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Simple and direct determination of complex refractive index and thickness of unsupported or embedded thin films by combined reflection and transmission ellipsometry at 45° angle of incidence

R. M.A. Azzam
University of New Orleans, razzam@uno.edu

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R. M. A. Azzam

Department of Electrical Engineering, University of New Orleans, Lakefront, New Orleans, Louisiana 70148

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Measurements of the polarization states (represented by complex numbers χ_r and χ_t, respectively) of light reflected and transmitted by an unsupported or embedded thin film, for totally polarized light (with nonzero p and s components) incident at 45°, permit simple, direct, and explicit determination of the film's complex refractive index N_1 *independently of film thickness or input polarization.* If $\alpha = \chi_r/\chi_t$, we find that $\alpha = r_s + r_s^{-1}$, where r_s is Fresnel's complex reflection coefficient of the ambient–film interface for the s polarization at 45° incidence. From α, r_s is determined, and from r_s we get $N_1 = N_0(1 + r_s^2)^{1/2}/(1 + r_s)$, where N_0 is the refractive index of the transparent medium surrounding the film. Knowledge of the incident polarization χ_i allows the film thickness to be determined, also explicitly, by using either of the ratios χ_i/χ_r or χ_i/χ_t.

INTRODUCTION

The inverse problem in ellipsometry,[1] in which ratios of complex reflection or transmission coefficients determined from polarization measurements are used to find unknown optical properties (including film thicknesses) of a stratified structure, is of long-standing interest. Even for the simple and often-adopted idealized three-phase (ambient–film–substrate) model (with homogeneous isotropic phases separated by sharp parallel-plane boundaries), the inverse problem defies analytical inversion in all but one known case. This is when the optical properties of all media are known and only the film's thickness is to be determined,[2] a case that is of limited practical interest.

In this Letter we show that combined reflection and transmission ellipsometry[3] at a 45° angle of incidence, on an absorbing thin film bounded by a transparent medium of known refractive index (i.e., a two-phase system that consists of a thin-film phase and a surrounding bulk phase), permits simple and explicit determination of the complex refractive index and thickness of the film.

METHOD

Figure 1 shows the reflection and transmission of collimated monochromatic totally polarized light by a plane-parallel thin film of thickness d, medium 1, surrounded by a transparent medium 0. Both media are assumed to be homogeneous and optically isotropic. ϕ is the angle of incidence (45°), and p and s indicate the linear polarizations parallel and perpendicular to the plane of incidence, respectively.

If we approximate the incident beam by a plane wave, the complex amplitude-reflection and -transmission coefficients for the ν polarization ($\nu = p, s$) are given by[4]

$$R_\nu = (r_{01\nu} + r_{10\nu}X)/(1 + r_{01\nu}r_{10\nu}X), \tag{1}$$

$$T_\nu = t_{01\nu}t_{10\nu}X^{1/2}/(1 + r_{01\nu}r_{10\nu}X), \tag{2}$$

where

$$X = \exp(-j2\pi d/D_\phi), \tag{3}$$

$$D_\phi = \frac{\lambda}{2}(N_1^2 - N_0^2 \sin^2\phi)^{-1/2}. \tag{4}$$

In Eqs. (1) and (2), $r_{mn\nu}$ and $t_{mn\nu}$ are Fresnel's reflection and transmission coefficients, respectively, of the mn interface for the ν polarization. In Eq. (4), λ is the free-space wavelength of light, and N_0 and N_1 are the real and complex refractive indices of the ambient and the film, respectively.

Fresnel's coefficients at the 01 and 10 interfaces are interrelated by[5]

$$r_{01\nu} = -r_{10\nu} = r_\nu, \tag{5}$$

$$t_{01\nu}t_{10\nu} = (1 - r_\nu^2). \tag{6}$$

Substitution of Eqs. (5) and (6) into Eqs. (1) and (2) gives

$$R_\nu = r_\nu(1 - X)/(1 - r_\nu^2X), \tag{7}$$

$$t_\nu = (1 - r_\nu^2)X^{1/2}/(1 - r_\nu^2X). \tag{8}$$

From Eqs. (7) and (8), the ratios of p and s reflection and transmission coefficients,

$$\rho_r = R_p/R_s, \qquad \rho_t = T_p/T_s, \tag{9}$$

are given by

$$\rho_r = (r_p/r_s)(1 - r_s^2X)/(1 - r_p^2X), \tag{10}$$

$$\rho_t = [(1 - r_p^2)/(1 - r_s^2)][(1 - r_s^2X)/(1 - r_p^2X)]. \tag{11}$$

In ellipsometry, measurements of the polarization states of the incident (i), reflected (r), and transmitted (t) waves determine ρ_r and ρ_t. Thus, if polarization is represented by the complex number[6]

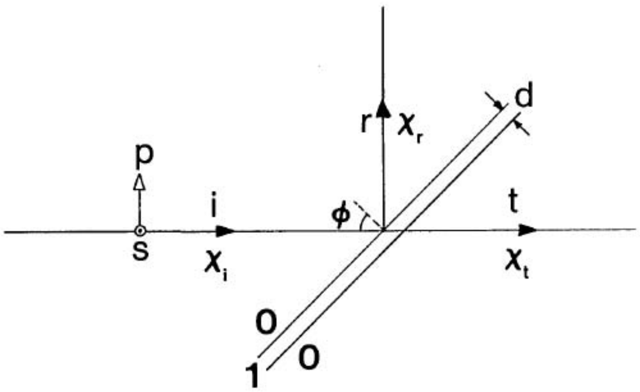

Fig. 1. A light beam (i) incident at $\phi = 45°$ is split into reflected (r) and transmitted (t) components by an unsupported or embedded thin film of thickness d, medium 1, which is in contact on both sides with a transparent medium 0. p and s indicate linear polarizations parallel and perpendicular to the plane of incidence. χ_i, χ_r, and χ_t represent the polarization states of the incident, reflected, and transmitted beams, respectively.

$$\chi_k = E_{ks}/E_{kp}, \qquad k = i, r, t, \tag{12}$$

where E_{ks} and E_{kp} are the s and p phasor components of the electric vector of the kth wave, we find that

$$\rho_r = \chi_i/\chi_r, \qquad \rho_t = \chi_i/\chi_t. \tag{13}$$

In general, Eqs. (10) and (11) overdetermine the film properties (its complex refractive index N_1 and thickness d) in terms of ρ_r and ρ_t. The inversion problem is greatly simplified if we form the ratio

$$\alpha = \rho_t/\rho_r \tag{14}$$

and substitute from Eqs. (10) and (11) to obtain

$$\alpha = (r_s/r_p)(r_p{}^2 - 1)/(r_s{}^2 - 1). \tag{15}$$

Significantly, α is independent of X and hence of film thickness. Furthermore, if Eqs. (13) are substituted into Eq. (14), we get

$$\alpha = \chi_r/\chi_t, \tag{16}$$

which shows that α is determined only by the reflected and transmitted polarization states, independently of the incident polarization. (The latter must, of course, have nonzero p and s components.)

For a known ambient refractive index N_0, Eq. (15) has the complex refractive index of the film, N_1, as its only unknown. The dependence of α on N_1 is implicit in the Fresnel coefficients r_p and r_s of the ambient–film interface.

To solve Eq. (15) for N_1, further simplification is achieved by choosing the angle of incidence $\phi = 45°$. At this angle, Abelès[7] has shown that

$$r_p = r_s{}^2. \tag{17}$$

Equation (17) reduces Eq. (15) to

$$\alpha = r_s + \frac{1}{r_s}, \tag{18}$$

from which

$$r_s = \frac{1}{2}[\alpha \pm (\alpha^2 - 4)^{1/2}]. \tag{19}$$

Of the two roots, only that for which $|r_s| < 1$ has physical significance.

It is interesting that the complex reflection coefficient for the s polarization, r_s, of the ambient–film interface can be determined from the measured polarization states of the reflected and transmitted waves. From r_s, we finally obtain[8]

$$N_1 = N_0(1 + r_s{}^2)^{1/2}/(1 + r_s). \tag{20}$$

To determine film thickness, the incident polarization χ_i must be known (e.g., $\chi_i = 1$, when a linear polarizer is placed in the incident beam with its transmission axis at 45° azimuth from the plane of incidence), so that ρ_r or ρ_t is determined by one of Eqs. (13). Subsequently, Eq. (10) or (11) is solved for X. With $r_p = r_s{}^2$ at $\phi = 45°$, Eq. (10) gives

$$X = (\rho_r - r_s)/r_s{}^3(\rho_r r_s - 1). \tag{21}$$

From ρ_r and r_s, complex X is calculated by using Eq. (21). Next, d is determined from X by Eq. (3):

$$d = (jD_\phi/2\pi)\ln X, \tag{22}$$

where D_ϕ is given by Eq. (4). For an absorbing film, Eq. (22) must provide a unique real value for d. For a transparent film, d is determined up to an integral multiple of the film-thickness period D_ϕ. Any small imaginary part of d must be considered an error caused by an error either of polarization measurements or of the model used to describe the film (e.g., the assumption of optical isotropy or homogeneity). An additional check is provided by determining d from ρ_t by solving Eq. (11) for X and using Eq. (22). Of course, the result must agree, to within an acceptable limit of error, with the determination of d from ρ_r.

SUMMARY

Combined reflection and transmission ellipsometry at a 45° angle of incidence permits simple, explicit, and direct determination of the complex refractive index and thickness of an absorbing thin film that is bounded on both sides by transparent media of the same known refractive index. Such films may be unsupported[9] (such as ultrathin metal foils[10]), immersed in a liquid (e.g., membranes), embedded in a surrounding solid phase, or deposited on a solid substrate and immersed in an index-matched liquid. Significantly, the film's complex refractive index is determined from the measured polarization states of the reflected and transmitted waves, independently of film thickness or incident polarization, and, from knowledge of the incident polarization (which must differ from the p or s state), the film thickness can be found.

In conclusion, we have found a simple solution to the inversion problem of reflection and transmission ellipsometry on an absorbing thin film surrounded by a transparent bulk phase.

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Note added in proof: Measurements on an ultrathin gold foil, kindly provided by Gary Reeves of Los Alamos National Laboratory, confirmed the validity of this technique. These results were presented at the Paris Ellipsometry Conference, June 7–10, 1983.

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5. Ref. 1, p. 283.
6. Ref. 1, Sec. 1.7. $\chi = (\tan\theta + j\tan\epsilon)/(1 - j\tan\theta\tan\epsilon)$, where θ and ϵ are the major-axis azimuth (measured from the p direction) and ϵ is the ellipticity angle of the polarization ellipse.
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10. Ultrathin sheets of gold foil of thickness less than 25 nm have recently been produced at Los Alamos National Laboratory. See Indust. Res. Develop. **25**(3), 43 (1983).