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Analytical determination of the complex dielectric function of an absorbing medium from two angles of incidence of minimum parallel reflectance

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The real and imaginary parts of the complex dielectric function (or complex refractive index) of an opaque substrate or a thick film can be determined from two pseudo-Brewster angles measured in two transparent incidence media of different refractive indices. This two-angle method is simple in that it involves no photometric or polarimetric analysis and in that the solution for the optical properties in terms of the measured angles is explicit, analytical, and direct (i.e. noniterative). The two-angle method is demonstrated for an opaque TiN film on a Cleartran ZnS substrate as a specific example. The effect of angle-of-incidence errors on the determination of the optical properties is investigated, and the domain of applicability of this new and interesting method is also delineated.

1. INTRODUCTION

Determination of the intrinsic optical properties of materials is important for solid-state physics¹ and for applications that involve optical coatings.² In the presence of strong absorption, these properties (specifically, the real and imaginary parts of the complex dielectric function $\epsilon = \epsilon_r + j\epsilon_i$) are determined mainly by use of spectral-reflectance³ or spectroscopic-ellipsometry⁴ methods.

In this paper a new and simple method is described for the determination of the complex ϵ of an optically isotropic (homogeneous, linear, and nonmagnetic) medium that differs from spectral-reflectance and spectroscopic-ellipsometry methods in that it does not require absolute or relative photometric measurement or polarization analysis of the reflected light. The method is based on a generalization of Brewster's law that determines ϵ_r of a transparent medium by $\epsilon_r = \tan^2 \phi_B$, in which ϕ_B is the angle of incidence of minimum (zero) reflectance for incident p -polarized light (R_p). For light reflection at the planar surface of an absorbing medium, R_p reaches a nonzero minimum at the so-called pseudo-Brewster angle ϕ_{pB} , which is a function of ϵ_r and ϵ_i . Two pseudo-Brewster angles ϕ_{pB1} and ϕ_{pB2} , measured with the p -polarized light incident from two different transparent media of known refractive indices, provide sufficient information to determine ϵ_r and ϵ_i . This nonphotometric, nonpolarimetric, two-angle method (TAM) has the further advantage that the inversion that determines the desired parameters from the measured data is analytical, explicit, and noniterative.

2. MATHEMATICAL BASIS OF THE TWO-ANGLE METHOD

The pseudo-Brewster angle is determined in terms of complex ϵ by solving the following cubic equation:

$$(2\epsilon_r + 2|\epsilon|^2)u^3 + (|\epsilon|^4 - 3|\epsilon|^2)u^2 - 2|\epsilon|^4u + |\epsilon|^4 = 0, \quad (1)$$

where $u = \sin^2 \phi$, and the subscript pB is dropped for sim-

licity. Equation (1) or equivalent equations have been derived by several authors.⁵⁻⁷ To develop the TAM, Eq. (1) is considered as a quadratic equation in $|\epsilon|^2$ whose solution can be manipulated to yield

$$y = [a + (a^2 - bx)^{1/2} - x^2]^{1/2}, \quad (2)$$

in which

$$x = \epsilon_r, \quad y = \epsilon_i, \quad (3)$$

$$a = \tan^4 \phi (1.5 - \sin^2 \phi), \quad (4a)$$

$$b = 2 \tan^4 \phi \sin^2 \phi. \quad (4b)$$

Equation (2) is important in that it gives the explicit Cartesian equation of the equi-pseudo-Brewster-angle contour in the complex ϵ plane. When ϕ is allowed to assume all possible values, a complete family of such curves is generated that fills the entire plane. The properties of these contours will be discussed in a forthcoming paper.

Measuring two pseudo-Brewster angles ϕ_1 and ϕ_2 in two transparent incidence media of different refractive indices $\epsilon_{01}^{1/2}$ and $\epsilon_{02}^{1/2}$ allows us to write Eq. (2) twice:

$$y_1 = [a_1 + (a_1^2 - b_1x_1)^{1/2} - x_1^2]^{1/2}, \quad (5)$$

$$y_2 = [a_2 + (a_2^2 - b_2x_2)^{1/2} - x_2^2]^{1/2}, \quad (6)$$

in which

$$x_1 + jy_1 = \epsilon/\epsilon_{01}, \quad (7)$$

$$x_2 + jy_2 = \epsilon/\epsilon_{02}. \quad (8)$$

Because ϵ_{01} and ϵ_{02} are real, Eqs. (7) and (8) define two parallel vectors; hence we can write

$$x_2 = x_1/\epsilon_0, \quad y_2 = y_1/\epsilon_0, \quad (9)$$

$$\epsilon_0 = \epsilon_{02}/\epsilon_{01}. \quad (10)$$

Substitution of Eqs. (9) into Eq. (6) gives

$$y_1 = [\epsilon_0^2 a_2 + (\epsilon_0^4 a_2^2 - \epsilon_0^3 b_2 x_1)^{1/2} - x_1^2]^{1/2}. \quad (11)$$

Equating the squares of the right-hand sides of Eqs. (5) and (11), we obtain, after considerable simplification,

$$x_1 = (4\epsilon_0^2) \frac{(\epsilon_0^2 a_2 - a_1)(\epsilon_0 a_1 b_2 - a_2 b_1)}{(b_1 - \epsilon_0^3 b_2)^2} \tag{12}$$

Equation (12) gives an explicit analytical solution for the real part of the complex dielectric function ϵ/ϵ_0 in terms of ϕ_1 , ϕ_2 , and ϵ_0 . The imaginary part, y_1 , is determined subsequently by direct substitution into the right-hand side of Eq. (5). ϵ is determined next by

$$\epsilon = \epsilon_0(x_1 + jy_1). \tag{13}$$

This completes the mathematical development of the TAM.

3. BULK AND THICK-FILM SAMPLES FOR THE TWO-ANGLE METHOD

To apply the TAM, the absorbing sample may have one planar surface available for specular reflection measurements. For this case of a bulk liquid or solid sample, the first pseudo-Brewster angle ϕ_1 may be measured with the light incident from vacuum, air, or an inert gas, so that $\epsilon_{01} = 1$. For ϕ_2 , a transparent and dense medium of incidence is required whose dielectric function ϵ_{02} is known or is determined separately. (It is also possible, and may even be desirable, to use two different dense media of incidence, e.g.,

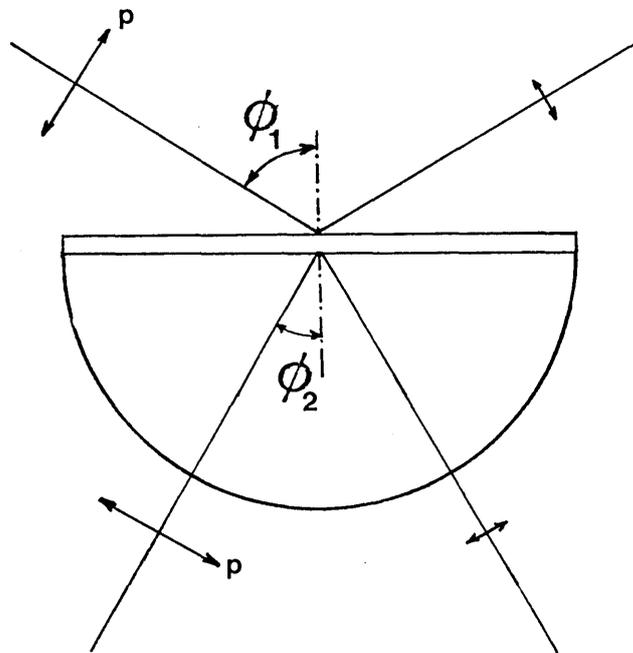


Fig. 2. Pseudo-Brewster angles ϕ_1 and ϕ_2 are measured in external and internal reflection, respectively, on an opaque film deposited upon a hemicylindrical or hemispherical transparent substrate.

two inert immersion liquids.) The situation is represented schematically in Fig. 1.

The preferred sample for the TAM is an opaque (optically thick) film with parallel-plane boundaries that is deposited (e.g., in vacuum) on a transparent substrate. The substrate takes the form of a hemicylinder or a hemisphere. ϕ_1 and ϕ_2 are measured in external and internal reflection, respectively, on the two parallel-plane faces of the film (Fig. 2). Ideally, ϕ_1 should be measured in vacuum (before the sample is removed from the deposition chamber) or in an inert gas, to avoid the formation of a thin contamination or oxide layer on the film face in contact with the ambient. The substrate refractive index should be high so that ϕ_1 and ϕ_2 are sufficiently far apart.

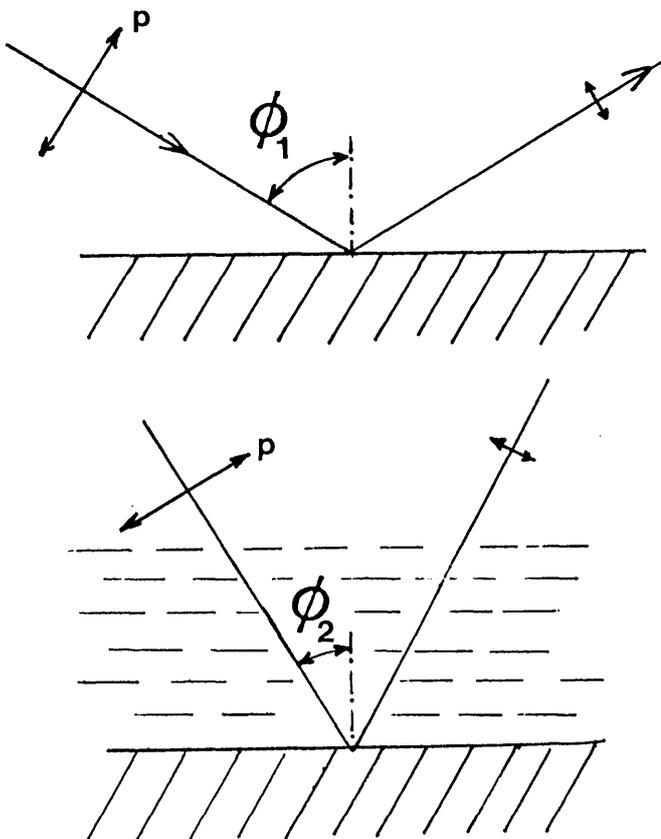


Fig. 1. Pseudo-Brewster angles ϕ_1 (upper panel) and ϕ_2 (lower panel) of minimum reflectance of p -polarized light are measured on the same surface of a bulk absorbing medium in two different media of incidence.

4. OPAQUE TiN FILM ON A ZnS SUBSTRATE: AN EXAMPLE

As a specific example, we consider an opaque film of TiN deposited on a hemicylindrical Cleartran (ZnS) substrate. The proposed method is tested by using the published values of the optical properties of TiN and ZnS (Refs. 8 and 9, respectively) and solving cubic Eq. (1) to determine the pseudo-Brewster angles ϕ_1 and ϕ_2 of external (in-air) and internal reflection, respectively, as functions of the wavelength of light λ in the 0.5–1.5- μm spectral range. The results thus obtained appear in Fig. 3 and simulate the actual data that one would obtain if the TAM were applied to the TiN–ZnS film–substrate system.

With the exact ϕ_1 and ϕ_2 derived from forward calculations, the known dielectric function of the transparent ZnS substrate, and working backwards using the analytical solution described in Section 2, we do obtain the same ϵ_r and ϵ_i of the opaque TiN film that we started with. This confirms the validity of the method. The spectra of ϵ_r and ϵ_i , retrieved

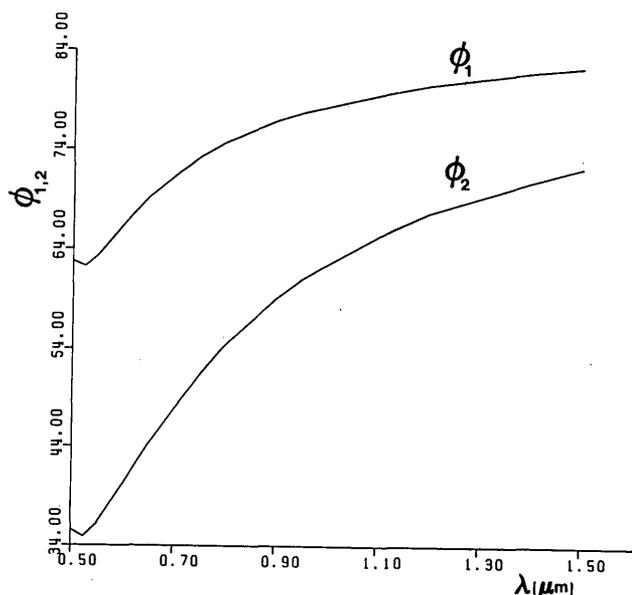


Fig. 3. Spectra of the pseudo-Brewster angles ϕ_1 and ϕ_2 of external (in-air) and internal reflection for a TiN film deposited on a Cleartran (ZnS) substrate. These curves are computed from Eq. (1) and the optical properties of TiN and ZnS given in Refs. 8 and 9, respectively.

from the pseudo-Brewster-angle spectra of Fig. 3, are shown in Fig. 4 by the continuous lines.

Because methods are available for the precise determination of the refractive index of a transparent substrate,¹⁰ the main source of error of the TAM is that associated with measurement of the angles of incidence of minimum parallel reflectance. To simulate the effect of angle-of-incidence errors, the exact pseudo-Brewster angles ϕ_1 and ϕ_2 are rounded off to the nearest 0.1° , which is equivalent to introducing a random error with a maximum possible value of 0.05° . Used in our analytical inversion method, these rounded-off ϕ_1 and ϕ_2 produce incorrect values of ϵ_r and ϵ_i that are represented by the crosses (X's) and pluses (+'s) in Fig. 4.

It is evident from Fig. 4 that the absolute errors $|\Delta\epsilon_r|$ and $|\Delta\epsilon_i|$ increase as $|\epsilon|$ itself increases, or, equivalently, as ϕ_1 and ϕ_2 increase well above 45° . This correlates well with the fact that, for large ϕ_1 and ϕ_2 , the associated constant-pseudo-Brewster-angle contours in the complex ϵ plane approximate semicircles centered on the origin, and the TAM loses its ability to resolve the argument of complex ϵ . Only an estimate of $|\epsilon|$ can be obtained ($\approx \tan^2 \phi_1$, in which ϕ_1 is the pseudo-Brewster angle measured in air or vacuum). Table 1 gives numerical data corresponding to some of the points in Fig. 4.

Another potential source of error is that the incident light may not be exactly p polarized. However, recent results¹¹ indicate that the presence of a small s -polarized component in the incident light has no effect or a negligible effect on the position of the pseudo-Brewster angle, to first order.

5. ERROR ANALYSIS

In Section 4 we have considered the effect of errors of ϕ_1 and ϕ_2 on the determination of ϵ_r and ϵ_i by the TAM, with specific

reference to a TiN film on a ZnS substrate (see Fig. 4 and Table 1). Because the TAM is analytical, it is possible to pursue an exact error analysis for this new method.

From Eq. (12) errors $d\phi_1$ and $d\phi_2$ cause a fractional error dx_1/x_1 of x_1 [$x_1 = \epsilon_r/\epsilon_0$ according to Eq. (7)], which is given by

$$dx_1/x_1 = F_1(da_1/a_1) + F_2(db_1/b_1) + F_3(da_2/a_2) + F_4(db_2/b_2), \quad (14)$$

in which

$$F_1 = [\epsilon_0 a_1 b_2 / (\epsilon_0 a_1 b_2 - a_2 b_1)] - [a_1 / (\epsilon_0^2 a_2 - a_1)], \quad (15a)$$

$$F_2 = [2b_1 / (\epsilon_0^3 b_2 - b_1)] - [a_2 b_1 / (\epsilon_0 a_1 b_2 - a_2 b_1)], \quad (15b)$$

$$F_3 = [\epsilon_0^2 a_2 / (\epsilon_0^2 a_2 - a_1)] - [a_2 b_1 / (\epsilon_0 a_1 b_2 - a_2 b_1)], \quad (15c)$$

$$F_4 = [\epsilon_0 a_1 b_2 / (\epsilon_0 a_1 b_2 - a_2 b_1)] - [2\epsilon_0^3 b_2 / (\epsilon_0^3 b_2 - b_1)]. \quad (15d)$$

Also, from Eqs. (4), an error of ϕ propagates into fractional errors of a and b given by

$$da/a = A(\phi) d\phi, \quad db/b = B(\phi) d\phi, \quad (16)$$

in which

$$A(\phi) = (\cos^2 2\phi + 4 \cos 2\phi + 7) / [\sin 2\phi(1 + 0.5 \cos 2\phi)], \quad (17a)$$

$$B(\phi) = (10 + 2 \cos 2\phi) / \sin 2\phi. \quad (17b)$$

Finally, from Eq. (5) an error dx_1 of x_1 causes an error dy_1 in the estimation of y_1 that is given by

$$dy_1/y_1 = G(dx_1/x_1), \quad (18)$$

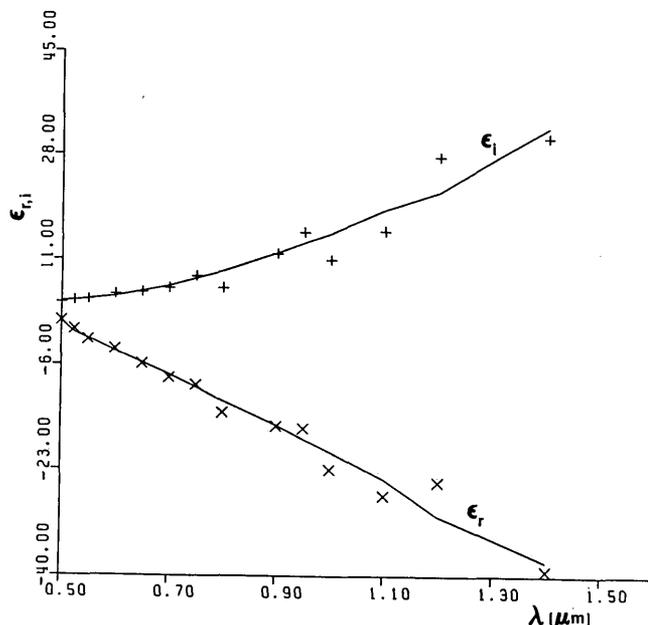


Fig. 4. Spectra of the real and imaginary parts, ϵ_r and ϵ_i , of the complex dielectric function of TiN (continuous lines) obtained by applying the TAM to the simulated pseudo-Brewster-angle data of Fig. 3. The crosses (X's) and pluses (+'s) represent ϵ_r and ϵ_i , respectively, as determined by the TAM when the exact pseudo-Brewster angles are rounded off to the nearest 0.1° . This procedure simulates the effect of angle-of-incidence errors.

Table 1. Application of the TAM to the TiN/ZnS Film-Substrate System^a

λ (μm)	$\epsilon_r(\text{TiN})$	$\epsilon_i(\text{TiN})$	$n(\text{ZnS})$	ϕ_1 ($^\circ$)	ϕ_2 ($^\circ$)	$\Delta\epsilon_r$	$\Delta\epsilon_i$
0.5	1.084	4.224	2.420	62.7945	35.6561	0.1171	-0.0480
0.6	-3.740	5.175	2.363	66.4406	40.1117	-0.1673	-0.0361
0.7	-7.475	6.890	2.332	71.3729	47.8874	-0.1809	-0.1553
0.8	-11.808	9.223	2.314	74.7611	54.4271	-1.8779	-2.7533
0.9	-15.875	12.296	2.301	76.9128	59.1369	-0.1236	-0.2195
1.0	-20.033	15.422	2.293	78.3832	62.5478	-2.8927	-4.4004

^a λ is the wavelength of light. $\epsilon_r(\text{TiN})$ and $\epsilon_i(\text{TiN})$ are real and imaginary parts of the complex dielectric function of the TiN film from Ref. 8. $n(\text{ZnS})$ is the refractive index of the ZnS substrate from Ref. 9. ϕ_1 and ϕ_2 are the pseudo-Brewster angles of external and internal reflection calculated from Eq. (1). $\Delta\epsilon_r$ and $\Delta\epsilon_i$ are the errors of ϵ_r and ϵ_i caused by rounding off ϕ_1 and ϕ_2 to the nearest 0.1° in the TAM. It is apparent that the TAM is reasonably accurate in the visible range but not so accurate in the IR for this particular system.

in which

$$G = -[x_1^2 X_1^{3/2} + (b_1/4)x_1]/[(a_1 - x_1^2)X_1^{3/2} + X_1^2], \quad (19)$$

$$X_1 = a_1^2 - b_1 x_1. \quad (20)$$

This completes the required error analysis for the TAM.

6. DOMAIN OF APPLICABILITY OF THE TWO-ANGLE METHOD

On the basis of the example given in Section 4 and others not present here, we can conclude that the TAM becomes inaccurate (or even unusable) when any one of the following conditions is met:

- (1) Both ϕ_1 and ϕ_2 are $\gg 45^\circ$. This is the case of large $|\epsilon|$, e.g., of high-reflectance metals in the IR.
- (2) $|\epsilon_i/\epsilon_r| \ll 1$; i.e., near an absorption edge or a plasma resonance. In this case ϵ_i is ill determined.
- (3) A high-index-of-refraction substrate (or immersion medium) is not available, so that the difference $\phi_1 - \phi_2$ is not sufficiently large (e.g., is $< 5^\circ$).

The error analysis of Section 5 can be used as a guide for deciding when to use or not to use the TAM.

7. CONCLUSION

In this paper a new and simple method has been introduced for determining the complex dielectric function ϵ , or complex refractive index $N = \epsilon^{1/2}$, of an opaque film or bulk substrate from two pseudo-Brewster angles measured in two different transparent media of incidence. This TAM has the following advantages:

- (1) No photometric or polarimetric measurements are required.
- (2) The optical properties are determined from the two measured angles analytically, explicitly, and noniteratively (see Section 2).
- (3) The effect of angle-of-incidence errors on the calcu-

lated optical properties is also analytically tractable (see Section 5).

Conditions under which the TAM may not be applicable have also been identified (see Section 6).

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