hat{\mathbf{j}} + k_x\hat{\mathbf{k}}), \tag{37b}$$

$$\hat{\mathcal{E}}_L = \frac{1}{k}(k_x\hat{\mathbf{i}} + k_z\hat{\mathbf{k}}). \tag{37c}$$

Their orthonormality condition is

$$\hat{\mathcal{E}}_\alpha \cdot \hat{\mathcal{E}}_\beta^* = \delta_{\alpha\beta} \quad (\alpha, \beta = +, -, L). \tag{38}$$

The 3×3 transformation matrix V that diagonalizes $\mathbf{K}^{(+)}$ also diagonalizes $\mathbf{K}^{(-)}$,

$$\mathbf{V}^{-1}\mathbf{K}^{(-)}\mathbf{V} = \mathbf{\Lambda}^{(-)}, \tag{39}$$

where

$$\mathbf{V} = \left[\hat{\mathcal{E}}_+, \hat{\mathcal{E}}_-, \hat{\mathcal{E}}_L\right] \tag{40}$$

because $\mathbf{K}^{(-)}$ is of higher symmetry than $\mathbf{K}^{(+)}$. The eigenvalues of $\mathbf{E}^{(-)}$ are thus found to be

$$\mathbf{\Lambda}^{(-)} = \begin{bmatrix} k_0^2 - k^2 & 0 & 0 \\ 0 & k_0^2 - k^2 & 0 \\ 0 & 0 & k_0^2 \end{bmatrix} \tag{41}$$

and the eigenvectors are again given by Eqs. (37). The fact that the transformation does not also diagonalize the surface field term in Eq. (32) is of no consequence to us. The object is only to find the coefficients of the eigenmodes of $\mathbf{E}^{(+)}(\mathbf{k})$ and $\mathbf{E}^{(-)}(\mathbf{k})$ which must vanish at the poles. These clearly are the diagonal elements of $\mathbf{\Lambda}^{(+)}$ and $\mathbf{\Lambda}^{(-)}$, respectively.

The vanishing of the diagonal elements of $\mathbf{\Lambda}^{(+)}$ gives the dispersion relation for each of the modes of the optically active medium and the location of the poles of $\mathbf{E}^{(+)}(\mathbf{k})$. The longitudinal mode dispersion relation simply is the vanishing of the dielectric constant $\kappa(\omega) = 0$ and does not involve optical activity. We thus do not consider it further here. The dispersion relations of the two circularly polarized propagating modes are

$$\kappa k_0^2 - k^2 \pm gk_0^2k = 0, \tag{42}$$

which yield

$$n_\pm = \frac{1}{2}(\pm gk_0 + \sqrt{4\kappa + g^2k_0^2}) \\ \cong \sqrt{\kappa} \pm gk_0/2, \tag{43}$$

where

$$k_\pm \equiv k_0n_\pm. \tag{44}$$

Since we see presently for a half space of matter bounded by the $z = 0$ plane, only the k_z component of the wave vector is a nontrivial variable in the $\mathbf{E}^{(+)}(\mathbf{k})$ transform, k_x and k_y being fixed by the problem definition and causality. Thus we express the matter dispersion relation as

$$k_z = \pm\sqrt{(k_\pm)^2 - k_x^2} \equiv \pm k_M^\pm, \tag{45}$$

where the prefactor of ±1 corresponds to the two directions of travel, not the $\pm$ mode designation. From this we see the association between the modes of the medium and the poles of $\mathbf{E}^{(+)}(\mathbf{k})$.

By a similar argument the diagonal elements of $\mathbf{\Lambda}^{(-)}$ must vanish to cancel poles corresponding to the propagating eigenmodes of $\mathbf{E}^{(-)}(\mathbf{k})$. Since k_0^2 cannot vanish, there is no pole corresponding to the longitudinal eigenvector and thus there is no propagating longitudinal mode in a vacuum, a well-known fact. The vanishing of the other two diagonal elements gives the dispersion relation of the vacuum,

$$k = k_0 \quad (\text{both} \pm \text{modes}). \tag{46}$$

We express the k_z component as

$$k_z = \pm\sqrt{k_0^2 - k_x^2} \equiv \pm k_V, \tag{47}$$

where the prefactor ±1 again refers to the two directions of travel, not the $\pm$ mode designation. From this we see the association between the modes of the vacuum and the poles of $\mathbf{E}^{(-)}(\mathbf{k})$.

With the poles of $\mathbf{E}^{(+)}(\mathbf{k})$ and $\mathbf{E}^{(-)}(\mathbf{k})$ now determined their functional forms can be written as

$$\mathbf{E}^{(-)}(\mathbf{k}) = -\frac{1}{2\pi i}\left\{\frac{a_+(k_x,k_y)\hat{\mathcal{E}}_+(\mathbf{k}^I)}{k_z - k_V + i\epsilon} + \frac{a_-(k_x,k_y)\hat{\mathcal{E}}_-(\mathbf{k}^I)}{k_z - k_V + i\epsilon} + \frac{b_+(k_x,k_y)\hat{\mathcal{E}}_+(\mathbf{k}^R)}{k_z + k_V + i\epsilon} + \frac{b_-(k_x,k_y)\hat{\mathcal{E}}_-(\mathbf{k}^R)}{k_z + k_V + i\epsilon}\right\}, \tag{48}$$

$$\mathbf{E}^{(+)}(\mathbf{k}) = \frac{1}{2\pi i}\left\{\frac{c_+(k_x,k_y)\hat{\mathcal{E}}_+(\mathbf{k}_+^T)}{k_z - k_M^+ - i\epsilon} + \frac{c_-(k_x,k_y)\hat{\mathcal{E}}_-(\mathbf{k}_-^T)}{k_z - k_M^- - i\epsilon} + \frac{d_+(k_x,k_y)\hat{\mathcal{E}}_+(\mathbf{k}_+^B)}{k_z + k_M^+ - i\epsilon} + \frac{d_-(k_x,k_y)\hat{\mathcal{E}}_-(\mathbf{k}_-^B)}{k_z + k_M^- - i\epsilon}\right\}, \tag{49}$$

where the incident, reflected, transmitted, and backward traveling wave vectors, $\mathbf{k}^I, \mathbf{k}^R, \mathbf{k}_\pm^T$, and $\mathbf{k}_\pm^B$, are not yet determined and $a_\pm, b_\pm, c_\pm$, and $d_\pm$ are as yet undetermined functions of k_x and k_y.

TRANSMISSION AND REFLECTION COEFFICIENTS

To proceed further we must specify the particular problem we wish to solve. Consider a plane wave impinging from the vacuum side at an angle θ_I to the outward normal of a half space of matter, the plane of incidence being the xz plane. Consider one pure circularly polarized incident mode at a time, say the + mode. Since the incident wave originates at $-\infty$, by causality it cannot cause the backward wave to come from $+\infty$. With this specification of the problem we can set

$$a_+ = E_0\delta(k_y)\delta(k_x - k_0\sin\theta_I), \tag{50a}$$

$$a_- = 0, \tag{50b}$$

$$b_\pm = r_\pm E_0\delta(k_y)\delta(k_x - k_0\sin\theta_R), \tag{50c}$$

$$c_\pm = t_\pm E_0\delta(k_y)\delta(k_x - k_\pm\sin\phi_\pm), \tag{50d}$$

$$d_\pm = 0, \tag{50e}$$

where the amplitude transmission coefficients $t_\pm$, the amplitude reflection coefficients $r_\pm$, the reflection angle θ_R, and the refraction angles $\phi_\pm$ are as yet unknown constants. In terms of these angles the wave vectors are given as

$$\mathbf{k}^I = k_0(\sin\theta_I, 0, \cos\theta_I), \tag{51a}$$

$$\mathbf{k}^R = k_0(\sin\theta_R, 0, -\cos\theta_R), \tag{51b}$$

$$\mathbf{k}^T_\pm = k_0 n_\pm(\sin\phi_\pm, 0, \cos\phi_\pm). \tag{51c}$$

The functions (48) and (49) with the specifications (50) and (51) are now substituted in the transformed wave equation (32). First, we note that all of the arguments of the Dirac δ functions involving k_x must be equal or a violation of causality would occur. For example, if integration over the small interval containing $k_x = k_0\sin\theta_+$ did not also contain $k_x = k_0\sin\theta_I$, then a transmitted wave would exist without an incident wave in the equation. By a similar argument $\mathbf{E}^{(0)}(k_x, k_y)$ must be proportional to the same product of Dirac δ functions. Therefore we have

$$k_x = k_0\sin\theta_I = k_0\sin\theta_R = k_0 n_\pm \sin\phi_\pm \equiv k_1, \tag{52}$$

which yields the reflection angle law

$$\theta_I = \theta_R \equiv \theta \tag{53}$$

and the refraction angle (Snell's) law

$$\sin\theta = n_\pm \sin\phi_\pm. \tag{54}$$

With these we can now write

$$k_V = k_0\cos\theta, \tag{55}$$

$$k_M^\pm = k_0 n_\pm \cos\phi_\pm. \tag{56}$$

The substitution into Eq. (32) gives three component equations which we consider individually. With the help of an identity,

$$k_M^\pm(k_0^2\kappa - k_1^2 - k_z^2) \pm k_0^2 g k_\pm k_z = (k_M^\pm - k_z)\left[k_M^\pm k_z + (k_M^\pm)^2 \mp k_0^2 g k_\pm\right], \tag{57}$$

the x component simplifies to a first-degree polynomial in k_z, the lone remaining variable in the equation. The equation can be satisfied only if the coefficient of k_z and the constant term separately vanish,

$$t_+\cos\phi_+ + t_-\cos\phi_- - (1 - r_- - r_+)\cos\theta = 0, \tag{58}$$

$$n_- t_+ + n_+ t_- - (1 + r_- + r_+) - A_1 = 0, \tag{59}$$

where A_1 is the $i = 1$ component of

$$A_i = \sqrt{2}\pi i k_0 g^S_{ij3} E_j^{(0)}/E_0 \tag{60}$$

and $\mathbf{E}^{(0)}$ is the amplitude coefficient of $\delta(k_y)\delta(k_x - k_1)$ in $\mathbf{E}^{(0)}(k_x, k_y)$. Similar handling of the y component also yields a first-degree polynomial in k_z, the vanishing of whose coefficients yields

$$-t_+ + t_- + 1 - r_- + r_+ = 0, \tag{61}$$

$$-n_-\cos\phi_+ t_+ + n_+\cos\phi_- t_- + \cos\theta(1 + r_- - r_+) + iA_2 = 0. \tag{62}$$

Similar handling of the z component yields only a constant equaling zero,

$$-t_+\cos\phi_+ - t_-\cos\phi_- + (1 - r_- - r_+)\cos\theta + \csc\theta A_3 = 0, \tag{63}$$

because the coefficient of k_z vanishes identically.

At this point it appears that we have five equations, (58), (59), (61) – (63), in seven unknowns, $t_\pm, r_\pm, A_1, A_2$, and A_3. However, $\mathbf{A}$ involves $\mathbf{E}^{(0)}$ which can be evaluated in terms of $t_\pm$ and $r_\pm$ by an equation analogous to Eq. (21). Such integrals converge only when interpreted within a long-wavelength theory as discussed before [20]. Substituting Eqs. (48) and (49) into the analog of Eq. (21) and following that procedure [20] yields

$$\mathbf{E}^{(0)}(k_x, k_y, \omega) = \frac{E_0}{4\pi}\{\hat{\mathcal{E}}_+(\mathbf{k}^I) + r_+\hat{\mathcal{E}}_+(\mathbf{k}^R) + r_-\hat{\mathcal{E}}_-(\mathbf{k}^R) + t_+\hat{\mathcal{E}}_+(\mathbf{k}^T_+) + t_-\hat{\mathcal{E}}_-(\mathbf{k}^T_-)\} \times\delta(k_y)\delta(k_x - k_1). \tag{64}$$

It is also important to interpret the surface optical activity tensor g^S_{ijk} from a long-wavelength point of view. As it stands, it has no interchange symmetry like that characterizing the bulk tensor of Eq. (28) as befits a tensor representing the first monolayer of an arbitrarily oriented crystal surface. If the surface layer that the surface field term represents were that thin, its effect on the long-wavelength transmission-reflection problem would be negligible. Its effect is likely to be measurable only if the surface layer has a finite thickness but necessarily small compared to a wavelength of the light. In this long-wavelength view the surface tensor would have the symmetry of the bulk crystal which we take to be cubic. Thus we first antisymmetrize g^S_{ijk} and then use Eqs. (28) and (31) to obtain

$$\begin{aligned} g^S_{ijk} &= \frac{1}{2}(g^S_{ijk} - g^S_{jik}) \\ &= \frac{1}{2} g_{ijk} \\ &= \frac{g}{2}\epsilon_{ijk}. \end{aligned} \tag{65}$$

A similar condition to this was found by Agranovich and Ginzburg [7,8] on their surface and bulk phenomenological constants as the only condition that would prevent energy deposition in the form of surface waves. We find no evidence of surface waves in this derivation leading to Eq. (65) which has not needed to use any energy argument. Instead Eq. (65) results here from a natural interpretation of this derivation in the long-wavelength sense.

With the use of Eqs. (60), (64), and (65) the components of **A** now become

$$\begin{aligned} A_1 &= \frac{k_0 g}{4}(-1 - r_+ + r_- - t_+ + t_-) \\ &= -\frac{k_0 g}{2}(t_+ - t_-), \end{aligned} \tag{66}$$

$$\begin{aligned} A_2 &= \frac{i k_0 g}{4}\left[\cos\theta(1 - r_+ - r_-) + t_+\cos\phi_+ + t_-\cos\theta_-\right] \\ &= \frac{i k_0 g}{2}(t_+\cos\phi_+ + t_-\cos\phi_-), \end{aligned} \tag{67}$$

$$A_3 = 0. \tag{68}$$

The necessity of A_3 vanishing is also apparent by combining Eqs. (58) and (63). With the use of Eq. (68) those two equations are now identical. Thus by combining Eqs. (58), (59), (61), (62), (66), and (67) we obtain four equations in the four unknowns $r_\pm$ and $t_\pm$,

$$t_+\cos\phi_+ + t_-\cos\phi + (r_+ + r_-)\cos\theta = \cos\theta, \tag{69a}$$

$$t_+ n + t_- n - r_+ - r_- = 1, \tag{69b}$$

$$t_+ - t_- - r_+ + r_- = 1, \tag{69c}$$

$$t_+ n\cos\phi_+ - t_- n\cos\phi_- + (r_+ - r_-)\cos\theta = \cos\theta, \tag{69d}$$

where, rather remarkably, the effect of the surface layer terms is to convert both n_+ and n_- in the equations to n where

$$n \equiv \sqrt{\kappa} = \frac{1}{2}(n_+ + n_-). \tag{70}$$

The solutions of Eqs. (69) are

$$t_+ = 2(n+1)\cos\theta(\cos\phi_- + \cos\theta)/D, \tag{71a}$$

$$t_- = 2(n-1)\cos\theta(\cos\phi_+ - \cos\theta)/D, \tag{71b}$$

$$r_+ = 2n(\cos\theta - \cos\phi_+)(\cos\theta + \cos\phi_-)/D, \tag{71c}$$

$$r_- = (n^2 - 1)\cos\theta(\cos\phi_+ + \cos\phi_-)/D, \tag{71d}$$

$$\begin{aligned} D \equiv\, & (n^2+1)\cos\theta(\cos\phi_+ + \cos\phi_-) \\ & + 2n(\cos\phi_+\cos\phi_- + \cos^2\theta). \end{aligned} \tag{71e}$$

These solutions apply to a + mode incident wave. An analogous derivation of a − mode incident wave yields solutions obtainable from Eqs. (71) by interchanging the + and − subscripts on *both* sides of the equations. Note that at normal incidence for an incident + mode

$$t_+ = 2/(n+1), \tag{72a}$$

$$t_- = r_+ = 0, \tag{72b}$$

$$r_- = (n-1)/(n+1). \tag{72c}$$

Thus in this special case there is mode preservation on transmission and mode reversal on reflection.

REAL-SPACE FIELDS

The real-space electric field can be found from the inverse transform (11) with the use of Eqs. (17), (48), and (49) to be

$$\begin{aligned} \mathbf{E}(\mathbf{x},\omega) = E_0\{ & \hat{\mathcal{E}}_+(\mathbf{k}^I)\theta(-z)e^{i(\mathbf{k}^I\cdot\mathbf{x}-\omega t)} + r_+\hat{\mathcal{E}}_+(\mathbf{k}^R)\theta(-z)e^{i(\mathbf{k}^R\cdot\mathbf{x}-\omega t)} + r_-\hat{\mathcal{E}}_-(\mathbf{k}^R)\theta(-z)e^{i(\mathbf{k}^R\cdot\mathbf{x}-\omega t)} \\ & + t_+\hat{\mathcal{E}}_+(\mathbf{k}^T_+)\theta(z)e^{i(\mathbf{k}^T_+\cdot\mathbf{x}-\omega t)} + t_-\hat{\mathcal{E}}_-(\mathbf{k}^T_-)\theta(z)e^{i(\mathbf{k}^T_-\cdot\mathbf{x}-\omega t)}\}, \end{aligned} \tag{73}$$

where the real part represents the physical field. Note that from the definition of $\theta(z)$, Eq. (13), the electric field of Eq. (73) is not defined at $z = 0$. The field at $z = 0$ obtained by taking the inverse transform of Eq. (64) is

$$\mathbf{E}^{(0)}(\mathbf{x},\omega) = \frac{E_0}{2}\{\hat{\mathcal{E}}_+(\mathbf{k}^I) + r_+\hat{\mathcal{E}}_+(\mathbf{k}^R) + r_-\hat{\mathcal{E}}_-(\mathbf{k}^R) + t_+\hat{\mathcal{E}}_+(\mathbf{k}^T_+) + t_-\hat{\mathcal{E}}_-(\mathbf{k}^T_-)\}e^{i(k_1 x-\omega t)}\delta(z)$$
$$= \mathbf{E}^{(0)}(x,\omega)\delta(z). \quad (74)$$

The presence of the $\delta(z)$ function represents the idealization of the surface layer into an infinitesimally thin layer and indicates that integration over this layer is intended and that the surface field value is thus the coefficient of $\delta(z)$. Continuity of the tangential components of $\mathbf{E}(\mathbf{x},\omega)$ now follows using Eqs. (69a) and (69c) to be

$$E_x(x,z\to 0^-,\omega) = E_x^{(0)}(x,\omega) = E_x(x,z\to 0^+,\omega), \quad (75a)$$

$$E_y(x,z\to 0^-,\omega) = E_y^{(0)}(x,\omega) = E_y(x,z\to 0^+,\omega). \quad (75b)$$

While the normal component of $\mathbf{E}(\mathbf{x},\omega)$ is not expected to be continuous, it is interesting that the surface field splits the difference,

$$E_z^{(0)}(x,\omega) = \frac{1}{2}\left[E_z(x,z\to 0^+,\omega) + E_z(x,z\to 0^-,\omega)\right]. \quad (76)$$

The magnetic induction field can be found from its inverse transform to be

$$\mathbf{B}(x,z,\omega) = \frac{E_0}{\omega}\{\mathbf{k}^I\times\hat{\mathcal{E}}_+(\mathbf{k}^I)\theta(-z)e^{i(\mathbf{k}^I\cdot\mathbf{x}-\omega t)} + r_+\mathbf{k}^R\times\hat{\mathcal{E}}_+(\mathbf{k}^R)\theta(-z)e^{i(\mathbf{k}^R\cdot\mathbf{x}-\omega t)}$$
$$+ r_-\mathbf{k}^R\times\hat{\mathcal{E}}_-(\mathbf{k}^R)\theta(-z)e^{i(\mathbf{k}^R\cdot\mathbf{x}-\omega t)} + t_+\mathbf{k}^T_+\times\hat{\mathcal{E}}_+(\mathbf{k}^T_+)\theta(z)e^{i(\mathbf{k}^T_+\cdot\mathbf{x}-\omega t)} + t_-\mathbf{k}^T_-\times\hat{\mathcal{E}}_-(\mathbf{k}^T_-)\theta(z)e^{i(\mathbf{k}^T_-\cdot\mathbf{x}-\omega t)}\} \quad (77)$$

and the surface magnetic field to be

$$\mathbf{B}^{(0)}(x,z,\omega) = \frac{E_0\nabla}{2i\omega}\times\{\hat{\mathcal{E}}_+(\mathbf{k}^I) + r_+\hat{\mathcal{E}}_+(\mathbf{k}^R) + r_-\hat{\mathcal{E}}_-(\mathbf{k}^R) + t_+\hat{\mathcal{E}}_+(\mathbf{k}^T_+) + t_-\hat{\mathcal{E}}_-(\mathbf{k}^T_-)\} \times e^{i(k_1 x-\omega t)}\delta(z). \quad (78)$$

The expected continuity of the normal component of $\mathbf{B}$ at the surface can now be shown as

$$B_z(x,z\to 0^-,\omega) = B_z^{(0)}(x,\omega) = B_z(x,z\to 0^+,\omega). \quad (79)$$

The $\mathbf{D}$ and $\mathbf{H}$ vectors can also be found by the inverse transform. As expected from the argument presented in the Introduction, it is found that the usual boundary conditions of continuity of normal $\mathbf{D}$ and tangential $\mathbf{H}$ are violated. The lack of continuity in each involves the part of the optical activity tensor that enters the respective constitutive relation [6] for $\mathbf{D}$ and for $\mathbf{H}$. It is the beauty of this method that the loss of those boundary conditions is inconsequential since this method uses no boundary conditions at all.

ENERGY CONSERVATION

Energy conservation in the transmission-reflection process is a fundamental requirement but has been difficult to satisfy in some calculations [7,15]. The problem arises from the debate on the proper form of the energy propagation vector in an optically active medium. The theory [17] that produced the fundamental constitutive derivation for optical activity [6] has also produced a fundamental and general derivation of the energy conservation law [18] for a very general dielectric medium. It includes energy propagation by light waves, acoustic waves, spin waves, polariton waves, etc. including nonlinear effects. It is easily specialized to optical activity.

Before, however, considering the medium let us find the Poynting vector for the incident and reflected waves on the vacuum side. It simply is

$$\langle\mathbf{S}\rangle = \frac{1}{2\mu_0}\mathrm{Re}\{\mathbf{E}\times\mathbf{B}^*\}, \quad (80)$$

where $\langle\,\rangle$ denotes the time average, Re denotes the "real part of," and the parts of $\mathbf{E}(\mathbf{x},\omega)$ and $\mathbf{B}(\mathbf{x},\omega)$ from Eqs. (73) and (77) that contain $\theta(-z)$ are used. The component of $\langle\mathbf{S}\rangle$ normal to the surface is found to be, again for a + mode incident wave,

$$\langle\mathbf{S}(z\to 0^-)\rangle\cdot\hat{\mathbf{k}} = \frac{\epsilon_0 c E_0^2}{2}(1 - r_+^2 - r_-^2)\cos\theta. \quad (81)$$

When the general derived energy flow vector [18] is specialized to propagation in an optically active medium, it takes the form

$$\langle S_k\rangle = \frac{1}{2}\mathrm{Re}\left\{\epsilon_{kj\ell}E_j\left(\frac{B_\ell^*}{\mu_0} - M_\ell^*\right) - E_\ell\frac{\partial Q_{\ell k}^*}{\partial t} - \sum_r \frac{\partial\rho^0\Sigma}{\partial\eta,_j^r}\frac{\partial\eta^{r*}}{\partial t}\right\}. \quad (82)$$

It is instructive to evaluate the $\mathbf{E}\times\mathbf{B}/\mu_0$ term first and then the remainder. Using now the parts of $\mathbf{E}(\mathbf{x},\omega)$ and $\mathbf{B}(\mathbf{x},\omega)$ from Eqs. (73) and (77) that contain $\theta(z)$, we find

$$\frac{1}{2\mu_0}\mathrm{Re}\{\mathbf{E}\times\mathbf{B}^*\}=\frac{\epsilon_0 c E_0^2}{2}\Bigg\{n_+t_+^2(\sin\phi_+\hat{\mathbf{i}}+\cos\phi_+\hat{\mathbf{k}})+n_-t_-^2(\sin\phi_-\hat{\mathbf{i}}+\cos\phi_-\hat{\mathbf{k}})$$
$$-t_+t_-\sin^2\left(\frac{\phi_+-\phi_-}{2}\right)\left[(n_+\sin\phi_++n_-\sin\phi_-)\hat{\mathbf{i}}+(n_+\cos\phi_++n_-\cos\phi_-)\hat{\mathbf{k}}\right]\Bigg\}. \quad (83)$$

It is easily shown that the terms proportional to t_+t_- are second order in the optical activity parameter gk_0 and so may be dropped.

With the use of the linearized forms of $\mathbf{M}$ and $\overleftrightarrow{\mathbf{Q}}$ from Eqs. (6) and (7), the real-space solution of Eq. (8) analogous to Eq. (25), and the part of $\mathbf{E}(\mathbf{x},\omega)$ from Eq. (73) that contains $\theta(z)$ we find successively

$$\frac{1}{2}\mathrm{Re}\left\{-\epsilon_{kj\ell}E_jM_\ell^*-E_\ell\frac{\partial Q_{\ell k}^*}{\partial t}-\sum_r\frac{\partial\rho^0\Sigma}{\partial\eta^r_{,k}}\frac{\partial\eta^{r*}}{\partial t}\right\}=-\frac{1}{2}\sum_{rs}\left[\frac{N^s q_{ki}^{sr}c_j^r}{\Omega_r^2-\omega^2}+\frac{c_i^r L_k^{rs}c_j^s}{(\Omega_r^2-\omega^2)(\Omega_s^2-\omega^2)}\right]\mathrm{Re}\left\{E_i\frac{\partial E_j^*}{\partial t}\right\}$$
$$=\frac{\epsilon_0 c E_0^2}{2}\left(\frac{k_0 g}{2}\right)\{-t_+^2(\sin\phi_+\hat{i}_k+\cos\phi_+\hat{k}_k)$$
$$+t_-^2(\sin\phi_-\hat{i}_k+\cos\phi_-\hat{k}_k)\}, \quad (84)$$

where terms second-order in k_0g have been dropped. Note now that when Eqs. (83) and (84) are added the terms in Eq. (84) are such as to convert both n_+ and n_- appearing in Eq. (83) to n of Eq. (70). This is similar to the effect of Eqs. (66)–(68) on Eqs. (69). The normal component of the sum of Eqs. (83) and (84) is now

$$\langle\mathbf{S}(z\to0^+)\rangle\cdot\hat{\mathbf{k}}=\frac{\epsilon_0 c E_0^2}{2}n(t_+^2\cos\phi_++t_-^2\cos\phi_-). \quad (85)$$

Equating Eq. (81) and Eq. (85) produces

$$(1-r_+^2-r_-^2)\cos\theta=n(t_+^2\cos\phi_++t_-^2\cos\phi_-). \quad (86)$$

Substitution of the solutions for $t_\pm$ and $r_\pm$ from Eqs. (71) into this equation now verifies energy conservation between the incident wave, the reflected waves, and the refracted waves,

$$\langle\mathbf{S}(z\to0^-)\rangle\cdot\hat{\mathbf{k}}=\langle\mathbf{S}(z\to0^+)\rangle\cdot\hat{\mathbf{k}}, \quad (87)$$

at the surface of an optically active (gyrotropic) medium. Note that Eq. (86), being a quadratic relationship, is a stringent test of the solution for r_+, r_-, t_+, and t_- which result from linear equations. Calculation of the energy propagation in the surface layer shows it to be second order in the optical activity parameter and thus negligible.

ENERGY DENSITY AND GROUP VELOCITY

The general energy conservation law [18] derived for dielectric media also produces an expression for the energy density which can be specialized to optical activity. For this case it becomes

$$H=\frac{1}{2}\sum_a\left(\frac{\partial\eta^a}{\partial t}\right)^2+\frac{1}{2}\sum_a\Omega_a^2(\eta^a)^2+\sum_{ab}L_k^{ab}\eta^a\eta^b{}_{,k}$$
$$+\frac{\epsilon_0E^2}{2}+\frac{B^2}{2\mu_0}, \quad (88)$$

where the terms in order are the kinetic energy of the optic modes, the linear restoring force energy of the optic modes, the correction to the restoring force energy arising from linear wave vector dispersion, the electric field energy, and the magnetic induction field energy. If a bulk crystal is considered, the propagating eigenmodes are given by

$$\mathbf{E}_\pm=E_0\hat{\mathcal{E}}_\pm(\mathbf{k}_\pm)e^{i(\mathbf{k}_\pm\cdot\mathbf{x}-\omega t)}, \quad (89)$$
$$\mathbf{B}_\pm=\mathbf{k}_\pm\times\mathbf{E}_\pm/\omega, \quad (90)$$

and the real-space solution of the optic mode equation (8) is

$$\eta^a=\frac{1}{\Omega_a^2-\omega^2}\Bigg\{\mathbf{c}^a\cdot\mathbf{E}_\pm+\sum_b\left[q_{ij}^{ab}N^b+\frac{(L_j^{ba}-L_j^{ab})c_i^b}{\Omega_b^2-\omega^2}\right]$$
$$\times(E_\pm)_{i,j}\Bigg\} \quad (91)$$

by using the iterative procedure used for Eq. (25). If the time average $\langle H\rangle$ is formed as in Eq. (80) and Eqs. (89)–(91) are substituted into Eq. (88), the time-averaged energy density

$$\langle H\rangle=\frac{\epsilon_0E_0^2}{2}\Bigg\{\kappa(\omega)+\frac{\omega}{2}\frac{\partial\kappa(\omega)}{\partial\omega}$$
$$\pm k_0\sqrt{\kappa(\omega)}\left[g(\omega)+\frac{\omega}{2}\frac{\partial g(\omega)}{\partial\omega}\right]\Bigg\} \quad (92)$$

is found for the two ($\pm$) circularly polarized modes. In contrast to the various phenomenological formulations, frequency dispersion is derived here and the dispersive corrections to the energy density arise naturally. The corresponding time-averaged energy flow vector is

$$\langle\mathbf{S}\rangle=\frac{1}{2}\epsilon_0cE_0^2\sqrt{\kappa(\omega)}. \quad (93)$$

Though $\langle\mathbf{S}\rangle$ does not depend on the optical activity parameter g, the ray velocity defined by

$$\mathbf{v}_r\equiv\frac{\langle\mathbf{S}\rangle}{\langle H\rangle} \quad (94)$$

does in view of Eq. (92). By a theorem [17] the ray velocity is equal to the group velocity $\mathbf{v}_g \equiv \nabla_{(\mathbf{k})}\omega$ for linear waves in homogeneous, conservative media.

CONCLUSIONS

We have brought to bear on the transmission-reflection problem of an optically active medium three important and new techniques: (1) a fundamental derivation of the constitutive relations that gives the mechanisms, the interchange symmetry, and the frequency and wave vector dispersion for both bulk and surface interactions, (2) a fundamental and general derivation of the energy flow vector in a dielectric that is easily specialized to optical activity, and (3) a wave vector space method for solving wave propagation problems in bounded media without the use of boundary conditions. Thus phenomenology has been avoided entirely.

As argued in the Introduction, the presence of a quadrupolarization contribution to the interaction is expected to cause the usual Maxwell boundary conditions of continuity of normal **D** and tangential **H** to fail [9]. Though no boundary conditions are used in the solution presented here, calculation using the solution shows the truth of these failures. On the other hand, we find that continuity of tangential **E** and normal **B** is obeyed. This is expected since **E** and **B** are the fundamental vacuum fields while the definitions of **D** and **H** are constitutive relations involving the material response functions of **P**, $\overleftrightarrow{\mathbf{Q}}$, and **M**.

The wave vector dispersive (nonlocal) nature of the optical activity interaction causes the interaction to be altered from its bulk crystal strength in a surface layer that is finite but thin compared to a wavelength, as realized also by others [7–10]. In the framework of the wave vector space method presented here the altered surface layer interaction arises naturally as surface distributions of the fields which can be evaluated as a part of the solution and which contribute importantly to that solution including the form of the transmission and reflection coefficients.

Energy conservation between the incident, reflected, and refracted waves is demonstrated in this theory. This means that there is no energy deposition in the surface layer in the form of surface waves as proposed by others [7,8]. Since the surface fields found in the present theory contain a propagating factor of $\exp i(k_1 x - \omega t)$, it might be thought that they represent surface waves. However, this factor can be seen to be simply the phase of the incident wave. A surface wave would have to have its own dispersion relation involving a special combination of material properties. No such relation arises.

We have used the new techniques to calculate the transmission and reflectivity coefficients for a half space of an optically active medium as a function of incidence angle and input polarization. Our results disagree with Silverman's calculations [15] using constitutive relations of Condon [3] and Born and Huang [5], though curiously there is agreement with part of the Condon model results. We remarked earlier that we believe Silverman's assumption of the usual Maxwell boundary conditions and the usual Poynting vector for energy flow are unjustified. Bokut' *et al.* [9] found the transmission and reflection coefficients for normal incidence and we are in agreement on them. Agranovich and Ginzburg [8] present the transmission and reflection coefficients of an optically active medium near a dipole resonance at normal incidence. The resonance gives rise to an extra propagating wave and caused them to invoke an "additional boundary condition" in finding the solution. Thus a direct comparison is not possible with our results.

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[1] We use the term *wave vector dispersion* in preference to *spatial dispersion* because of its analogy to the term *frequency dispersion* which is universally preferred over *temporal dispersion*.

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