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ELLIPSOMETRY OF UNSUPPORTED AND EMBEDDED THIN FILMS

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Résumé - Nous décrivons ici une méthode explicite nouvelle de détermination simultanée de l'indice complexe et de l'épaisseur des couches minces absorbantes sans support par ellipsométrie de réflexion et transmission. Cette technique a été appliquée au cas d'une lame d'or.

Abstract - A technique for the simultaneous and explicit determination of the complex refractive index and thickness of an unbacked or embedded thin film by combined reflection and transmission ellipsometry at any angle of incidence is presented and applied to an ultrathin gold foil.

Recently we have shown that it is possible to determine, *simply and directly*, both the complex refractive index and thickness of absorbing, unsupported (unbacked) or embedded, thin films by combined reflection and transmission ellipsometry at 45° angle of incidence [1]. To our knowledge, this is the only case that permits *explicit* (non-iterative) inversion to obtain all three parameters of the film, which is assumed to be in contact on both sides with the same transparent medium, or by different media of the *same* refractive index. This paper has two objectives. First, we have found, to our own surprise, that the technique remains as simple at *any* angle of oblique incidence ϕ as it is at 45°. This important generalization is discussed. Second, we present results of the successful application of the method to ultrathin gold foils.

THEORY

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 an arbitrary angle of incidence and p and s represent the linear polarizations parallel and perpendicular to the plane of incidence, respectively. The ellipsometric ratios of p and s complex reflection and transmission coefficients are given by [1]:

$$\rho_r = (r_p/r_s) (1 - r_s^2 X)/(1 - r_p^2 X), \quad (1)$$

$$\rho_t = (1 - r_p^2) (1 - r_s^2 X)/(1 - r_s^2) (1 - r_p^2 X), \quad (2)$$

$$X = \exp \{-j4\pi (d/\lambda) S_1\}, \quad (3)$$

$$S_1 = (N_1^2 - N_0^2 \sin^2 \phi)^{1/2}. \quad (4)$$

Fig. 1 Reflection and transmission of light by an unsupported thin film.

r_p and r_s are the 0-1 (ambient-film) interface reflection coefficients, N_1 is the complex refractive index of the film and N_0 is the real refractive index of the surrounding medium at the wavelength λ of the incident light. From Eqs. (1) and (2), we find that the ratio

$$\alpha = \rho_t / \rho_r = (r_s / r_p) (1 - r_p^2) / (1 - r_s^2), \quad (5)$$

is independent of λ , hence of film thickness. It can also be readily verified /1/ that α is determined only by the reflected and transmitted polarization states, independently of the incident polarization, provided that the latter has nonzero p and s components.

With N_0 known, Eq. (5) has N_1 as its only unknown. To simplify the solution of Eq. (5) for N_1 , we had originally suggested /1/ that ϕ be chosen at 45° , so that Abelès condition /2/ ($r_p = r_s^2$) could be used. Later, we discovered that exactly the same simplicity is attained at every angle of oblique incidence. At any ϕ , Abelès condition is replaced by a more general relationship /3/:

$$r_p = r_s (r_s - \cos 2\phi) / (1 - r_s \cos 2\phi). \quad (6)$$

When Eq. (6) is substituted into Eq. (5), a quartic equation in r_s is obtained with coefficients determined by α and ϕ . Fortuitously, the quartic equation is reducible to the following quadratic equation:

$$r_s^2 - \mu r_s + 1 = 0, \quad (7)$$

where

$$\mu = [2 \cos 2\phi + \alpha (1 + \cos^2 2\phi)] / (1 + \alpha \cos 2\phi). \quad (8)$$

Of the two roots of Eq. (7),

$$r_s = \frac{1}{2} [\mu \pm (\mu^2 - 4)^{1/2}], \quad (9)$$

that for which $|r_s| < 1$ is selected. Next N_1 is obtained from r_s by /3/:

$$N_1 = [\sin^2 \phi + \cos^2 \phi \left(\frac{1 - r_s}{1 + r_s} \right)^2]^{1/2}. \quad (10)$$

To determine the thickness d , Eq. (1) (or, equivalently, Eq. (2)) is solved for X :

$$X = (1 / r_p r_s) (\rho_r r_s - r_p) / (\rho_r r_p - r_s). \quad (11)$$

Two values for d can be obtained from complex X . To see this, Eq. (3) is rewritten as:

$$X = \exp(-d/D_a) \exp(-j2\pi d/D_p), \quad (12)$$

where

$$D_a = \lambda / (4\pi |\operatorname{Im} S_1|), \quad D_p = \lambda / (2\operatorname{Re} S_1) \quad (13)$$

represent attenuation- and phase-thickness parameters, respectively. From Eq. (12), a unique thickness,

$$d = d_a = -(\ln|X|)D_a, \quad (14)$$

and a series of thicknesses,

$$d_p = -(\arg X / 2\pi) D_p + m D_p, \quad (15)$$

are obtained, where m is an integer. Ideally, only one value of d_p , at a certain m , must agree with d_a .

APPLICATION TO ULTRATHIN GOLD FOIL

Self-supporting, light-transmitting, thin films of gold have been prepared and extensively characterized, using transmission interferometry, by Casset /4/, and have received prominent attention recently /5/. Here we present the first ellipsometric study of such films using the simple and novel method we have just described.

Two-zone null ellipsometry /6/ was performed using He-Ne laser light ($\lambda = 6328\text{\AA}$) reflected and transmitted by a non-opaque and unbacked gold foil kindly supplied by Gary Reeves of Los Alamos National Laboratory. The film was mounted on a brass O ring of about 5mm inside diameter. The measured reflection and transmission ellipsometric parameters (ψ_r, Δ_r) and (ψ_t, Δ_t) of the film are given in Table 1 at three angles on incidence: 45, 60, 70°. (Recall that the complex ratio of p and s reflection or transmission coefficients is given by $\rho_k = \tan \psi_k \exp(j\Delta_k)$ where $k = r$ or t .) We have also measured the intensity transmittance of the film at normal incidence and found it to be 0.03866 (i.e. $\approx 4\%$).

Reducing the data of Table 1 gave the results of Table 2. In this table, n and k are the refractive index and extinction coefficient of the film ($N_1 = n - jk$), as determined by Eqs. (8)-(10), where $\alpha = \rho_t/\rho_r$. d_a is the film thickness as obtained from Eq. (14). Equation (15) failed to supply any reasonable value of film thickness; it returned only unacceptably large values of d_p that indicate that the film is opaque when it is not. This discrepancy between theory and experiment can be explained as follows. For the present film, $|X|$, as determined by Eq. (11), is less than 0.02 at all angles of incidence. When the absolute value of a complex quantity is small, the uncertainty of its argument becomes large. (Of course, in the limit $|X| = 0$, $\arg X$ is indeterminate.) Because d_p is obtained from $\arg X$, it, too, becomes uncertain. Equivalently, the calculated phase-thickness parameter D_p is too large ($> 10000\text{\AA}$ at all angles) to be useful as a sensible "yardstick" with which to measure a thickness of the order of a few hundred $\text{\AA}$. By comparison, D_a ($= 174\text{\AA}$ at 45°, $169\text{\AA}$ at 60° and $165\text{\AA}$ at 70°) is of the correct size to suit the purpose film-thickness determination. (Remember also that we are assuming an *idealized model* of a homogeneous and isotropic film with parallel-plane boundaries.)

Because the intensity transmittance at normal incidence T_0 is a sensitive function of film thickness, we decided to improve upon our film-thickness determination by changing d from d_a to minimize the difference, $|T_{0m} - T_{0c}|$, between the measured, $T_{0m} = 0.03866$, and calculated, T_{0c} , values of T_0 . The resulting thicknesses are simply denoted by d in Table 2. Interestingly and significantly, the degree of fit to ellipsometric data, represented by the sum of squares,

$$SSQ = (\psi_{rm} - \psi_{rc})^2 + (\Delta_{rm} - \Delta_{rc})^2 + (\psi_{tm} - \psi_{tc})^2 + (\Delta_{tm} - \Delta_{tc})^2, \quad (16)$$

did not change appreciably. This lends considerable credibility to the newly determined film thickness d . In Eq. (16), the subscripts m and c refer to

Table 1. Ellipsometric Parameters (in deg.) of an Ultrathin Au Foil

ϕ	ψ_r	Δ_r	ψ_t	Δ_t
45°	43.432	153.474	59.321	-25.116
60°	42.455	127.560	69.620	-49.070
70°	41.952	101.318	76.246	-73.448

Table 2. Film Characteristics and Degree-of-Fit Parameters

ϕ	n	k	$d_a(\text{\AA})$	$d(\text{\AA})$	SSQ(deg. ²)	$ T_{om}-T_{oc} $
45°	0.311	-2.800	713	642	0.429	0.25×10^{-4}
60°	0.314	-2.846	777	629	0.364	0.36×10^{-4}
70°	0.282	-2.899	654	616	0.303	1.1×10^{-4}

measured and calculated values, respectively. Table 2 lists the residual $|T_{om}-T_{oc}|$ and SSQ. Averaging the results in Table 2, we obtain the following film parameters:

$$n = 0.302 (+0.02), k = -2.848 (+0.05), d = 629 (+13)\text{\AA}. \quad (17)$$

For comparison, we have determined (using two-zone null ellipsometry) n and k of a *thick* (opaque) gold film deposited on glass; we obtained $n = 0.333$, $k = -2.974$ at $\lambda = 6328\text{\AA}$. Because the optical properties of evaporated films depend on conditions of their preparation, the differences between n and k of the foil and those of the thick film on glass are considered reasonable and not unexpected.

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