Inversion of the *s* and *p* reflectances of absorbing media

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Simple analytic formulas are given for the real and imaginary parts of the dielectric function of an absorbing medium in terms of the TE and TM reflectances R_s and R_p. An analysis of the formulas shows zero/zero instability at 0°, 45°, and 90° angles of incidence. The instability (extreme sensitivity to experimental error) at 45° is related to the result that $R_p = R_s{}^2$ at 45° incidence, for all absorbing or nonabsorbing media. It is shown that for materials of large refractive index the deduced values of the real and imaginary parts of the dielectric function are very sensitive to experimental error, even at the optimum angle of incidence. © 1997 Optical Society of America [S0740-3232(97)02106-6]

1. INTRODUCTION

The optical properties of homogeneous absorbing media are characterized by the real and imaginary parts n_r and n_i of the refractive index $n = n_r + in_i$, or equivalently by the real and imaginary parts of the dielectric function $\epsilon = \epsilon_r + i\epsilon_i = n^2$, so that

$$\epsilon_r = n_r{}^2 - n_i{}^2, \qquad \epsilon_i = 2n_r n_i. \tag{1}$$

At a given angle of incidence, measurement of the *s* (TE) and *p* (TM) reflectances R_s and R_p gives two numeric values, which can be matched to the two unknowns ϵ_r and ϵ_i (or n_r and n_i). To date, this inversion of the reflectance data to obtain the optical constants has been done numerically,[1] although an approximate analytic inversion near glancing incidence has been given.[2] Here we obtain simple analytic formulas for ϵ_r and ϵ_i in terms of R_s, R_p and the angle of incidence and use these to determine the sensitivity of ϵ_r and ϵ_i to experimental error.

The normal component q of the wave vector in the absorbing medium is complex, with[3,4]

$$(cq/\omega)^2 = \epsilon - \epsilon_1 \sin^2\theta = \epsilon_r - \epsilon_1 \sin^2\theta + i\epsilon_i \tag{2}$$

where ϵ_1 is the dielectric constant of the medium of incidence and θ is the angle of incidence. We write $q = q_r + iq_i$, so that

$$q_r{}^2 - q_i{}^2 = \left(\frac{\omega}{c}\right)^2 (\epsilon_r - \epsilon_1 \sin^2\theta), \tag{3}$$

$$2q_r q_i = \epsilon_i (\omega/c)^2. \tag{4}$$

Thus q_r and q_i contain a square root within a square root; for example,

$$q_r = \frac{\omega}{c}\left\{\frac{1}{2}\left[\epsilon_r - \epsilon_1 \sin^2\theta + \sqrt{(\epsilon_r - \epsilon_1 \sin^2\theta)^2 + \epsilon_i{}^2}\right]\right\}^{1/2}. \tag{5}$$

The *s* and *p* reflectances are given by[3,4]

$$R_s = \frac{(q_1 - q_r)^2 + q_i{}^2}{(q_1 + q_r)^2 + q_i{}^2}, \tag{6}$$

$$R_p = \frac{(Q_1 - Q_r)^2 + Q_i{}^2}{(Q_1 + Q_r)^2 + Q_i{}^2}, \tag{7}$$

where $q_1 = (\omega/c)\cos\theta$ is the normal component of the wave vector in medium 1 and $Q_1 = q_1/\epsilon_1$, $Q = q/\epsilon$, so that[4]

$$Q_r = \frac{\epsilon_r q_r + \epsilon_i q_i}{\epsilon_r{}^2 + \epsilon_i{}^2}, \qquad Q_i = \frac{\epsilon_r q_i - \epsilon_i q_r}{\epsilon_r{}^2 + \epsilon_i{}^2}. \tag{8}$$

The difficulty in analytically solving for ϵ_r and ϵ_i in terms of R_s and R_p is that the unknown real and imaginary parts of ϵ are contained in q_r and q_i within the square roots. Section 2 shows how the square roots can be eliminated, and analytic formulas found for ϵ_r and ϵ_i. The intermediate expressions are complicated, but the final results are not. Only the method and the solution will be given; the details of the intermediate steps are omitted.

2. INVERSION METHOD

We form the quantities

$$s = \frac{1 - R_s}{1 + R_s} = \frac{2q_1 q_2}{q_1^2 + q_r{}^2 + q_i{}^2}, \tag{9}$$

$$p = \frac{1 - R_p}{1 + R_p} = \frac{2Q_1 Q_r}{Q_1^2 + Q_r{}^2 + Q_i{}^2}, \tag{10}$$

and note that s^2 and p^2 contain only squares of q_r and q_i after Eq. (4) is used to substitute for the product $q_r q_i$ in $Q_r{}^2$:

$$Q_r{}^2 = \frac{\epsilon_r{}^2 q_r{}^2 + \epsilon_i{}^2 q_i{}^2 + \epsilon_r \epsilon_i{}^2 (\omega/c)^2}{(\epsilon_r{}^2 + \epsilon_i^2)^2}. \tag{11}$$

Thus s^2 and p^2 are each equal to expressions that contain a single square root,

$$\rho = \sqrt{(\epsilon_r - \epsilon_1 \sin^2 \theta)^2 + \epsilon_i^2}. \tag{12}$$

From each of s^2 and p^2 we obtain expressions for ρ, namely ρ_s and ρ_p. Then we eliminate the square roots altogether in two ways: We form two functions that are identically zero,

$$z_1(\epsilon_r, \epsilon_i, s^2, p^2, \theta) = \rho_s - \rho_p \tag{13}$$

and, from Eq. (12),

$$z_2(\epsilon_r, \epsilon_i, s^2, \theta) = (\epsilon_r - \epsilon_1 \sin^2 \theta)^2 + {\epsilon_i}^2 - {\rho_s}^2. \tag{14}$$

On setting both functions z_1 and z_2 equal to zero, and eliminating square roots by solving for them and squaring, we obtain purely algebraic equations for ϵ_r and ϵ_i. Both are quadratic in ${\epsilon_i}^2$. The condition that the two quadratics have a common root is expressible in terms of the coefficients of the two quadratics [see Eq. (14) of Ref. 5]. This condition is in the form $Z = 0$, and here Z factors into two expressions, both linear in ϵ_r, one of which contains the physical solution. The expression for the common root of the two quadratics [Eq. (15) of Ref. 5] then gives ${\epsilon_i}^2$. The results for ϵ_r and ϵ_i are, expressed in terms of s, p and $C = \cos^2 \theta$,

$$\frac{\epsilon_r}{\epsilon_1} = \frac{c_0 + c_1 C + c_2 C^2 + c_3 C^3}{2C[s(1 - sp) - p(1 - s^2)C]^2},$$

$$c_0 = s^2(p - s)^2,$$

$$c_1 = 2s(1 - s^2)(2s - p - sp^2),$$

$$c_2 = 2(1 - s^2)[p(p - s) - 2s^2(1 - p^2)],$$

$$c_3 = 2p(1 - s^2)[2s - p(1 + s^2)], \tag{15}$$

$$\frac{\epsilon_1}{\epsilon_1} = \frac{s(p - s)|1 - 2C|[d_0 + d_1 C + d_2 C^2 + d_3 C^3 + d_4 C^4]^{1/2}}{2C[s(1 - sp) - p(1 - s^2)C]^2},$$

$$d_0 = -s^2(p - s)^2,$$

$$d_1 = 4s(1 - s^2)(p - s),$$

$$d_2 = 4(1 - s^2)[s^2(1 + p^2) - p(s + p)],$$

$$d_3 = 8p^2(1 - s^2)^2,$$

$$d_4 = -4p^2(1 - s^2)^2. \tag{16}$$

Note that the solutions for ϵ_1 and ϵ_i have the same denominator, which goes to zero as $C = \cos^2 \theta$ goes to zero, namely at grazing incidence. Since ϵ_r and ϵ_i are fixed quantities, there must be a corresponding zero in the numerator. When $C = 0$ there are, in fact, two factors in the numerators of ϵ_r and ϵ_i that tend to zero at grazing incidence, namely, s and $s - p$ (both R_s and R_p tend to unity at grazing incidence so s and p both tend to zero). Physically, no information can be obtained from measurements at glancing incidence, since R_s and R_p are equal to unity there, for all media and, in fact, for all reflecting stratifications (Secs. 2 and 3 of Ref. 4). Mathematically, this manifests itself as a zero/zero instability.[6] Section 3 analyzes the consequence of two other zero/zero instabilities and looks at the effect of experimental error on the deduced optical constants ϵ_r and ϵ_i.

3. SENSITIVITY TO EXPERIMENTAL ERROR

Experimental measurements of R_s and R_p carry some small but finite uncertainty, as does the measurement of the angle of incidence. What is the effect of experimental error on the deduced values of ϵ_r and ϵ_i? We saw at the end of the last section that these values become more sensitive to error as glancing incidence is approached. There are two more values of angle of incidence that should be avoided: 0° and 45°.

At normal incidence $R_s = R_p$ (there is no physical difference at normal incidence between the TE and TM polarizations when the media are isotropic), and when $C = \cos^2 \theta$ equals unity the numerator and denominator of the ϵ_r expression in Eq. (15) both have $(p - s)^2$ as a factor. Likewise, the numerator and denominator of the ϵ_i expression in Eq. (16) both have $(p - s)^4$ as a factor. Thus both expressions show a zero/zero instability at normal incidence, as expected.

The fact that the inversion fails at normal and glancing incidence is expected on physical grounds. Less obvious is the failure at 45° incidence. It is known[7–9] that $R_p = {R_s}^2$ at 45° angle of incidence. The form of Eq. (16) shows that this must be so: the numerator has the factor $1 - 2\cos^2 \theta$, which is zero at 45°. We thus expect a corresponding zero in the denominator when $C = \cos^2 \theta = 1/2$. This leads to $p = 2s/(1 + s^2)$, which implies $R_p = {R_s}^2$. It is straightforward to verify from Eqs. (6) and (7) that at $\theta = 45°$ R_p does equal ${R_s}^2$. The value of R_s at 45° is

$$R_s(45°) = \frac{8\epsilon_1 {\epsilon_i}^2 + 2\epsilon_r v^2 - v^3}{8\epsilon_1 {\epsilon_i}^2 + 2\epsilon_r v^2 + v^3}$$

$$v^2 = 2\epsilon_1\{2\epsilon_r - \epsilon_1 + [(2\epsilon_r - \epsilon_1)^2 + {\epsilon_i}^2]^{1/2}\}. \tag{17}$$

We note in passing that the result $R_p = {R_s}^2$ at 45° angle of incidence does not hold for reflection from basal planes of anisotropic crystals (for which the TE and TM characterizations suffice; see for example Sec. 7–12 of Ref. 4). In that case we find, for a uniaxial crystal with ordinary and extraordinary indices n_o and n_e, on using the results of Sec. 5.1 of Ref. 10, that at 45° incidence

$${R_s}^2 - R_p = \frac{16 n_1 n_o (m_e n_o - m_o n_e)({n_o}^3 n_e - {n_1}^2 m_o m_e)}{(n_o n_e + n_1 m_e)^2 (n_1 + m_o)^4}, \tag{18}$$

where

$${m_o}^2 = 2{n_o}^2 - {n_1}^2 \qquad {m_e}^2 = 2{n_e}^2 - {n_1}^2. \tag{19}$$

The inversion solutions for ϵ_r and ϵ_i, Eqs. (15) and (16), become identities when the exact values of $s = (1 - R_s)/(1 + R_s)$ and $p = (1 - R_p)/(1 + R_p)$ are substituted. (This statement implies, incidentally, that the expression $d_0 + d_1 C + d_2 C^2 + d_3 C^3 + d_4 C^4$ in Eq. (16) is identically zero for nonabsorbing media, for which $\epsilon_i = 0$. The numerator N_r of the right-hand side of Eq. (15) is thus $\epsilon_r \epsilon_1$ times the denominator D, and likewise the numerator N_i of the right-hand side of Eq. (16) is $\epsilon_i \epsilon_1$ times the same denominator. We have seen above that the common denominator D is zero at 0°, 45°, and 90° angle of incidence, leading to a zero/zero instability.

Any region in which D is small is to be avoided, since there is cancellation between terms of order unity to produce a small quantity, with a large uncertainty. Thus the reciprocal of D acts as an error multiplier.

Figure 1 shows the reflectivities R_s and R_p for glass, silicon, and aluminum at 633 nm. Figure 2 depicts D for those materials. We see that D can be small even at its peak, with maximum values of approximately 9.0×10^{-3} at 71° for glass ($\epsilon = 2.25$), 4.1×10^{-4} at 72° for Si($\epsilon \approx 15 + 0.15i$) and 4.9×10^{-7} at 76° for Al($\epsilon \approx -56 + 21i$). (The dielectric function values for Al and Si are taken from Ref. 11, pp. 405 and 565.) For large values of $|\epsilon|$ there is almost complete cancellation of the terms in D and corresponding cancellation in the numerators of the expressions for ϵ_r and ϵ_i. The terms that cancel are of order unity and experimentally come from measurements of R_s, R_p and the angle of incidence. The insensitivity of reflectivities to refractive-index values when these are large has been noted (see, for example, Fig. 3 of Ref. 6). Conversely, the deduced values of ϵ_r and ϵ_i from measurements of R_s, R_p, and θ are very sensitive to experimental error.

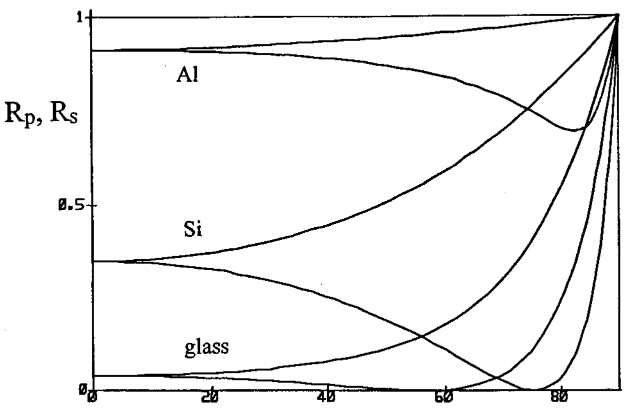

Fig. 1. Reflectances R_s and R_p for glass ($\epsilon = 2.25$), silicon ($\epsilon = 15 + 0.15i$), and aluminum ($\epsilon = -56 + 21i$), versus the angle of incidence θ. In all cases the s and p reflectances are equal at normal incidence, and $R_s \geqslant R_p$ at all angles incidence.

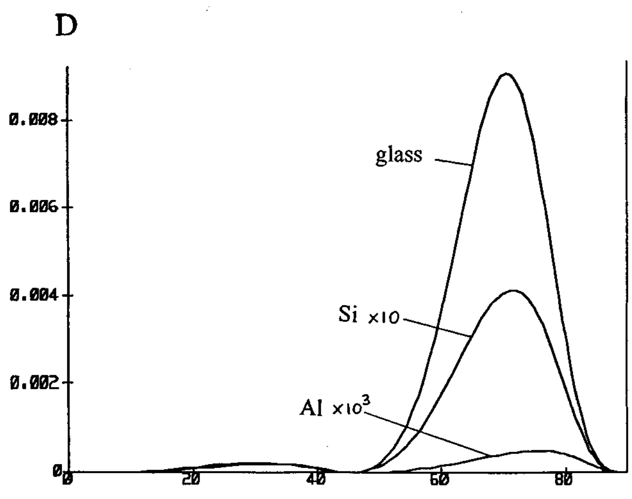

Fig. 2. Common denominator $D = 2C[s(1 - sp) - p(1 - s^2)C]^2$ of inversion formulas (15) and (16) as a function of the angle of incidence for Al($\epsilon \approx -56 + 21i$), Si($\epsilon \approx 15 + 0.15i$), and glass ($\epsilon = 2.25$). The reciprocal of D is a measure of sensitivity to error; note the zeros at 0°, 45°, and 90°.

A more precise measure of the sensitivity to experimental error is provided by the derivatives of the expressions for ϵ_r and ϵ_i with respect to the variables $C = \cos^2\theta$, $p = (1 - R_p)/(1 + R_p)$ and $s = (1 - R_s)/(1 + R_s)$. For example, writing r for ϵ_r/ϵ_1, we have

$$\mathrm{d}r = \frac{\partial r}{\partial C}\mathrm{d}C + \frac{\partial r}{\partial p}\mathrm{d}p + \frac{\partial r}{\partial s}\mathrm{d}s. \tag{20}$$

On the assumption that the errors in C, p, and s are uncorrelated and random, the root mean square of $\mathrm{d}r$ gives the uncertainty in ϵ_r/ϵ_1:

$$\langle(\mathrm{d}r)^2\rangle = \left[\left(\frac{\partial r}{\partial C}\right)^2\langle(\mathrm{d}C)^2\rangle + \left(\frac{\partial r}{\partial p}\right)^2\langle(\mathrm{d}p)^2\rangle + \left(\frac{\partial r}{\partial s}\right)^2\langle(\mathrm{d}s)^2\rangle\right]. \tag{21}$$

A given experiment will have random errors in C, p, and s of different magnitudes, and each varying with the angle of incidence. To obtain a simple measure of the effect of random errors on the deduced value of $r = \epsilon_r/\epsilon_1$, we take the random errors in C, p, and s to be of the same order of magnitude, and calculate

$$\Delta_r = \left[\left(\frac{\partial r}{\partial C}\right)^2 + \left(\frac{\partial r}{\partial p}\right)^2 + \left(\frac{\partial r}{\partial s}\right)^2\right]^{1/2}. \tag{22}$$

A logarithmic plot of Δ_r versus angle of incidence is shown in Fig. 3, for the range $\theta \geqslant 55°$ of practical interest. The physical meaning of Δ_r is that of an error multiplier: for a given common value of the root-mean-square error in C, p, or s, the error in $r = \epsilon_r/\epsilon_1$ will be Δ_r times this error (under the assumptions given above). We see that glass has a minimum error multiplier of $\sim$27 near $\theta = 68°$, Si $\sim$157 near 78° and Al $\sim 2.1 \times 10^4$ near 83°. The reason for the dip in the Δ_r curve for Si is that $\partial r/\partial C$ is close to zero near 78°: for nonabsorbing materials ($\epsilon_i = 0$), the derivative $\partial\epsilon_r/\partial C$ is zero at $C = 2\epsilon_1/3\epsilon_r + O(\epsilon_1/\epsilon_r)^3$. For large ϵ_r/ϵ_1 this zero is at approximately $(2\epsilon_1/3\epsilon_r)^{1/2}$ rad from glancing incidence. For $\epsilon_r/\epsilon_1 = 15$ this gives $\theta \approx 78°$.

The error multiplier Δ_I for $I = (\epsilon_i/\epsilon_1)^2$, namely,

$$\Delta_I = \left[\left(\frac{\partial I}{\partial C}\right)^2 + \left(\frac{\partial I}{\partial p}\right)^2 + \left(\frac{\partial I}{\partial s}\right)^2\right]^{1/2}, \tag{23}$$

is shown in Fig. 4. Its minimum values are for glass $\sim$56 near 66°, for Si $\sim 2.1 \times 10^3$ near 76°, and for Al $\sim 2.8 \times 10^5$ near 86°. The dip in the silicon curve is due to $\partial\epsilon_i^2/\partial C$ passing through zero when ϵ_i is zero. The derivative of ϵ_i^2 with respect to C is then

$$\frac{1}{\epsilon_1^2}\left(\frac{\partial\epsilon_i^2}{\partial C}\right)_{\epsilon_i=0} = \frac{(r-1)(1-r-C)(rC+C-1)}{C(1-2C)(1-C)}. \tag{24}$$

We note that, as expected, this is infinite at $C = 1$, 1/2, and 0 ($\theta = 0°$, 45°, and 90°). The derivative is zero when

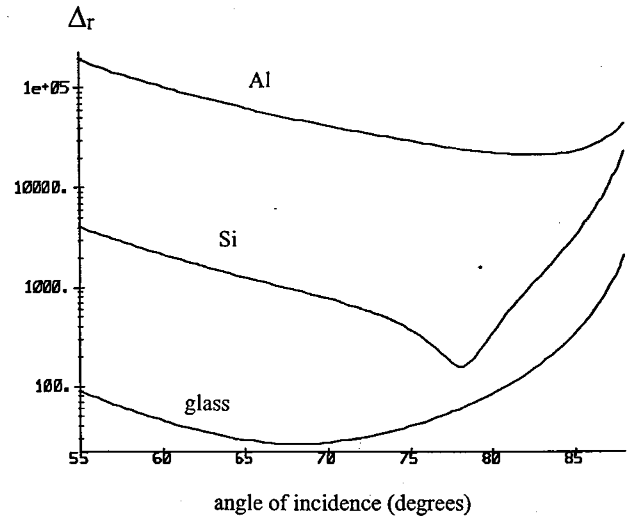

Fig. 3. Plot of the error multiplier Δ_r, which multiplies experimental uncertainties in $C = \cos^2\theta, p = (1 - R_p)/(1 + R_p)$, and $s = (1 - R_s)/(1 + R_s)$ to estimate the uncertainty in ϵ_r/ϵ_1.

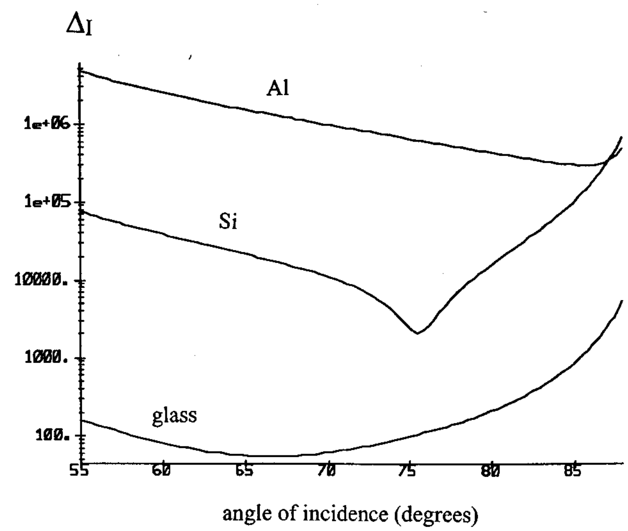

Fig. 4. Plot of the error multiplier Δ_I, which gives the uncertainty in $\epsilon_i{}^2 2/\epsilon_1{}^2$ for glass, silicon, and aluminum.

$C = 1 - \epsilon_r/\epsilon_1$ and $C = \epsilon_1/(\epsilon_r + \epsilon_1)$. For silicon the latter expression puts the minimum in Δ_I at 75.5°, very close to the actual location at 75.6°.

4. CONCLUSIONS

Section 3 has demonstrated that extraction of ϵ_r and ϵ_i from experimental reflectance values is impossible near $\theta = 0°$, 45°, and 90° and that the error multipliers D^{-1}, Δ_r, and Δ_I can be large, even at the optimum angle of incidence. This problem is moderate for glasses (ϵ_i zero or very small, $\epsilon_r \sim 2$ or 3) and more pronounced for materials with large real part of the dielectric constant, particularly if the imaginary part is also large. A typical metallic reflector such as aluminum has very large error multipliers, so large that one might well be skeptical about optical constants derived from inversion of θ, R_p, and R_s measurements.

My conclusions are that accurate inversion of s and p reflectances is possible for materials of moderate absorption but that the choice of the range of angle of incidence is very important, particularly for reflectors with large real part of the dielectric function. In the latter case the angle of incidence should be close to $\arccos(2\epsilon_1/3\epsilon_r)^{1/2}$ or $\arccos[\epsilon_1/(\epsilon_r + \epsilon_1)]^{1/2}$ for determination of ϵ_r and ϵ_i, respectively. In all cases the angle of incidence should be large, from 60° upward.

The error analysis presented above has made assumptions about the nature of the errors and about the relative magnitudes of the uncertainties in the determination of θ, R_s, and R_p. To avoid the latter assumptions, the experimenter may wish to estimate random error simply by repeated measurements within a range of angles of incidence, and substitution of the measured values into formulas (15) and (16).

REFERENCES

1. L. Ward, *The Optical Constants of Bulk Materials and Films* (Hilger, Bristol, UK, 1988), Sec. 2.6.
2. R. M. A. Azzam, "Grazing-incidence differential-reflectance method for explicit determination of the complex dielectric function of an isotropic absorbing medium," Rev. Sci. Instrum. **54**, 853–855 (1983).
3. M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1965), Chap. 13.
4. J. Lekner, *Theory of Reflection* (Nijhoff/Kluwer, Dordrecht, The Netherlands, 1987), Chap. 8.
5. J. Lekner, "Inversion of reflection ellipsometric data," Appl. Opt. **33**, 5159–5165 (1994).
6. J. Lekner, "Determination of complex refractive index and thickness of a homogeneous layer by combined reflection and transmission ellipsometry," J. Opt. Soc. Am. A **11**, 2156–2158 (1994).
7. S. P. F. Humphreys-Owen, "Comparison of reflection methods for measuring optical constants without polarimetric analysis, and proposal for new methods based on the Brewster angle," Proc. Phys. Soc. London **77**, 949–957 (1961).
8. D. W. Berreman, "Simple relation between reflectances of polarized components of a beam when the angle of incidence is 45°," J. Opt. Soc. Am. **56**, 1784 (1966).
9. R. M. A. Azzam, "On the reflection of light at 45° angle of incidence," Opt. Acta **26**, 113–115, 301 (1979).
10. J. Lekner, "Reflection and refraction by uniaxial crystals," J. Phys., Condens. Matter **3**, 6121–6133 (1991).
11. E. D. Palik, ed., *Handbook of Optical Constants of Solids* (Academic, Orlando, Fla., 1985).