Ellipsometry of a thin film between similar media

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The conventional formula for the ellipsometric ratio \( \rho = r_p/r_s \) diverges in the limit when the dielectric constants on either side of an inhomogeneous layer become equal, \( \epsilon_1 = \epsilon_2 \). The general case, including \( \epsilon_1 = \epsilon_2 \), necessitates going to second order in the layer thickness. A formula is derived that includes the \( \epsilon_1 = \epsilon_2 \) case without divergence; the predicted maximum in the imaginary part of \( \rho \) when \( \epsilon_1 = \epsilon_2 \) indicates that index matching (of the bounding media) can significantly increase the ellipsometric signal.

1. INTRODUCTION

Recent work of Beaglehole\(^1\) has brought into focus a long-standing problem in the ellipsometry of thin films. This problem is that the first-order (in the film thickness) correction to the Fresnel formulas gives a divergent result for the ellipsometric ratio \( \rho = r_p/r_s \) when the bounding media have equal dielectric constants. The equality of the dielectric constants \( \epsilon_1, \epsilon_2 \) of the bounding media was shown some years ago\(^2\) to give a finite \( \rho \), and, in fact, a zero \( \rho \) (\( \rho \) is defined as the value of the imaginary part of \( \rho \) at the principal angle, where the real part of \( \rho \) is zero). What has emerged from the calculations of Beaglehole of \( \rho \) for a uniform film is that as \( \epsilon_1 \rightarrow \epsilon_2 \) the magnitude of \( \rho \) first increases before going to zero at \( \epsilon_1 = \epsilon_2 \), reaching a maximum for \( \epsilon_2 \) close to \( \epsilon_1 \). Because a maximum in the magnitude of \( \rho \) is of practical importance in polarization modulation ellipsometry,\(^3,4\) we have developed a theory for the general case (encompassing all of \( \epsilon_1 \neq \epsilon_2, \epsilon_1 = \epsilon_2 \), and \( \epsilon_1 = \epsilon_2 \)). This is given in Section 3. Before that, the conventional first-order theory is reviewed in Section 2.

2. THE FIRST-ORDER EXPRESSION FOR \( \rho, \epsilon_1 \neq \epsilon_2 \)

Consider an inhomogeneous layer of thickness \( \Delta z \), of dielectric function \( \epsilon(z) \), bounded by media of dielectric constants \( \epsilon_1 \) and \( \epsilon_2 \). Light, of angular frequency \( \omega \) and speed \( \epsilon \) (in vacuum), is incident from medium 1. In the absence of the interfacial layer, the \( s \) and \( p \) polarization reflection amplitudes would be

\[
\begin{align*}
    r_{s0} &= \frac{q_1 - q_2}{q_1 + q_2}, \\
    r_{p0} &= \frac{Q_2 - Q_1}{Q_2 + Q_1},
\end{align*}
\]

where \( q_1 \) and \( q_2 \) are the normal components of the wave vectors in media 1 and 2 and \( Q_1 = q_1/\epsilon_1, Q_2 = q_2/\epsilon_2 \). The \( q \)'s are given by

\[
q_1^2 = \epsilon_1 \frac{\omega^2}{c^2} - K^2, \quad q_2^2 = \epsilon_2 \frac{\omega^2}{c^2} - K^2.
\]

Here \( K \) is the (invariant) component of the wave vectors along the interface,

\[
(cK/\omega)^2 = \epsilon_1 \sin^2 \theta_1 = \epsilon_2 \sin^2 \theta_2,
\]

where \( \theta_1 \) and \( \theta_2 \) are the angles of incidence and transmission.

The presence of the layer modifies the Fresnel reflection amplitudes [Eq. (1)]. The modification can be expressed as a power series in the layer thickness,

\[
\begin{align*}
    r_p &= r_{p0} + r_{p1} + r_{p2} + \cdots, \\
    r_s &= r_{s0} + r_{s1} + r_{s2} + \cdots,
\end{align*}
\]

where subscript \( n = (0, 1, 2, \ldots) \) denotes terms that are \( n \)th order in \( \Delta z/\epsilon \).

Now

\[
r_{s0} = \frac{q_1^2 - q_2^2}{(q_1 + q_2)^2} = \frac{A \omega^2/c^2}{(q_1 + q_2)^2};
\]

where \( A = \epsilon_1 - \epsilon_2 \) and, provided that \( \Delta \epsilon \neq 0, r_{s0} \neq 0 \) and

\[
\begin{align*}
    r_p &= r_{p0} + r_{p1} r_{s0} - r_{p0} r_{s1} + r_{p2} r_{s2} + \cdots, \\
    r_s &= r_{s0} + r_{s1} r_{p0} - r_{s1} r_{p1} + r_{s2} r_{p2} + \cdots.
\end{align*}
\]

Long-wave perturbation theory,\(^5,6\) which is reviewed in Chap. 3 of Ref. 7, gives the corrections \( r_{p0} \) and \( r_{s0} \) to the Fresnel reflection amplitudes. The first-order corrections are

\[
\begin{align*}
    r_{s0} &= \frac{2i q_1 \omega^2/c^2}{(q_1 + q_2)^2} \lambda_1, \\
    r_{p1} &= \frac{2i Q_1}{(Q_1 + Q_2)^2} \left( Q_2^2 \lambda_1 - K^2 \right) / \epsilon_1 \epsilon_2 \lambda_1,
\end{align*}
\]

where the integrals \( \lambda_1 \) and \( \lambda_1 \) are the first in the sets

\[
\lambda_n = \int_{-\infty}^{\infty} dz (\epsilon - \epsilon_0) z^{n-1},
\]

\[
\Lambda_n = \epsilon_1 \epsilon_2 \int_{-\infty}^{\infty} dz \left( \frac{1}{\epsilon_0} - \frac{1}{\epsilon} \right) z^{n-1}.
\]

Here \( \delta_0(z) \) is the step function representing a sharp transition between media 1 and 2, \( \delta_0(z) = \epsilon_1 \) for \( z < 0, \delta_0(z) = \epsilon_2 \) for \( z > 0 \). From Eqs. (6)–(8) we find after some manipulation that...
3. SECOND-ORDER THEORY FOR GENERAL $\Delta e$

We will calculate $r_p/r_s$, avoiding division by $r_{s0}$ or $r_{p0}$ so as to include the possibility of $\epsilon_1 = \epsilon_2$. We use the form

$$
\frac{r_p}{r_s} = \frac{r_{p0} + r_{p1} + r_{p2} + \ldots}{r_{s0} + r_{s1} + r_{s2} + \ldots}
$$

(19)

The first-order terms $r_{p1}$ and $r_{s1}$ have been given above. The second-order terms are [Ref. 2, Eqs. (15) and (29)]

$$
\begin{align*}
\frac{r_p}{r_s} &= -2iQ_1\omega^2/2^2
\end{align*}
$$

(20)

We use the form

$$
\begin{align*}
\Delta \epsilon = J_0 + \left(1 + \frac{1}{\epsilon_1}\right)J_1
\end{align*}
$$

(21)

where $J$ is related to a second-order integral invariant $J_2$ [Ref. 2, Eq. (B7)] by

$$
\Delta \epsilon = J_2 + \left(1 + \frac{1}{\epsilon_1}\right)J_1.
$$

(22)

For nonabsorbing layers the first-order terms $r_{s1}$ and $r_{p1}$ are imaginary, and the second-order terms $r_{s2}$ and $r_{p2}$ are real. We multiply the numerator and denominator of Eq. (19) by the complex conjugate of the denominator. After a lengthy rearrangement of terms, the ratio $r_p/r_s$ can be expressed in terms of the three integral invariants $I_1$, $J_2$, and $i_2$; $i_2$ is defined as

$$
i_2 = 2\Delta \epsilon \lambda_2 - \lambda_1^2.
$$

(23)

(These three integral invariants characterize the reflectivities $|r_p|^2$ and $|r_s|^2$ and the ellipsometric ratio $r_p/r_s$ to second order in the layer thickness, for any layer. Their properties are discussed and their functional forms are tabulated for six profiles in Secs. 3–6 of Ref. 7.) The result is

$$
\begin{align*}
\frac{r_p}{r_s} &= \frac{(q_1 + q_2)^2}{\epsilon_i^2 Q_1 + Q_2^2}
\end{align*}
$$

(24)

where $\theta_0$ is the common value of $\theta_1$ and $\theta_2$ when $\epsilon_1 = \epsilon_2$. The value of $\phi$ is thus zero, not infinity, to lowest order in the interface thickness. The fact that the simple theory gives a divergence as $\Delta \epsilon \rightarrow 0$, whereas the $\Delta \epsilon = 0$ value is zero, suggests the existence of a maximum for small $\Delta \epsilon$. This turns out to be true, as we will see in Section 3.
tric constants of the bounding media. At normal incidence (K = 0), we find \( r_p/r_s \to -1 \), as it must, since there is then no physical difference between the s and p waves. At grazing incidence (\( q_1, Q_1 \to 0 \)), \( r_p/r_s \to -1 \), in accord with a general theorem of reflection (Ref. 7, Sec. 2-3). When \( \Delta \epsilon = 0 \) the invariants \( i_0 \) and \( j_2 \) take the values \(-\lambda_1^2 \) and \(-2\lambda_1 \lambda_1/\epsilon_0 \), respectively, and \( r_p/r_s \) reduces to the value given in Eq. (18).

We note in passing that Eq. (18) tends not to the correct order theory. The points are calculated from the exact reflection invariants theorem of reflection (Ref. 7, Sec. 2-3). When \( \Delta \epsilon = 0 \) the real part of \( \Delta \epsilon \Delta z/\epsilon \) is correct to lowest order in the film thickness and accurate away from grazing incidence.

Thus, when \((\epsilon_0/\epsilon)^{1/2} \cos \theta_0 \) is much less than \( \lambda_1/\epsilon \) and \( \lambda_1 \epsilon/\epsilon \) respectively, \( r_s \to -1 \) and \( r_p \to -1 \), as required. For thin films this limit is reached, however, only for \( \theta_0 \) close to \( 90^\circ \). When \( \epsilon_1 = \epsilon_2 \) the formula \( r_p/r_s \approx \cos^2 \theta_0 - (\lambda_1/\lambda_1) \sin^2 \theta_0 \), derived as in Eq. (18) or from Eq. (24), is correct to lowest order in the film thickness and accurate away from grazing incidence.

Of particular interest is the value of \( \hat{\rho} \). From Eq. (24) we find that the real part of \( r_p/r_s \) equals zero at an angle \( \theta_0 \) that differs in second order in the film thickness from the Brewster angle \( \theta_B = \arctan(\epsilon_0/\epsilon_1)^{1/2} \). Approximating \( \theta_0 \) by \( \theta_B \) and using Eqs. (15) and (16) in Eq. (24), we find that

\[
\hat{\rho} \approx -\frac{1}{2} \Delta \epsilon (\epsilon_1 + \epsilon_2)^{1/2} \frac{\omega}{c} I_1.
\]

This tends to zero as \( \Delta \epsilon \to 0 \), as required. The exact \( \hat{\rho} \) (to second order, but not assuming \( \theta_p = \theta_B \)) is also zero when \( \Delta \epsilon = 0 \) because the functional form \( \Delta I_1/[(\Delta \epsilon)^2 - 4q_1 q_2] \) is retained.

For a uniform layer,

\[
I_1 = \frac{(\epsilon_1 - \epsilon_2)(\epsilon - \epsilon_2)}{\epsilon} \Delta z, \quad i_2 = (\epsilon_1 - \epsilon)(\epsilon - \epsilon_2)\Delta z^2,
\]

\[
\hat{\rho}_m \approx \frac{\epsilon_1(\epsilon_1 - \epsilon)}{4\epsilon}.
\]

Note that \( \hat{\rho}_m \) is independent of the film thickness: index matching \( \epsilon_2 \) close to \( \epsilon_1 \) can give the large ellipsometric signal [expression (28)] even for very thin films.

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**REFERENCES**