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We extend ellipsometry to the direct measurement of small perturbations of the Jones matrix of any linear non-depolarizing optical sample (system) subjected to a modulating stimulus such as temperature, stress, or electric or magnetic field. The methodology of this technique, to be called Modulated Generalized Ellipsometry (MGE), is presented. First an ellipsometer with arbitrary polarizing and analyzing optics is assumed, and subsequently the discussion is specialized to a conventional ellipsometer having either the polarizer-sample-analyzer (PSA) or the polarizer-compensator-sample-analyzer (PCSA) arrangement. MGE provides the tool for the systematic study of thermo-optical, piezo-optical, electro-optical, magneto-optical, and other allied effects for both isotropic and anisotropic materials that may be examined in either transmission or reflection. MGE is also applicable to (1) modulation spectroscopy of anisotropic media, (2) the study of electrochemical reactions on optically anisotropic electrodes, and (3) the extension of AIDER (angle-of-incidence-derivative ellipsometry and reflectometry) to the determination of the optical properties of anisotropic film-substrate systems.

I. INTRODUCTION

Conventional ellipsometry is concerned with measuring the ratio of reflection coefficients of an optically isotropic surface for the $p$ and $s$ polarizations of an obliquely incident monochromatic plane wave of light. The extension of ellipsometry to the determination of the normalized Jones matrix of any linear nondepolaring optical system in general, and the reflection matrix of an anisotropic surface in particular, is now known as generalized ellipsometry (GE). Measurement of small changes of the ellipsometric $(\psi, \Delta)$ and reflectance $(\rho)$ parameters of an optically isotropic surface induced by a modulating field has been termed modulated ellipsometry (ME). In this paper we further extend ellipsometry to the measurement of small perturbations (typically sinusoidal) of the Jones matrix of any linear nondepolaring optical sample (system). This new technique is called modulated generalized ellipsometry (MGE).

Consider the interaction of polarized light with a linear nondepolaring optical sample $S$, Fig. 1. Such interaction is described by a Jones matrix $J$ characteristic of $S$. In Fig. 1, $p$, $s$ represent mutually orthogonal axes that are transverse to, and constitute a right-handed coordinate system with, the direction of propagation; $p$ is parallel and $s$ is perpendicular to the plane of the wave vectors of the incident and emergent beams (the plane of incidence in case of light reflection from a surface). In the $p$s coordinate system, $J$ is generally nondiagonal:

$$J = \begin{bmatrix} J_{pp} & J_{ps} \\ J_{sp} & J_{ss} \end{bmatrix}.$$  \hspace{1cm} (1)

Provided that

$$J_{ss} \neq 0,$$  \hspace{1cm} (2)

Eq. (1) can be cast in the form

$$J = J_{ss} J_n,$$  \hspace{1cm} (3)

where $J_n$ is the normalized Jones matrix of the sample:

$$J_n = \begin{bmatrix} \rho_1 & \rho_2 \\ \rho_3 & 1 \end{bmatrix},$$  \hspace{1cm} (4)

$$\rho_1 = J_{pp}/J_{ss}, \quad \rho_2 = J_{ps}/J_{ss}, \quad \rho_3 = J_{sp}/J_{ss}.$$

It is convenient to write

$$\rho_i = \tan \psi_i e^{i \Delta_i}, \quad i = 1, 2, 3,$$  \hspace{1cm} (6)

and

$$|J_{ss}|^2 = \sigma_{ss}.$$  \hspace{1cm} (7)

$J_n$, Eq. (4), defines the polarization response of the sample (i.e., its effect on the ellipse of polarization alone) and $\sigma_{ss}$, Eq. (7), determines its absolute intensity response. The angle of $J_{ss} \arg(J_{ss})$ represents an absolute phase shift, a quantity measurable only by interferometry.

If a time-harmonic modulation that varies as $\sin \omega t$ is applied to the sample, its normalized Jones matrix $J_n$ and intensity response parameter $\sigma_{ss}$ will execute small oscillations $(\delta J_n, \delta \sigma_{ss})$ of amplitudes $(\delta J_n, \delta \sigma_{ss})$ around quiescent (average) values $(\bar{J}_n, \bar{\sigma}_{ss})$ so that

$$J_n = \bar{J}_n + \delta J_n,$$

$$\sigma_{ss} = \bar{\sigma}_{ss} + \delta \sigma_{ss},$$

to first order, where

$$\delta J_n = \bar{J}_n \sin \omega t,$$

$$\delta \sigma_{ss} = \bar{\sigma}_{ss} \sin \omega t.$$  \hspace{1cm} (9)

Fig. 1. A beam of polarized light interacts with a linear nondepolaring optical sample $S$ with Jones matrix $J$. $ps$ is a reference Cartesian coordinate system defined in the same manner for both the input $(i)$ and output $(o)$ beams.
and intensity response parameter \( \sigma_{\text{ss}} \) are perturbed by small amounts \( \delta J_n \) and \( \delta \sigma_{\text{ss}} \). This induces a change of the detector signal \( \delta s_D \), which can be obtained from Eq. (13) by logarithmic differentiation:

\[
\delta s_D/s_D = (\delta \sigma_{\text{ss}}/\sigma_{\text{ss}}) + \sum_{i=1}^{3} (\alpha_{\psi i} \delta \psi_i + \alpha_{\Delta i} \delta \Delta_i).
\]  

In Eq. (14), \( \alpha_{\psi i} \) and \( \alpha_{\Delta i} \) define psi and delta sensitivity functions that relate \( \delta \psi_i \) and \( \delta \Delta_i \) to \( \delta s_D \); they are given by

\[
\alpha_{\psi i} = (1/f) \partial f/\partial \psi_i, \quad \alpha_{\Delta i} = (1/f) \partial f/\partial \Delta_i,
\]  

\( i = 1, 2, 3 \).

When the applied stimulus is sinusoidal, \( \delta \psi_i \), \( \delta \Delta_i \), and \( \delta \sigma_{\text{ss}} \) will also become sinusoidal, Eqs. (9) and (10). Because \( \delta \psi_i \), \( \delta \Delta_i \), \( \delta \sigma_{\text{ss}} \), and \( \delta s_D \) have the same frequency and are in phase, Eq. (14) can be used to relate their amplitudes:

\[
\delta s_D/\bar{f} = (\delta \sigma_{\text{ss}}/\sigma_{\text{ss}}) + \sum_{i=1}^{3} (\overline{\alpha_{\psi i}} \delta \psi_i + \overline{\alpha_{\Delta i}} \delta \Delta_i),
\]  

where \( \overline{\alpha_{\psi i}} \) and \( \overline{\alpha_{\Delta i}} \) are obtained from Eqs. (15) as

\[
\overline{\alpha_{\psi i}} = (1/f) \partial f/\partial \psi_i |_{\varphi_i}, \quad \overline{\alpha_{\Delta i}} = (1/f) \partial f/\partial \Delta_i |_{\varphi_i}.
\]  

The left-hand side of Eq. (16) represents the ac/dc signal ratio, or modulation depth, \( \delta m \):

\[
\delta m = \delta s_D/\bar{f}.
\]  

If \( \delta m \) is measured at seven different settings of the ellipsometer optics, we can use Eq. (16) to yield seven linear equations. These can be combined together in matrix form as

\[
\delta m = I \delta S,
\]  

where \( \delta m \) is a 7×1 measurement vector whose elements are the seven values of \( \delta m \):

\[
\delta m = \begin{bmatrix} \delta m_1 \\
\delta m_2 \\
\vdots \\
\delta m_7 \end{bmatrix}
\]  

\( I \) is a 7×7 instrument matrix that is specified by the ellipsometer and its chosen seven settings, and also by the average (without modulation) Jones matrix \( \overline{J} \) of the sample:

\[
I = \begin{bmatrix}
1 & \overline{\alpha_{\psi 1}} & \overline{\alpha_{\psi 2}} & \overline{\alpha_{\psi 3}} & \overline{\alpha_{\Delta 1}} & \overline{\alpha_{\Delta 2}} & \overline{\alpha_{\Delta 3}} \\
1 & \overline{\alpha_{\psi 1}} & \overline{\alpha_{\psi 2}} & \overline{\alpha_{\psi 3}} & \overline{\alpha_{\Delta 1}} & \overline{\alpha_{\Delta 2}} & \overline{\alpha_{\Delta 3}} \\
\vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\
1 & \overline{\alpha_{\psi 7}} & \overline{\alpha_{\psi 7}} & \overline{\alpha_{\psi 7}} & \overline{\alpha_{\Delta 7}} & \overline{\alpha_{\Delta 7}} & \overline{\alpha_{\Delta 7}}
\end{bmatrix}
\]  

\( \delta S \) is a 7×1 sample-perturbation vector whose elements are the seven sample-perturbation parameters.
where $I$ is the inverse of $I$ given by Eq. (21).

Equation (23) represents the basis of MGE. It shows how the seven sample-perturbation parameters $\delta \sigma_{\alpha\beta}/\sigma_{\alpha\beta}$, and $(\delta \phi_{\alpha}, \delta \Delta_{\alpha})$, $i = 1, 2, 3$, can be obtained from seven measurements of the ac/dc signal ratio $\delta m$ at seven different settings of the ellipsometer optics.

### III. EVALUATION OF THE INSTRUMENT MATRIX $I$ FOR THE PCSA AND PSA ELLIPSOMETER ARRANGEMENTS

In the previous section we presented the methodology of MGE assuming an ellipsometer with arbitrary polarizing and analyzing optics. We have found [Eq. (23)] that the sample-perturbation vector $\delta S$ [Eq. (22)] can be determined by premultiplying the measurement vector $6m$ [Eq. (20)] by the inverse $I^{-1}$ of the instrument matrix $I$ [Eq. (21)]. Therefore the use of an ellipsometer for MGE requires the evaluation of $I$. In this section, we evaluate $I$ for the PSA (polarizer-sample-analyzer) and PCSA (polarizer-compensator-sample-analyzer) commonly used ellipsometer arrangements. The Jones vector of light incident on the sample is

$$E_i = \begin{bmatrix} E_{ip} \\ E_{is} \end{bmatrix},$$

(24)

where

$$E_{ip} = \cos P, \quad E_{is} = \sin P,$$

(25a)

if a polarizer alone is used, and

$$E_{ip} = \cos C \cos (P - C) - \rho_C \sin C \sin (P - C),$$

$$E_{is} = \sin C \cos (P - C) + \rho_C \cos C \sin (P - C),$$

(25b)

if both polarizer and compensator are used, $P$ and $C$ are the azimuth angles of the transmission axis of the polarizer and the fast axis of the compensator, respectively. From the $p$ axis positive in a counterclockwise sense looking into the beam. $\rho_C$ is the slow-to-fast complex-amplitude relative transmittance of the compensator. The Jones vector of light incident on the photodetector, referenced to the transmission and extinction axes of the analyzer, is related to $E_i$ by

$$E_d = T_A R(A) J E_i,$$

(26)

where $J$, $R(A)$, and $T_A$ are the Jones matrices of the sample, rotation $A$, and ideal linear analyzer, respectively. $A$ is the azimuth angle of the transmission axis of the analyzer measured from the $p$ axis positive in a counterclockwise sense looking into the beam. If we substitute $E_i$ from Eq. (24), $J$ from Eqs. (3), (4), and (22), and

$$R(A) = \begin{bmatrix} \cos A & \sin A \\ -\sin A & \cos A \end{bmatrix}, \quad T_A = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix},$$

(27)

into the right-hand side of Eq. (26), we obtain

$$E_d = L,$$

(28)

where

$$L = J s \{ (\rho_1 \cos A + \rho_3 \sin A) E_{ip} + (\rho_2 \cos A + \sin A) E_{is} \}.$$
TABLE I. Partial derivatives required for the evaluation of the instrument matrix $I$ for the PSA ellipsometer using Eqs. (35) and (21).

<table>
<thead>
<tr>
<th>$\phi_1/\lambda_1$</th>
<th>$s_1/\lambda_1$</th>
<th>$s_2/\lambda_1$</th>
<th>$s_3/\lambda_1$</th>
<th>$s_4/\lambda_1$</th>
<th>$s_5/\lambda_1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t_1$</td>
<td>$\cos^4 \phi_1 \sec^2 \psi_1 \cos \Delta_1$</td>
<td>$- \cos^2 \phi_1 \tan \psi_1 \sin \Delta_1$</td>
<td>$\cos^4 \phi_1 \sec^2 \psi_1 \cos \Delta_1$</td>
<td>$- \cos^2 \phi_1 \tan \psi_1 \sin \Delta_1$</td>
<td>$\cos^4 \phi_1 \sec^2 \psi_1 \cos \Delta_1$</td>
</tr>
<tr>
<td>$t_2$</td>
<td>$\cos^4 \phi_1 \sec^2 \psi_1 \sin \Delta_1$</td>
<td>$\cos^2 \phi_1 \tan \psi_1 \cos \Delta_1$</td>
<td>$\cos^4 \phi_1 \sec^2 \psi_1 \sin \Delta_1$</td>
<td>$\cos^2 \phi_1 \tan \psi_1 \cos \Delta_1$</td>
<td>$\cos^4 \phi_1 \sec^2 \psi_1 \sin \Delta_1$</td>
</tr>
</tbody>
</table>

sults are summarized in Table I (PSA) and Table II (PCSA). With these tables, the analytical procedure to evaluate $I$ becomes complete. Notice that $I$, Eq. (21), is expressed in terms of $\alpha_{\Delta_1}$ and $\alpha_{\Delta_2}$, which are the values of $\alpha_{\Delta_1}$ and $\alpha_{\Delta_2}$ when $\psi_1=\psi_2$ and $\Delta_1=\Delta_2$ (see Eqs. (15) and (17)). Therefore, $\alpha_{\Delta_1} i=1,2,3$ (or, equivalently $J_n$) have to be either known or, more likely, should be measured. This means that both $GE$ (measurement of $J_n$) and MGE (measurement of $\delta J_n$ and $\delta J_n/\delta \varphi$) are expected to be conducted jointly. This is discussed in the following section.

IV. COMBINED GE AND MGE

Measurements of the average (dc) and alternating (ac) components of the detector signal $s_d$ at seven different settings of the ellipsometer polarizer and analyzer optics provide adequate information to determine both $J_n$ (GE) and $\delta J_n$, $\delta J_n/\delta \varphi$ (MGE). From Eq. (13), we can write

$$s_{dn} = K \alpha_{\varphi} \alpha_{\Delta i} * (\hat{\psi}_i, \hat{\Delta}_i), \quad n=1,2,\ldots,7,$$

which relates the dc signal $s_{dn}$ at the $n$th measurement to $(\hat{\psi}_i, \hat{\Delta}_i)$, $i=1,2,3$, and the corresponding ellipsometer settings $\alpha_{\varphi i}$, $\alpha_{\Delta i}$. Division of six of Eqs. (36) by the seventh yields six equations that can be solved for the six unknowns $(\hat{\psi}_i, \hat{\Delta}_i)$, $i=1,2,3$. Thus $J_n$ is determined. From measurements of $\delta J_n$ [Eq. (18)] at the seven settings and $J_n$, we can calculate $\delta J_n$ and $\delta J_n/\delta \varphi$ as explained in Secs. II and III.

The above procedure represents a practically attractive method of combined GE and MGE. The principal disadvantage of this method lies in the difficulty of extracting $(\hat{\psi}_i, \hat{\Delta}_i)$, $i=1,2,3$, (hence $J_n$) from the dc-signal data using the set of six equations mentioned above. In addition, the accuracy of the photometric measurement of $J_n$ may be unacceptable, although small errors of $J_n$ do not affect the determination of the perturbation parameters $\delta J_n$ and $\delta J_n/\delta \varphi$ to first order.

An alternate procedure for combined GE and MGE is to measure $J_n$ by a null method5,4 (from the mapping of three different polarization states by the sample), then use $J_n$ to determine $\delta J_n$ and $\delta J_n/\delta \varphi$ as explained in Secs. II and III. To minimize the number of adjustments of the ellipsometer components, a null may first be reached and the appropriate data (nonphotometric, usually azimuth angles) is recorded at that null; then the polarizer or analyzer is offset to two different settings (e.g., in opposite directions) away from the null and the ac and dc photometric signals are recorded. With three nulls, six measurements can be obtained in this manner. The seventh required measurement can be taken by choosing a third different off-null setting at any one (e.g., the last) of the three nulls.

In this section we have briefly mentioned only two modes of carrying out combined GE and MGE. Obviously, other modes of operation of the ellipsometer for this type of application are possible. The choice of a mode that realizes optimum accuracy and/or precision for a given sample has not been discussed. Generally, such an investigation will be based on the psi and delta sensitivity functions $\alpha_{\Delta i}$ and $\alpha_{\varphi i}$ (obtained, e.g., by use of Tables I and II) and would be directed to an evaluation of the accuracy and precision of inverting the instrument matrix $I$ [Eq. (21)]. Another important practical consideration is the use of more than the theoretical minimum amount of data required to determine $J_n$ and $\delta J_n$, $\delta J_n/\delta \varphi$. These and other extensions of the present work (such as automation) are beyond the scope of this paper.

V. APPLICATIONS OF MGE

Up to this point we have not attempted to specify the nature of the sample, the type of modulation, or the manner in which light interacts with the sample. This leaves the scope of MGE wide open. The following are broad areas of application:

1. the study of thermo-optical, piezo-optical, electro-optical, and magneto-optical effects (among others) in reflection and transmission for samples that are initially anisotropic ($\alpha_{\Delta i}$ is nondiagonal), or that become anisotropic as a result of the application of modulation, (e.g., stress, electric or magnetic field);

2. the systematic extension of modulation spectroscopy22 to the study of band structure of anisotropic crystals from spectra of $(\delta \psi_i, \delta \Delta_i)$, $i=1,2,3$, and $\delta \psi_i/\delta \varphi_i$ measured as functions of photon energy;

3. the study electrochemical processes11,12 on optically anisotropic electrodes under periodic potentials; and

TABLE II. Partial derivatives required for the evaluation of the instrument matrix $I$ for the PCSA ellipsometer using Eqs. (35) and (21).

<table>
<thead>
<tr>
<th>$\psi_1/\phi_1$</th>
<th>$\psi_1/\Delta_1$</th>
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<td>$\cos^4 \phi_1 \sec^2 \psi_1 \cos \Delta_1$</td>
</tr>
<tr>
<td>$t_2$</td>
<td>$\cos^4 \phi_1 \sec^2 \psi_1 \cos \Delta_1$</td>
<td>$- \cos^2 \phi_1 \tan \psi_1 \sin \Delta_1$</td>
<td>$\cos^4 \phi_1 \sec^2 \psi_1 \sin \Delta_1$</td>
<td>$\cos^2 \phi_1 \tan \psi_1 \cos \Delta_1$</td>
<td>$\cos^4 \phi_1 \sec^2 \psi_1 \sin \Delta_1$</td>
</tr>
</tbody>
</table>

R. M. A. Azzam
(4) the generalization of the recently introduced technique of AIDER\(^3\) (angle-of-incidence-derivative ellipsometry and reflectometry) to anisotropic surfaces. In this case we determine \(R_0\), \(\partial R_0/\partial \varphi\), and \(\partial \ln R_{ss}/\partial \varphi\), where \(R_0\) is the normalized reflection matrix, \(R_{ss} = |R_{ss}|^2\), and \(\varphi\) is the angle of incidence. This allows the determination of up to thirteen optical parameters that characterize an anisotropic film-substrate system, e.g., all of the optical constants and film thickness of an absorbing biaxial film on a biaxial substrate.\(^24\)

\(^{*}\)Supported by the National Science Foundation.


\(^{6}\)M. Elshazly-Zaghloul, R. M. A. Azzam, and N. M. Bashara, in Ref. 5.

\(^{7}\)P. S. Hauge, in Ref. 5.

\(^{8}\)D. J. De Smet, in Ref. 5.


\(^{10}\)If \(J_{ss} = 0\), the Jones matrix [Eq. (1)] can still be normalized by factoring out another nonzero element, e.g., \(J_{pp}\). The subsequent development has to be modified accordingly.

\(^{11}\)This choice of time dependence is adopted only for simplicity. The analysis is applicable to any waveform of periodic modulation \(M(t)\) impressed on the sample given that the resulting optical perturbations \((\delta \varphi_4, \delta \varphi_5, \delta \Delta)\) also have the same waveform as, and are in phase with, the modulation. Thus, we may replace \(sin \omega t\) by \(M(t)\) in Eqs. (9) and (10). Furthermore, the step that leads from Eq. (14) to Eq. (16) remains valid when \(M(t)\) replaces \(sin \omega t\). The caret over a quantity that varies as \(M(t)\) signifies the peak value of \(M(t)\).

\(^{12}\)They include the azimuth angles and optical properties of the individual elements of \(P\) and \(A\).

\(^{13}\)A lock-in amplifier can be used for the precise measurement of the small ac signal received by the photodetector (see Fig. 2). The reference signal to the lock-in amplifier is derived from the same source of modulation \(M\) applied to the sample. The procedure is similar to that discussed in Refs. 9 and 10.

\(^{14}\)The determination of \((\delta \varphi_4, \delta \Delta)\), \(i = 1, 2, 3\) (or \(\delta \varphi_5\)) can be separated from the determination of \(\delta \varphi_4/\delta \varphi_5\) by subtracting one (e.g., the seventh) of the seven equations represented by Eq. (16) from the remaining six. This gives a matrix equation of the form \(\delta \Phi' = \Phi' \delta S'\). \(\Phi'\) is a modified \(6 \times 6\) measurement vector with elements \((\delta \phi_4 - \delta \phi_5, \delta \phi_5, \delta \Delta_1, \delta \Delta_2, \delta \Delta_3, \delta \phi_4, \delta \phi_5, \delta \Delta_1, \delta \Delta_2, \delta \Delta_3)\); \(I'\) is a modified \(6 \times 6\) instrument matrix with elements \((\delta \phi_{4k} - \delta \phi_{5k}, \delta \phi_{4k} - \delta \phi_{5k}, \delta \phi_{4k} - \delta \phi_{5k}, \delta \phi_{4k} - \delta \phi_{5k}, \delta \phi_{4k} - \delta \phi_{5k}, \delta \phi_{4k} - \delta \phi_{5k})\), \(I'_{k}\) is a \(6 \times 1\) sample-perturbation vector with elements \(\delta \phi_4, \delta \phi_5, \delta \Delta_1, \delta \Delta_2, \delta \Delta_3, \delta \phi_4, \delta \phi_5, \delta \Delta_1, \delta \Delta_2, \delta \Delta_3\); and \(\delta S'\) is a \(6 \times 1\) sample-perturbation vector with elements \(\delta \phi_4, \delta \phi_5, \delta \Delta_1, \delta \Delta_2, \delta \Delta_3, \delta \phi_4, \delta \phi_5, \delta \Delta_1, \delta \Delta_2, \delta \Delta_3\), and hence \(\delta \Phi'\) can be obtained from \(\delta \Phi' = (I')^{-1} \delta S',\) where \((I')^{-1}\) is the inverse of \(I'\). Once \(\delta S'\) has been determined, \(\delta \varphi_4/\delta \varphi_5\) can be calculated from any one of Eqs. (16). This procedure has the obvious advantage of requiring the inversion of a \(6 \times 6\) matrix \((I')\) instead of a \(7 \times 7\) matrix (1).

\(^{15}\)Sec, for example, F. L. McCrackin, J. Opt. Soc. Am. 60, 57 (1970). Notice that we assume unit amplitude for the electric vibration of the light leaving the polarizer.

\(^{16}\)Extension to the general case of arbitrary values of \(\rho_0\) and \(C\) is straightforward but the results are quite complicated.


\(^{19}\)This assumes that the principal axes of the dielectric tensors of the substrate and film have arbitrary but known orientation.