Inversion of transmission ellipsometric data for transparent films

John Lekner

Transmission ellipsometry measures the real and imaginary parts of the ratio $\tau = t_p/t_s$, where t_p and t_s are the transmission amplitudes for the p and s polarizations. For a homogeneous layer, the unknowns to be determined are the layer dielectric constant $\varepsilon = n^2$ and the layer thickness Δz. For nonabsorbing films the thickness can be eliminated, and an algebraic equation for ε results. This equation is reduced to a quadratic equation. The thickness is then analytically determined also. The effect of measurement errors on the deduced dielectric constant and layer thickness is discussed. Inversion of thin-film data is also considered.

1. Introduction

Azzam[1] showed that at a 45° angle of incidence, transmission ellipsometric data on a transparent unsupported or embedded layer can be inverted analytically. Here a solution for the general case is presented, at all angles of incidence and for a film that may be supported on a substrate different from the medium of incidence. The methods are similar to those used recently in the analytic reduction of reflection ellipsometric data.[2] A layer is considered that is of thickness Δz and refractive index n, between a medium of incidence of refractive index n_1 and a substrate of refractive index n_2. (The dielectric constants $\varepsilon_1 = n_1^2$, $\varepsilon = n^2$, and $\varepsilon_2 = n_2^2$ are also made use of.) Monochromatic light of angular frequency ω is incident at angle θ_1.

The transmission ellipsometric ratio is given by[3–5]

$$\tau = \frac{t_p}{t_s} = \frac{n_1}{n_2}\frac{(1-p_1)(1-p_2)}{(1+s_1)(1+s_2)}\frac{1+s_1s_2Z}{1+p_1p_2Z}, \tag{1}$$

where

$$s_1 = \frac{q_1-q}{q_1+q}, \qquad s_2 = \frac{q-q_2}{q+q_2},$$
$$p_1 = \frac{Q-Q_1}{Q+Q_1}, \qquad p_2 = \frac{Q_2-Q}{Q_2+Q} \tag{2}$$

are the Fresnel reflection amplitudes of the s and p polarizations at the first and second interfaces, and

$$Z = \exp(2iq\Delta z). \tag{3}$$

The q's are the normal components of the wave vector in the three media:

$$q_1^2 = \varepsilon_1\omega^2/c^2 - K^2, \qquad q^2 = \varepsilon\omega^2/c^2 - K^2,$$
$$q_2^2 = \varepsilon_2\omega^2/c^2 - K^2, \tag{4}$$

where K is the (invariant) tangential component of the wave vector:

$$K = n_1(\omega/c)\sin\theta_1 = n_2(\omega/c)\sin\theta_2. \tag{5}$$

Finally, the Q's are defined by

$$Q_1 = q_1/\varepsilon_1, \qquad Q = q/\varepsilon, \qquad Q_2 = q_2/\varepsilon_2. \tag{6}$$

Transmission ellipsometry measures the real and imaginary parts of τ. Section 2 shows how the unknowns ε and Δz can be determined analytically from Eq. (1) and the experimental data.

2. Inversion of the Transmission Ellipsometry Data

For mathematical convenience the known factor n_1/n_2 is removed from Eq. (1), and we consider as our data the real and imaginary parts of

$$\frac{n_2}{n_1}\tau = t = x + iy. \tag{7}$$

Let us also define the functions

$$f = \frac{(1-p_1)(1-p_2)}{(1+s_1)(1+s_2)}, \qquad P = p_1p_2, \qquad S = s_1s_2. \tag{8}$$

The author is with the Department of Physics, Victoria University of Wellington, Wellington, New Zealand.

Received 12 August 1993; revised manuscript received 3 December 1993.
0003-6935/94/225108-03$06.00/0.

Then Eq. (1) can be written as

$$t = x + iy = f\frac{1 + SZ}{1 + PZ}. \qquad (9)$$

For nonabsorbing layers, Z lies on the unit circle. Because Eq. (9) is a linear equation in Z, we can solve for Z and eliminate it by using $ZZ^* = 1$. This gives us [compare Eq. (13) of Ref. 5], on writing $x^2 + y^2 = r^2$,

$$(1 - P^2)r^2 - 2f(1 - SP)x + f^2(1 - S^2) = 0. \qquad (10)$$

The unknown in Eq. (10) is the dielectric constant of the layer, ε. This appears in q (in s_1 and s_2) and in $Q = q/\varepsilon$ (in p_1 and p_2). Because

$$\varepsilon\frac{\omega^2}{c^2} = q^2 + K^2, \qquad (11)$$

we see that Eq. (10) is an algebraic equation in the unknown q. After some algebraic manipulation this reduces to a quadratic in q^2, and thus to a quadratic in ε.

The algebra and the final equation are simplified by the introduction of dimensionless variables u, v, and w, defined by [2]

$$q^2 = q_1q_2u, \qquad K^2 = q_1q_2v, \qquad q_2 = q_1w. \qquad (12)$$

The unknown ε resides in u, which satisfies the equation

$$au^2 + bu + c = 0,$$

where

$$\begin{aligned} a &= [(1 + v)r^2 - (2 + v)(v + w)x + (v + w)^2]w, \\ b &= (1 + v)[v + w + vw(2 + v + w)]r^2 \\ &\quad - (v + w)[v + 2w + vw(4 + 2v + w)]x \\ &\quad + (1 + v)(v + w)^2w, \\ c &= [(1 + v)vr^2 - (1 + 2v)(v + w)x + (v + w)^2]vw. \end{aligned} \qquad (13)$$

The solution of Eqs. (13) gives us two values of u, and thus two values of

$$\begin{aligned} \varepsilon &= \left(\frac{c}{\omega}\right)^2(q^2 + K^2) = \left(\frac{c}{\omega}\right)^2 q_1q_2(u + v) \\ &= n_1n_2\cos\theta_1\cos\theta_2(u + v). \end{aligned} \qquad (14)$$

The physical root must give all the Fresnel reflection amplitudes a modulus not exceeding unity. If measurements at two angles of incidence are available, the physical roots will agree, and the spurious roots will not. When u (and thus ε) are known, the Fresnel coefficients can be computed, and the thickness can be found from Eq. (9) by means of

$$\exp(2iq\Delta z) = Z = \frac{f - t}{tP - fS}, \qquad (15)$$

which leads to

$$2q\Delta z = \arctan[yf(S - P), xf(S + P) - f^2S - r^2P] \quad (\text{mod } 2\pi), \qquad (16)$$

where arctan (β, α) stands for the arctangent of β/α, placed in the correct quadrant according to the signs of α and β.

3. Effect of Experimental Errors

Figure 1 shows the physical and spurious solutions for the dielectric constant ε and the thickness Δz of a layer of water resting on glass ($n_1 = 1$, $n = 4/3$, $n_2 = 3/2$, $\omega\Delta z/c = 0.5$). The true values of x and y as defined in Eq. (7) were substituted into Eqs. (13), which were solved for u and thus for ε. The two values of ε are shown in Fig. 1 by solid and dashed curves. For a given ε, Eq. (16) then gives the thickness value. Again, the two values of $\omega\Delta z/c$ are plotted as solid and dashed curves. Note that there is a rapid variation with angle of incidence of the spurious value of the thickness, despite a weak variation in the spurious value of ε. Thus, if there is doubt about which root is the physical one, measurements at two angles of incidence will decide between them. The figure also shows the effect of introducing errors δx and δy into the real and imaginary parts of t. Both δx and δy have been taken to be 0.0002 in magnitude, which is approximately ten times the actual experimental error. The effect of such errors (of either sign) is shown in the figure by dotted curves.

We see from Fig. 1 that the effect of errors is largest at small angles of incidence. This is because at normal incidence there is no physical difference between the s and p polarizations (for isotropic media) and $t_s = t_p$, for any layer (or indeed for any isotropic

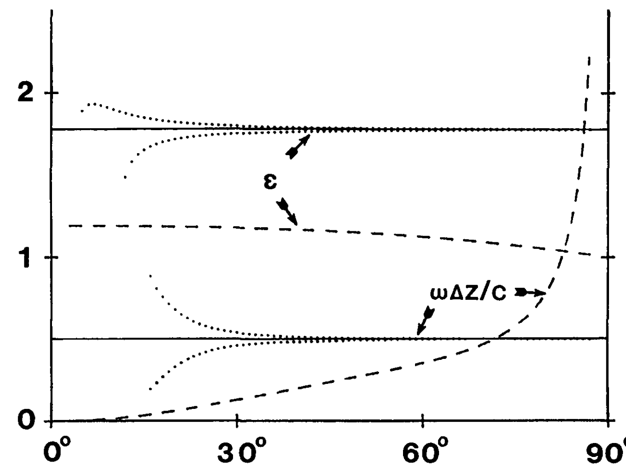

Fig. 1. Physical (solid curves) and spurious (dashed curves) values of ε and $\omega\Delta z/c$ obtained from Eqs. (13) and (16), shown as a function of the angle of incidence. Dotted curves show the result of measurement errors (see text).

stratification). Thus no information can be deduced from the measurement of t_p/t_s at normal incidence. The same is true for reflection ellipsometry of isotropic media.[6] (The upper dotted ε curve appears to dip toward the true value of ε at small angles, but in fact it passes through the true value and rapidly diverges from it as the angle of incidence decreases.)

For small angles of incidence, $\tau = t_p/t_s$ is unity plus a term of order K^2. This follows from the fact that τ depends on $K = n_1(\omega/c)\sin\theta_1$ through q_1, q, and q_2, the squares of each being linear in K^2. There are two types of term of order K^2: those originating in the Fresnel coefficients, for which the appropriate dimensionless parameter is $(cK/\omega)^2/\varepsilon_1 = \sin^2\theta_1$, and those originating in $Z = \exp(2iq\Delta z)$, which are characterized by the dimensionless parameter $K^2\Delta z/(n\omega/c) = (\varepsilon_1/n)\sin^2\theta_1(\omega\Delta z/c)$. Large angles of incidence give the best accuracy for both ε and Δz.

4. Thin Films

Roughly speaking, films can be considered thin when their thickness is small compared with the probe wavelength. More precisely, the thickness enters as $q\Delta z$, and this dimensionless parameter varies from $2\pi n\Delta z/\lambda$ at normal incidence to $(2\pi\Delta z/\lambda)(n^2 - n_1{}^2)^{1/2}$ at grazing incidence. The film is thin when $q\Delta z \ll 1$.

At a fixed angle of incidence, the ratio of transmission amplitudes $\tau = t_p/t_s$ moves on a circle in the complex plane, because it is related to $Z = \exp(2iq\Delta z)$ by means of a bilinear transformation. For different refractive indices the paths are different circles, but all these circles pass through a fixed point on the real axis:

$$\tau_+ = \tau(Z = +1) = \frac{n_1 n_2 \omega^2/c^2}{q_1 q_2 + K^2}. \tag{17}$$

This point must be independent of the layer characteristics, because zero thickness of the layer gives $Z = 1$. All thicknesses Δz such that $q\Delta z$ is an integer multiple of π will also give $\tau = \tau_+$.

The other real value of τ occurs when $Z = -1$: from Eq. (1) we find

$$\tau_- = \tau(Z = -1) = \frac{q_1 q_2 + q^2}{n_1 n_2 \varepsilon(Q_1 Q_2 + Q^2)}. \tag{18}$$

The center of the τ circle (for a given dielectric constant of the layer and a given angle of incidence) lies on the real axis at $(\tau_+ + \tau_-)/2$, and its radius is one half the modulus of

$$\tau_+ - \tau_- = \frac{K^2(\varepsilon_1 - \varepsilon)(\varepsilon - \varepsilon_2)\omega^2/c^2}{(q_1 q_2 + K^2)\varepsilon^2 n_1 n_2 (Q_1 Q_2 + Q^2)}. \tag{19}$$

Because at given angle of incidence the loci of τ for layers with different refractive indices all pass through τ_+, it appears that no information can be obtained concerning the refractive index of a thin film. This is not so: If we expand Z as $1 + 2iq\Delta z + \cdots$ we find that, to first order in Δz,

$$\tau = \frac{n_1 n_2 \omega^2/c^2}{q_1 q_2 + K^2}\left[1 + \frac{iK^2 I_1}{\varepsilon_1\varepsilon_2(Q_1 + Q_2)} + \cdots\right], \tag{20}$$

where I_1 is the homogeneous layer form of a first-order integral invariant:

$$I_1 = \frac{(\varepsilon_1 - \varepsilon)(\varepsilon - \varepsilon_2)}{\varepsilon}\Delta z. \tag{21}$$

The identity $\varepsilon_1\varepsilon_2(Q_1 + Q_2)\omega^2/c^2 = (q_1 + q_2)(q_1 q_2 + K^2)$ of Ref. 7 is used in reducing the first-order term. The same integral invariant I_1 enters into the first-order expression for the reflection ellipsometric ratio.[7,4] As in the reflection case, transmission ellipsometry of thin films can obtain the quantity I_1, but not ε or Δz separately [compare Section V of Ref. 2]. The thickness at which ε and Δz can both be obtained varies with the accuracy of the ellipsometric data; it is roughly one tenth of a wavelength for most experiments.

5. Summary

One can easily invert the transmission ellipsometry data for a transparent film (by solving a quadratic equation) to find the dielectric constant ε and thickness Δz of the layer. This analytic inversion is much simpler than that in the reflection ellipsometry case, which requires the solution of a quintic equation. Errors in measurement cause errors in the deduced refractive index and thickness, and these diverge as normal incidence is approached. For thin films, transmission ellipsometry can determine $I_1 = \Delta z(\varepsilon_1 - \varepsilon)(\varepsilon - \varepsilon_2)/\varepsilon$, but not ε and Δz separately.

References

1. R. M. A. Azzam, "Transmission ellipsometry on transparent unbacked or embedded thin films with application to soap films in air," Appl. Opt. **30,** 2801–2806 (1991).
2. J. Lekner, "On the inversion of ellipsometric data," Appl. Opt. **33, X–X** (1994).
3. R. M. A. Azzam and N. M. Bashara, *Ellipsometry and Polarized Light* (North-Holland, Amsterdam, 1977), Sec. 4.4.2.
4. J. Lekner, *Theory of Reflection* (Nijhoff/Kluwer, Dordrecht, The Netherlands, 1987), Sect. 2-4.
5. M. C. Dorf and J. Lekner, "Reflection and transmission ellipsometry of a uniform layer," J. Opt. Soc. Am. A **4,** 2096–2100 (1987).
6. J. Lekner, "Analytic inversion of ellipsometric data for an unsupported nonabsorbing uniform layer," J. Opt. Soc. Am. A **7,** 1875–1877 (1990).
7. J. Lekner, "Invariant formulation of the reflection of long waves by interfaces," Physica (The Hague) **128A,** 229–252 (1984).