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Modulated generalized ellipsometry

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We extend ellipsometry to the direct measurement of small perturbations of the Jones matrix of any linear nondepolarizing 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[1,2] 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 nondepolarizing optical system in general,[3] and the reflection matrix of an anisotropic surface in particular,[4] is now known as *generalized ellipsometry* (GE).[5-8] Measurement of small changes of the ellipsometric (ψ, Δ) and reflectance ($\mathcal{R}$) parameters of an optically isotropic surface induced by a modulating field[9-12] has been termed *modulated ellipsometry* (ME).[9,10] In this paper we further extend ellipsometry to the measurement of small perturbations (typically sinusoidal) of the Jones matrix of any linear nondepolarizing optical sample (system). This new technique is called *modulated generalized ellipsometry* (MGE).

Consider the interaction of polarized light with a linear nondepolarizing optical sample S, Fig. 1. Such interaction is described by a Jones matrix[13,14] $\mathbf{J}$ 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 ps coordinate system, $\mathbf{J}$ is generally nondiagonal:

$$\mathbf{J}=\begin{bmatrix} J_{pp} & J_{ps} \\ J_{sp} & J_{ss} \end{bmatrix}. \tag{1}$$

Provided that[15]

$$J_{ss}\neq 0, \tag{2}$$

Eq. (1) can be cast in the form

$$\mathbf{J}=J_{ss}\mathbf{J}_n, \tag{3}$$

where $\mathbf{J}_n$ is the normalized Jones matrix of the sample:

$$\mathbf{J}_n=\begin{bmatrix} \rho_1 & \rho_2 \\ \rho_3 & 1 \end{bmatrix}, \tag{4}$$

$$\rho_1=J_{pp}/J_{ss}, \quad \rho_2=J_{ps}/J_{ss}, \quad \rho_3=J_{sp}/J_{ss}. \tag{5}$$

It is convenient to write

$$\rho_i=\tan\psi_i\, e^{j\Delta_i}, \quad i=1, 2, 3, \tag{6}$$

and

$$|J_{ss}|^2=\sigma_{ss}. \tag{7}$$

$\mathbf{J}_n$, Eq. (4), defines the polarization response of the sample (i.e., its effect on the ellipse of polarization alone) and σ_{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[16] modulation that varies as $\sin\omega t$ is applied to the sample, its normalized Jones matrix $\mathbf{J}_n$ and intensity response parameter σ_{ss} will execute small oscillations ($\widetilde{\delta\mathbf{J}}_n$, $\widetilde{\delta\sigma}_{ss}$) of amplitudes ($\widehat{\delta\mathbf{J}}_n$, $\widehat{\delta\sigma}_{ss}$) around quiescent (average) values ($\bar{\mathbf{J}}_n$, $\bar{\sigma}_{ss}$) so that

$$\begin{aligned}\mathbf{J}_n&=\bar{\mathbf{J}}_n+\widetilde{\delta\mathbf{J}}_n,\\ \sigma_{ss}&=\bar{\sigma}_{ss}+\widetilde{\delta\sigma}_{ss},\end{aligned} \tag{8}$$

to first order, where

$$\begin{aligned}\widetilde{\delta\mathbf{J}}_n&=\widehat{\delta\mathbf{J}}_n\sin\omega t,\\ \widetilde{\delta\sigma}_{ss}&=\widehat{\delta\sigma}_{ss}\sin\omega t.\end{aligned} \tag{9}$$

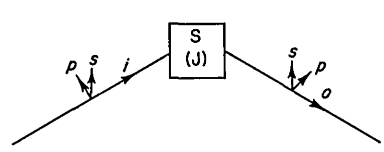

FIG. 1. A beam of polarized light interacts with a linear nondepolarizing optical sample S with Jones matrix $\mathbf{J}$. ps is a reference Cartesian coordinate system defined in the same manner for both the input (i) and output (o) beams.

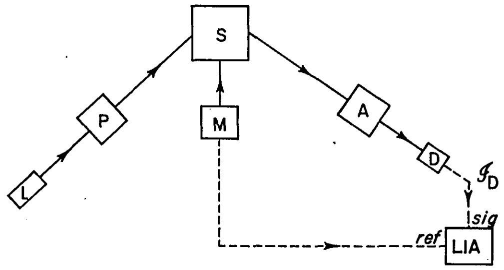

FIG. 2. A general ellipsometer arrangement with arbitrary polarizing (P) and analyzing (A) optics. S is the sample under measurement by MGE, and M is the source of modulation applied to S. The signal output $\mathcal{S}_D$ of the photodetector D can be measured by a lock-in amplifier LIA, which receives its reference signal from M. L represents the light source.

With $\mathbf{J}_n$ expressed in the form of Eq. (4) and ρ_i given by Eq. (6), the presence of modulation causes ψ_i, Δ_i to vary as

$$\begin{aligned} &\psi_i = \bar{\psi}_i + \widetilde{\delta\psi}_i, \qquad \Delta_i = \bar{\Delta}_i + \widetilde{\delta\Delta}_i, \\ &\widetilde{\delta\psi}_i = \widehat{\delta\psi}_i \sin\omega t, \quad \widetilde{\delta\Delta}_i = \widehat{\delta\Delta}_i \sin\omega t, \\ &i = 1, 2, 3. \end{aligned} \tag{10}$$

In Eqs. (8)–(10), the bar ($\bar{\ }$) over a symbol indicates an average value, and the tilde ($\widetilde{\ }$) and caret ($\widehat{\ }$) indicate a sinusoidal quantity and its amplitude, respectively. Notice that we have assumed $\widetilde{\delta\mathbf{J}}_n$ (hence $\widetilde{\delta\psi}_i$, $\widetilde{\delta\Delta}_i$) and $\widetilde{\delta\sigma}_{ss}$ to be in phase with the applied modulation, which is justified as long as $\omega \ll \omega_{\text{opt}}$, where ω_{opt} is the optical frequency.

The amplitude of the normalized-Jones-matrix perturbation $\widehat{\delta\mathbf{J}}_n$ can be constructed from $(\widehat{\delta\psi}_i, \widehat{\delta\Delta}_i)$ by

$$\widehat{\delta\mathbf{J}}_n = \begin{bmatrix} \widehat{\delta\rho}_1 & \widehat{\delta\rho}_2 \\ \widehat{\delta\rho}_3 & 0 \end{bmatrix}, \tag{11}$$

$$\begin{aligned} &\widehat{\delta\rho}_i = (\sec^2\bar{\psi}_i \widehat{\delta\psi}_i + j\tan\bar{\psi}_i\, \widehat{\delta\Delta}_i)\, e^{j\bar{\Delta}_i}, \\ &i = 1, 2, 3. \end{aligned} \tag{12}$$

Equations (11) and (12) follow from Eqs. (4) and (6), respectively.

MGE is defined as the measurement of $\widehat{\delta\mathbf{J}}_n$ and $\widehat{\delta\sigma}_{ss}$ that result from the application of sinusoidal modulation to the sample S. The methodology of MGE is explained in the following section.

II. METHODOLOGY OF MGE

We assume a general ellipsometer with arbitrary polarizing (P) and analyzing (A) optics, Fig. 2. The signal received by the photodetector D can be written as

$$\mathcal{S}_D = K\sigma_{ss} f(\psi_1, \Delta_1, \psi_2, \Delta_2, \psi_3, \Delta_3; a_\mu, b_\nu), \tag{13}$$

where K is a proportionality factor, f is a function whose form depends on the ellipsometer, and a_μ, b_ν ($\mu, \nu = 1, 2, 3, \ldots,$) are two sets of parameters that characterize A and P, respectively.[17]

When a modulation (from the source M in Fig. 2) is applied to the sample, its normalized Jones matrix $\mathbf{J}_n$ and intensity response parameter σ_{ss} are perturbed by small amounts $\delta\mathbf{J}_n$ and $\delta\sigma_{ss}$. This induces a change of the detector signal $\delta\mathcal{S}_D$ that can be obtained from Eq. (13) by logarithmic differentiation:

$$\delta\mathcal{S}_D/\mathcal{S}_D = (\delta\sigma_{ss}/\sigma_{ss}) + \sum_{i=1}^{3}(\alpha_{\psi i}\delta\psi_i + \alpha_{\Delta i}\delta\Delta_i). \tag{14}$$

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 $\delta\mathcal{S}_D$; they are given by

$$\begin{aligned} &\alpha_{\psi i} = (1/f)\, \partial f/\partial\psi_i, \\ &\alpha_{\Delta i} = (1/f)\, \partial f/\partial\Delta_i, \\ &i = 1, 2, 3. \end{aligned} \tag{15}$$

When the applied stimulus is sinusoidal, $\delta\psi_i$, $\delta\Delta_i$, and $\delta\sigma_{ss}$ will also become sinusoidal, Eqs. (9) and (10). Because $\widetilde{\delta\psi}_i$, $\widetilde{\delta\Delta}_i$, $\widetilde{\delta\sigma}_{ss}$, and $\widetilde{\delta\mathcal{S}}_D$ have the same frequency and are in phase, Eq. (14) can be used to relate their amplitudes:

$$\widehat{\delta\mathcal{S}}_D/\bar{\mathcal{S}}_D = (\widehat{\delta\sigma}_{ss}/\bar{\sigma}_{ss}) + \sum_{i=1}^{3}(\bar{\alpha}_{\psi i}\widehat{\delta\psi}_i + \bar{\alpha}_{\Delta i}\widehat{\delta\Delta}_i), \tag{16}$$

where $\bar{\alpha}_{\psi i}$ and $\bar{\alpha}_{\Delta i}$ are obtained from Eqs. (15) as

$$\begin{aligned} &\bar{\alpha}_{\psi i} = (1/f)\, \partial f/\partial\psi_i|_{\bar{\psi}_i, \bar{\Delta}_i}, \\ &\bar{\alpha}_{\Delta i} = (1/f)\, \partial f/\partial\Delta_i|_{\bar{\psi}_i, \bar{\Delta}_i}. \end{aligned} \tag{17}$$

The left-hand side of Eq. (16) represents the ac/dc signal ratio,[18] or modulation depth, δm:

$$\delta m = \widehat{\delta\mathcal{S}}_D/\bar{\mathcal{S}}_D. \tag{18}$$

If δm 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

$$\delta\mathbf{m} = \mathbf{I}\delta\mathbf{S}, \tag{19}$$

where $\delta\mathbf{m}$ is a 7×1 measurement vector whose elements are the seven values of δm:

$$\delta\mathbf{m} = \begin{bmatrix} \delta m_1 \\ \delta m_2 \\ \vdots \\ \delta m_7 \end{bmatrix}; \tag{20}$$

$\mathbf{I}$ 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 $\bar{\mathbf{J}}$ of the sample:

$$\mathbf{I} = \begin{bmatrix} 1 & \bar{\alpha}_{\psi 11} & \bar{\alpha}_{\Delta 11} & \bar{\alpha}_{\psi 21} & \bar{\alpha}_{\Delta 21} & \bar{\alpha}_{\psi 31} & \bar{\alpha}_{\Delta 31} \\ 1 & \bar{\alpha}_{\psi 12} & \bar{\alpha}_{\Delta 12} & \bar{\alpha}_{\psi 22} & \bar{\alpha}_{\Delta 22} & \bar{\alpha}_{\psi 32} & \bar{\alpha}_{\Delta 32} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ 1 & \bar{\alpha}_{\psi 17} & \bar{\alpha}_{\Delta 17} & \bar{\alpha}_{\psi 27} & \bar{\alpha}_{\Delta 27} & \bar{\alpha}_{\psi 37} & \bar{\alpha}_{\Delta 37} \end{bmatrix}; \tag{21}$$

$\delta\mathbf{S}$ is a 7×1 sample-perturbation vector whose elements are the seven sample-perturbation parameters:

$$\delta S = \begin{bmatrix} \widehat{\delta\sigma}_{ss}/\bar{\sigma}_{ss} \\ \widehat{\delta\psi}_1 \\ \widehat{\delta\Delta}_1 \\ \widehat{\delta\psi}_2 \\ \widehat{\delta\Delta}_2 \\ \widehat{\delta\psi}_3 \\ \widehat{\delta\Delta}_3 \end{bmatrix} . \tag{22}$$

Inverting Eq. (19), we obtain

$$\delta S = I^{-1}\,\delta m , \tag{23}$$

where I^{-1} is the inverse of I given by Eq. (21).

Equation (23) represents the basis of MGE. It shows how the seven sample-perturbation parameters[19] $\widehat{\delta\sigma}_{ss}/\bar{\sigma}_{ss}$ and $(\widehat{\delta\psi}_i, \widehat{\delta\Delta}_i)$, $i=1, 2, 3$, can be obtained from seven measurements of the ac/dc signal ratio δm at seven different settings of the ellipsometer optics.

III. EVAULATION 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 δS [Eq. (22)] can be determined by premultiplying the measurement vector δm [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-compensater-sample-analyzer) commonly used ellipsometer arrangements. The Jones vector of light incident on the sample is

$$E_i = \begin{bmatrix} E_{ip} \\ E_{is} \end{bmatrix}, \tag{24}$$

where

$$E_{ip} = \cos P, \quad E_{is} = \sin P , \tag{25a}$$

if a polarizer alone is used, and

$$\begin{aligned} E_{ip} &= \cos C \cos(P-C) - \rho_C \sin C \sin(P-C) , \\ E_{is} &= \sin C \cos(P-C) + \rho_C \cos C \sin(P-C) , \end{aligned} \tag{25b}$$

if both polarizer and compensator are used.[20] P and C are the azimuth angles of the transmission axis of the polarizer and the fast axis of the compensator, respectively, measured from the p axis positive in a counterclockwise sense looking into the beam. ρ_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

$$E_D = T_A R(A)\, J E_i , \tag{26}$$

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[13,14]

$$R(A) = \begin{bmatrix} \cos A & \sin A \\ -\sin A & \cos A \end{bmatrix}, \quad T_A = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} , \tag{27}$$

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

$$E_D = \begin{bmatrix} L \\ 0 \end{bmatrix} , \tag{28}$$

where

$$L = J_{ss}\left[(\rho_1 \cos A + \rho_3 \sin A) E_{ip} + (\rho_2 \cos A + \sin A) E_{is}\right] . \tag{29}$$

The detected signal is given by

$$\mathcal{I}_D = K |L|^2 , \tag{30}$$

where K is a proportionality constant. If we substitute ρ_i from Eq. (6) and E_{ip}, E_{is} from Eqs. (25a) into Eq. (29), and use the result in Eq. (30), we obtain

$$\mathcal{I}_D = K\sigma_{ss} f(\psi_1, \Delta_1, \psi_2, \Delta_2, \psi_3, \Delta_3; A, P) \tag{31}$$

for the PSA ellipsometer, where σ_{ss} is defined by Eq. (7), and

$$f = l_1^2 + l_2^2 , \tag{32}$$

$$\begin{aligned} l_1 &= \cos A \cos P\,(\tan\psi_1 \cos\Delta_1 + \tan P \tan\psi_2 \cos\Delta_2 \\ &\quad + \tan A \tan\psi_3 \cos\Delta_3 + \tan A \tan P) , \\ l_2 &= \cos A \cos P\,(\tan\psi_1 \sin\Delta_1 + \tan P \tan\psi_2 \sin\Delta_2 \\ &\quad + \tan A \tan\psi_3 \sin\Delta_3) . \end{aligned} \tag{33}$$

For the PCSA ellipsometer, we pursue only the special case[21] when the compensator acts as a quarter-wave retarder ($\rho_C = -j$) and the azimuth of its fast axis is set at $C = \pi/4$. With $\rho_C = -j$ and $C = \pi/4$, Eqs. (25b) yield $E_{ip} = \exp\{j[P - (\pi/4)]\}$ and $E_{is} = \exp\{-j[P-(\pi/4)]\}$. Substituting these values of E_{ip} and E_{is} into Eq. (29), with ρ_i given by Eq. (6), and using the result in Eq. (30), we obtain equations that are identical in form with Eqs. (31) and (32), except that

$$\begin{aligned} l_1 &= \cos A[\tan\psi_1 \sin(2P + \Delta_1) + \tan\psi_2 \cos\Delta_2 \\ &\quad + \tan A \tan\psi_3 \sin(2P + \Delta_3) + \tan A] , \\ l_2 &= \cos A[\tan\psi_1 \cos(2P + \Delta_1) - \tan\psi_2 \sin\Delta_2 \\ &\quad + \tan A \tan\psi_3 \cos(2P + \Delta_3)] . \end{aligned} \tag{34}$$

To evaluate I, Eq. (21), we need to determine $\alpha_{\psi i}$ and $\alpha_{\Delta i}$. From Eqs. (15) and (32), we have

$$\begin{aligned} \alpha_{\psi i} &= (2l_1 \partial l_1/\partial\psi_i + 2l_2 \partial l_2/\partial\psi_i)/(l_1^2 + l_2^2) , \\ \alpha_{\Delta i} &= (2l_1 \partial l_1/\partial\Delta_i + 2l_2 \partial l_2/\partial\Delta_i)/(l_1^2 + l_2^2) , \\ i &= 1, 2, 3. \end{aligned} \tag{35}$$

The twelve partial derivatives $\partial l_1/\partial\psi_i$, $\partial l_1/\partial\Delta_i$, $\partial l_2/\partial\psi_i$, and $\partial l_2/\partial\Delta_i$ $(i = 1, 2, 3)$ that appear in Eqs. (35) can be readily derived from Eqs. (33) and Eqs. (34) for the PSA and PCSA ellipsometers, respectively. The re-

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

	$\partial l/\partial\psi_1$	$\partial l/\partial\Delta_1$	$\partial l/\partial\psi_2$	$\partial l/\partial\Delta_2$	$\partial l/\partial\psi_3$	$\partial l/\partial\Delta_3$
l_1	$\cos A\cos P\sec^2\psi_1\cos\Delta_1$	$-\cos A\cos P\tan\psi_1\sin\Delta_1$	$\cos A\sin P\sec^2\psi_2\cos\Delta_2$	$-\cos A\sin P\tan\psi_2\sin\Delta_2$	$\sin A\cos P\sec^2\psi_3\cos\Delta_3$	$-\sin A\cos P\tan\psi_3\sin\Delta_3$
l_2	$\cos A\cos P\sec^2\psi_1\sin\Delta_1$	$\cos A\cos P\tan\psi_1\cos\Delta_1$	$\cos A\sin P\sec^2\psi_2\sin\Delta_2$	$\cos A\sin P\tan\psi_2\cos\Delta_2$	$\sin A\cos P\sec^2\psi_3\sin\Delta_3$	$\sin A\cos P\tan\psi_3\cos\Delta_3$

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 $\bar{\alpha}_{\psi i}$ and $\bar{\alpha}_{\Delta i}$, which are the values of $\alpha_{\psi i}$ and $\alpha_{\Delta i}$ when $\psi_i = \bar{\psi}_i$ and $\Delta_i = \bar{\Delta}_i$ [see Eqs. (15) and (17)]. Therefore $(\bar{\psi}_i, \bar{\Delta}_i)$, $i = 1, 2, 3$, (or, equivalently $\bar{\mathbf{J}}_n$) have to be either known or, more likely, should be measured. This means that both GE (measurement of $\bar{\mathbf{J}}_n$) and MGE (measurement of $\widehat{\delta\mathbf{J}}_n$ and $\widehat{\delta\sigma}_{ss}/\bar{\sigma}_{ss}$) 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 $\mathcal{I}_D$ at seven different settings of the ellipsometer polarizer and analyzer optics provide adequate information to determine both $\bar{\mathbf{J}}_n$(GE) and $\widehat{\delta\mathbf{J}}_n$, $\widehat{\delta\sigma}_{ss}/\bar{\sigma}_{ss}$ (MGE). From Eq. (13), we can write

$$\bar{\mathcal{I}}_{Dn} = K\bar{\sigma}_{ss} f(\bar{\psi}_1, \bar{\Delta}_1, \bar{\psi}_2, \bar{\Delta}_2, \bar{\psi}_3, \bar{\Delta}_3, a^n_\mu, b^n_\nu), \quad n = 1, 2, \ldots, 7, \tag{36}$$

which relates the dc signal $\bar{\mathcal{I}}_{Dn}$ at the nth measurement to $(\bar{\psi}_i, \bar{\Delta}_i)$, $i = 1, 2, 3$, and the corresponding ellipsometer settings a^n_μ, b^n_ν. Division of six of Eqs. (36) by the seventh yields six equations that can be solved for the six unknowns $(\bar{\psi}_i, \bar{\Delta}_i)$, $i = 1, 2, 3$. Thus $\bar{\mathbf{J}}_n$ is determined. From measurements of δm [Eq. (18)] at the seven settings and $\bar{\mathbf{J}}_n$, we can calculate $\widehat{\delta\mathbf{J}}_n$ and $\widehat{\delta\sigma}_{ss}/\bar{\sigma}_{ss}$ as explained in Secs. II and III.

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

An alternate procedure for combined GE and MGE is to measure $\bar{\mathbf{J}}_n$ by a null method[3,4] (from the mapping of three different polarization states by the sample), then use $\bar{\mathbf{J}}_n$ to determine $\widehat{\delta\mathbf{J}}_n$ and $\widehat{\delta\sigma}_{ss}/\bar{\sigma}_{ss}$, 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 $\alpha_{\psi i}$ and $\alpha_{\Delta i}$ (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 $\bar{\mathbf{J}}_n$ and $\widehat{\delta\mathbf{J}}_n$, $\widehat{\delta\sigma}_{ss}/\bar{\sigma}_{ss}$. 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 ($\bar{\mathbf{J}}$ 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 spectroscopy[22] to the study of band structure of anisotropic crystals from spectra of $(\widehat{\delta\psi}_i, \widehat{\delta\Delta}_i)$, $i = 1, 2, 3$, and $\widehat{\delta\sigma}_{ss}/\bar{\sigma}_{ss}$ measured as functions of photon energy;

(3) the study electrochemical processes[11,12] 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).

	$\partial l/\partial\psi_1$	$\partial l/\partial\Delta_1$	$\partial l/\partial\psi_2$	$\partial l/\partial\Delta_2$	$\partial l/\partial\psi_3$	$\partial l/\partial\Delta_3$
l_1	$\cos A\sec^2\psi_1\sin(2P+\Delta_1)$	$\cos A\tan\psi_1\cos(2P+\Delta_1)$	$\cos A\sec^2\psi_2\cos\Delta_2$	$-\cos A\tan\psi_2\sin\Delta_2$	$\sin A\sec^2\psi_3\sin(2P+\Delta_3)$	$\sin A\tan\psi_3\cos(2P+\Delta_3)$
l_2	$\cos A\sec^2\psi_1\cos(2P+\Delta_1)$	$-\cos A\tan\psi_1\sin(2P+\Delta_1)$	$-\cos A\sec^2\psi_2\sin\Delta_2$	$-\cos A\tan\psi_2\cos\Delta_2$	$\sin A\sec^2\psi_3\cos(2P+\Delta_3)$	$-\sin A\tan\psi_3\sin(2P+\Delta_3)$

(4) the generalization of the recently introduced technique of AIDER[23] (angle-of-incidence-derivative ellipsometry and reflectometry) to anisotropic surfaces. In this case we determine R_n, $\partial \mathrm{R}_n/\partial\varphi$, and $\partial\, ln\, \mathcal{R}_{ss}/\partial\varphi$, where R_n is the normalized reflection matrix, $\mathcal{R}_{ss} = |R_{ss}|^2$, and φ 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]

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[12] J. Horkans, B. D. Cahan, and E. Yeager, Surface Sci. **46**, 1 (1974).

[13] W. A. Shurcliff, *Polarized Light* (Harvard University, Cambridge, 1962).

[14] D. Clarke and J. F. Grainger, *Polarized Light and Optical Measurement* (Pergamon, New York, 1971).

[15] 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.

[16] 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 ($\delta\sigma_{ss}$, $\delta\psi_i$, $\delta\Delta_i$) 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 $M(t)$.

[17] They include the azimuth angles and optical properties of the individual elements of P and A.

[18] 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.

[19] The determination of $(\widehat{\delta\psi_i}, \widehat{\delta\Delta_i})$, $i = 1, 2, 3$, (or $\widehat{\delta \mathbf{J}_n}$) can be separated from the determination of $\widehat{\delta\sigma_{ss}}/\bar{\sigma}_{ss}$ 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 $\delta \mathrm{m}' = \mathbf{I}'\delta \mathbf{S}'$. $\delta\mathrm{m}'$ is a modified 6×1 measurement vector with elements $(\delta m_k - \delta m_7)$ $(k = 1, 2, \ldots, 6)$; $\mathbf{I}'$ is a modified 6×6 instrument matrix with elements $[(\bar{\alpha}_{\psi 1k} - \bar{\alpha}_{\psi 17}), (\bar{\alpha}_{\Delta 1k} - \bar{\alpha}_{\Delta 17}), (\bar{\alpha}_{\psi 2k} - \bar{\alpha}_{\psi 27}), (\alpha_{\Delta 2k} - \alpha_{\Delta 27}), (\bar{\alpha}_{\psi 3k} - \bar{\alpha}_{\psi 37}), (\bar{\alpha}_{\Delta 3k} - \bar{\alpha}_{\Delta 37})]$ in the kth row $(k = 1, 2, \ldots, 6)$; and $\delta\mathbf{S}'$ is a 6×1 sample-perturbation vector with elements $\delta\psi_1$, $\delta\Delta_1$, $\delta\psi_2$, $\delta\Delta_2$, $\delta\psi_3$, $\delta\Delta_3$. $\delta\mathbf{S}'$, and hence $\widehat{\delta\mathbf{J}_n}$, can be obtained from $\delta\mathbf{S}' = [\mathbf{I}']^{-1}\delta\mathrm{m}'$, where $[\mathbf{I}']^{-1}$ is the inverse of $\mathbf{I}'$. Once $\delta\mathbf{S}'$ has been determined, $\widehat{\delta\sigma_{ss}}/\bar{\sigma}_{ss}$ can be calculated from any one of Eqs. (16). This procedure has the obvious advantage of requiring the inversion of a 6×6 matrix $(\mathbf{I}')$ instead of a 7×7 matrix $(\mathbf{I})$.

[20] See, 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.

[21] Extension to the general case of arbitrary values of ρ_C and C is straightforward but the results are quite complicated.

[22] M. Cardona, *Modulation Spectroscopy* (Academic, New York, 1969).

[23] R. M. A. Azzam, Opt. Commun. **16**, 153 (1976).

[24] This assumes that the principal axes of the dielectric tensors of the substrate and film have arbitrary *but known* orientation.