# Reflection and transmission ellipsometry of a uniform layer 

Michael C. Dorf and John Lekner<br>Department of Physics, Victoria University of Wellington, Private Bag, Wellington, New Zealand

Received February 18, 1987; accepted June 3, 1987


#### Abstract

We examine the general properties of the ellipsometric quantities $\rho=r_{p} / r_{s}$ and $\tau=t_{p} / t_{s}$ for a nonabsorbing uniform film. At fixed angle of incidence and for variable thickness, $\tau$ moves periodically along circles (as is known) and $\rho$ along quartics in the complex plane. For the special case of a film between like media we derive envelopes (and corresponding bounds on $\tau$ and $\rho$ ) as functions of the dielectric constants of the layer and its bounding medium. In the case of unlike media on either side of the film, bounds are found for $\operatorname{Re} \tau$ and $\operatorname{Im} \tau$; for $\epsilon^{2}>\epsilon_{1} \epsilon_{2}$ (where $\epsilon_{1}$, $\epsilon$, and $\epsilon_{2}$ are, respectively, the dielectric constants of the medium from which light is incident, of the film, and of the substrate), we find an approximate upper bound for the ellipsometric quantity $\bar{\rho}$ (defined as $\operatorname{Im} \rho$ when $\operatorname{Re} \rho=0$ ).


## 1. INTRODUCTION

Consider a uniform nonabsorbing layer of thickness $\Delta z$ and dielectric constant $\epsilon$, bounded by uniform media of dielectric constants $\epsilon_{1}=n_{1}{ }^{2}$ and $\epsilon_{2}=n_{2}{ }^{2}$. In what follows we take light to be incident from medium 1 and partially transmitted into medium 2. The optical properties of this system have been well studied, ${ }^{1-4}$ but some properties of interest have not been determined. Here we focus on the ellipsometric quantities $\rho$ $=r_{p} / r_{s}$ and $\tau=t_{p} / t_{s}$, where $r$ and $t$ are the reflection and transmission amplitudes and the subscripts $s$ and $p$ refer to TE and TM polarizations, respectively. By examining the behavior of $\tau$ and $\rho$ for a fixed angle of incidence we are able to place restrictions on the values of $\tau$ and $\rho$ as complex functions of $\epsilon_{1}, \epsilon$, and $\epsilon_{2}$. These restrictions will be of use in making and interpreting ellipsometric measurements.

In the notation of a recent book on reflection ${ }^{4}$ (cited here as TR), the ellipsometric ratios are given, for example, from Azzam and Bashara, ${ }^{3}$ Sec. 4.4, or from TR, Sec. 2-4, by

$$
\begin{align*}
& \rho=\frac{p+p^{\prime} Z}{1+p p^{\prime} Z} \frac{1+s s^{\prime} Z}{s+s^{\prime} Z}  \tag{1}\\
& \tau=\frac{n_{1}}{n_{2}} \frac{(1-p)\left(1-p^{\prime}\right)}{(1+s)\left(1+s^{\prime}\right)} \frac{1+s s^{\prime} Z}{1+p p^{\prime} Z} \tag{2}
\end{align*}
$$

where
$s=\frac{q_{1}-q}{q_{1}+q}, \quad s^{\prime}=\frac{q-q_{2}}{q+q_{2}}, \quad p=\frac{Q-Q_{1}}{Q+Q_{1}}, \quad p^{\prime}=\frac{Q_{2}-Q}{Q_{2}+Q}$
are the reflection amplitudes of the $s$ and $p$ polarizations at the first and second interfaces and

$$
\begin{equation*}
Z=\exp (2 i q \Delta z) \tag{4}
\end{equation*}
$$

The $q$ 's are the normal components of the wave vector in the three media, and $Q_{1}=q_{1} / \epsilon_{1}, Q=q / \epsilon, Q_{2}=q_{2} / \epsilon_{2}$.

Some general properties of $\rho$ and $\tau$ follow immediately from Eqs. (1) and (2). When there is no absorption within the layer and $\sin ^{2} \theta_{1}<\epsilon / \epsilon_{1}, q$ is real and $Z$ moves along the unit circle in the complex plane. The motion is periodic in $\Delta z$, with period $\pi / q ; \rho$ and $\tau$ correspondingly move on closed curves in the complex plane. At normal incidence $p=s$ and $p^{\prime}=s^{\prime}$, so $\rho=1$ and $\tau=1$.

At grazing incidence $q_{1}=n_{1}(\omega / c) \cos \theta_{1}$ tends to zero, and $s \rightarrow-1, p \rightarrow 1$; thus $\rho \rightarrow-1$, in accord with a general result proved in TR, Sec. 2-3. To obtain the limiting value of $\tau$ at grazing incidence, let $\gamma=\pi / 2-\theta_{1}$ be the grazing angle. Then, if $\epsilon \neq \epsilon_{1}$ and $\epsilon_{2} \neq \epsilon_{1}$,

$$
\begin{align*}
q_{1} c / \omega= & n_{1} \cos \theta_{1}=n_{1} \sin \gamma=n_{1} \gamma+O\left(\gamma^{3}\right), \\
q c / \omega= & \left(\epsilon-\epsilon_{1} \sin ^{2} \theta_{1}\right)^{1 / 2}=\left(\epsilon-\epsilon_{1}\right)^{1 / 2} \\
& \times\left[1+\gamma^{2} \epsilon_{1} / 2\left(\epsilon-\epsilon_{1}\right)\right]+O\left(\gamma^{4}\right) \\
q_{2} c / \omega= & \left(\epsilon_{2}-\epsilon_{1} \sin ^{2} \theta_{1}\right)^{1 / 2}=\left(\epsilon_{2}-\epsilon_{1}\right)^{1 / 2} \\
& \times\left[1+\gamma^{2} \epsilon_{1} / 2\left(\epsilon_{2}-\epsilon_{1}\right)\right]+O\left(\gamma^{4}\right) \tag{5}
\end{align*}
$$

It follows that at grazing incidence

$$
\begin{align*}
& s=-1+\frac{2 n_{1} \gamma}{\mu}+O\left(\gamma^{2}\right), \quad p=1-\frac{2 \epsilon \gamma}{n_{1} \mu}+O\left(\gamma^{2}\right)  \tag{6}\\
& s^{\prime}=\frac{\mu-\mu_{2}}{\mu+\mu_{2}}+O\left(\gamma^{2}\right), \quad p^{\prime}=\frac{\mu_{2} / \epsilon_{2}-\mu / \epsilon}{\mu_{2} / \epsilon_{2}+\mu / \epsilon}+O\left(\gamma^{2}\right) \tag{7}
\end{align*}
$$

where $\mu=\left(\epsilon-\epsilon_{1}\right)^{1 / 2}$ and $\mu_{2}=\left(\epsilon_{2}-\epsilon_{1}\right)^{1 / 2}$. The consequent limiting value of $\tau$ is

$$
\begin{equation*}
\tau \rightarrow \frac{\epsilon}{n_{1} n_{2}} \frac{1-p^{\prime}}{1+s^{\prime}} \frac{1-s^{\prime} Z}{1+p^{\prime} Z} \tag{8}
\end{equation*}
$$

in which $s^{\prime}$ and $p^{\prime}$ stand for the leading terms in Eqs. (7). This limiting value depends on $\Delta z$ (however, $\tau$ tends to a value independent of $\Delta z$, namely, $\epsilon / \epsilon_{1}$, in the special case $\epsilon_{1}=$ $\epsilon_{2}$; this case is discussed in Section 2).

## 2. UNIFORM LAYER BETWEEN LIKE MEDIA

We consider first the special case where $\epsilon_{1}=\epsilon_{2}$, for example, a soap film in air or a layer of dielectriclike glass immersed in a fluid dielectric. Then $q_{2}=q_{1}$ and $s^{\prime}=-s, p^{\prime}=-p$. The ellipsometric ratios reduce to

$$
\begin{equation*}
\rho=\frac{p}{s} \frac{1-S Z}{1-P Z}, \quad \tau=\frac{1-P}{1-S} \frac{1-S Z}{1-P Z} \tag{9}
\end{equation*}
$$

where

$$
\begin{equation*}
S=s^{2}, \quad P=p^{2} \tag{10}
\end{equation*}
$$

Equations (9) are both bilinear (or fractional) transformations. Such conformal mappings have the property that circles in the $Z$ plane transform to circles in the complex plane of the new variable. We can see this, and at the same time obtain the center and radius of the transformed circle, as follows. Both $\rho$ and $\tau$ have the form

$$
\begin{equation*}
w=f \frac{1-S Z}{1-P Z} . \tag{11}
\end{equation*}
$$

Thus

$$
\begin{equation*}
(w P-f S) Z=w-f \tag{12}
\end{equation*}
$$

On multiplying Eq. (12) by its complex conjugate and using $Z Z^{*}=1$, we find that

$$
\begin{equation*}
\left(1-P^{2}\right) w w^{*}-f(1-S P)\left(w+w^{*}\right)+f^{2}\left(1-S^{2}\right)=0 \tag{13}
\end{equation*}
$$

(We have assumed that $\epsilon \epsilon_{1}$, so that $s, p$, and all coefficients derived from them are real at all angles.) This is the equation of a circle: setting $w=x+i y$, we can write it in the form

$$
\begin{equation*}
\left(x-x_{0}\right)^{2}+y^{2}=r^{2}, \tag{14}
\end{equation*}
$$

where

$$
\begin{equation*}
x_{0}=f \frac{1-S P}{1-P^{2}}, \quad r=f \frac{S-P}{1-P^{2}} \tag{15}
\end{equation*}
$$

These results may be obtained more directly by using the fact that $w$ moves on a circle whose center lies on the real axis. Setting $Z= \pm 1$, we obtain the points $w_{ \pm}$on the real axis of the $w$ plane. Then $x_{0}=\left(w_{-}+w_{+}\right) / 2$ and $r=$ $\left(w_{-}-w_{+}\right) / 2$.

For the transmission ellipsometric ratio, $f=(1-P) /$ ( $1-S$ ) and

$$
\begin{equation*}
x_{\tau}=\frac{1-S P}{(1-S)(1+P)}, \quad r_{\tau}=\frac{S-P}{(1-S)(1+P)} \tag{16}
\end{equation*}
$$

We note that for all circles in the $\tau$ plane

$$
\begin{equation*}
x_{\tau}-r_{\tau}=1 \tag{17}
\end{equation*}
$$

At normal incidence the center is at $(1,0)$ and the radius tends to zero. At grazing incidence we take the limit as $\gamma \rightarrow 0$ in Eqs. (16), using Eqs. (6), and find that $x_{\tau}$ tends to $\left(\epsilon+\epsilon_{1}\right) / 2 \epsilon_{1}$ and $r_{\tau}$ tends to $\left(\epsilon-\epsilon_{1}\right) / 2 \epsilon_{1}$. Thus the loci of $\tau$ are nested circles bounded by the largest, for which

$$
\left(x-\frac{\epsilon+\epsilon_{1}}{2 \epsilon_{1}}\right)^{2}+y^{2}=\left(\frac{\epsilon-\epsilon_{1}}{2 \epsilon_{1}}\right)^{2},
$$

as shown in Fig. 1.
For the reflection ellipsometric ratio $\rho=r_{p} / r_{s}, f=p / s$, and

$$
\begin{equation*}
x_{\rho}=\frac{p}{s} \frac{1-S P}{1-P^{2}}, \quad r_{\rho}=\frac{p}{s} \frac{S-P}{1-P^{2}} . \tag{18}
\end{equation*}
$$

[For determination of the envelope, discussed below, it is convenient to treat the radius as an algebraic quantity, allowing it to change sign with $p$ at $\theta_{B}=\arctan \left(\epsilon / \epsilon_{1}\right)^{1 / 2}$. A discontinuity in the slope of $r_{\rho}$ is thus avoided.] At normal incidence the center lies at $(1,0)$ and the radius tends to zero. At grazing incidence $x_{\rho}$ tends to $-\left(\epsilon+\epsilon_{1}\right) / 2 \epsilon$ and $r_{\rho}$ to $\left(\epsilon-\epsilon_{1}\right) /$ $2 \epsilon$. The circle approached at grazing incidence intersects the point $(-1,0)$. Note that both the center and the radius are zero at $\theta_{B}=\arctan \left(\epsilon / \epsilon_{1}\right)^{1 / 2}$, the Brewster angle for a


Fig. 1. Paths of $\tau=x+i y$ in the complex plane for fixed angles of incidence and variable thickness of a layer between like media. The largest circle is approached at grazing incidence. Thus $\operatorname{Re} \tau$ must lie between unity and $\epsilon / \epsilon_{1}$, while $|\operatorname{Im} \tau|$ cannot exceed $\left(\epsilon-\epsilon_{1}\right) / 2 \epsilon_{1}$. The loci are drawn for $\epsilon_{1}=1$ and $\epsilon=(3 / 2)^{2}$, representing glass in air.
boundary between semi-infinite media with dielectric constants $\epsilon_{1}$ and $\epsilon$.

The envelope of a set of curves $\phi(x, y, \alpha)=0$ for variable parameter $\alpha$ is given by the simultaneous solution of $\phi=0$ and $\partial \phi / \partial \alpha=0$ (see, for example, Ref. 5, articles 139 and 140). Here $\alpha$ can be the angle of incidence $\theta_{1}$ or some function of $\theta_{1}$ that increases or decreases monotonically, such as $s$. For a family of circles with center $x_{0}$ and radius $r$,

$$
\begin{align*}
\phi & =\left(x-x_{0}\right)^{2}+y^{2}-r^{2}  \tag{19}\\
\frac{\partial \phi}{\partial \alpha} & =-2\left(x-x_{0}\right) \frac{\mathrm{d} x_{0}}{\mathrm{~d} \alpha}-2 r \frac{\mathrm{~d} r}{\mathrm{~d} \alpha} \tag{20}
\end{align*}
$$

Setting $\phi$ and $\partial \phi / \partial \alpha$ equal to zero, we obtain the parametric equations of the envelope:

$$
\begin{equation*}
x=x_{0}-r \frac{\mathrm{~d} r}{\mathrm{~d} x_{0}}, \quad y= \pm r\left[1-\left(\frac{\mathrm{d} r}{\mathrm{~d} x_{0}}\right)^{2}\right]^{1 / 2} \tag{21}
\end{equation*}
$$

For given dielectric constants $\epsilon_{1}$ and $\epsilon, x_{0}$ and $r$ are specified by the angle of incidence $\theta_{1}$. Since the center and the radius are specified in Eqs. (18) in terms of the reflection amplitudes $s$ and $p$, it is convenient to use one of these as the parameter. The other amplitude can be eliminated by the use of Eqs. (3), which implies that

$$
\begin{equation*}
\frac{1+s}{1-s}=\frac{q_{1}}{q}, \quad \frac{1+p}{1-p}=\frac{Q}{Q_{1}}=\frac{\epsilon_{1}}{\epsilon} \frac{q}{q_{1}} . \tag{22}
\end{equation*}
$$

These relations, and those consequent, are due to Azzam. ${ }^{6}$ The apparent difference between these and Azzam's is due to the use of different conventions: Azzam has $p=-s$ at normal incidence; we have $p=s$. (The relationship between $s$ and $p$ and the electric- and magnetic-field components is discussed in TR, p. 7.) From Eqs. (22) we obtain the identity

$$
\begin{equation*}
\frac{1+p}{1-p}=\frac{\epsilon_{1}}{\epsilon} \frac{1-s}{1+s} \tag{23}
\end{equation*}
$$

which gives $p$ in terms of $s$ or vice versa:


Fig. 2. Paths of $\rho$ in the complex plane for fixed angles of incidence and variable thickness of a layer between like media. The paths are circles, distorted to ellipses by enlargement of the vertical scale. The dashed curve is the envelope. The values of $\epsilon_{1}$ and $\epsilon$ are for air and glass, as in Fig. 1.

$$
\begin{equation*}
p=\frac{\epsilon_{1}-\epsilon-\left(\epsilon_{1}+\epsilon\right) s}{\epsilon_{1}+\epsilon-\left(\epsilon_{1}-\epsilon\right) s}, \quad s=\frac{\epsilon_{1}-\epsilon-\left(\epsilon_{1}+\epsilon\right) p}{\epsilon_{1}+\epsilon-\left(\epsilon_{1}-\epsilon\right) p} . \tag{24}
\end{equation*}
$$

The two reflection amplitudes are symmetrically related by a bilinear transformation. If we denote the transformation by $T$, we see that $T[T(z)]=z$ for all complex values of $z$. In general, a transformation will have this property if there exists a relation between $z$ and $T(z)$ that is symmetric with respect to the interchange of $z$ and $T(z)$. In the case of the reflection amplitudes, Eq. (23) is such an expression.

The derivative of $p$ with respect to $s$ is

$$
\begin{equation*}
\frac{\mathrm{d} p}{\mathrm{~d} s}=\frac{-4 \epsilon_{1} \epsilon}{\left[\epsilon_{1}+\epsilon-\left(\epsilon_{1}-\epsilon\right) s\right]^{2}} \tag{25}
\end{equation*}
$$

and, from Eqs. (18),
$\frac{\mathrm{d} r}{\mathrm{~d} x_{0}}$

$$
\begin{equation*}
=\frac{\frac{s}{p} \frac{\mathrm{~d} p}{\mathrm{~d} s}\left[S-3 P+P^{2}(3 S-P)\right]+\left(1-P^{2}\right)(S+P)}{\frac{s}{p} \frac{\mathrm{~d} p}{\mathrm{~d} s}\left[1+P\left(3 P-3 S-P^{2} S\right)\right]-\left(1-P^{2}\right)(1+S P)} . \tag{26}
\end{equation*}
$$

The envelope of $\rho=x+i y$ is the concatenation of two curves. Equations (21), (25), and (26) define the envelope from $x=1$ until this curve is tangent to the circle for grazing incidence. This circle then continues the envelope, which is shown in Fig. 2.

## 3. UNIFORM LAYER BETWEEN UNLIKE MEDIA

We now consider the case in which $\epsilon_{1} \neq \epsilon_{2}$, starting with the transmission ellipsometric ratio, which is simpler. Equation (2) is a bilinear transformation of $Z=\exp (2 i q \Delta z)$, which for fixed angle of incidence and variable thickness moves on the unit circle; thus $\tau$ moves on a circle in the complex plane, as noted by Azzam. ${ }^{7}$ From Eq. (2) and the analogs of Eqs. (11) and (15), the center and the radius of the circle in the $\tau$ plane are

$$
\begin{equation*}
x_{0}=f \frac{1-s s^{\prime} p p^{\prime}}{1-P P^{\prime}}, \quad r=f \frac{p p^{\prime}-s s^{\prime}}{1-P P^{\prime}}, \tag{27}
\end{equation*}
$$

where

$$
\begin{equation*}
f=\frac{n_{1}}{n_{2}} \frac{(1-p)\left(1-p^{\prime}\right)}{(1+s)\left(1+s^{\prime}\right)} \tag{28}
\end{equation*}
$$

At normal incidence, $f \rightarrow 1, x_{0} \rightarrow 1$, and $r \rightarrow 0$. At grazing incidence,

$$
\begin{equation*}
f \rightarrow \frac{\epsilon}{n_{1} n_{2}} \frac{1-p^{\prime}}{1+s^{\prime}}, \quad x_{0} \rightarrow f \frac{1+s^{\prime} p^{\prime}}{1-P^{\prime}}, \quad r \rightarrow f \frac{p^{\prime}+s^{\prime}}{1-P^{\prime}} \tag{29}
\end{equation*}
$$

where $s^{\prime}$ and $p^{\prime}$ are given by the leading terms in Eqs. (7). The limit point (largest value of $\operatorname{Re} \tau$ ) attained at grazing incidence is $x_{0}+r$. When $\epsilon>\epsilon_{2}$, it follows from Eqs. (7) that $s^{\prime}+p^{\prime}>0$ at grazing incidence, so $x_{0}+r \rightarrow \epsilon / n_{1} n_{2}$. (In the case of like media, this gives $\epsilon / \epsilon_{1}$, as before.) When $\epsilon<\epsilon_{2}$, $s^{\prime}+p^{\prime}<0$ and $x_{0}+r \rightarrow n_{2} / n_{1}$ at grazing incidence. Figure 3 shows $\tau$ for a layer of water on glass.

The reflection ellipsometric ratio, given by Eq. (1), no longer has circles as the loci for fixed angle of incidence when $\epsilon_{1} \neq \epsilon_{2}$. The paths are quartic curves, as we shall now show. Equation (1) is a quadratic in $Z$, which we write as

$$
\begin{equation*}
\alpha Z^{2}+\beta Z+\gamma=0, \tag{30}
\end{equation*}
$$

where the coefficients $\alpha, \beta$, and $\gamma$ all depend linearly on $\rho=$ $r_{p} / r_{s}$ :

$$
\begin{gather*}
\alpha=s^{\prime} p^{\prime}(\rho p-s), \quad \beta=\rho\left(s^{\prime}+s p p^{\prime}\right)-p^{\prime}-p s s^{\prime} \\
\gamma=\rho s-p . \tag{31}
\end{gather*}
$$

To find the loci at fixed angle of incidence, we eliminate $Z$ from Eq. (30) and the complex conjugate of Eq. (30). Multiplying Eq. (30) by $Z^{*}$ and the conjugate of Eq. (30) by $Z$ and using $Z Z^{*}=1$, we obtain

$$
\begin{equation*}
\alpha Z+\beta+\gamma Z^{*}=0, \quad \alpha^{*} Z^{*}+\beta^{*}+\gamma^{*} Z=0 \tag{32}
\end{equation*}
$$

Thus

$$
\begin{equation*}
Z=\frac{\beta^{*} \gamma-\alpha^{*} \beta}{|\alpha|^{2}-|\gamma|^{2}} \tag{33}
\end{equation*}
$$

and again using $Z Z^{*}=1$ gives


Fig. 3. Paths of $\tau$ in the complex plane for fixed angles of incidence and variable film thickness. The refractive indices used are $n_{1}=1$, $n=4 / 3, n_{2}=3 / 2$.


Fig. 4. Paths of $\rho$ in the complex plane for fixed angles of incidence and variable film thickness. The curves are quartics, which are very nearly circles, as illustrated in Fig. 5. The dielectric constants are as in Fig. 3.


Fig. 5. The $60^{\circ}$ quartic of Fig. 4 (solid curve) compared with a circle passing through the same points on the real axis (dashed curve).

$$
\begin{equation*}
\left(|\alpha|^{2}-|\gamma|^{2}\right)^{2}-|\beta|^{2}\left(|\alpha|^{2}+|\gamma|^{2}\right)+2 \operatorname{Re}\left(\alpha^{*} \beta^{2} \gamma^{*}\right)=0 \tag{34}
\end{equation*}
$$

Since $\alpha, \beta$, and $\gamma$ are linear in $\rho=x+i y$, this is a quartic in $x$ and $y$, of the form

$$
\begin{equation*}
a\left(x^{2}+y^{2}\right)^{2}+2 b\left(x^{2}+y^{2}\right) x+c\left[\left(x-x_{0}\right)^{2}+y^{2}\right]=c r^{2} \tag{35}
\end{equation*}
$$

The coefficients in Eq. (35) are determined by Eqs. (31) and (34); they depend in a complicated manner on $\epsilon / \epsilon_{1}, \epsilon_{2} / \epsilon_{1}$, and $\theta_{1}$. Because we make no use of the general coefficients, we omit them. In the degenerate case where $\epsilon_{1}=\epsilon_{2}, a$ and $b$ are zero, and Eq. (35) reduces to the equation of a circle with center $x_{\rho}$ and radius $r_{\rho}$ given by Eqs. (18). Since Eq. (35) contains no terms of odd power in $y$, the quartic curves have reflection symmetry about the $x$ axis.

When $Z= \pm 1$ ( $2 q \Delta z$ respectively an even or an odd multiple of $\pi$ ), $\rho$ is real if the reflection amplitudes $s, s^{\prime}, p$, and $p^{\prime}$ are real. (This holds in the absence of absorption within the layer and the substrate for $\epsilon$ and $\epsilon_{2}$ greater than $\epsilon_{1} \sin ^{2} \theta_{1}$.) We call $\rho_{ \pm}$the values of $\rho$ corresponding to $Z= \pm 1$ :

$$
\begin{equation*}
\rho_{ \pm}=\frac{p \pm p^{\prime}}{1 \pm p p^{\prime}} \frac{1 \pm s s^{\prime}}{s \pm s^{\prime}} \tag{36}
\end{equation*}
$$

In terms of these we can define a generalized center $x_{0}$ and radius $r$ by

$$
\begin{equation*}
x_{0}=\left(\rho_{-}+\rho_{+}\right) / 2, \quad r=\left(\rho_{-}-\rho_{+}\right) / 2 \tag{37}
\end{equation*}
$$

These are

$$
\begin{align*}
x_{0} & =\frac{p s\left(1-S^{\prime}\right)\left(1-P^{\prime}\right)-p^{\prime} s^{\prime}(1-S)(1-P)}{\left(1-P P^{\prime}\right)\left(S-S^{\prime}\right)}  \tag{38}\\
r & =\frac{p s^{\prime}(1-S)\left(1-P^{\prime}\right)-p^{\prime} s\left(1-S^{\prime}\right)(1-P)}{\left(1-P P^{\prime}\right)\left(S-S^{\prime}\right)} \tag{39}
\end{align*}
$$

Note that the denominator common to Eqs. (38) and (39) goes to zero when $S^{\prime}=S\left(s^{\prime}= \pm s\right)$. The condition $s^{\prime}=-s$ can be satisfied only if $\epsilon_{1}=\epsilon_{2}$; in this case there is no divergence, as Eqs. (38) and (39) reduce to Eqs. (18). The other possibility, $s^{\prime}=s$, is satisfied when $q^{2}=q_{1} q_{2}$. This is one of the conditions for zero reflection of the $s$ polarization. At normal incidence it holds if $\epsilon^{2}=\epsilon_{1} \epsilon_{2}$. At oblique incidence it can be satisfied only if $\epsilon^{2}<\epsilon_{1} \epsilon_{2}$, and then it holds at the angle of incidence (see TR, p. 46)

$$
\begin{equation*}
\theta_{1}=\arcsin \left[\frac{\epsilon_{1} \epsilon_{2}-\epsilon^{2}}{\epsilon_{1}\left(\epsilon_{1}+\epsilon_{2}-2 \epsilon\right)}\right]^{1 / 2} \tag{40}
\end{equation*}
$$

Thus, when $\epsilon^{2}<\epsilon_{1} \epsilon_{2}$, no bounds (independent of angle of incidence and layer thickness) can be put on $\rho=r_{p} / r_{s}$. On the other hand, when $\epsilon^{2}>\epsilon_{1} \epsilon_{2}$, as for a layer of water on glass (illustrated in Fig. 4), the trajectories of $\rho$ are contained in a bounded region.
As illustrated in Fig. 5, the quartics are often closely approximated by circles. From the equation of these circles we deduce some approximate properties of the $\rho$ curve. The circles are determined by the two points $\rho_{ \pm}$corresponding to $Z= \pm 1$, as given in Eq. (26). As $Z=\exp (2 i q \Delta z)$ moves on the unit circle, the circle in the $\rho$ plane that passes through the points $\rho_{ \pm}$at $Z= \pm 1$ is

$$
\begin{equation*}
\rho=1 / 2\left(\rho_{+}+\rho_{-}\right)+1 / 2\left(\rho_{+}-\rho_{-}\right) Z . \tag{41}
\end{equation*}
$$

We write this as $\rho=x_{0}-r Z$, with center $x_{0}$ and radius $r$ given by Eqs. (38) and (39).
Polarization modulation ellipsometry ${ }^{8}$ has particular in-


Fig. 6. Maximum value of $\bar{\rho}$, the value of $\operatorname{Im} \rho$ when $\operatorname{Re} \rho=0$. The curve results from approximating the quartic loci of $\rho=r_{p} / r_{s}$ by circles. The points are exact values of $\bar{\rho}_{\max }$. The figure is drawn for $\epsilon_{1}=1$ and $\epsilon_{2}=(3 / 2)^{2}$.
terest in the angle at which $\operatorname{Re} \rho=0$ (the principal angle) and in $\bar{\rho}$, the value of $\operatorname{Im} \rho$ at this angle. When the paths of $\rho$ are approximated by circles, the envelope of $\rho$ crosses the imaginary axis when $x_{0}=0$, at the points $\pm r i$. Thus an approximate upper bound on $\bar{\rho}$ (denoted by $\bar{\rho}_{\text {max }}$ ) is the value of $|r|$ evaluated at the angle $\theta_{0}$ that gives $x_{0}=0$. From Eq. (38), this condition leads to a quadratic in $u=\epsilon_{1} \sin ^{2} \theta_{0}$, namely,

$$
\begin{align*}
&\left(\frac{\epsilon_{1}+\epsilon_{2}}{\epsilon}-1-\frac{\epsilon^{2}}{\epsilon_{1} \epsilon_{2}}\right) u^{2}+\left(\epsilon^{2}-\epsilon_{1} \epsilon_{2}\right) \\
& \times\left(\epsilon^{-1}+\epsilon_{1}^{-1}+\epsilon_{2}^{-1}\right) u-\left(\epsilon^{2}-\epsilon_{1} \epsilon_{2}\right)=0 \tag{42}
\end{align*}
$$

Substitution of the solution of Eq. (42) into Eq. (39) gives the required upper bound on $\bar{\rho}$. Figure 6 illustrates the behavior of $\bar{\rho}_{\max }$ as a function of $\epsilon$ for fixed $\epsilon_{1}$ and $\epsilon_{2}$, for $\epsilon^{2}>$ $\epsilon_{1} \epsilon_{2}$. (For $\epsilon^{2}<\epsilon_{1} \epsilon_{2}, \bar{\rho}_{\text {max }}$ diverges, as noted above; moreover, in this regime the quartics are not well approximated by circles.)

As $\epsilon \rightarrow\left(\epsilon_{1} \epsilon_{2}\right)^{1 / 2}=n_{1} n_{2}$ from above, $\bar{\rho}_{\max } \rightarrow 1$. This result follows from solving Eq. (42) in the above limit: we find that $\theta_{0} \rightarrow 0$ and $|r| \rightarrow 1$. By definition, $r_{p}=r_{s}$ at normal incidence, and indeed the point $x=1$ lies on the unit circle. As $\epsilon \rightarrow n_{1} n_{2}$ and $\theta_{1} \rightarrow 0$, this becomes the only point on the $\rho$ trajectory, in the sense that for $\epsilon$ just greater than $n_{1} n_{2}$, and near normal incidence, $\rho$ varies very slowly with layer thickness near $x=1$ and very rapidly elsewhere. For $\epsilon^{2} \gg \epsilon_{1} \epsilon_{2}$, the angle resulting from Eq. (42) tends to grazing incidence. Again $|r|$ approaches unity, but now the density of points on the trajectory of $\rho$ is concentrated at $x=-1$, consistent with the general result (TR, Section 2-3) that $r_{p} / r_{s} \rightarrow-1$ at
grazing incidence. We conclude that for all positive real values of $\epsilon_{1}, \epsilon$, and $\epsilon_{2}$ such that $\epsilon_{1}<\left(\epsilon_{1} \epsilon_{2}\right)^{1 / 2}<\epsilon, \bar{\rho}_{\max }<1$.

When absorption occurs within the film, $q$ has a positive imaginary part, and $Z=\exp (2 i q \Delta z)$ spirals into the origin as the thickness $\Delta z$ of the film increases. As a result, $\rho$ and $\tau$ do not describe closed curves. However, absorption within the substrate ( $\epsilon_{2}$ and $q_{2}$ complex) does not alter the qualitative behavior described above. The loci of $\tau$ in general, and of $\rho$ in the case of like media, are still circles, and $\rho$ still follows quartics in the unlike-media case. The approximation of these quartics by circles is no longer valid: for example, the points $\rho_{+}$and $\rho_{-}$corresponding to $Z= \pm 1$ no longer lie on the real axis.

## REFERENCES

1. M. Born and E. Wolf, Principles of Optics (Pergamon, Oxford, 1965).
2. D. E. Aspnes, "Spectroscopic ellipsometry of solids," in Optical Properties of Solids, B. O. Seraphin, ed. (North-Holland, Amsterdam, 1976), Chap. 15.
3. R. M. A. Azzam and N. M. Bashara, Ellipsometry and Polarized Light (North-Holland, Amsterdam, 1977).
4. J. Lekner, Theory of Reflection of Electromagnetic and Particle Waves (Nijhoff, Dordrecht, The Netherlands, 1987).
5. H. Lamb, Infinitesimal Calculus (Cambridge U. Press, Cambridge, 1924).
6. R. M. A. Azzam, "Relationship between the $p$ and $s$ Fresnel reflection coefficients of an interface independent of angle of incidence," J. Opt. Soc. Am. A 3, 928-929 (1986).
7. R. M. A. Azzam, "Polar curves for transmission ellipsometry," Opt. Commun. 14, 145-147 (1975).
8. D. Beaglehole, "Ellipsometry of liquid surfaces," J. Phys. (Paris) C 10, 147-154 (1983).
