Optical Properties of Four Phase Systems

Amir Bazdar , Saeid Rafizadeh

***Abstract*— This paper shows that the four optical parameters [n(metal), k(metal), n(film), and thickness of film] of any metal, semiconductor, or dielectric material covered by a transparent thin film can be determined from ellipsometric data taken at two or more angles of incidence. The method is convenient, non-destructive, and accurate. As an example, an aluminum mirror with a silicon oxide protective coating was characterized. Results are analyzed and probable accuracy determined. The overall range and accuracy to be expected of the method are presented and discussed.**

***Index Terms*— Ellipsometry , Spectroscopy , Palomar Mirror**

I. INTRODUCTION

THERE is often a need to determine the optical constants (or dielectric function) of conductor and semiconductor surfaces. It is difficult, however, to prepare and maintain such a surface free of tickle contaminants. On the other hand, it is relatively easy to maintain such a surface in a constant clean condition if it is covered by a hard durable transparent film, such as a tough oxide. The coating may itself be easily cleaned and maintained. Therefore, there is a need to nondestructively characterize the conductor surface in the presence of its protective cover. There is also a need to determine thickness and refractive index of the covering layer, which is generally unknown and which may be of great interest in its own right. Such a case is represented by a dielectric coated mirror. This paper describes in detail the method of determining all four optical parameters simultaneously, i.e., a method for determining refractive index and thickness of the layer, and the complex index of refraction of the underlying material.

II. TYPICAL EXPERIMENTAL PROCEDURE AND RESULT

In general, upon reflection at any surface, the state of polarization of the incident light changes. Ellipsometry derives its name from the measurement of elliptically polarized light that results from optical reflection. Ellipsometry is a very sensitive method for analysis of specularly reflecting surfaces or of surfaces plus films deposited on them [1]. The change in the state of polarization due to reflection is measured and interpreted in terms of the optical properties of the reflecting surface. In other words, the technique enables one to characterize a surface or thin film in terms of complex refractive index and thickness of the film [2].

A single set of ellipsometric measurements Δ and ψ [3] is sufficient to determine the complex index of refraction of a film whose thickness is known, assuming isotropy and homogeneity. However, in the case of the more general problem involving a thin transparent film and an absorbing substrate, at least four data are needed. A solution to the problem is to increase the number of independent experimental measurements by a method such as varying the angle of incidence [4] or changing the refractive index of the first phase.

A computer problem based on the least square method was developed to invert a system of nonlinear complex equations such as the following for a three-phase system [5,6] to determine n_2 and h_2/λ (thickness/wavelength is dimensionless and mathematically convenient to use) of the transparent film, n_3 and k_3 of the highly absorbing and infinitely thick substance from ellipsometric measurements.

$$r_{\parallel} = \frac{r_{\parallel 12} + r_{\parallel 23}e^{2i\beta}}{1 + r_{\parallel 12}\; r_{\parallel 23}e^{2i\beta}} \tag{1.1}$$

$$r_{\perp} = \frac{r_{\perp 12} + r_{\perp 23}e^{2i\beta}}{1 + r_{\perp 12}\; r_{\perp 23}e^{2i\beta}} \tag{1.2}$$

$$R_{\perp} = \; |r_{\perp}|^2 \quad , \quad R_{\parallel} = \; |r_{\parallel}|^2 \tag{2}$$

$$\tan\psi = (R_{\parallel})^{1/2}/(R_{\perp})^{1/2} \;,\; \Delta = \delta_{\parallel} - \delta_{\perp}$$

$$\delta_{\parallel} = \arg r_{\parallel} \quad , \quad \delta_{\perp} = \arg r_{\perp} \tag{3}$$

and:

$$\beta_j = 2\pi\,(h/\lambda)\,/\,\xi_j \quad , \quad \xi_j = \hat{n}_j \cos\theta_j$$

where: $\hat{n}_j = n_j + ik_j$

As an example of a sample, a silicon oxide coated aluminum mirror was used, commercially known as a *palomar* mirror. It consists of an aluminum film deposited on a glass substrate with the exposed side of the aluminum covered by a thin silicon monoxide film (see Figure 1). The aluminum is highly absorbent and thick such that we can assume it to be homogeneous and infinitely thick. The entire sample will work as a three phase system: The first phase is

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Amir Bazdar is with the Department of Electronic ,I.A.U Urmia Branch (phone: 0098-441-3440680; fax: 0098-441-3440680; e-mail: a.bazdar@iaurmia.ac.ir).

Saeid Rafizadeh is with the Department of Electronic, I.A.U Urmia Branch (e-mail: saeidrafizadeh@gmail.com).

air or some other transparent substrate used. The second phase is the SiO_x film, and the third phase is aluminum. The glass substrate has no optical effects.

The instrument used was a Rudolph Research Model RR 2000 automatic ellipsometer. Major parts of the instrument are shown in Figure 2.

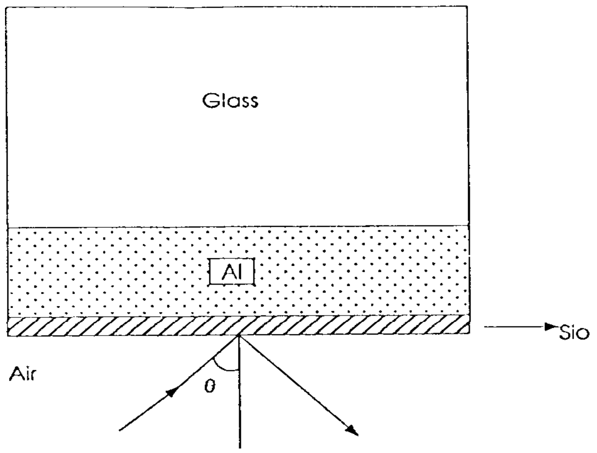

Fig. 1: Schematic of a Palomar mirror.

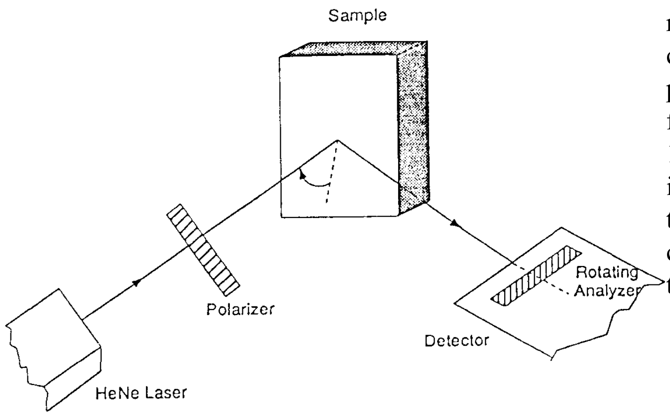

Fig. 2: Schematic of an ellipsometer.

The ellipsometric quantities to be determined are the ratio of the reflection coefficients of the parallel and perpendicular components after reflection (i.e., $\tan\psi = \left| r_{\parallel} \right| / \left| r_{\perp} \right|$), and the phase difference between the components (i.e., $\Delta = \delta_{\parallel} - \delta_{\perp}$). In practice these two quantities are obtained by calculation from two other measurement quantities named the azimuth of the elliptically polarized light, *α,* and its ellipticity, ϵ using equations:

$$\tan\Delta = \tan(2\varepsilon) / \sin(2a) \qquad (4.1)$$

$$\tan(2\psi) = \tan(2a) / \cos(\Delta) \qquad (4.2)$$

The monochromic light source is a laser with λ = 6328 Å. The incident beam becomes linearly polarized when it passes through the polarizer. The reflected beam from the sample surface is elliptically polarized. Measurements were made at six different angles of incidence. The quantities ϵ and *α were* taken from each angle of incidence direction from the instrument. The magnitudes of Δ and *ψ* were calculated from ϵ and *α* readings, using the set of equations (4). Results are shown in Table 1.

Table 1: Ellipsometry parameters (first phase is air).

λ	a	ϵ	Δ	ψ
45°	134.04	2.675	185.343	44.044
50°	133.79	6.68	193.372	43.823
52°	133.69	8.75	197.517	43.751
55°	133.51	12.37	204.769	43.649
60°	133.20	19.625	219.305	43.606
65°	132.88	27.95	235.973	43.812

The least square computer program was used to invert the values of Δ and *ψ* for the data to calculate n_2, h_2/λ, n_3, and k_3. Results are shown in Table 3. The experiment was repeated using mineral oil instead of air as a first phase. A convenient set up, illustrated in Figure 3, was used for this purpose. A thin glass container in circular cross-section was filled with mineral oil, which has the index of refraction 1.4771 at λ = 6328 Å. An important point in this procedure is that the sample should be placed in mineral oil such that the incidence point of the light beam must coincide with the center of the container. When this is true it is easy to set up the device at any desired angle.

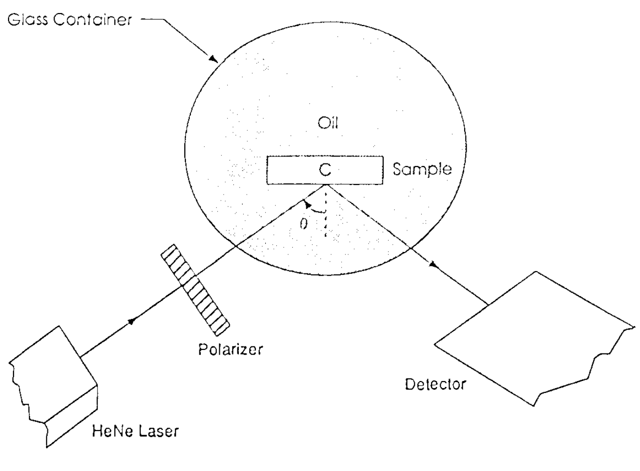

Fig. 3: The unit of internal reflection measurement.

The results are shown in Tables 2 and 3. Comparison of the different sets of measurements shows good agreement in calculated optical parameters, but not as good as within each set. The sample was also measured by reflection spectroscopy [2]. All reflection measurements were made in a Cary-14 double beam spectrophometer with expanded scale slide wire. Measurements were made at different angles of incidence and polarization. The data and calculated optical parameters are given in Table 4.

Table 2: Ellipsometry parameters (first phase mineral oil).

λ	a	ϵ	Δ	ψ
45°	133.42	9.455	161.063	43.505
50°	132.92	12.240	155.463	43.116
55°	132.27	15.53	148.35	42.662
60°	131.32	19.43	140.909	42.138
65°	129.83	24.03	131.474	41.555
70°	127.00	29.75	119.539	40.976

First Phase	Calculated Optical Parameters			
	n_2	h_2/λ	n_3	k_3
Air	1.485	0.7010	1.028	6.2620
Mineral Oil	1.480	0.690	0.997	5.9881

Table 3: Values of optical parameters of palomar mirror.

Table 4: Optical parameters using spectroscopy measurements.

Spectroscopyrements Measurements	Calculated Optical Parameter			
	n_2	h_2/λ	n_3	k_3
$R_{\parallel}$ (45) = 0.838228 $R_{\perp}$ (45) = 0.92080 $R_{\parallel}$ (55.48) = 0.804716 $R_{\perp}$ (55.48) = 0.9348 $R_{\parallel}$ (65.47) = 0.773575 $R_{\perp}$ (65.47) = 0.9523	1.49	0.73	1.13	7.1.5

Table 5: Calculated optical parameters of aluminum bulk (two phase system, first phase air)

Angle of Incidence	Optical Parameters	
	n	k
45°	0.9966	5.9979
65°	0.9966	5 9979
Combination of 45° and 65°	0.9966	5.9979

A number of points are illustrated by comparison of data in Tables 4, 5 and 6.

a. The accuracy of determining the four parameters is very good, comparable to that for the well-known case of two parameter determination.

b. Adding more data of the same variance will increase the accuracy of the overall method, but it will never decrease the accuracy.

c. Reflectance measurements are very poor for determining four optical parameters.

Combination of Measurements	Calculated Optical Parameters			
	n_2	h_2/λ	n_3	k_3
$6E_{in}$	1.4801	0.6897	0.9971	6.005
$6E_{ext}$	1.4853	0.7004	1.0283	6.0899
$6E_{in}$ + $6E_{ext}$	1.4801	0.7011	0.9966	6.0184

Table 6: Values of optical parameters, calculated from different combinations of measurements.

III. CONCLUSION

For the typical case of a transparent film protected metal or semiconductor, the four parameter determination is surprisingly accurate, comparable to the accuracy that can be expected for determining n + ik of a clean bare surface. There is the tremendous advantage, however, in not having to prepare and maintain the clean bare surface (this is very difficult for a metal-like uranium, or even for aluminum). The range of applicability is wide indeed, including all materials with k of the order of unity or greater. For very thin protective films of 50 Å, the method does not work well unless augmented by more information, or the addition of another known film.

A method of error analysis based on least-squares fitting of an arbitrary function has been used to determine the general usefulness of the method. This has proved to be a powerful and convenient approach to quickly ascertain the sensitivity and overall accuracy to he expected for any particular case. This method is recommended for anyone desiring to optimize his results.

This approach to discovering and proving needed new methods tor optical characterization seems to be new in the literature. Perhaps further useful procedures will result from similar approaches.

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