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

Karim Javily

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Total refraction at oblique incidence by a transparent bilayer coating on a high-index transparent or absorbing substrate

R. M. A. Azzam and Karim Javily

Transparent bilayer coatings that produce total refraction (TR) of obliquely incident monochromatic light into an underlying substrate are considered. When the substrate is transparent, it is shown that TR takes place without any accompanying change of polarization. Totally refracting bilayers are realizable in the IR where high-refractive-index substrates are available. This is illustrated by a BaF2-ZnSe bilayer on a Ge substrate at a 10.6-um (CO₂-laser) wavelength and 45° angle of incidence. Limited changes of the angle of incidence, wavelength, and refractive indices and thicknesses of the two films of the bilayer are introduced, and their effects on the condition of TR are determined. TR (hence absorption) is also possible for absorbing (semiconductor or metallic) substrates using transparent bilayers of films of nonquarter-wave optical thickness, as is further demonstrated in this paper.

1. Introduction

For a given transparent or absorbing substrate at a given angle of (oblique) incidence, it is possible to suppress completely the reflection of parallel p-or perpendicularly s-polarized incident monochromatic light using a transparent single-layer coating.' However, when the incident light has nonzero p and *s* components (e.g., with equal mix as in the case of unpolarized or circularly polarized light), the reflectance of the surface can only be reduced by a singlelayer coating to a nonzero minimum.^{2,3} Complete and simultaneous antireflection of the p and s polarizations at oblique incidence (a condition that we shall conveniently refer to as total refraction) requires at least two layers, as was first noted by Baumeister.4 When the substrate is transparent, each one of the two layers is of quarter-wave optical thickness at the given angle of incidence. For a glass substrate (with refractive index of \sim 1.5), the required refractive index of the outer layer (in contact with the air) is too low (1.08) to make total refraction using a bilayer practical.4

In this paper we examine realizable bilayer coatings on a high-index (e.g., Ge) substrate that produce total

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refraction (TR) of incident (IR) monochromatic light at oblique incidence. The sensitivity of the design to changes of the angle of incidence, wavelength, film refractive indices, and film thicknesses is considered. We also prove that TR occurs without any change of polarization. Furthermore, we find that TR (hence total absorption) continues to be possible with transparent bilayer coatings when the substrate is absorbing (e.g., semiconducting or metallic) over a wide range of the substrate extinction coefficient. In the latter case, the two films of the bilayer no longer have quarterwave optical thicknesses.

II. Bilayer Coatings that Produce Total Refraction

Consider a monochromatic light beam of wavelength λ traveling in an ambient (medium 0, usually air) which is reflected at an angle ϕ by two transparent thin films (films 1 and 2) on a substrate (medium 3). All (bulk and thin-flim) media are assumed to be homogeneous, optically isotropic, linear, nonmagnetic, and separated by sharp parallel-plane interfaces. The complex amplitude-reflection coefficients of such a system are given by⁵

$$
R_{\nu} = \frac{r_{01\nu} + r_{12\nu}X_1 + r_{01\nu}r_{12\nu}r_{23\nu}X_2 + r_{23\nu}X_1X_2}{1 + r_{01\nu}r_{12\nu}X_1 + r_{12\nu}r_{23\nu}X_2 + r_{01\nu}r_{23\nu}X_1X_2}, \ \nu = p,s,
$$
 (1)

where

$$
X_i = \exp(-j2\pi \zeta_i), \qquad i = 1, 2,
$$
 (2)

and *rim,,* is Fresnel's reflection coefficient of the *im* interface for the v polarization. In Eq. (2), ζ_i is the normalized thickness of the ith film,

The authors are with University of New Orleans, Lakefront, Department of Electrical Engineering, New Orleans, Louisiana 70148. Received 12 July 1985.

$$
\zeta_i = d_i/D_{\phi i}, \qquad i = 1, 2, \tag{3}
$$

where d_i is the actual (metric) thickness, and $D_{\phi i}$ is the associated film thickness period,

$$
D_{\phi i} = \frac{\lambda}{2} \left(N_i^2 - N_0^2 \sin^2 \phi \right)^{-1/2}, \qquad i = 1, 2. \tag{4}
$$

 N_i is the refractive index of the *i*th medium ($i = 0,1,2,3$). The ambient and two films are transparent, so that N_0 , N_1 , and N_2 are real, whereas the substrate is in general absorbing with complex refractive index *N3* $= n_3 - j k_3.$

For a transparent substrate coated by two layers of quarterwave optical thicknesses $({\zeta_1} = {\zeta_2} = 1/2)$, antireflection of the ν polarization occurs when

$$
r_{01\nu} - r_{12\nu} + r_{23\nu} = r_{01\nu} r_{12\nu} r_{23\nu}, \tag{5}
$$

which is readily obtained by setting the numerator of Eq. (1) equal to zero and $X_1 = X_2 = -1$.

For the $\nu = s$ polarization, we substitute $r_{lms} = (S_l S_m$ / $(S_l + S_m)$ in Eq. (5) and obtain (after considerable simplification)

$$
S_0 S_2^2 = S_1^2 S_3,\tag{6}
$$

where
$$
\sum_{i=1}^{n} x_i
$$

$$
S_i = (N_i^2 - N_0^2 \sin^2 \phi)^{1/2}, \qquad i = 0, 1, 2, 3. \tag{7}
$$

Similarly, for the $\nu = p$ polarization, substitution of $r_{lmp} = (\dot{N}_m^2 S_l - N_l^2 S_m) / (\dot{N}_m^2 S_l + N_l^2 S_m)$ into Eq. (5) yields

$$
N_1^4 N_3^2 S_0 S_2^2 = N_0^2 N_2^4 S_1^2 S_3. \tag{8}
$$

It is convenient to define the relative refractive indices, $n_i = N_i/N_0$ (here $i = 1,2,3$), and introduce the variable $u = \sin^2 \phi$.

Concomitant antireflection of the p and s polarizations (hence total refraction) is possible when Eqs. (6) and (8) are satisfied simultaneously. When Eq. (6) is satisfied, Eq. (8) will also be satisfied if $N_1^4 N_3^2$ = $N_0^2 N_2^4$, or

$$
n_2^4 = n_1^4 n_3^2. \tag{9}
$$

Equation (6) [with S_i given by Eq. (7)] and Eq. (9) are sufficient to determine the required refractive indices n_1 and n_2 of the two films for given n_3 and ϕ . By manipulating these equations, we get the following solution:

$$
n_1^2 = \{(n_3 + u) + [(n_3 + u)^2 - (n_3 + 1)^2 u]^{1/2}\}/(n_3 + 1). \tag{10}
$$

After determining n_1 from Eq. (10), n_2 is subsequently obtained from Eq. (9).

It is instructive to consider some limit of Eq. (10). When $n_3 \gg 1$ (very-high-index substrate), we obtain

$$
n_1 = \sqrt{2} \cos(\phi/2), \tag{11}
$$

which is independent of n_3 . Thus the refractive index of the outer layer (next to air) is always $\langle \sqrt{2} = 1.414$. As $\phi \to 0$, $u \to 0$, and the normal-incidence limit of n_1^2 from Eq. (10) is

$$
n_1^2 = 2n_3/(n_3 + 1). \tag{12}
$$

As $\phi \rightarrow 90^{\circ}$, $u \rightarrow 1$, and the grazing-incidence limit of n_1^2 is found to be 1.

The complex amplitude-transmission coefficients of the bilayer of quarterwave films $(X_1 = X_2 = -1)$ for the *s* and p polarizations are given by⁶

$$
t_s = -2S_0S_1S_2/(S_0S_2^2 + S_1^2S_3),\tag{13}
$$

$$
t_p = -2n_1^2n_2^2n_3S_0S_1S_2/(n_1^4n_3^2S_0S_2^2 + n_2^4S_1^2S_3). \tag{14}
$$

From Eq. (9) we have $n_2^4 = n_1^4 n_3^2 = n_1^2 n_2^2 n_3$. Substitution of these relations into Eq. (14) and comparing the result with Eq. (13) lead to

$$
t_p = t_s. \tag{15}
$$

From Eq. (15) we reach the important conclusion that the state of polarization of the totally refracted light is identical to that of the incident light for all incident polarizations. In other words, the bilayer produces total refraction without change of polarization.

III. Transparent Bilayer on a Ge Substrate Producing Total Refraction of 1⁰ . 6-/im Radiation at 450 Angle of Incidence

Bilayer coatings that produce total refraction at oblique incidence are practical in the IR because highindex transparent substrates are available in this part of the spectrum. As an example, consider the reflection in air $(N_0 = 1)$ of 10.6- μ m CO₂-laser radiation by a Ge substrate with refractive index ⁷ 4 at **450** angle of incidence. To obtain the refractive indices of the two films of the TR bilayer, $n_3 = 4$ and $u = \frac{1}{2}$ are substituted in Eq. (10) to give

$$
n_1 = (0.9 + \sqrt{0.31})^{1/2} = 1.20697, \tag{16a}
$$

which is identical to the index obtained from Eq. (22) of Ref. 4. A thin-film coating material with this low refractive index at $\lambda = 10.6 \mu m$ is BaF₂, as has been reported by Kemeny.8 Equation (9) subsequently gives

$$
n_2 = 2.41394.\t(16b)
$$

A possible coating material with this index⁹ at 10.6 μ m is ZnSe. It should be remembered that the refractive indices of thin films significantly depend on conditions of their preparation which must then be controlled to thicknesses are equal to one-half of the corresponding thickness periods; from Eqs. (4) and (16), we obtain

$$
d_1 = 2.70920 \,\mu\text{m}, \qquad d_2 = 1.14815 \,\mu\text{m}. \tag{17}
$$

We now examine the behavior of this design as a function of angle of incidence ϕ and wavelength λ . Figure 1 shows the p and s intensity reflectances plotted vs ϕ in the range $0 \le \phi \le 50^{\circ}$ for the Ge substrate which is coated with the totally refracting bilayer whose properties are specified by Eqs. (16) and (17). The reflectance \mathcal{R}_{ν} for the ν polarization is determined by

$$
\mathcal{R}_{\nu} = |R_{\nu}|^2, \tag{18}
$$

where R_{ν} is given by Eq. (1). For $\phi \neq 45^{\circ}$, both layers are no longer of quarterwave optical thickness, and X_1 , $X_2 \neq -1$. For $\phi < 45^{\circ}$, \mathcal{R}_p and \mathcal{R}_s are $\lt 1.4 \times 10^{-3}$, which is the normal-incidence reflectance. Increasing ϕ from 45 to 50° increases \mathcal{R}_p from 0 to 8.5 \times 10⁻⁵ and

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Fig. 1. p and s intensity reflectances (on a logarithmic scale) plotted vs the angle of incidence ϕ (in degrees) for a bilayer of two films of refractive indices $n_1 = 1.20697$ and $n_2 = 2.41394$ and thicknesses $d_1 =$ 2.70920 μ m and d_2 = 1.14815 μ m, respectively, on a Ge substrate of refractive index $n_3 = 4$. A 10.6- μ m (CO₂-laser) beam is assumed to be incident from air $(N_0 = 1)$. At $\phi = 45^{\circ}$, $\mathcal{R}_p = \mathcal{R}_s = 0$, and total refraction takes place.

Fig. 2. p and s intensity reflectances (on a logarithmic scale) vs wavelength λ (in microns) at 45° angle of incidence for a bilayer of two films of refractive indices $n_1 = 1.20697$ and $n_2 = 2.41394$ and thicknesses $d_1 = 2.70920 \ \mu \text{m}$ and $d_2 = 1.14815 \ \mu \text{m}$, respectively, on a Ge substrate of refractive index $n_3 = 4$. At the design wavelength λ = 10.6 μ m, \mathcal{R}_p = \mathcal{R}_s = 0, and total refraction takes place from air into Ge. The effect of material dispersion is ignored.

 \mathcal{R}_s from 0 to 4.4 \times 10⁻⁴. This indicates that the condition of total refraction, which is satisfied exactly at $\phi = 45^{\circ}$, continues to hold with little error over a wide range of incidence angle, $0 \le \phi \le 50^{\circ}$.

Figure 2 shows the variation of \mathcal{R}_p and \mathcal{R}_s with wavelength λ from 8 to 12 μ m. In this calculation, the refractive indices of the two films and of the substrate are kept constant independent of λ (i.e., material dispersion is neglected). $\bar{R}_p = R_s = 0$ at the design wavelength of $10.6 \mu m$, and both reflectances rise steeply as λ shifts from this value. However, \mathcal{R}_p and \mathcal{R}_s remain below 1%, for $9 \le \lambda \le 12 \mu m$, indicating acceptable performance over an appreciable spectral band.

The sensitivity of the design to small changes (± 0.1) of the refractive index of each of films 1 and 2 is illustrated in Figs. 3 and 4, respectively. In Fig. $3 n_1$ is allowed to vary by ± 0.1 around the design index of 1.20697, although positive errors are more likely to take place in this case. In the presence of an error Δn_1 $= \pm 0.02$, the reflectances \mathcal{R}_p and \mathcal{R}_s remain $\leq 10^{-3}$.

Figure 4 indicates that the design is more tolerant to equal errors (± 0.1) of the higher refractive index n_2 of the inner film (that is first deposited on the Ge substrate). If n_2 is shifted by $\Delta n_2 = \pm 0.02$, we still have $\mathcal{R}_p, \mathcal{R}_s < 1.5 \times 10^{-4}$

Finally, Figs. 5 and 6 describe the sensitivity of the design to small errors ($\pm 0.01 \mu m = \pm 10 \text{ nm}$) of the thickness of films 1 and 2, respectively. Figure 5 indicates good tolerance to ± 10 -nm errors of thickness of the outer layer, because \mathcal{R}_p and \mathcal{R}_s remain $\leq 10^{-5}$. For equal errors of thickness of the inner high-index layer (film 2) Fig. 6 shows that \mathcal{R}_p , $\mathcal{R}_s < 1.5 \times 10^{-3}$.

To summarize this section, we have demonstrated that totally refracting (polarization-preserving) transparent bilayers are possible on high-index substrates (e.g., Ge) in the IR (e.g., at $\lambda = 10.6 \,\mu\text{m}$). Furthermore, the design is found to be reasonably tolerant to changes of incidence angle, wavelength, film refractive indices, and film thicknesses.

IV. Totally Refracting Bilayers on Absorbing Substrates

Intuitively, total refraction and absorption of obliquely incident monochromatic light should also be possible using a transparent bilayer on an absorbing substrate. However, in this case it is expected that the layers would no longer be of quarterwave optical thickness. For an absorbing substrate with complex refractive index $N_3 = n_3 - j k_3$, the analysis of Sec. II that begins with Eq. (5) is not applicable. We have adopted a numerical approach as follows. The reflectance of the bilayer-substrate structure for incident unpolarized or circularly polarized light,

$$
\mathcal{R}_u = \frac{1}{2} \left(\mathcal{R}_p + \mathcal{R}_s \right),\tag{19}
$$

is considered as a function of the system parameters:

$$
\mathcal{R}_u = f(n_1, \zeta_1, n_2, \zeta_2; n_3, k_3, \phi). \tag{20}
$$

For a given substrate at a given angle of incidence, n_3 , k_3 , and ϕ are fixed, and \mathcal{R}_u is a function of the refractive indices n_1, n_2 and normalized thicknesses ζ_1, ζ_2 of the bilayer. A computer program was developed that

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Fig. 3. p and s reflectances (on a logarithmic scale) vs the refractive-index n_1 of the first film with thickness $d_1 = 2.70920 \ \mu \text{m}$. The second film has $n_2 = 2.41394$ and $d_2 = 1.14815 \mu m$. Films 1 and 2 form a bilayer on a Ge substrate ($n_3 = 4$) on which 10.6- μ m radiation is incident from air $(N_0 = 1)$ at 45° angle of incidence. When $n_1 =$ 1.20697, $\mathcal{R}_p = \mathcal{R}_s = 0$, and total refraction occurs.

Fig.4. Same as in Fig.3 except that now the refractive index of film 2 is being shifted (by \pm 0.1) around the design value $n_2 = 2.41394$.

iterates on all four variables $n_1, n_2, \zeta_1, \zeta_2$ to minimize \mathcal{R}_u . Total refraction is considered accomplished when the minimum reflectance $\mathcal{R}_{\text{umin}}$ at the end of iteration is $<$ 10^{-12}.

As an example, we considered a substrate with complex refractive index $N_3 = 4 - jk_3$, where the extinction coefficient k_3 is allowed to increase from 0 to 5, and an angle of incidence of 45°. Figure 7 shows the (numeri-

Fig. 5. p and s intensity reflectances (on a logarithmic scale) vs the thickness d_1 of film 1 of a bilayer $(n_1 = 1.20697, n_2 = 2.41394, d_2 =$ 1.4815 μ m) on a Ge substrate ($n_3' = 4$) on which 10.6- μ m radiation is incident from air $(N_0 = 1)$ at 45° angle of incidence. When $d_1 =$ 2.70920 μ m, $\mathcal{R}_p = \mathcal{R}_s = 0$, and total refraction takes place.

Fig. 6. Same as in Fig. 5 except that the thickness of film 2 is now being shifted (by $\pm 0.01 \mu$ m) around the design value $d_2 = 1.14815 \mu$ m.

cally determined) refractive indices n_1 and n_2 of the bilayer plotted against k_3 . As k_3 increases from 0 to 5, *n1* increases slowly and monotonically from 1.20697 to 1.27259, whereas n_2 increases appreciably and monotonically from 2.41394 to 4.83923.

Figure 8 shows the corresponding variation of the normalized thicknesses ζ_1 and ζ_2 of the two films of the bilayer with k_3 . Both ζ_1 and ζ_2 decrease monotonically

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Fig. 7. Refractive indices n_1 and n_2 of the two films of a totally refracting bilayer on an absorbing substrate with complex refractive index $N_3 = 4 - jk_3$ plotted vs the extinction coefficient k_3 . Light is assumed to be incident from air $(N_0 = 1)$ at 45° angle of incidence.

Fig. 8. Normalized thicknesses ζ_1 and ζ_2 of the two films of a totally refracting bilayer (with refractive indices given by Fig. 7) on an absorbing substrate with complex refractive index $N_3 = 4 - jk_3$ plotted vs the extinction coefficient *k3 .* Light is assumed to be incident from air $(N_0 = 1)$ at 45° angle of incidence.

as k_3 increases from the common value of 0.5 at $k_3 = 0$ to 0.483405 and 0.302413, respectively. Thus the deviation from the quarterwave optical thickness increases as the substrate absorption increases and is much more pronounced for the inner film (film 2) than it is for the outer film (film 1).

Results similar to those shown in Figs. 7 and 8 at other angles of incidence were obtained but are not included here. However, for further illustration, Table I provides numerical data for totally refracting bilayers on a Ti substrate with complex refractive in- $\text{dex}^{10}N_3 = 4-j2$ for 10.6- μ m CO₂-laser radiation at three angles of incidence $\phi = 30, 45,$ and 60°. Table I lists the bilayer refractive indices (n_1, n_2) and the normalized (ζ_1, ζ_2) and actual (d_1, d_2) film thicknesses. In all cases the computed minimum reflectance for incident unpolarized light of the bilayer-substrate system is well below 10^{-12} .

Table II gives the characteristics of totally refracting transparent bilayers on a Si substrate at **450** angle of incidence for five lines of the A_r ⁺ laser and at the 632.8nm wavelength of the He-Ne laser. Listed in that table are the complex refractive index N_3 of Si, obtained from Aspnes and Studna¹¹ by interpolation, the calculated refractive indices n_1 and n_2 , the normalized thicknesses ζ_1 and ζ_2 , and actual thicknesses d_1 and d_2 of the two films of the bilayer. For all cases shown $\mathcal{R}_{\textit{umin}}$ < 10^{-12} is achieved. The refractive index n_1 of film 1 is realizable by low-index fluoride coating materials¹² such as CaF₂, whereas n_2 of film 2 can be obtained by controlling the evaporation conditions of TiO₂ as indicated by Pulker *et al.*¹³

The substrate complex refractive index and angle of incidence determine whether totally refracting bilayers are possible. For certain substrates, we have found that $\mathcal{R}_{\text{umin}}$ cannot be reduced to zero for realistic film refractive indices. For example, for an Al substrate with $N_3 = 1.212 - 6.924$ at $\lambda = 632.8$ mn, and with the search range of n_1 and n_2 limited to ≤ 7 , the lowest value of \mathcal{R}_u at $\phi = 45^\circ$ is 3.13% and is attained when n_1 $= 7, n_2 = 3.76422, \zeta_1 = 0.49184, \text{ and } \zeta_2 = 0.85006.$ This solution, of course, is unacceptable, and three or more layers may be needed for total refraction at oblique incidence.

V. Summary

Practical totally refracting transparent bilayers on transparent and absorbing substrates for monochromatic light at oblique incidence have been considered. When the substrate is transparent, the totally refract-

Table I. CharacterIstics of Totally Refracting Transparent Bilayer Coatings on a TI Substrate (N3 = 4-12) for 10.6-μm CO₂-Laser Radiation at Three Angles of Incidence^a

ϕ (deg)	n,	$\scriptstyle{n_2}$	51	52	$d_1 \,(\mu \text{m})$	$d_2(\mu m)$
30	1.27008 1.23052	3.02252 2.91216	0.48388 0.48552	0.36073 0.36981	2.19661 2.55520	0.64138 0.69380
45 60	1.17336	2.75678	0.48792	0.38174	3.26638	0.77304

 a ϕ is the angle of incidence. n_1 and n_2 are the refractive indices of films 1 and 2 of the bilayer, whereas ζ_1 , ζ_2 and d_1 , d_2 are the corresponding normalized and actual film thicknesses, respectively.

Table II. Characteristics of Totally Refracting Transparent Bilayer Coatings on a Si Substrate at 45° Angle of Incidence for Five Ar+-Laser Lines and the 632.8-nm He-Ne-Laser Linea

N_3						
(λ)	n_{1}	n ₂	ζ_1	ζ_2	d_1 (nm)	d_2 (nm)
$5.063 - j3.218$ (325 nm)	1.25157	3.54333	0.48759	0.37408	76.73	17.51
$4.775 - j0.167$ (442 nm)	1.22145	2.67145	0.49894	0.49061	110.71	42.09
$4.639 - j0.141$ (454.5 nm)	1.21921	2.62776	0.49905	0.49163	114.18	44.14
$4.375 - j0.080$ (488 nm)	1.21452	2.54098	0.49939	0.49469	123.40	49.46
$4.168 - i0.053$ (528.7 nm)	1.21050	2.47163	0.49955	0.49614	134.41	55.38
$3.884 - i0.020$ (632.8 nm)	1.20439	2.37365	0.49980	0.49835	162.20	69.59

 a N_3 is the complex refractive index of Si at wavelength λ obtained by interpolation of the data of Ref. 11. n_1 and n_2 are the refractive indices of films 1 and 2 of the bilayer, whereas ζ_1 , ζ_2 and d_1 , d_2 are the corresponding normalized and actual film thicknesses, respectively.

ed wave has the same state of polarization as the incident wave. In this case, the substrate refractive index must be sufficiently high to raise the refractive index of the low-index outer layer to a value that can be realized by a thin solid film. This is illustrated by a Ge substrate with a refractive index of 4 for 10.6- μ m radiation at 45° angle of incidence. In this case a BaF₂-ZnSe bilayer can produce total refraction. The behavior of this bilayer-substrate system as a function of the various design parameters (angle of incidence, wavelength, refractive indices, and thicknesses of the two films) is also studied. The solution to the problem of total refraction by a transparent bilayer is analytically obtained when the substrate is transparent. However, when the substrate is absorbing, the problem is more complicated, and a numerical approach is used. Total refraction is considered attained when the reflectance larized light is reduced to $\langle 10^{-12}$. This was demonstrated for the case of an absorbing substrate with complex refractive index $N_3 = 4 - jk_3$, where the extinction coefficient k_3 is allowed to vary from 0 to 5 to represent a range of semiconductors and metals. The normalized thicknesses of the two films of the bilayer differ from 0.5. Hence the films are not of quarterwave of optical thickness, a condition that holds only when the substrate is transparent $(k_3 = 0)$. Specific examples for which results appear in tables include a Ti substrate 10.6- μ m (CO₂-laser) wavelength at three angles of incidence and a Si substrate at several $(A_r^+$ and He-Ne-laser) wavelengths at **450** angle of incidence.

At the present time, R. M. A. Azzam is at the Département de Physique du Solide, Universite de Provence, Marseille, France on a sabbatical leave as a Fulbright Senior Research Scholar and Professeur Associ6.

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