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Polarization-dependent optical parameters of arbitrarily anisotropic homogeneous layered systems

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We present a unified theoretical approach to electromagnetic plane waves reflected or transmitted at arbitrarily anisotropic and homogeneous layered systems. Analytic expressions for the eigenvalues for the four-wave components inside a randomly oriented anisotropic medium are reported explicitly. As well, the partial transfer matrix for a slab of a continuously twisted biaxial material at normal incidence is described. Transition matrices for the incident and exit media are presented. Hence, a complete analytical 4×4 matrix algorithm is obtained using Berreman's 4×4 differential matrices [D. W. Berreman, J. Opt. Soc. Am. **62**, 502 (1972)]. The algorithm has a general approach for materials with linear optical response behavior and can be used immediately, for example, to analyze ellipsometric investigations.

I. INTRODUCTION

During the last decade polarization-dependent optical investigations have become standard methods to explore the properties of solids and liquids.[1] In particular, because of the rapid development of computational capabilities as well as the requirement for nondestructive methods to examine epitaxial layer systems, spectroscopic ellipsometry has become an important technique. The optical constants of various semiconductor materials have been extracted over a wide spectral range using standard models for light propagation in isotropic media.[1–3] On the other hand, investigations on arbitrarily anisotropic layered systems have been restricted mainly to data collection and qualitative analysis. This is because of the complex formulas associated with anisotropic systems, and the large number of unknown parameters such as the orientation of the crystal axes and the three direction-dependent complex refractive indices for each material. For isotropic layered media matrix methods are well known that involve 2×2 matrices.[2,3] The p (electric-field vector **E** parallel to the plane of incidence) and s (electric-field vector **E** perpendicular to the plane of incidence) modes of plane parallel electromagnetic waves are independent of each other (uncoupled modes). Thus for each mode the wave propagation can be described separately. For each component for the layered media such as ambient, slab, . . . , slab, substrate, there exists a matrix that contains the ratio of the incident and emerging parts of each mode with respect to an appropriate coordinate system. All multiple reflections between the slab interfaces that may occur in the case of transparent layers are retained in a self-consistent way. From the product of all matrices one can easily compute the reflection and transmission coefficients for the p and s modes, respectively.

In the case of birefringent epitaxial systems, the four magnetic and electric parts of the plane wave are no longer spatially independent of each other, and a so-called mode coupling appears. This will happen inside the anisotropic material during the propagation of light. Consequently 4×4 matrices are needed in order to establish a similar matrix method. Dealing with first-order Maxwell equations, Berreman showed a general way to calculate the reflection and transmission coefficients of an anisotropic slab from a wave transfer matrix of rank 4. In a similar manner to the isotropic case these matrices connect the in-plane components of all modes of the plane waves at opposite interfaces. They also include the effects of all multiple reflections if a part of the wave is traveling along a direction with no or weak absorption. In principle, the media considered are allowed to be gyrotropic as well as magnetic.[4] Unfortunately, in order to obtain this matrix, a power series has to be expanded as a function of $(\omega/c)d$ (where ω stands for frequency of the plane wave, c for the vacuum velocity of light, and d for the thickness of the slab), that depends on the spectral wavelength. Also explicit matrix expressions involving isotropic or anisotropic incident and exit media are not given. Wöhler *et al.* applied the Caley-Hamilton theorem to avoid especially the row summation in the Berreman algorithm.[5] But this faster algorithm becomes singular if the dielectric tensor reduces to a scalar. Moreover, the eigenvalues of the wave transfer matrix are needed but not presented for a general case. In general, anisotropic or isotropic slabs will be embedded between isotropic or anisotropic ambient and substrate media. Therefore, to obtain a complete matrix method, incident and exit transition matrices for the ambient (incident medium) and the substrate (exit medium) have to be introduced. Then a general wave transfer matrix can be found that contains all polarization-dependent optical parameters. Yeh reported a 4×4 matrix algebra including those transition matrices solving Maxwell's equations in **k** space.[6] Unfortunately, already for the case of an isotropic ambient, his method is not generally useful since all treated materials

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have to be anisotropic. Otherwise the algorithm becomes singular.[7]

The aim of the present paper is to report a completed algebra to calculate all the measurable polarization-dependent parameters for arbitrarily anisotropic and homogeneous layered systems including twisted media. We present explicit expressions for the four eigenvalues inside a randomly oriented biaxial material. Using these expressions we derive also a particular solution for a continuously twisted biaxial material at normal incidence. As well, we give a rigorous derivation of the incident and exit matrices. Not done so far in such a closed and complete analytical form, the algebra is already seen as a suitable method for computational data analysis of experimental results obtained from polarization-dependent measurement techniques such as generalized ellipsometry. This modern approach of ellipsometry involves the determination of three predefined and normalized optical system Jones matrix elements and has been improved very recently.[8] The Jones matrices connect the field amplitudes of plane waves before and after an optical system, and contain information about the optical-transfer function. In three follow-up publications we report on how to measure and analyze these matrices from layered anisotropic samples combining generalized ellipsometry with our matrix algebra. (i) We obtain the dielectric function tensor of uniaxial TiO_2. (ii) Through a small optical anisotropy we show evidence of the relation between the band-gap reduction and valence-band splitting in ordered $Ga_{1-x}In_xP$. (iii) We obtain directly the temperature dependence of the dielectric function tensor of nematic liquids.[8–10]

A short review of the propagation of plane waves in homogeneous layered media, and the definition of the general transfer matrix is given in Sec. II. Following Yeh's notation the derivation of the polarization-dependent parameters from the general transfer matrix is briefly reviewed in Sec. III. Here we also give a definition of the parameters to determine by generalized ellipsometry. The complete analytical algorithm for the calculation of the partial transfer matrices for anisotropic slabs is shown in Sec. IV A. The treatment of isotropic layers is explained in Appendix A. If the dielectric function tensor varies sinusoidally along the sample normal, a particular solution for the partial transfer matrix at normal incidence is reported in Sec. IV B. The incident and exit matrices are then introduced in Sec. IV C. In Appendixes B and C we demonstrate the validity of the algebra through known analytic expressions for a biaxial film-substrate system with its main axes aligned parallel to the laboratory system.

II. GENERAL TRANSFER MATRIX

Consider a layered system with plane parallel interfaces. Assume an incident light wave with wave vector $\mathbf{k}_a$ coming from the incident medium (ambient, index a, $-\infty<z<0$, complex index of refraction n_a) at an angle of incidence Φ_a (Fig. 1).[11] Then $\mathbf{k}_a$ and the wave vector of the reflected wave $\mathbf{k}_a'$ from the plane of incidence. Let A_p, A_s, and B_p, B_s, denote the complex amplitudes of the p and s modes of the incident and reflected waves, respectively. The exit medium (substrate, index f, $z_N<z<\infty$) does not include a back side. Hence there exist only two amplitudes for the

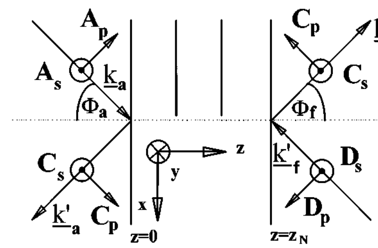

FIG. 1. Incident, reflected, and transmitted p and s modes of a plane wave with their wave vectors $\mathbf{k}_a$, $\mathbf{k}_a'$, $\mathbf{k}_f$, and $\mathbf{k}_f'$, respectively. D_s and D_p indicate the modes of the back-traveling waves inside the substrate. If the exit medium is anisotropic there may exist four different wave vectors $\mathbf{k}_f$.

transmitted p and s modes, C_p and C_s, respectively (back-traveling waves are not permitted, $D_p=0$ and $D_s=0$).[12] The Cartesian laboratory coordinate system is then defined when the plane of incidence coincides with the x-z plane, where the origin is set at the interface of the ambient and the stratified media.[13] Without loss of generality the wave vector $\mathbf{k}_a$ does not have a component parallel to the y direction.[14] In order to connect the four wave amplitudes inside the incident medium (left side of the structure in Fig. 1) with the two transmitted amplitudes inside the exit medium (right side of Fig. 1) a general transfer matrix can be defined for any given layered structure:

$$\begin{pmatrix} A_s \\ B_s \\ A_p \\ B_p \end{pmatrix} = \mathbf{T} \begin{pmatrix} C_s \\ D_s \\ C_p \\ D_p \end{pmatrix} = \begin{pmatrix} T_{11} & T_{12} & T_{13} & T_{14} \\ T_{21} & T_{22} & T_{23} & T_{24} \\ T_{31} & T_{32} & T_{33} & T_{34} \\ T_{41} & T_{42} & T_{43} & T_{44} \end{pmatrix} \begin{pmatrix} C_s \\ 0 \\ C_p \\ 0 \end{pmatrix}. \quad (1)$$

Note that $\mathbf{T}$ is just the matrix $\mathbf{M}$ used by Yeh [Eq. (22) in Ref. 6]. The tangential components of the electric- and magnetic-field vectors are continuous across the interface between two media. (If the surface current density $\mathbf{K}$ and the surface charge density σ vanish.) Thus a 4×4 matrix algebra that describes the propagation of monochromatic plane waves through the entire layered system can be introduced. If d_i is the thickness of the ith layer, a partial transfer matrix $\mathbf{T}_{\mathrm{ip}}$ that connects the in-plane wave components at the interface at $z=z_i$ with those at the next interface at $z=z_i+d_i$ can be defined for both isotropic and anisotropic layers. Hence the ordered product of all partial matrices from all N layers transfers the in-plane components at the first interface at $z=0$ to the last interface at $z=z_N$. Likewise, the incident matrix $\mathbf{L}_a$ projects the in-plane wave components of the incident and reflected waves through to the first interface. The exit matrix $\mathbf{L}_f$ projects the transmitted amplitudes from the last interface through to the exit medium that may be isotropic or anisotropic. The general transfer matrix $\mathbf{T}$ as defined in Eq. (1) is then most easily obtained from the product of all inverted matrices $\mathbf{T}_{\mathrm{ip}}$ for each layer, as well as the incident and exit matrices in the order of their appearance:

$$\mathbf{T}=\mathbf{L}_a^{-1}\prod_{i=1}^{N} \mathbf{T}_{\mathrm{ip}}(d_i)]^{-1}\mathbf{L}_f=\mathbf{L}_a^{-1}\prod_{i=1}^{N} \mathbf{T}_{\mathrm{ip}}(-d_i)\mathbf{L}_f. \quad (2)$$

Note that due to the symmetry of the coordinate system the inversion of $\mathbf{T}_p$ that is indicated in Eq. (2) does not require a matrix inversion calculation.

III. POLARIZATION-DEPENDENT OPTICAL PARAMETERS

In the following subsection we show how the measurable polarization-dependent optical parameters can be obtained immediately from the general transfer matrix and therefore through the algebra discussed here. Note that the expressions mentioned or derived here constitute the connection to the respective experiment.

A. Transmission and reflection coefficients

The transmission and reflection coefficients of layered systems are traditionally defined as the ratios of the amplitudes of the incident and reflected or transmitted waves, respectively. They can be expressed in terms of the elements of the general transfer matrix $\mathbf{T}$. Consider Eq. (1) as a system of four linear relations between the p and s components on the left and right sides of Fig. 1. As an example, one might choose the Jones reflection and transmission coefficients r_{ss} and t_{sp}, respectively.[15] Inside the substrate only transmitted waves are allowed. Therefore the ratios can be found as follows:

$$r_{ss}\equiv\left(\frac{B_s}{A_s}\right)_{A_p=0}=\frac{T_{21}T_{33}-T_{23}T_{31}}{T_{11}T_{33}-T_{13}T_{31}},$$

$$t_{sp}\equiv\left(\frac{C_p}{A_s}\right)_{A_p=0}=\frac{-T_{31}}{T_{11}T_{33}-T_{13}T_{31}}. \quad (3)$$

Note that all eight conceivable quotients of the incident and emerging wave parts can be expressed in terms of the elements of the matrix $\mathbf{T}$.[6]

B. Generalized ellipsometric parameters

The complex reflectance ratio ρ has been traditionally defined as

$$\rho=\tan\Psi e^{i\Delta}\equiv\left(\frac{B_p}{A_p}\right)\left(\frac{B_s}{A_s}\right)^{-1}, \quad (4)$$

and can be expressed through the ratios of the amplitudes of the incident and reflected waves. Let χ be the ratio of the incident modes A_p and A_s, then the ellipsometric ratio is obtained from Eq. (1) as follows:

$$\rho=\frac{1}{\chi}\,\frac{T_{41}(T_{33}-\chi T_{13})+T_{43}(\chi T_{11}-T_{31})}{T_{21}(T_{33}-\chi T_{13})+T_{23}(\chi T_{11}-T_{31})}, \quad \chi\equiv\frac{A_p}{A_s}, \quad (5)$$

or, using the expression for the transmission and reflection coefficients derived from $\mathbf{T}$,

$$\rho=[r_{pp}+r_{sp}(\chi)^{-1}](r_{ss}+r_{ps}\chi)^{-1}, \quad (6)$$

or, in a slightly different form,

$$\rho=\left(\frac{r_{pp}}{r_{ss}}+\frac{r_{sp}}{r_{ss}}(\chi)^{-1}\right)\left(1+\frac{r_{pp}}{r_{ss}}\frac{r_{ps}}{r_{pp}}\chi\right)^{-1}. \quad (7)$$

As seen in the latter equation the complex reflectance ratio ρ is then a combination of three ratios formed by the elements of the Jones reflection matrix, and depends on the ratio of the incident wave amplitudes A_p and A_s. The basis of generalized ellipsometry is to define and determine three linear independent normalized reflection matrix elements. Defining a set of those normalized elements

$$\frac{r_{pp}}{r_{ss}}\equiv R_{pp}, \quad \frac{r_{ps}}{r_{pp}}\equiv R_{ps}, \quad \frac{r_{sp}}{r_{ss}}\equiv R_{sp} \quad (8)$$

results in Eq. (7):

$$\rho=[R_{pp}+R_{sp}(\chi)^{-1}](1+R_{pp}R_{ps}\chi)^{-1}. \quad (9)$$

Equation (1) simplifies in cases where only isotropic materials are included in the layered system or in some special cases where, e.g., the Cartesian principal axes of the material layers are all oriented parallel to the axes of the laboratory coordinate system. The special behavior of the partial transfer matrix when the principal axes of the crystal system coincide with the laboratory system is shown through Appendixes B and C. In this case the ellipsometric ratio holds:

$$\rho^{is}=\frac{T_{43}T_{11}}{T_{33}T_{21}}, \quad R_s=\frac{T_{21}}{T_{11}} \quad \text{and} \quad R_p=\frac{T_{43}}{T_{33}}. \quad (10)$$

IV. CALCULATION OF THE GENERAL TRANSFER MATRIX

The attention can generally be restricted to cases where all material layers are homogeneous, so that the optical properties are independent of z except at step-function changes across interfaces. Inhomogeneous layered media can be treated in the same way if they are approximated as a stack of piecewise homogeneous layers. In Sec. IV A we report analytic expressions for the eigenvalues inside a randomly oriented anisotropic medium. Thus a fully analytic algorithm for the calculation of the partial transfer matrix for those media is available. In order to make the algorithm more transparent we give a brief review of the derivation of the initial differential equation originally reported in Ref. 4. In Sec. IV B we derive a special solution for the case of a sinusoidally varying dielectric function tensor at normal incidence using the results from Sec. IV A. The incident and exit matrices are then introduced and discussed in Sec. IV C.

A. Partial transfer matrix for anisotropic slabs

From first-order Maxwell equations Berreman derived the following set of four differential equations for the in-plane components of the electric and magnetic fields in Gaussian units and Cartesian coordinates:

$$\partial_z\Psi(z)=ik_0\Delta(z)\Psi(z),$$

$$\Psi(z)=(E_x,E_y,H_x,H_y)^T(z), \quad k_0\equiv\frac{\omega}{c}, \quad (11)$$

where ω is the angular frequency, c is the vacuum velocity of light and $(\ )^T$ denotes the transposed vector.[4] The media are assumed to be nonmagnetic ($\mu=\mathbf{E}$, where $\mathbf{E}$ is the unit matrix) and nongyrotropic ($\boldsymbol{\rho}=\mathbf{0}$).[16] The dielectric tensor ε

contains the main values of the orientationally dependent dielectric functions ε_{0x}, ε_{0y}, and ε_{0z} that belong to the intrinsic Cartesian principle axes of the anisotropic material. They are in general different from the laboratory coordinate axes. Thus a simple rotation about the three Euler angles φ, Ψ, and θ describes the orientation of the Cartesian crystal coordinate system with respect to the laboratory coordinate system:

$$\varepsilon=\mathbf{A}\begin{pmatrix}\varepsilon_{0x} & 0 & 0\\ 0 & \varepsilon_{0y} & 0\\ 0 & 0 & \varepsilon_{0z}\end{pmatrix}\mathbf{A}^{-1}, \tag{12}$$

where $\mathbf{A}$ is the orthogonal rotation matrix.[17] It should be pointed out that in general the Euler angles may vary with the angular frequency ω.

The matrix Δ defined in Eq. (11) depends on the dielectric tensor ε and the x component k_x of the wave vector $\mathbf{k}_a$:

$$\Delta=\begin{pmatrix} -k_x\dfrac{\varepsilon_{31}}{\varepsilon_{33}} & -k_x\dfrac{\varepsilon_{32}}{\varepsilon_{33}} & 0 & 1-\dfrac{k_x^2}{\varepsilon_{33}}\\ 0 & 0 & -1 & 0\\ \varepsilon_{23}\dfrac{\varepsilon_{31}}{\varepsilon_{33}}-\varepsilon_{21} & k_x^2-\varepsilon_{22}+\varepsilon_{23}\dfrac{\varepsilon_{32}}{\varepsilon_{33}} & 0 & k_x\dfrac{\varepsilon_{23}}{\varepsilon_{33}}\\ \varepsilon_{11}-\varepsilon_{13}\dfrac{\varepsilon_{31}}{\varepsilon_{33}} & \varepsilon_{12}-\varepsilon_{13}\dfrac{\varepsilon_{32}}{\varepsilon_{33}} & 0 & -k_x\dfrac{\varepsilon_{13}}{\varepsilon_{33}}\end{pmatrix},$$

$$k_x\equiv n_a\sin\Phi_a. \tag{13}$$

As long as the medium is homogeneous the matrix Δ does not depend on z, and the solution of Eq. (11) can be written formally by defining the partial transfer matrix $\mathbf{T}_p$ as follows:

$$\Psi(z+d)=\exp\left\{i\frac{\omega}{c}\Delta d\right\}\Psi(z)=\mathbf{T}_p\Psi(z),$$

$$\mathbf{T}_p\equiv\exp\left\{i\frac{\omega}{c}\Delta d\right\}. \tag{14}$$

This matrix connects the in-plane components of the electric and magnetic fields at interfaces separated by a distance d. Note that $\mathbf{T}_p$ includes the effects of all multiple reflections if a part of the wave is traveling along a direction with no or weak absorption. It may also be noted that $\mathbf{T}_p$ is unitary if the medium is nonabsorptive in any direction of propagation. This can be shown to be a direct consequence of the conservation of energy.[18] Otherwise the squares of $\Psi(z+d)$ and $\Psi(z)$ may not be equal. It should be pointed out that the partial transfer matrix $\mathbf{T}_p$ depends on the distance from the layer interfaces. If the thickness d of the layer can be determined independently then there are nine unknowns for each wavelength, including six for the main dielectric functions and three for the Euler angles.

There exist different methods to determine the partial transfer matrix $\mathbf{T}_p$ as a function of the wave transfer matrix Δ. Requiring sufficiently small thickness d, the exponential function can be expanded in a common series in the spectrally varying factor $(\omega/c)d$. However, Wöhler *et al.* showed a faster way to calculate the partial transfer matrix applying the theorem of Cayley-Hamilton.[5] The matrix function can then be expressed by a finite series expansion up to the power of $n-1$, where n is the rank of the matrix:

$$\mathbf{T}_p\equiv\exp\left\{i\frac{\omega}{c}\Delta d\right\}=\beta_0\mathbf{E}+\beta_1\Delta+\beta_2\Delta^2+\beta_3\Delta^3. \tag{15}$$

Note that for the latter equation small thicknesses are no longer a requirement to obtain $\mathbf{T}_p$. The scalars β_i must obey the following set of equations:

$$\exp\left\{i\frac{\omega}{c}q_k d\right\}=\sum_{j=0}^{3}\beta_j q_k^j,\quad k=1,\ldots,4. \tag{16}$$

(This can be read more explicitly in Ref. 5.) Here q_i are the eigenvalues of the matrix Δ. Each solution is associated with one of the four plane waves existing in a homogeneous medium. Two solutions have a positive real part and constitute the forward-traveling plane waves with respect to the chosen laboratory coordinate system. The other solutions with negative real parts are due to the back-traveling wave components.

Here we report complete analytic expressions for the eigenvalues for a randomly oriented biaxial material. For simplicity we restrict ourselves to dielectric media that obey the symmetry property $\varepsilon_{ij}=\varepsilon_{ji}$. We transform both sides of Eq. (11) into a more appropriate form, applying a unitary matrix Γ defined as follows:

$$\tilde{\Psi}(\zeta)\equiv\Gamma\Psi(\zeta),\quad \Gamma=\begin{pmatrix}1 & i & 0 & 0\\ 1 & -i & 0 & 0\\ 0 & 0 & 1 & i\\ 0 & 0 & 1 & -i\end{pmatrix}, \tag{17}$$

that results in

$$\tilde{\Psi}(\zeta)=(E^+,E^-,H^+,H^-)^T(\zeta),$$

$$H^\pm=H_x\pm iH_y,\quad E^\pm=E_x\pm iE_y. \tag{18}$$

The vector $\tilde{\Psi}$ now contains the in-plane field amplitudes of two elliptically polarized modes. Although of the same form as Eq. (11), the coefficients of the differential equation result in a changed wave transfer matrix $\tilde{\Delta}$:

$$\tilde{\Delta}\equiv\Gamma\Delta\Gamma^{-1},$$

$$\tilde{\Delta}=\begin{pmatrix} v_+ & v_- & -i\left(1-\dfrac{1}{2\varepsilon_{33}}k_x^2\right) & -\dfrac{i}{2\varepsilon_{33}}k_x^2\\ v_+ & v_- & \dfrac{i}{2\varepsilon_{33}}k_x^2 & i\left(1-\dfrac{1}{2\varepsilon_{33}}k_x^2\right)\\ f & s_+ & v_- & -v_-\\ s_- & -f & -v_+ & v_+\end{pmatrix}, \tag{19}$$

with their elements

$$v_{\pm}=\frac{1}{2\varepsilon_{33}}(-\varepsilon_{13}\pm i\varepsilon_{23})k_x,$$

$$f=-\frac{i}{2\varepsilon_{33}}(\varepsilon_{13}^2+\varepsilon_{23}^2-\varepsilon_{33}[\varepsilon_{11}+\varepsilon_{22}-k_x^2]),$$

$$s_{\pm}=\frac{1}{2\varepsilon_{33}}(2[\varepsilon_{13}\varepsilon_{23}-\varepsilon_{12}\varepsilon_{33}]) \pm i[\varepsilon_{23}^2+\varepsilon_{33}(\varepsilon_{11}-\varepsilon_{22}+k_x^2)-\varepsilon_{13}^2]. \quad (20)$$

Though not affected through the latter unitary transformation, the eigenvalues can now be found more simple from $\tilde{\Delta}$ as follows:

$$q_{1/2}^{+}=\frac{1}{2}\left\{-k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}-\left[-\frac{2}{3}t_1+\left(k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}\right)^2+\Sigma\right]^{1/2}\pm\left[-\frac{4}{3}t_1+2\left(k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}\right)^2-\Sigma-\frac{s_3}{4\left[-\frac{2}{3}t_1+\left(k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}\right)^2+\Sigma\right]^{1/2}}\right]^{1/2}\right\},$$

$$q_{1/2}^{-}=\frac{1}{2}\left\{-k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}+\left[-\frac{2}{3}t_1+\left(k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}\right)^2+\Sigma\right]^{1/2}\pm\left[-\frac{4}{3}t_1+2\left(k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}\right)^2-\Sigma+\frac{s_3}{4\left[-\frac{2}{3}t_1+\left(k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}\right)^2+\Sigma\right]^{1/2}}\right]^{1/2}\right\} \quad (21)$$

where the following abbreviations are used:

$$\Sigma=\tfrac{1}{3}[s_1(\tfrac{1}{2}\{s_2+\sqrt{s_2^2-4s_1^3}\})^{-1/3} +(\tfrac{1}{2}\{s_2+\sqrt{s_2^2-4s_1^3}\})^{1/3}],$$

$$s_1=t_1^2+12\left(k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}t_2+t_3\right),$$

$$s_2=2t_1^3+36k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}t_1t_2+108\left(t_2^2+\left[k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}\right]^2t_3\right)-72t_1t_3,$$

$$s_3=-8k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}\left(\left[k_x\frac{\varepsilon_{13}}{\varepsilon_{33}}\right]^2-t_1\right)+16t_2,$$

and

$$t_1=\frac{1}{\varepsilon_{33}}\left\{\varepsilon_{13}^2+\varepsilon_{23}^2-\varepsilon_{33}\left[\varepsilon_{11}+\varepsilon_{22}-k_x^2\left(1+\frac{\varepsilon_{11}}{\varepsilon_{33}}\right)\right]\right\},$$

$$t_2=\frac{k_x}{\varepsilon_{33}}(\varepsilon_{13}\varepsilon_{22}-\varepsilon_{12}\varepsilon_{23}-\varepsilon_{13}k_x^2), \quad (22)$$

$$t_3=i\left\{4fv_+v_-+2[s_-v_-v_--s_+v_+v_+] -\left(1-\frac{k_x^2}{\varepsilon_{33}}\right)[s_+s_-+ff]\right\}.$$

Here $q_{1/2}^{+}(q_{1/2}^{-})$ are the two eigenvalues that refer to the forward- (backward-) traveling waves, respectively. The latter formulas provide complete analytic solutions for the matrix equation Eq. (14) together with the coefficients β_i which can now be evaluated immediately following Eq. (16). The explicit expressions for eigenvalues inside an arbitrary oriented biaxial medium can be used for further discussions, for example, for cases of vanishing anisotropy, and to avoid the use of numerical root-finding algorithms. Naturally, the eigenvalues reported so far for some special orientations of uniaxial media are retained here as particular solutions.[19]

B. Partial transfer matrix for continuously twisted biaxial materials

The future application of generalized ellipsometry consists in the investigation of optical systems with increasing complexity. As an example of a layered system we report a special solution of the partial transfer matrix for a slab of a continuously twisted biaxial material. Here the dielectric function tensor depends on the spatial position with respect to the z axis. Such a homogeneous twisted medium consists of a helical structure with periodicity along the z direction. Hence the matrix Δ depends on z at each spatial position inside the slab. Let $\mathbf{n}$ be a unit vector that is oriented parallel to the z axes of the crystal coordinate system in each virtual plane formed by the twisted medium. Then P is the distance between one full turn of the vector $\mathbf{n}$ around the z axis of the laboratory coordinate system. We can express the z dependence of the dielectric function tensor that then describes a spiral per length P. For simplicity we suggest to use the elements of the dielectric function tensor appearing in Eq. (13) as those that describe the orientation and optical properties of the biaxial material at the lowest boundary of the slab. The helicoidal rotation of the vector $\mathbf{n}$ along the z axis is then described by a rotation matrix $\mathbf{B}$ that depends only on the turn per unit length P:

$$\mathbf{B}=\begin{pmatrix}\cos\zeta & -\sin\zeta & 0\\ \sin\zeta & \cos\zeta & 0\\ 0 & 0 & 1\end{pmatrix}, \quad \zeta=\frac{2\pi}{P}z, \quad (23)$$

and the elements of the dielectric function tensor at each z position are given by

$$\varepsilon(\zeta)\equiv\mathbf{B}(\zeta)\begin{pmatrix}\varepsilon_{11} & \varepsilon_{12} & \varepsilon_{13}\\ \varepsilon_{12} & \varepsilon_{22} & \varepsilon_{23}\\ \varepsilon_{13} & \varepsilon_{23} & \varepsilon_{33}\end{pmatrix}\mathbf{B}(\zeta)^{-1}. \tag{24}$$

Very similarly to what was done in Sec. IV A, we transform both sides of Eq. (11) into a more appropriate form applying the unitary matrix Γ. Next we use a matrix $\mathbf{F}(\zeta)$ to transfer the helicoidal dependence of the vector $\tilde{\Psi}$:

$$\mathbf{F}(\zeta)\equiv\mathrm{diag}(\exp\{i\zeta\},\exp\{-i\zeta\},\exp\{i\zeta\},\exp\{-i\zeta\}), \tag{25}$$

where diag( ) indicates the diagonal 4×4 matrix, introducing a vector $\tilde{\Phi}$:

$$\tilde{\Psi}(\zeta)=\mathbf{F}(\zeta)\tilde{\Phi}(\zeta). \tag{26}$$

If we substitute the last equation into Eq. (11) and carry out the derivative of $\mathbf{F}(\zeta)$ with respect to the variable ζ, we obtain another differential equation system. Again, although of the same form as Eq. (11), the coefficients result in a changed wave transfer matrix $\tilde{\Delta}$:

$$\partial_z\tilde{\Phi}(\zeta)=ik_0\tilde{\Delta}(\zeta)\tilde{\Phi}(\zeta), \quad \tilde{\Delta}(\zeta)=\mathbf{U}+k_x\mathbf{V}(\zeta)+\frac{k_x^2}{2}\mathbf{W}(\zeta). \tag{27}$$

As indicated in the latter equation the new wave transfer matrix $\tilde{\Delta}$ can be written as a sum of three matrices. The first matrix $\mathbf{U}$ is now constant with respect to the variable ζ, whereas the terms $\mathbf{V}$ and $\mathbf{W}$ do depend on ζ. Hence for vanishing incidence angles (vanishing x component of the incident wave vector) the wave transfer matrix $\tilde{\Delta}$ becomes independent of ζ, and the solution of the differential equation can then be found in our standard way. The most explicit form of the matrix $\tilde{\Delta}$ $(k_x=0)$ is given below

$$\tilde{\Delta}(k_x=0)\equiv\mathbf{U}=\begin{pmatrix}-n & 0 & -i & 0\\ 0 & n & 0 & i\\ f(k_x=0) & s_+(k_x=0) & -n & 0\\ s_-(k_x=0) & -f(k_x=0) & 0 & n\end{pmatrix}, \tag{28}$$

with

$$n=\frac{2\pi}{k_0P}. \tag{29}$$

The eigenvalues are found immediately using the expressions from the last section as follows:

$$q_\pm\equiv\sqrt{n^2-if\pm\chi}, \quad \chi=\sqrt{s_+s_--i4n^2f} \quad (k_x\equiv0), \tag{30}$$

and refer to both elliptical eigenmodes traveling inside the twisted medium. Now we can apply the theorem of Caley-Hamilton in order to solve the matrix expression mentioned in Eq. (14). A set of four complex coefficients β_i need to be found so that:

$$\exp\{ik_0q_kd\}=\beta_jq_k^j, \quad j,k=1,\ldots,4,$$

$$q_{1/2}=\pm q_+, \quad q_{3/4}=\pm q_+. \tag{31}$$

Here again d indicates the distance between the lower and the upper interface of the slab. The last equations lead to

$$\beta_0=\frac{1}{2\chi}\{q_+^2\cos\kappa_--q_-^2\cos\kappa_+\},$$

$$\beta_1=i\frac{1}{2\chi}\{(q_+^2/q_-)\sin\kappa_--(q_-^2/q_+)\sin\kappa_+\},$$

$$\beta_2=\frac{1}{2\chi}\{\cos\kappa_+-\cos\kappa_-\},$$

$$\beta_3=i\frac{1}{2\chi}\{(1/q_+)\sin\kappa_+-(1/q_-)\sin\kappa_-\}, \tag{32}$$

and the definition of the phase thickness $\kappa_\pm$ that are similar to those from the p and s modes of plane waves inside a homogeneous biaxial medium that is oriented parallel to the laboratory coordinate axes (see Appendix C):

$$\kappa_\pm\equiv k_0dq_\pm. \tag{33}$$

In order to obtain the transfer matrix that applies to the slab given in coordinates of the laboratory coordinate system, we finally perform the necessary back transformation that yields

$$\mathbf{T}_p(d)=\Gamma^{-1}\mathbf{F}\left(\zeta=\frac{2\pi}{P}d\right)(\beta_0\mathbf{E}+\beta_1\mathbf{U}+\beta_2\mathbf{U}^2+\beta_3\mathbf{U}^3)\Gamma. \tag{34}$$

Note that we need the inverse of $\mathbf{T}_p$ as indicated in Eq. (2). Yet is does not require a matrix inversion algorithm because of the property

$$\mathbf{T}_p(-d)=\Gamma^{-1}(\beta_0\mathbf{E}+\beta_1\mathbf{U}+\beta_2\mathbf{U}^2+\beta_3\mathbf{U}^3)\times\mathbf{F}\left(\zeta=-\frac{2\pi}{P}d\right)\Gamma. \tag{35}$$

Note that all formulas are valid for complex director optical constants. Note also that no approximations were included in the derivation of the transfer matrix of the helicoidal medium, and, hence, this approach is exact and general as long as we treat the case of normal incidence.

C. Transition matrices

The general transfer matrix $\mathbf{T}$ defined in Eq. (1) connects the four wave amplitudes inside the incident medium with the two transmitted amplitudes inside the exit medium. The incident wave may travel at an oblique angle of incidence. Therefore the p and s modes of the incident, reflected, and transmitted waves A_p, A_s, B_p, B_s, C_p, and C_s are in general not equal to the tangential field amplitudes at the surface of the sample at $z=0$, and at the last interface at $z=z_N$, respectively. Thus the p and s modes inside the ambient and the substrate must be connected with the in-plane wave components of the electric- and magnetic-field vectors at $z=0$ and $z=z_N$, respectively. The transition matrix $\mathbf{L}_a$ (incident matrix) projects the tangential parts of the waves existing in the incident medium through to the first interface where the transition matrix $\mathbf{L}_f$ (exit matrix) projects the tangential electric and magnetic fields from the last interface into the exit medium. Through the following sections we give a rigorous and clear derivation of these matrices with respect to our chosen laboratory coordinate system.

1. Incident matrix

Let Ψ_a be the vector of the p and s modes A_p, A_s, B_p, and B_s, respectively. The vectors Ψ_{inc} and Ψ_{ref} may contain in-plane electric- and magnetic-field components at $z=0$ associated with the incident and reflected waves, respectively. Then the incident matrix $\mathbf{L}_a$ is defined by the following equation:

$$\mathbf{L}_a\Psi_a=\Psi_{\mathrm{inc}}(z=0)+\Psi_{\mathrm{ref}}(z=0),\quad \Psi_a\equiv(A_s,B_s,A_p,B_p)^T. \tag{36}$$

In homogeneous, nonmagnetic, and isotropic media, the magnetic-field components are simply related to their connected orthogonal electric-field amplitudes through the complex index of refraction n. Hence (using simple geometry and Fig. 1) the projection of the incident wave onto the surface yields

$$\vec{H}_s(\vec{E}_p)\cdot\vec{e}_y=H_y=n_aA_p,\quad \vec{E}_s\cdot\vec{e}_y=E_y=A_s,$$
$$\vec{H}_p(\vec{E}_s)\cdot\vec{e}_x=H_x=-n_aA_s\cos\Phi_a,\quad \vec{E}_p\cdot\vec{e}_x=E_x=A_p\cos\Phi_a, \tag{37}$$

where $\vec{e}_x$ and $\vec{e}_y$ are the unit vectors of the x-y plane. Therefore, for Ψ_{inc} one has

$$\Psi_{\mathrm{inc}}=(A_p\cos\Phi_a,A_s,-n_aA_s\cos\Phi_a,n_aA_p)^T, \tag{38}$$

where the elements of Ψ_{ref} are obtained quite similarly,

$$\Psi_{\mathrm{ref}}=(-B_p\cos\Phi_a,B_s,n_aB_s\cos\Phi_a,n_aB_p)^T. \tag{39}$$

The inverse of the incident matrix $\mathbf{L}_a$ is required to obtain Ψ_a–that is, the left side of Eq. (1)–and to calculate the general transfer matrix $\mathbf{T}$. Thus the explicit expression of $\mathbf{L}_a^{-1}$ can be found comparing both sides of Eq. (36) and solving the associated algebraic equation system as follows:

$$\mathbf{L}_a^{-1}=\frac{1}{2}\begin{pmatrix} 0 & 1 & -1/n_a\cos\Phi_a & 0\\ 0 & 1 & 1/n_a\cos\Phi_a & 0\\ 1/\cos\Phi_a & 0 & 0 & 1/n_a\\ -1/\cos\Phi_a & 0 & 0 & 1/n_a \end{pmatrix}. \tag{40}$$

Note that the incident matrix $\mathbf{L}_a$ depends only on the angle of incidence Φ_a and the complex index of refraction n_a of the ambient material.

2. Exit matrix

If the substrate material is isotropic, the exit matrix that describes the projection from the in-plane components at the last interface into the exit medium can be found in the same manner as described in Sec. IV C 1. Let Ψ_f be the vector associated with the p and s modes of the transmitted waves inside the substrate, C_p and C_s, respectively. Apply the matrix $\mathbf{L}_f$ to the vector Ψ_f. Thus the vector Ψ_{trans} whose components contain the tangential field amplitudes at $z=z_N$ can be described as

$$\Psi_{\mathrm{trans}}(z=z_N)=\mathbf{L}_f\Psi_f=\mathbf{L}_f(C_s,0,C_p,0)^T. \tag{41}$$

On the other hand, the projection of the p and s modes onto the x-y plane yields, for Ψ_{trans},

$$\Psi_{\mathrm{trans}}=(C_p\cos\Phi_f,C_s,-n_fC_s\cos\Phi_f,n_fC_p)^T, \tag{42}$$

where n_f is the complex refractive index of the substrate material. It follows that the 4×4 exit matrix $\mathbf{L}_f$ in the case of an isotropic exit medium is given by

$$L_{f21}=1,\quad L_{f31}=-n_f\cos\Phi_f,$$
$$L_{f13}=\cos\Phi_f,\quad L_{f43}=n_f,\quad \text{else}\ L_{fij}=0. \tag{43}$$

The angle Φ_f is then obtained applying Snell's[3] law:

$$\cos\Phi_f=\sqrt{1-[(n_a/n_f)\sin\Phi_a]^2}. \tag{44}$$

In the case of an anisotropic exit medium both transmitted plane waves are no longer necessarily decoupled. In order to project the in-plane components at $z=z_N$ again through to the substrate, it is sufficient to assume that the transmitted field vector Ψ_f consists only of a linear combination of the eigenvectors Ξ_i of the wave transfer matrix Δ:

$$\Psi_f=\sum_{i=1}^{4}c_i\Xi_i(q_i). \tag{45}$$

There exist only transmitted components. Therefore, the eigenvectors that belong to back-traveling waves are not permitted. Hence the two eigenvectors $\Xi_i(q_i)$ with $\mathrm{Re}\{q_i\}>0$ must be separated, and may be labeled as Ξ_1 and Ξ_3. If they are unit vectors the coefficients c_1 and c_3 are equal to the amplitudes of the p and s modes C_p and C_s, respectively, which must be determined experimentally. Therefore the exit matrix for anisotropic substrate materials $\mathbf{L}_f$ can be written as follows:

$$L_{fjk}=\Xi_{jk},\quad L_{fj(k+1)}=0,\quad j=1\cdots4,\quad k=1,3. \tag{46}$$

Note that the incident and exit matrices for isotropic incident and exit media as discussed above can also be derived directly from the wave transfer matrix Δ using their eigenvectors and the assumption from Eq. (45) without geometrical considerations.

V. CONCLUSIONS

A systematic procedure has been presented for obtaining analytic expressions for the transmission and reflection coefficients of monochromatic plane waves traveling at an oblique angle of incidence to arbitrarily anisotropic layered materials systems. 4×4 matrices introduced by Berreman are used to describe propagation through plane-parallel anisotropic or isotropic slabs. Analytic expressions for the eigenvalues of plane waves in homogeneous biaxial media are reported explicitly. In addition, a particular solution for the partial transfer matrix for homogeneous twisted biaxial materials at normal incidence is derived. Incident and exit matrices for the ambient and substrate side are introduced. Therefore, a complete matrix algebra to calculate the optical parameters of layered systems is now available. Consequently, all polarization-dependent parameters can be calculated directly from the product of all matrices. The parameters determined by generalized ellipsometry are of particular interest.

The 4×4 matrix algebra presented here is a general approach applicable to all homogeneous media with a linear dielectric response and for monochromatic plane waves. The construction set to calculate the general transfer matrix permits a systematic treatment of various special configurations and a gradual derivation of the optical behavior of several parts of a given sample. Hence the algebra is very useful for computational applications. Furthermore, in principle the magnetic and gyrotropic properties of all media can be handled in the same way, including their gyrotropic and magnetic tensors.[4] Finally, the algorithm is still valid for conceivably singular situations such as normal or glancing angles of incidence, vanishing anisotropy, and transparent layers.

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APPENDIX A: PARTIAL TRANSFER MATRIX FOR ISOTROPIC SLABS

The set of the eigenvalues of the wave transfer matrix Δ for isotropic materials is given by

$$q_1=q_2=-q_3=-q_4\equiv q=\sqrt{\varepsilon-k_x^2}=\sqrt{n^2-n_a^2\sin^2\Phi_a}. \tag{A1}$$

The solution of Eq. (16) fails. However, the expansion of the exponential series in Eq. (14) and the separation of the associated sums for the cos and sin functions result directly in

$$T_p=E\left\{1+\frac{(ik_0dq)^2}{2!}+\frac{(ik_0dq)^4}{4!}+\cdots\right\}+i\frac{\Delta}{q}\left\{\frac{(k_0dq)}{1!}-\frac{(k_0dq)^3}{3!}+\cdots\right\}, \tag{A2}$$

$$T_p=E\,\cos(k_0dq)+i\frac{\Delta}{q}\sin(k_0dq), \tag{A3}$$

$$T_p=\begin{pmatrix} \cos k_0dq & 0 & 0 & i\dfrac{q}{\varepsilon}\sin k_0dq \\ 0 & \cos k_0dq & -\dfrac{i}{q}\sin k_0dq & 0 \\ 0 & -iq\sin k_0dq & \cos k_0dq & 0 \\ i\dfrac{\varepsilon}{q}\sin k_0dq & 0 & 0 & \cos k_0dq \end{pmatrix}. \tag{A4}$$

The identities: $\Delta^2=q^2\mathbf{E}$ and $\Delta^3=q^2\Delta$ are used. If the differences between the main values of the dielectric tensor are small the solvability of Eq. (16) can be used to decide either to calculate the matrix for an anisotropic or an isotropic material.

APPENDIX B: $\mathbf{L}_f$ ($\varepsilon_{ij}=0\forall i\neq j$, $\varepsilon_{11}\neq\varepsilon_{22}\neq\varepsilon_{33}$)

The aim here is to demonstrate how the matrix algebra discussed above can be applied to a straightforward derivation of any analytic expression for a given sample. For simplicity, consider a biaxial film-substrate system with film thickness d, and an isotropic ambient where both crystal systems are aligned parallel to the Cartesian laboratory system axes. All possible combinations of biaxial, uniaxial, or isotropic film-substrate configurations as collected in Ref. 3 can be derived from this example by changing the meaning of the refractive indices in the apparent formulas.

The exit matrix $\mathbf{L}_f$ is found using Eq. (46). The dielectric function tensor ε is diagonal and contains three different optical constants for the respective direction of light propagation n_x^2, n_y^2, and n_z^2, respectively. The nontrivial elements of Δ are found as follows:

$$\Delta_{14}=\cos^2\Phi_z,\quad \Delta_{23}=-1,$$

$$\Delta_{32}=-n_y^2\cos^2\Phi_y,\quad \Delta_{41}=n_x^2. \tag{B1}$$

The eigenvalues follow from Eq. (21):

$$q_{1/2}=\pm N_{xz}\,,\quad q_{3/4}=\pm N_{yy}\,,$$

$$N_{ij}\equiv n_i\cos\Phi_j=n_i\sqrt{1-[(n_a/n_j)\sin\Phi_a]^2}. \tag{B2}$$

The eigenvectors that refer only to transmitted waves ($\mathrm{Re}\{q_1,q_3\}>0$) are then determined up to a constant factor as

$$\Xi_1(q_1)=(0,1,-n_y\cos\Phi_y\,,0)^T,$$

$$\Xi_3(q_3)=(\cos\Phi_z,0,0,n_x)^T, \tag{B3}$$

and $\mathbf{L}_f$ of the biaxial exit medium can be obtained most easily by inserting the eigenvectors into Eq. (46) as the first and third columns:

$$L_{f13}=\cos\Phi_z\,,\quad L_{f21}=1,$$

$$L_{f31}=-n_y\cos\Phi_y\,,\quad L_{f43}=n_x\,. \tag{B4}$$

APPENDIX C: $\mathbf{T}_p$ ($\varepsilon_{ij}=0\,\forall i\neq j,\ \varepsilon_{11}\neq\varepsilon_{22}\neq\varepsilon_{33}$)

With the solutions of the eigenequation [Eq. (A6)] the coefficients β_i are found explicitly to be

$$\beta_0^{bi}=\{N_{xz}^2\cos(k_0dN_{yy})-N_{yy}^2\cos(k_0dN_{xz})\}/D,$$

$$\beta_1^{bi}=i\{(N_{xz}^2/N_{yy})\sin(k_0dN_{yy})-(N_{yy}^2/N_{xz})\sin(k_0dN_{xz})\}/D,$$

$$\beta_2^{bi}=\{\cos(k_0dN_{xz})-\cos(k_0dN_{yy})\}/D, \tag{C1}$$

$$\beta_3^{bi}=i\{\sin(k_0dN_{xz})/N_{xz}-\sin(k_0dN_{yy})/N_{yy}\}/D,$$

$$D=N_{xz}^2-N_{yy}^2\,.$$

The nature of the partial transfer matrix is the same as for an isotropic layer, as seen expanding the power series in Δ with the coefficients β_i given above. Defining phase thicknesses for the p and s polarizations as κ_p and κ_s, respectively,

$$\kappa_p^{bi}\equiv k_0dN_{xz}\,,\quad \kappa_s^{bi}\equiv k_0dN_{yy}\,, \tag{C2}$$

$\mathbf{T}_p$ for the biaxial film is obtained as

$$T_p^{bi}=\begin{pmatrix} \cos\kappa_p & 0 & 0 & i(N_{xz}/n_x^2)\sin\kappa_p \\ 0 & \cos\kappa_s & -i(1/N_{yy})\sin\kappa_s & 0 \\ 0 & -iN_{yy}\sin\kappa_s & \cos\kappa_s & 0 \\ i(n_x^2/N_{xz})\sin\kappa_p & 0 & 0 & \cos\kappa_p \end{pmatrix}. \tag{C3}$$

Let $n_{\bar{x}}$, $n_{\bar{y}}$, and $n_{\bar{z}}$ denote the complex refractive indices for the substrate material, and n_x, n_y, and n_z for the biaxial slab, respectively. The ellipsometric ratio ρ, for example, is given by Eq. (10) using the elements of the general transfer matrix. After multiplying $\mathbf{T}_p$ according to Eq. (2) from the left side with $\mathbf{L}_a^{-1}$, and from the right side with $\mathbf{L}_f$ from the biaxial substrate [Eq. (B4)], $\mathbf{T}$ results in

$$T_{11}=(T_{p22}^{bi}-T_{p23}^{bi}N_{\overline{yy}})-(T_{p32}^{bi}-T_{p33}^{bi}N_{\overline{yy}})/N_{aa}\,,$$

$$T_{21}=(T_{p22}^{bi}-T_{p23}^{bi}N_{\overline{yy}})+(T_{p32}^{bi}-T_{p33}^{bi}N_{\overline{yy}})/N_{aa}\,,$$

$$\begin{aligned}T_{33}&=(T_{p41}^{bi}\cos\Phi_{\bar{z}}+T_{p44}^{bi}n_{\bar{x}})/n_a\\&\quad-(T_{p11}^{bi}\cos\Phi_{\bar{z}}+T_{p14}^{bi}n_{\bar{x}})/\cos\Phi_a\,,\end{aligned} \tag{C4}$$

$$\begin{aligned}T_{34}&=(T_{p41}^{bi}\cos\Phi_{\bar{z}}+T_{p44}^{bi}n_{\bar{x}})/n_a\\&\quad+(T_{p11}^{bi}\cos\Phi_{\bar{z}}+T_{p14}^{bi}n_{\bar{x}})/\cos\Phi_a\,,\end{aligned}$$

$$T_{ij}=0\quad \text{else.}$$

With the partial reflection coefficients for the p and s polarization at the 0-1 (ambient-film) and 1-2 (film-substrate) interfaces r_{01pp}, r_{01ss}, r_{12pp}, and r_{12ss}, respectively,

$$r_{01pp}^{bi-bi}=\frac{N_{xa}-N_{az}}{N_{xa}+N_{az}},\quad r_{01ss}^{bi-bi}=\frac{N_{aa}-N_{yy}}{N_{aa}+N_{yy}},$$

$$r_{12pp}^{bi-bi}=\frac{N_{\bar{x}z}-N_{x\bar{z}}}{N_{\bar{x}z}-N_{x\bar{z}}},\quad r_{12ss}^{bi-bi}=\frac{N_{yy}-N_{\overline{yy}}}{N_{yy}-N_{\overline{yy}}}, \tag{C5}$$

the ellipsometric ratio can be written as follows:

$$\rho=\left(\frac{r_{01pp}+r_{12pp}e^{+i2\kappa_p}}{1+r_{01pp}r_{12pp}e^{+i2\kappa_p}}\right)\left(\frac{1+r_{01ss}r_{12ss}e^{+i2\kappa_s}}{r_{01ss}+r_{12ss}e^{+i2\kappa_s}}\right)^{-1}. \tag{C6}$$

Because of the symmetry the off-diagonal reflection coefficients vanish, as seen again from the general transfer matrix $\mathbf{T}$. A detailed treatment of Δ reveals that the associated elements of $\mathbf{T}$ appear only if at least one of the crystal axes is nonparallel to any of the laboratory coordinate axes or if the optical axes of uniaxial media do not lie in the y-z plane. As long as Δ has the diagonal form of Eq. (B1), all partial transfer matrices have the same vanishing elements as $\mathbf{T}_p$ from an isotropic slab. The patterns of vanishing elements of the partial transition matrices, incident and exit matrices reveal that after multiplication $\mathbf{T}$ still has only four nonvanishing elements.

Note that in spite of the notation recommended by the 1968 International Conference on Ellipsometry at the University of Nebraska,[20] the time dependence of the harmonic fields used here is given by $\exp\{-i\omega t\}$. This results in positive imaginary parts of the dielectric functions of all treated materials and changes the sign in the phase thicknesses, here defined as κ_p and κ_s, in comparison with the formulas given in Ref. 3.

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[11] All conceivable combinations of isotropic and anisotropic materials can be considered even for the case of very small anisotropy. Sandwich the layers between the incident and exit media. Note that the coherent length of the light beam must be greater than a characteristic sample length such as the sum of all layer thicknesses. This may be very important if a thick slab is considered for optical transmission investigations. Here the meanings of the terms ''substrate'' and ''exit medium'' are the same.

[12] Note that for transmission measurements one must also include the incident medium as the exit medium. The exit medium should be the first material in the light path that does not contain back-traveling waves. Perhaps this is the material that supports the layer stack, or else, and in case of transmission arrangement, it is the medium between the sample and the detector.

[13] According to the coordinate system in Berreman's original publication (see Ref. 4).

[14] With this restriction one is committed to a specific set of the Euler angles that describe the orientations of the principal crystal axes.

[15] See for example, *Ellipsometry and Polarized Light* (Ref. 3).

[16] Note the sign change of H_x in Eq. (23) of Ref. 4.

[17] See, for example, H. Goldstein, *Classical Mechanics* (Addison-Wesley, Reading, MA, 1965). Note that the matrix **A** given there is just the inverse of the matrix **A** mentioned here. The Euler angles describe the rotation from the laboratory Cartesian coordinate system into the crystal main axes system. For the classification of anisotropic media (crystals) and their dielectric tensors and symmetries, see, for example, P. Yeh, *Optical Waves in Layered Media*, Wiley Series in Pure and Applied Optics (Wiley, New York, 1988).

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