Optical degeneracies in anisotropic layered media: Treatment of singularities in a 4×4 matrix formalism

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4×4 matrices have been used extensively to study the propagation of light in anisotropic layered systems whose principal optic axes have arbitrary orientation. We present a general theory for the propagation of light in arbitrarily anisotropic layered systems that is particularly suited for treating optical degeneracies that arise (1) when light propagates in an isotropic medium embedded within the anisotropic layers or (2) when light propagates along one of the optic axes in an anisotropic layer. Boundary conditions are applied explicitly to the electric and magnetic fields at each interface, and transfer matrices that relate the transmitted and reflected fields to the optical properties of the system are developed. Criteria are given for identifying the mathematical singularities caused by the degeneracies described above, and a method for treating the singularities in the relevant expressions is presented.

I. INTRODUCTION

A number of authors[1–11] have used a 4×4-matrix formalism to study the propagation of light in arbitrarily anisotropy layered systems. The mathematical singularities that arise in these studies, caused by optical degeneracies when an isotropic layer is embedded within the anisotropic layers or when light propagates along an optic axis of an anisotropic layer, have not been treated systematically to date. We briefly review electromagnetic wave propagation in anisotropic media, explicitly apply boundary conditions to each interface in the system, calculate the transfer matrix, and systematically treat singularities that arise under the conditions stated above.

II. THEORY

A. Eigenmodes

Consider an electromagnetic plane wave propagating in a semi-infinite ambient medium (isotropic or anisotropic), and let the wave be incident on a layered anisotropic system whose optic axes have arbitrary orientation. The wave vector of the incident wave is $\mathbf{k}_0$, and the plane of incidence is the x-z plane. Phase continuity at each of the interfaces gives the following equations:

$$k_{iy}=0,\ \ k_{ix}=k_{0x}=\omega n_0 \sin\theta/c=\omega q_0/c, \quad \text{and}$$

$$k_{iz}=\omega q_i/c, \tag{1}$$

where ω is the angular frequency of the wave, n_0 is the refractive index that characterizes the incident wave, and the subscript i refers to quantities in layer i. If each layer is homogeneous and described by the dielectric tensor (ε_{ikm}), then Maxwell's equations can be written as

$$\begin{pmatrix} \mu_i\varepsilon_{i11}-q_i^2 & \mu_i\varepsilon_{i12} & \mu_i\varepsilon_{i13}+q_0q_i \\ \mu_i\varepsilon_{i21} & \mu_i\varepsilon_{i22}-q_0^2-q_i^2 & \mu_i\varepsilon_{i23} \\ \mu_i\varepsilon_{i31}+q_0q_i & \mu_i\varepsilon_{i32} & \mu_i\varepsilon_{i33}-q_0^2 \end{pmatrix}\begin{pmatrix} E_{ix} \\ E_{iy} \\ E_{iz}\end{pmatrix}=0, \tag{2}$$

where μ_i is the magnetic permeability (assumed here to be a scalar) for layer i, and E_{ix}, E_{iy}, and E_{iz} are the x,y, and z components of the electric fields, respectively, for layer i. No restrictions have been imposed on (ε_{ikm}). For nontrivial solutions to exist,

$$\begin{vmatrix} \mu_i\varepsilon_{i11}-q_i^2 & \mu_i\varepsilon_{i12} & \mu_i\varepsilon_{i13}+q_0q_i \\ \mu_i\varepsilon_{i21} & \mu_i\varepsilon_{i22}-q_0^2-q_i^2 & \mu_i\varepsilon_{i23} \\ \mu_i\varepsilon_{i31}+q_0q_i & \mu_i\varepsilon_{i32} & \mu_i\varepsilon_{i33}-q_0^2 \end{vmatrix}=0. \tag{3}$$

The four roots of this fourth-order polynomial equation correspond to the z components of the four possible directions of the eigenmodes of the fields. The four values for the electric field eigenmodes are obtained by substituting the roots for q_i, denoted as q_{ij}, $j=1$ to 4, into Eq. (2) and solving for $\mathbf{E}_i=(E_{ix},E_{iy},E_{iz})^T$, where the superscript T denotes the transpose. Expressions for $\mathbf{E}_i$ are derived in the following section. An equivalent expression for q_{ij} for $j=1$ to 4 is given by Schubert.[10]

B. Boundary conditions

The boundary conditions on the electric and magnetic fields yield the following equations between media $i-1$ and i, denoted as surface i, provided no free charge density or surface current exists

$$(\mathbf{E}_{(i-1)1}+\mathbf{E}_{(i-2)2}+\mathbf{E}_{(i-1)3}+\mathbf{E}_{(i-1)4}-\mathbf{E}_{i1}-\mathbf{E}_{i2}-\mathbf{E}_{i3}-\mathbf{E}_{i4})\times\mathbf{n}=0, \tag{4}$$

$$\frac{1}{\mu_{i-1}}(\mathbf{k}_{(i-1)1}\times\mathbf{E}_{(i-1)1}+\mathbf{k}_{(i-1)2}\times\mathbf{E}_{(i-1)2}+\mathbf{k}_{(i-1)3}\times\mathbf{E}_{(i-1)3}+\mathbf{k}_{(i-1)4}\times\mathbf{E}_{(i-1)4})\times\mathbf{n}=\frac{1}{\mu_i}(\mathbf{k}_{i1}\times\mathbf{E}_{i1}+\mathbf{k}_{i2}\times\mathbf{E}_{i2}+\mathbf{k}_{i3}\times\mathbf{E}_{i3}+\mathbf{k}_{i4}\times\mathbf{E}_{i4})\times\mathbf{n}, \quad (5)$$

where $\mathbf{n}$ is the surface normal pointing from medium i to medium $i-1$, and the subscripts 1, 2, 3, and 4 indicate the eigenmodes of fields. We assume that the fields and $\mathbf{k}$ vectors can be written as

$$\mathbf{E}_{ij}=E_{ij}(\gamma_{ij1},\gamma_{ij2},\gamma_{ij3}) \text{ and } \mathbf{k}_{ij}=(k_{0x},0,k_{ijz}), \quad \text{with } j=1 \text{ to } 4, \quad (6)$$

where E_{ij} are the common factors of the three components of $\mathbf{E}_{ij}$. By substituting the values for q_{ij} ($j=1$ to 4) into Eq. (2), values for γ_{ijl} ($j=1$ to 4 and $l=1$ to 3) are found to be given by

$$\gamma_{i11}=\gamma_{i22}=\gamma_{i42}=-\gamma_{i31}=1, \quad (7)$$

$$\gamma_{i12}=\frac{\mu_i\varepsilon_{i23}(\mu_i\varepsilon_{i31}+q_0q_{i1})-\mu_i\varepsilon_{i21}(\mu_i\varepsilon_{i33}-q_0^2)}{(\mu_i\varepsilon_{i33}-q_0^2)(\mu_i\varepsilon_{i22}-q_0^2-q_{i1}^2)-\mu_i^2\varepsilon_{i23}\varepsilon_{i32}}, \quad (8)$$

$$\gamma_{i13}=-\frac{\mu_i\varepsilon_{i31}+q_0q_{i1}}{\mu_i\varepsilon_{i33}-q_0^2}-\frac{\mu_i\varepsilon_{i32}}{\mu_i\varepsilon_{i33}-q_0^2}\gamma_{i12}, \quad (9)$$

$$\gamma_{i21}=\frac{\mu_i\varepsilon_{i32}(\mu_i\varepsilon_{i13}+q_0q_{i2})-\mu_i\varepsilon_{i12}(\mu_i\varepsilon_{i33}-q_0^2)}{(\mu_i\varepsilon_{i33}-q_0^2)(\mu_i\varepsilon_{i11}-q_{i2}^2)-(\mu_i\varepsilon_{i13}+q_0q_{i2})(\mu_i\varepsilon_{i31}+q_0q_{i2})}, \quad (10)$$

$$\gamma_{i23}=-\frac{\mu_i\varepsilon_{i31}+q_0q_{i2}}{\mu_i\varepsilon_{i33}-q_0^2}\gamma_{i21}-\frac{\mu_i\varepsilon_{i32}}{\mu_i\varepsilon_{i33}-q_0^2}, \quad (11)$$

$$\gamma_{i32}=\frac{\mu_i\varepsilon_{i21}(\mu_i\varepsilon_{i33}-q_0^2)-\mu_i\varepsilon_{i23}(\mu_i\varepsilon_{i31}+q_0q_{i3})}{(\mu_i\varepsilon_{i33}-q_0^2)(\mu_i\varepsilon_{i22}-q_0^2-q_{i3}^2)-\mu_i^2\varepsilon_{i23}\varepsilon_{i32}}, \quad (12)$$

$$\gamma_{i33}=\frac{\mu_i\varepsilon_{i31}+q_0q_{i3}}{\mu_i\varepsilon_{i33}-q_0^2}+\frac{\mu_i\varepsilon_{i32}}{\mu_i\varepsilon_{i33}-q_0^2}\gamma_{i32}, \quad (13)$$

$$\gamma_{i41}=\frac{\mu_i\varepsilon_{i32}(\mu_i\varepsilon_{i13}+q_0q_{i4})-\mu_i\varepsilon_{i12}(\mu_i\varepsilon_{i33}-q_0^2)}{(\mu_i\varepsilon_{i33}-q_0^2)(\mu_i\varepsilon_{i11}-q_{i4}^2)-(\mu_i\varepsilon_{i13}+q_0q_{i4})(\mu_i\varepsilon_{i31}+q_0q_{i4})}, \quad (14)$$

$$\gamma_{i43}=-\frac{\mu_i\varepsilon_{i31}+q_0q_{i4}}{\mu_i\varepsilon_{i33}-q_0^2}\gamma_{i41}-\frac{\mu_i\varepsilon_{i32}}{\mu_i\varepsilon_{i33}-q_0^2}. \quad (15)$$

Here, we have used the convention that q_{i1} and q_{i2} represent forward traveling modes, and q_{i3} and q_{i4} represent backward traveling modes.

It should be noted that each γ_{ij} is unique only up to a constant factor, since the three equations represented by Eq. (2) are homogeneous. However, this factor can be absorbed by E_{ij}. Therefore, this uncertainty has no effect on the later discussion.

In the method above, we assumed that two of the equations from Eq. (2) are independent. However, this assumption may not be valid when the eigenfields are degenerate. These degeneracies may lead to zero denominators in the expressions for γ_{ijl} ($j=1$ to 4, $l=1$ to 3), thus giving rise to singularities. To give a systematic, comprehensive treatment of the singularities, two problems must be considered. First, the singularities must be identified, and second, the singularities must be removed. Using linear algebra, it can be proved that when all roots in Eq. (3) are different, the rank of the matrix of Eq. (2) is two. Two equations can always be chosen from Eq. (2) to solve for γ_{ijl}, giving Eqs. (7)–(15) or equivalent expressions obtained by Cramer's rule, and no singularities arise. When repeated roots occur, however, there are only two cases to consider. For the first case, if the materials are lossless *or* if they can be represented by diagonal dielectric tensors relative to the laboratory coordinate system, the rank of the 3×3 matrix in Eq. (2) becomes one, and singularities occur. For the second case, if the materials are absorptive *and* the dielectric tensor contains off-diagonal elements, no generalizations can be made at this time, and singularities must be determined by examining individual γ_{ijl}'s. In *all* situations where singularities occur, however, only one of the three equations represented by Eq. (2) is independent, the solution of which consists of two independent vectors, corresponding to two eigenmodes of the fields. In what follows, we demonstrate how to deal with these singularities.

In general, the diagonal entries are not zero, and they are the dominant terms in the dielectric tensor. Usually $\mu_i\varepsilon_{i33}-q_0^2\neq 0$, and we would choose the third equation from Eq. (2) as the independent equation from which to develop new expressions for γ_{ijl}. If $q_{i1}=q_{i2}$, then the values obtained for γ_{ijl} in Eqs. (8)–(15) can be replaced by the following values for γ_{ijl} ($j=1,2$; $l=1,2,3$), which are two independent vectors of the solution of Eq. (2):

$$\gamma_{i11}=\gamma_{i22}=1, \tag{16}$$

$$\gamma_{i12}=\gamma_{i21}=0, \tag{17}$$

$$\gamma_{i13}=-\frac{\mu_i\varepsilon_{i31}+q_0q_{i1}}{\mu_i\varepsilon_{i33}-q_0^2}, \tag{18}$$

$$\gamma_{i23}=-\frac{\mu_i\varepsilon_{i32}}{\mu_i\varepsilon_{i33}-q_0^2}. \tag{19}$$

Any other two independent vectors that are the linear combination of the above two vectors are equally valid.

Similarly, if $q_{i3}=q_{i4}$, then the values of γ_{ijl} in Eqs. (8)–(15) can be replaced by the following expressions for γ_{ijl} ($j=3,4$; $l=1,2,3$):

$$\gamma_{i31}=-\gamma_{i42}=-1, \tag{20}$$

$$\gamma_{i32}=\gamma_{i41}=0, \tag{21}$$

$$\gamma_{i33}=\frac{\mu_i\varepsilon_{i31}+q_0q_{i3}}{\mu_i\varepsilon_{i33}-q_0^2}, \tag{22}$$

$$\gamma_{i43}=-\frac{\mu_i\varepsilon_{i32}}{\mu_i\varepsilon_{i33}-q_0^2}. \tag{23}$$

Equations (16)–(23) are the replacements for Eqs. (7)–(15) regardless of whether the singularities arise from the first case or from the second case.

Using the expressions for γ_{ijl}, Eqs. (4) and (5) can be rewritten in matrix form as

$$A_{i-1}E_{i-1}=A_iE_i, \tag{24}$$

where

$$A_i=\begin{pmatrix} \gamma_{i11} & \gamma_{i21} & \gamma_{i31} & \gamma_{i41} \\ \gamma_{i12} & \gamma_{i22} & \gamma_{i32} & \gamma_{i42} \\ \frac{1}{\mu_i}(q_{i1}\gamma_{i11}-q_0\gamma_{i13}) & \frac{1}{\mu_i}(q_{i2}\gamma_{i21}-q_0\gamma_{i23}) & \frac{1}{\mu_i}(q_{i3}\gamma_{i31}-q_0\gamma_{i33}) & \frac{1}{\mu_i}(q_{i4}\gamma_{i41}-q_0\gamma_{i43}) \\ \frac{1}{\mu_i}q_{i1}\gamma_{i12} & \frac{1}{\mu_i}q_{i2}\gamma_{i22} & \frac{1}{\mu_i}q_{i3}\gamma_{i32} & \frac{1}{\mu_i}q_{i4}\gamma_{i42} \end{pmatrix}$$

and

$$E_i=(E_{i1},E_{i2},E_{i3},E_{i4})^T.$$

Equation (24) can be written as

$$E_{i-1}=L_iE_i, \tag{25}$$

where

$$L_i=A_{i-1}^{-1}A_i. \tag{26}$$

Since the boundary conditions are independent, A_i is not singular. Its inverse exists, and L_i is well defined.

C. Transfer matrix

Using the boundary conditions, we can find the transfer matrix. For a system with n layers, in which a layer may be either isotropic or anisotropic, if we define a four component field vector as

$$\psi=(\psi_1,\psi_2,\psi_3,\psi_4)^T, \tag{27}$$

then the transfer matrix T relates the field ψ_0 in the ambient to the field ψ_{n+1} in the substrate by the following relation:

$$\psi_0=T\psi_{n+1}=P_0XP_{n+1}^{-1}\psi_{n+1}, \tag{28}$$

where P_0 projects the fields in the ambient to the eigenmodes in the first layer, and P_{n+1}^{-1} projects the fields in the substrate back to the form of ψ. These two matrices depend on the representation of ψ. The following relation can be derived using a similar procedure as that in Ref. 4:

$$X=L_1K_1^{-1}L_2K_2^{-1}\dots L_nK_n^{-1}L_{n+1}, \tag{29}$$

which is independent of the representation of ψ. The K_l matrix describes the transmission of the eigenfields from the top of layer l to its bottom.

$$(K_l)_{jk}=\begin{cases}0, & j\neq k\\ \exp(i\omega q_{lj}h_l/c), & j=k.\end{cases} \tag{30}$$

The q_{lj}, $j=1$ to 4, are determined from Eq. (3) for layer l.

X is a characteristic matrix of the system. If all the dielectric tensors relative to the laboratory coordinates are known, the matrix X can be straightforwardly calculated before a measurement is made. On the other hand, if the dielectric tensor is the quantity to be determined, a suitable format of ψ can be chosen to simplify P_0 and P_{n+1} for extraction of X from experimental data. In this way, the technique we have given for removing singularities applies to any stratified anisotropic system and for any representation of field vectors.

D. Analysis for $\mu_i \varepsilon_{i33} - q_0^2 \leqslant 0$

In the discussion above, we assumed that $\mu_i \varepsilon_{i33} - q_0^2 \neq 0$ and found explicit expressions for γ_{ijl}, since it is the usual case. It is still possible that $\mu_i \varepsilon_{i33} - q_0^2 = 0$. However, it should be noted that the occurrence of singularities in the expressions for the eigenfields does not depend on whether $\mu_i \varepsilon_{i33} - q_0^2 = 0$. In this section, we discuss this problem for isotropic and anisotropic media separately. For clarity, in the following, we assume the materials involved are lossless.

1. Isotropic media

Case 1. $\mu_i \varepsilon_{i33} - q_0^2 = 0$. In this case, from Eq. (3), $q_{ij} = 0$, $j = 1$ to 4. The total internal reflection occurs in layer i. The fields in the layer travel along the interface and $K_i = I$, the identity matrix. Since there is no decay in the fields, physically the thickness of the layer does not affect the fields coupled in the next layer. The fields in this layer need not be calculated. This can also be seen from Eqs. (26) and (29):

$$
\begin{aligned}
X &= L_1 K_1^{-1} \ldots A_{i-1}^{-1} A_i I^{-1} A_i^{-1} A_{i+1} \ldots L_n K_n^{-1} \dot{L}_{n+1} \\
&= L_1 K_1^{-1} \ldots A_{i-1}^{-1} A_{i+1} \ldots L_n K_n^{-1} L_{n+1}. \qquad (31)
\end{aligned}
$$

Case 2. $\mu_i \varepsilon_{i33} - q_0^2 < 0$. In this case, from Eq. (3), q_{ij}, $j = 1$ to 4, are pure imaginary numbers. They correspond to evanescent fields (leaky modes) in the layer. Physically, some energy may be radiated along the interface if the thickness of the layer is not too large. A more detailed discussion on leaky modes can be found elsewhere.[12–15]

2. Anisotropic media

In the following cases, we assume that $\mu_i \varepsilon_{i33} - q_0^2 = 0$. Then the constant term in Eq. (3) is

$$
\begin{aligned}
C = \mu_i^2 [&\mu_i \varepsilon_{i12} \varepsilon_{i23} \varepsilon_{i31} + \mu_i \varepsilon_{i13} \varepsilon_{i32} \varepsilon_{i21} - (\mu_i \varepsilon_{i22} - q_0^2) \varepsilon_{i13} \varepsilon_{i31} \\
&- \mu_i \varepsilon_{i11} \varepsilon_{i23} \varepsilon_{i32}]. \qquad (32)
\end{aligned}
$$

Case 3. If $\varepsilon_{ij} = \varepsilon_{ji}$, and $\varepsilon_{13} = \varepsilon_{23} = 0$. In this case, $C = 0$. From Eq. (3), one obtains

$$
q_{i1,3} = 0, \quad q_{i2,4} = \pm \sqrt{\mu_i \varepsilon_{i22} - q_0^2}. \qquad (33)
$$

One of the forward traveling fields is totally reflected, and depending on the value of ε_{i22}, the other one may be a transmitted or evanescent field.

Case 4. $C < 0$. In this case, since $C = \Pi_{j=1}^{4} q_{ij}$, $q_{ij} \neq 0$, $j = 1$ to 4. The only possibility for q_{ij} is that $q_{i1} < 0, q_{i2,4}$ are a pair of conjugate, pure imaginary numbers, and $q_{i3} > 0$, because if all q_{ij} are real, two of them must be negative and the other positive, making $C > 0$. Therefore, one forward mode is evanescent while the other is regular. The same is true for the backward modes.

Case 5. $C > 0$. In this case, q_{ij} may be either real or complex. A complex q_{ij} corresponds to an evanescent field.

For anisotropic media, when $\mu_i \varepsilon_{i33} - q_0^2 = 0$ and no degeneracies occur, using Cramer's rule, we choose two equations from Eq. (2) to avoid having $\mu_i \varepsilon_{i33} - q_0^2$ appear in the denominators in the expressions for γ_{ijl}. When degenacies occur, e.g., in case 3 or 5, we choose the first or second equation from Eq. (2) to solve for γ_{ijl}.

III. SUMMARY

We have developed a general theory for the propagation of light in anisotropic materials whose optic axes have an arbitrary orientation, which is particularly suited for the treatment of degeneracies that cause singularities in matrix formalism. We have identified specific cases where singularities arise in the matrix treatment, developed a general method for treating singularities, and derived mathematical expressions that allow for removal of the singularities to ensure that all the matrices are invertible.

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