Reflection Characterization of Nanoscopically Stratified Surface Structures and Optical Probing of Anisotropic Nanolayers

Peep Adamson
Institute of Physics, University of Tartu, 142 Riia, Tartu, 51014, Estonia
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The reflection of linearly polarized light from a layered nanoscopic system of anisotropic dielectric films on homogeneous isotropic substrate is investigated in the long-wavelength limit. The expressions for the reflectances and ellipsometric angles of an N-layer system of anisotropic ultrathin films are derived. For absorbing substrate mathematical relationships take a relatively simple additive form. In comparison with absorbing substrate the fully transparent system is more complicated and in this case an ultrathin film in multilayer system is sensitive to the presence of other ultrathin films (an interaction between them emerges) and the contribution of each layer to the reflection coefficient is not expressed in a purely additive form. The analytical results are supported by computer-aided analysis made on the basis of general wave propagation theory for anisotropic layered media. The most useful feature of obtained approximate expressions is that they are simply invertible, allowing a direct calculation of the parameters of ultrathin layers. [DOI: 10.1380/ejssnt.2010.167]

Keywords: Semi-empirical models and model calculations; Nano-films, stacks, and other nano materials; Insulating films; Ellipsometry; Reflection spectroscopy

I. INTRODUCTION

Layered systems have grown in importance in the technology of micro- and nanoelectronic devices [1, 2]. As a consequence the diagnostics of anisotropic thin films and multilayers has been the subject of much concentrated attention. Particularly, polarization-dependent optical reflection techniques, which are fast, noninvasive, and inexpensive, are of considerable interest for investigating layered systems. For determining optical constants of ultrathin films and surface layers the classical reflection methods suffer many disadvantages. Because of that in the special case of ultrathin layers, it is best to apply the differential reflection methods, which are founded on the direct measurement of the contribution of an ultrathin layer to the reflection coefficient or ellipsometric angles. By now the corresponding differential methods are adequately elaborated for isotropic ultrathin films [3-13].

A purpose of this paper is to study the reflection characteristics analytically in a long-wavelength limit for an N-layer system of biaxially anisotropic ultrathin dielectric layers upon an isotropic and homogeneous substrate. The remarkable advantage of the analytical approach lies in the fact that the relatively simple final expressions for reflection characteristics not only give a physical insight into the reflection problem of anisotropic ultrathin layered structures but also are especially advantageous for tackling the inverse problem. This is because the creation a solution for the inversion problem of an anisotropic layered system is rather intricate process owing to the complexity of the corresponding exact reflection equations. In general, these equations cannot analytically be inverted and the solution of such problems can only be found by the use of numerical methods. However, these methods, as a rule, come across serious difficulties, the main two being the instability and the non-uniqueness of the solution. In addition, it is important to note that the analytical algorithms for calculating the parameters of interest directly from the measured data are very fast in comparison with classical, e.g., regressive type of algorithms. Due to this fact, approximate analytical techniques are also used to provide initial guesses at the values of the variable parameters, and regression techniques are then used to fine-tune the desired parameters.

II. REFLECTION CHARACTERISTICS

Assuming that all the media are nonmagnetic, we consider the reflection of s- and p-polarized time-harmonic (the complex representation is taken in the form exp(-i\omega t), where \omega = 2\pi\nu/\lambda, and \lambda is a vacuum wavelength) electromagnetic plane waves in an ambient medium with real isotropic and homogeneous dielectric constant \varepsilon_a \equiv n_a^2 from a layered system with plane parallel interfaces consisting of N anisotropic homogeneous dielectric films of thickness d_i \ll \lambda with real principal dielectric-tensor components in the crystal-coordinate system \varepsilon_{xx} = \langle n_{xx}\rangle^2, \varepsilon_{yy} = \langle n_{yy}\rangle^2, and \varepsilon_{zz} = \langle n_{zz}\rangle^2. The anisotropic films are numbered from ambient to substrate, i.e., index i = 1, ..., N so that the last film with index i = N is located upon a semi-infinite non-absorbing, isotropic, and homogeneous substrate with real dielectric constant \varepsilon_s = n_s^2. The orientations of the crystal axes are described by the Euler angles \theta_i, \varphi_i, and \psi_i with respect to a fixed \{x, y, z\} coordinate system (the Cartesian laboratory coordinate system). The laboratory x, y, and z axes are defined as follows. The reflecting surface is the xy plane, and the plane of incidence is the xz plane, with the z axis normal to the surface of the layered medium and directed into it. The incident light beam in the ambient medium makes an angle \phi_a with the z axis. The dielectric tensor for i-th anisotropic layer in the \{x, y, z\}
coordinate system is given by

$$
\begin{bmatrix}
\varepsilon_{11}^{(i)} & \varepsilon_{12}^{(i)} & \varepsilon_{13}^{(i)} \\
\varepsilon_{21}^{(i)} & \varepsilon_{22}^{(i)} & \varepsilon_{23}^{(i)} \\
\varepsilon_{31}^{(i)} & \varepsilon_{32}^{(i)} & \varepsilon_{33}^{(i)}
\end{bmatrix} = A \begin{bmatrix}
\varepsilon_{xx}^{(i)} & 0 & 0 \\
0 & \varepsilon_{yy}^{(i)} & 0 \\
0 & 0 & \varepsilon_{zz}^{(i)}
\end{bmatrix} A^{-1},
$$

(1)

where \( A \) is the orthogonal coordinate rotation matrix [14]. Therefore,

$$
\varepsilon_{11}^{(i)} = -\Gamma_1^{(i)} \sin 2\varphi_i \sin 2\psi_i \cos \theta_i + \Gamma_2^{(i)} \cos^2 \varphi_i + \Gamma_3^{(i)} \sin^2 \varphi_i \cos^2 \theta_i + \varepsilon_{11}^{(i)} \sin^2 \varphi_i \sin^2 \theta_i,
$$

(2)

$$
\varepsilon_{22}^{(i)} = \Gamma_1^{(i)} \sin 2\varphi_i \sin 2\psi_i \cos \theta_i + \Gamma_2^{(i)} \sin^2 \varphi_i + \cos^2 \varphi_i (\Gamma_3^{(i)} \cos^2 \theta_i + \varepsilon_{22}^{(i)} \sin^2 \theta_i),
$$

(3)

$$
\varepsilon_{33}^{(i)} = \Gamma_3^{(i)} \sin^2 \theta_i + \varepsilon_{33}^{(i)} \cos^2 \theta_i,
$$

(4)

$$
\varepsilon_{12}^{(i)} = \varepsilon_{21}^{(i)} = \Gamma_1^{(i)} \cos 2\varphi_i \sin 2\psi_i \cos \theta_i + \sin 2\varphi_i (\Gamma_3^{(i)} - \varepsilon_{33}^{(i)} \sec^2 \theta_i)/2,
$$

(5)

$$
\varepsilon_{13}^{(i)} = \varepsilon_{31}^{(i)} = \sin \theta_i (\Gamma_3^{(i)} \cos \varphi_i \sin 2\psi_i - \sin \varphi_i \cos \theta_i (\Gamma_3^{(i)} - \varepsilon_{33}^{(i)})),
$$

(6)

$$
\varepsilon_{23}^{(i)} = \varepsilon_{32}^{(i)} = \sin \theta_i \Gamma_3^{(i)} \sin \varphi_i \sin 2\psi_i + \cos \varphi_i \cos \theta_i (\Gamma_3^{(i)} - \varepsilon_{33}^{(i)}),
$$

(7)

where

$$
\Gamma_1^{(i)} \equiv (\varepsilon_{xx}^{(i)} - \varepsilon_{yy}^{(i)})/2,
$$

$$
\Gamma_2^{(i)} \equiv \varepsilon_{xx}^{(i)} \cos^2 \psi_i + \varepsilon_{yy}^{(i)} \sin^2 \psi_i,
$$

$$
\Gamma_3^{(i)} \equiv \varepsilon_{xx}^{(i)} \sin^2 \psi_i + \varepsilon_{yy}^{(i)} \cos^2 \psi_i.
$$

In a similar manner to the isotropic case we use the matrix method for calculating the contributions of ultrathin layers to the reflection characteristics. Since \( s \) and \( p \) modes are no longer spatially independent of each other (a so-called mode coupling appears) in the anisotropic medium, then, consequently, \( 4 \times 4 \) matrices are needed in order to establish a similar matrix method. Dealing directly with first-order Maxwell equations, Berremann [15] showed a general way to calculate the reflection characteristics of an anisotropic layered system from a wave transfer matrix of rank 4. For transparent substrates the reflectances \( r_{pp} \) and \( r_{ss} \) are equal to zero in the first order in \( d_i/\lambda \). The second-order formulas take the form:

$$
R_{pp}^{(N)} = R_p^{(0)} \left[ 1 + \frac{16\pi^2 n_a n_s \cos \phi_a \cos \phi_s}{\varepsilon_a \cos^2 \phi_a - \varepsilon_s \cos^2 \phi_s} \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} (c_{21}^{(i)} c_{12}^{(j)} - c_{21}^{(j)} c_{12}^{(i)}) (d_i d_j / \lambda^2)
\right]
\times
\sum_{i=1}^{N} \sum_{j=1}^{N} \left[ (\varepsilon_{hh}^{(i)} - \varepsilon_{ss}^{(i)} \cos^2 \phi_a) (\varepsilon_{hh}^{(j)} - \varepsilon_{ss}^{(j)} \cos^2 \phi_a)
\right] (d_i d_j / \lambda^2),
$$

(8)

$$
R_{ss}^{(N)} = R_s^{(0)} \left[ 1 + \frac{16\pi^2 n_a n_s \cos \phi_a \cos \phi_s}{\varepsilon_a - \varepsilon_s} \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} (c_{43}^{(i)} - c_{43}^{(j)}) (d_i d_j / \lambda^2)
\right]
\times
\sum_{i=1}^{N} \sum_{j=1}^{N} \left[ (\varepsilon_{hh}^{(i)} - \varepsilon_{ss}^{(i)} \cos^2 \phi_a) (\varepsilon_{hh}^{(j)} - \varepsilon_{ss}^{(j)} \cos^2 \phi_a)
\right] (d_i d_j / \lambda^2),
$$

(9)

$$
R_p^{(N)} = \frac{16\pi^2 \varepsilon_a \cos^2 \phi_a}{(n_a \cos \phi_a + n_s \cos \phi_s) (n_a \cos \phi_a + n_s \cos \phi_s)} \times
\sum_{i=1}^{N} \left[ (c_{23}^{(i)} \cos \phi_a + P_s c_{13}^{(i)} \sigma) (d_i / \lambda)^2
\right],
$$

(10)

where

$$
\begin{align}
\varepsilon_{23}^{(i)} &= 1 - \varepsilon_a \sin^2 \phi_a / \varepsilon_{33}^{(i)}, \\
\varepsilon_{13}^{(i)} &= 1 - \varepsilon_a \sin^2 \phi_a / \varepsilon_{33}^{(i)}, \\
\varepsilon_{12}^{(i)} &= 1 - \varepsilon_a \sin^2 \phi_a / \varepsilon_{33}^{(i)}, \\
\varepsilon_{21}^{(i)} &= 1 - \varepsilon_a \sin^2 \phi_a / \varepsilon_{33}^{(i)}, \\
\varepsilon_{31}^{(i)} &= 1 - \varepsilon_a \sin^2 \phi_a / \varepsilon_{33}^{(i)}, \\
\varepsilon_{32}^{(i)} &= 1 - \varepsilon_a \sin^2 \phi_a / \varepsilon_{33}^{(i)}, \\
\varepsilon_{12}^{(i)} &= 1 - \varepsilon_a \sin^2 \phi_a / \varepsilon_{33}^{(i)}, \\
\varepsilon_{21}^{(i)} &= 1 - \varepsilon_a \sin^2 \phi_a / \varepsilon_{33}^{(i)},
\end{align}
$$

(11)

and

$$
\begin{align}
P_s &= +1, \\
\sigma &= sp \text{ or } ps, \text{ and } \Gamma_{pp} = -1 \text{ (in this paper the first subscript indicates the incident light). Since in anisotropic samples the Jones matrix [16] contains off-diagonal terms, a so-called generalized ellipsometry is designed for anisotropic systems [17]. Use of multiple input polarization states enables determination of the off-diagonal elements and thus to collect information regarding the anisotropy from all Jones matrix elements. As in the case with conventional ellipsometry, the absolute values of the Jones matrix elements are not determined. There are three normal-}
\end{align}
$$
ized complex ratios, which may be chosen for measurement [18, 19]. For transparent substrates ellipsometric quantities take the form:

\[
\delta \Delta^{(N)}_{pp} = 4\pi n_a \cos \phi_a (\varepsilon_a - \varepsilon_s)^{-1} \sum_{i=1}^{N} \left[ (\epsilon_{a i}^{(i)} \cos^2 \phi_a - \epsilon_{c 12}^{(i)} \epsilon_{s i}^{(i)})(\cos^2 \phi_a - \varepsilon_a \varepsilon_s - \sin^2 \phi_a)^{-1}
- \epsilon_{c 41}^{(i)} + \varepsilon_s \cos^2 \phi_a \right] (d_i/\lambda),
\]

if \( \phi_a \neq \phi_B = \arctan(n_s/n_a) \) (for \( \phi_a = \phi_B \), we have \( \delta \Delta^{(r)}_{pp} = \pi/2 \)), and

\[
\delta \Psi^{(N)}_{pp} = \pi (\varepsilon_a + \varepsilon_s)^{1/2} \sum_{i=1}^{N} \left[ (\epsilon_{a i}^{(i)} \cos \phi_a - \epsilon_{c 12}^{(i)} \epsilon_{s i}^{(i)})(\varepsilon_a - \varepsilon_s)^{-1} \right] (d_i/\lambda),
\]

if \( \phi_a = \phi_B \) (for \( \phi_a \neq \phi_B \) the quantity \( \delta \Psi^{(r)}_{pp} \sim (d_i/\lambda)^2 \)). The ellipsometric angles \( \Delta^{(r)} \approx \pi/2 \) in the first order in \( d_i/\lambda \) and the quantities \( \tan \Psi^{(r)}_a \) can be expressed in the form:

\[
\tan \Psi^{(N)}_a = 4\pi n_a \cos \phi_a (n_a \cos \phi_a + n_s \cos \phi_s)(n_a \cos \phi_a + n_s \cos \phi_a)^{-1}
\times \sum_{i=1}^{N} (\epsilon_{a i}^{(i)} \cos \phi_a + P_s n_s \epsilon_{s i}^{(i)})(\varepsilon_a \cos^2 \phi_a - \varepsilon_s \cos^2 \phi_a)^{-1} (d_i/\lambda).
\]

Approximate analytical results obtained above are correlated with the exact computer solution of the reflection problem for a multilayer system of anisotropic homogeneous films. The main question is, what is the accuracy of approximate formulas? As illustrated (Fig. 1), the governing factor, which determines the accuracy of approximate formulas, is the small parameter \( d_i/\lambda \). Broadly speaking, the error of approximate equations does not exceed a few percent if \( \sum_{i=1}^{N} d_i/\lambda \leq 10^{-2} \). However, the accuracy of approximate equations for given values of \( d_i/\lambda \) depends on the values of material dielectric constants as well. Because of this, it is difficult to indicate explicitly the value of \( \sum_{i=1}^{N} d_i/\lambda \) where the long-wavelength approximation is broken down. Notice that for ultrathin films with nanometric thickness, the developed approximation theory is highly accurate in the far-infrared part of the electromagnetic spectrum \((d_i/\lambda \leq 10^{-3})\). The dependence of the accuracy of the approximate formulas for ellipsometric quantities on the angle of incidence \( \phi_a \) is shown in Fig. 2. One can see that the approximate formulas are inapplicable in the neighborhood of point where these quantities as a function of \( \phi_a \) changes its sign passing through the zero value because it is insufficient to restrict oneself to terms of the first order in the expansion in \( d_i/\lambda \) in the vicinity of this point.

### III. REFLECTION DIAGNOSTICS

Let us consider potential applicability of approximate expressions obtained above for the optical diagnostics of an ultrathin biaxially anisotropic film. In this work we shall restrict our consideration to the case where the thickness \( d \) of an anisotropic ultrathin film is known. In the case of a transparent substrate, for instance, on the basis of the measurements of \( R_{ps}, R_{sp}, \) and \( \Delta R_{pp}/R_{pp}^{(0)} \equiv (R_{pp} - R_{p}^{(0)})/R_{pp}^{(0)} \) at the two incident angles \( \phi_a = 0^\circ \) and \( \phi_a^{(i)} \neq 0^\circ \) we can determine the quantity \( \varepsilon_{11} - \varepsilon_{22}/\varepsilon_{33} \equiv x \) from the equation

\[
x = (\varepsilon_a + \varepsilon_s)/2 \pm ((\varepsilon_a + \varepsilon_s)^2/4 + P_0 - \varepsilon_a \varepsilon_s)^{1/2},
\]

where

\[
P_0 = \left[ \frac{\Delta R_{pp}(0^\circ)(\varepsilon_a - \varepsilon_s)^2}{R_{pp}^{(0)}(0^\circ)} \right] \frac{n_a - n_s}{n_s} \left( \frac{\lambda}{d} \right)^2,
\]

\[
S_0 = \frac{(n_a + n_s)^4}{16\pi^2 \varepsilon_a} [R_{ps}(0^\circ)R_{sp}(0^\circ)],
\]

and the quantity \( \varepsilon_{33}^{-1} \equiv y \) from the equation

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where

\[ a_{11} = \cos^2 \phi_0^1 \cos^2 \phi_0^1, \quad a_{12} = \varepsilon_0^2 \varepsilon_s \sin^4 \phi_0^1, \quad a_{13} = \varepsilon_0 \sin^2 \phi_0^1 (\varepsilon_0 \cos^2 \phi_0^1 + \varepsilon_s \cos^2 \phi_0^1), \]

\[ a_{14} = -\varepsilon_0 \sin^2 \phi_0^1 + \varepsilon_s \cos^2 \phi_0^1, \quad a_{15} = -2 \varepsilon_0^2 \varepsilon_s \sin^2 \phi_0^1, \quad a_{16} = \varepsilon_0 \varepsilon_s - P, \]

\[ P_i = \left( \frac{\varepsilon_0^2 \cos^2 \phi_0^1 - \varepsilon_0 \cos^2 \phi_0^1}{16 \pi^2 n_a n_s \cos \phi_0^1 \cos \phi_0^1} \right) \Delta R_{pp} (\phi_0^1) - \frac{S_i (\varepsilon_0 \cos^2 \phi_0^1 - \varepsilon_0 \cos^2 \phi_0^1)}{(n_a n_s \cos \phi_0^1 \cos \phi_0^1 + \varepsilon_s \cos^2 \phi_0^1)} \right] \left( \frac{\lambda}{d} \right)^2.

If both incident angles are not equal to zero, i.e., \( \phi_0^1 \neq 0 \) and \( \phi_0^2 \neq 0 \), then we can determine \( x \) and \( y \) from the following system of two equations:

\[ a_{11} x^2 + a_{12} y^2 + a_{13} x y + a_{14} x + a_{15} y + a_{16} = 0, \]

where \( i = 1, 2 \). This system can be solved with a computer. On the other hand, rather than solve the nonlinear system of two equations, the problem can be reduced to a quartic equation for one unknown. This approach has an advantage over the first method because for solving the quartic equations foolproof methods exist. For unknown \( y \), for instance, one can obtain the following equation:

\[ A y^4 + B y^3 + C y^2 + D y + F = 0, \]

where

\[ A = a_{11} f_1^2 + a_{12} f_2^2 - a_{13} f_1 f_2, \]

\[ B = 2(a_{11} f_1 f_2 + a_{12} f_1 f_3) - a_{13} f_2 f_4 + f_1 f_5 \]

\[ -a_{14} f_1 f_4 + a_{15} f_1^2, \]

\[ C = a_{11} (f_2^2 + 2 f_3 f_4) + a_{12} f_2^2 - a_{13} (f_3 f_4 + f_2^2) \]

\[ -a_{14} (f_2 f_4 + f_1 f_3 + 2 a_{15} f_4 f_5 + a_{16} f_4^2), \]

\[ D = 2 a_{15} f_2 f_3 - a_{13} f_3 f_5 - a_{14} (f_4 f_2 + f_1 f_5) \]

\[ + a_{15} f_2^2 + 2 a_{16} f_4 f_5, \]

\[ F = a_{11} f_2^2 - a_{14} f_2 f_3 + a_{16} f_3^2, \]

\[ f_1 = a_{11} a_{21} - a_{11} a_{22}, \quad f_2 = a_{15} a_{21} - a_{11} a_{25}, \quad f_3 = a_{15} a_{26}, \quad f_4 = a_{15} a_{24} - a_{11} a_{24}, \quad f_5 = a_{14} a_{21} - a_{11} a_{21}. \] If \( y \) is known, then for the second unknown \( x \) can be obtained a simple quadratic equation as seen from Eq. (16).

Next we consider the determination of the quantities \( \varepsilon_{22} - \varepsilon_{23}^2 / \varepsilon_{33} \), \( \varepsilon_{12} - \varepsilon_{13}^2 / \varepsilon_{33} \), and \( \varepsilon_{22} - \varepsilon_{23}^2 / \varepsilon_{33} \). More simple way for determining \( \varepsilon_{22} - \varepsilon_{23}^2 / \varepsilon_{33} \) is using ellipsometric quantity \( \delta \Delta_{pp} \). From Eq. (11) we obtain that

\[ \varepsilon_{22} - \varepsilon_{23}^2 / \varepsilon_{33} = \varepsilon_0 \sin^2 \phi_0 + \varepsilon_s \cos^2 \phi_0 \]

\[ + \frac{(\varepsilon_1 - \varepsilon_3) \sin^2 \phi_0 - (1 - \varepsilon_0 \sin^2 \phi_0 \varepsilon_{33}) \varepsilon_s \cos^2 \phi_0 - \varepsilon_0 \varepsilon_s \sin^2 \phi_0}{4 \pi n_a \cos \phi_0 / d}. \]

The quantities \( \varepsilon_{12} - \varepsilon_{13} \sin \varepsilon_{23} / \varepsilon_{33} \) and \( \varepsilon_{23} / \varepsilon_{33} \) can be determined from the measurements of \( R_0 \) or general ellipsometric parameters in the reflection or transmission mode. For example, on the basis of Eqs. (10) and (13) one can obtain that

\[ \varepsilon_{12} - \varepsilon_{13} \sin \varepsilon_{23} / \varepsilon_{33} = \frac{K_{ps} + K_{sp}}{2 \cos \phi_0}, \]

\[ \varepsilon_{23} / \varepsilon_{33} = \frac{K_{sp} - K_{ps}}{2 n_a n_s \sin \phi_0}, \]

where

\[ K_{ps} = \frac{\sqrt{\varepsilon_0 n_a \cos \phi_0 + n_s \cos \phi_0} (\varepsilon_0 n_a \cos \phi_0 + n_s \cos \phi_0)}{4 \pi n_a \cos \phi_0}, \]

or

\[ K_{sp} = \pm \tan \psi \]
The solution of this system has the form:

\[
\begin{align*}
\epsilon_{xx} &= \epsilon_{11}, \\
\epsilon_{yy} &= \epsilon_{22} - \epsilon_{33} - \epsilon_{zz}, \\
\epsilon_{zz} &= \frac{[(\epsilon_{22} + \epsilon_{33}) \pm (\epsilon_{22} - \epsilon_{33})^2 + 4\epsilon_{13}^2]^{1/2}}{2}, \quad (25) \\
\theta &= \arcsin[\epsilon_{22} - \epsilon_{yy}] / (\epsilon_{zz} - \epsilon_{yy})^{1/2}. \quad (26)
\end{align*}
\]

Analogously it can be shown that if \( \theta = \psi = 0 \) then

\[
\begin{align*}
\epsilon_{xx} &= \epsilon_{11} + \epsilon_{22} \pm \frac{[(\epsilon_{22} - \epsilon_{33})^2 + 4\epsilon_{13}^2]^{1/2}}{2}, \quad (27) \\
\epsilon_{yy} &= \epsilon_{11} + \epsilon_{22} - \epsilon_{xx}, \quad (28) \\
\epsilon_{zz} &= \epsilon_{33}, \quad (29) \\
\varphi &= \arcsin[(\epsilon_{11} - \epsilon_{xx}) / (\epsilon_{yy} - \epsilon_{xx})]^{1/2}. \quad (30)
\end{align*}
\]

Next, if only one angle is known, for instance, the angle \( \psi \) (for simplicity we suppose that \( \psi = 0 \)), then one...
The solution of this system has the form:

\[ \varepsilon_{zz} = \varepsilon_{33}, \]
\[ \varepsilon_{xx} + \varepsilon_{yy} = \varepsilon_{11} + \varepsilon_{22}, \]
\[ \varepsilon_{xx} \sin^2(\varphi + \psi) + \varepsilon_{yy} \cos^2(\varphi + \psi) = \varepsilon_{22}, \]
\[ (\varepsilon_{xx} - \varepsilon_{yy}) \sin(2(\varphi + \psi)) = 2\varepsilon_{12}. \]

Solving the system (37) gives:

\[ \varepsilon_{xx} = \frac{\{\varepsilon_{11} + \varepsilon_{22} \pm \sqrt{[\varepsilon_{11} - \varepsilon_{22}]^2 + 4\varepsilon_{12}^2}\}^{1/2}}{2}, \]
\[ \varepsilon_{yy} = \frac{\varepsilon_{11} + \varepsilon_{22} - \varepsilon_{xx}}, \]
\[ \varphi + \psi = \arcsin[\frac{\varepsilon_{22} - \varepsilon_{yy}}{\varepsilon_{xx} - \varepsilon_{yy}}]^{1/2}. \]

As seen from Eq. (40) in this instance (\(\theta = 0\)) we can determine only the sum of two angles \(\varphi\) and \(\psi\).

As a practical matter, it is also interesting to clear up the impact of an error in thickness upon the accuracy of approximate relations. Note that in the preceding calculations (Fig. 3) the thickness of an ultrathin film is supposed to be exactly known in advance. A computer simulation for the possible errors of approximate relations, involving an error of thickness as well, shows that an error in film thickness introduces practically the same uncertainty for desired dielectric tensor as errors of reflection characteristics. In other words, an instrumental error on determining the film thickness is not bound to be smaller than an instrumental error of reflection characteristics.

**IV. CONCLUSIONS**

The analytical approach developed in this paper not only provides insight into the nature of reflection problem for anisotropic ultrathin films on isotropic substrates but also furnishes the methods for determining the optical constants of such films. The essential property of the obtained approximate formulas is that they are relatively easily invertible allowing direct calculations of optical constants of uniform ultrathin anisotropic film upon isotropic substrate on the basis of differential measurements. For instance, the successful method for this purpose is the combination of ellipsometry with differential reflectance. Concurrently performed numerical calculations show that the accuracy of the long-wavelength approximation is reasonable if \(d/\lambda \leq 10^{-2}\). Therefore, in the infrared region of wavelengths the suggested methods can also be used for optical diagnostics of anisotropic films with layer thicknesses close to visual wavelengths.

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