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# Calibration procedures for a two-modulator generalized ellipsometer

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## ABSTRACT

A Two-Modulator Generalized Ellipsometer (2-MGE) has been extremely useful in characterizing optical properties of uniaxial bulk materials, thin films and diffraction gratings. The instrument consists of two polarizer-photoelastic modulator pairs, one operating as the polarization state generator and the other as the polarization state detector. Each photoelastic modulator operates at a different resonant frequency (such as 50 kHz and 60 kHz), making it possible to measure eight elements of the reduced sample Mueller matrix simultaneously. In certain configurations, light reflection from non-depolarizing anisotropic samples can be completely characterized by a single measurement, and the entire reduced Jones matrix can be determined, including the cross polarization coefficients. The calibration of the instrument involves the measurement of the azimuthal angle of the polarizer with respect to the modulator, the modulation amplitude, and the modulator strain for each polarizer photoelastic modulator pair, where the last two are functions of wavelengths. In addition, it is essential to calibrate the azimuthal angles of the polarization state generator and the polarization state detector with respect to the plane of incidence in the ellipsometry configuration that is used in the measurements. Because two modulators operating at different frequencies are used, these calibrations are actually easier and more accurate than for one modulator ellipsometers. In this paper, we will discuss these calibrations and the resultant accuracy limitations of the 2-MGE.

**Keywords:** Ellipsometry, generalized ellipsometry, uniaxial materials, cross polarization, photoelastic modulator

## 1. INTRODUCTION

The two modulator generalized ellipsometer (2-MGE)<sup>1</sup> is an especially powerful instrument in that it can completely measure the polarization-dependent optical properties of many reflective or transmissive samples. The instrument consists of two polarizer-photoelastic modulator (PEM) pairs, where the 2 PEM's are operating at different resonant frequencies (~50 kHz and ~60 kHz in our case). For non-depolarizing samples that can be described with a Mueller-Jones matrix, the complete Mueller matrix can be determined with a single measurement of the 2-MGE. If all 16 elements are required, then four separate measurements at different azimuthal orientations of the polarization state generator (PSG) and the polarization state detector (PSD) must be made.

This instrument has been successfully used to measure the optical functions of a variety of uniaxial crystals,<sup>2-6</sup> including rutile (TiO<sub>2</sub>), zinc oxide (ZnO), bismuth triiodide (BiI<sub>3</sub>), and a series of rare earth phosphates. Some of these samples were so small (<1mm<sup>2</sup>) that the conventional optical path had to be modified with focussing lenses. These lenses are strained, so the ellipsometric measurements had to be corrected for their strain-induced birefringence of the lenses.<sup>7</sup>

As with any ellipsometer, accurate measurements require accurate calibrations. Since the accuracies desired of the ellipsometric parameters are typically  $\pm 0.001$ - $0.003$ , the azimuthal angles must be determined to  $\sim \pm 0.015^\circ$  and other parameters must be determined to 0.1-0.3%. Fortunately, many of these calibrations are actually *EASIER* with the 2-MGE than for other instruments using photoelastic modulators. As we will show in this paper, certain errors often are solely responsible for a particular ellipsometric parameter being different from zero. Hence, the error can then be unambiguously eliminated by just nulling the appropriate ellipsometric parameter.

## 2. PHOTOELASTIC MODULATOR

The basic principle of photoelastic modulation is that a light beam passes through an optical element that is mechanically stressed by the acoustic wave generated by an oscillating piezoelectric transducer. The oscillating stress (typically at 20-80 kHz) generates an oscillating optical anisotropy by the photoelastic effect, creating a wave plate with a time-dependent retardation. The retardation of the PEM is normally expressed as<sup>8</sup>

$$\delta = A \sin(\omega t + \phi) + \delta_o, \quad (1)$$

where  $A$  is the amplitude of the modulation,  $2\pi\omega$  is the frequency of the modulator,  $\phi$  is the phase of the modulator, and  $\delta_o$  is the static retardation of the PEM. It is normally assumed that the static retardation can be expressed as a linear correction (as done in Eq. 1), which implies that the static strain in the PEM is collinear with one of the major oscillating axes of the PEM.<sup>9, 10</sup> If this assumption cannot be made, then the analysis becomes extremely complicated since the directions of the major axes of the PEM vary through the oscillation cycle of the optical element.

Typically, the modulation amplitude  $A$  is controlled by a modulator voltage  $V_m$ , which is given by<sup>8</sup>

$$A = \frac{d}{\lambda} \pi n^3(\lambda) K Q(\lambda) V_m = \frac{d}{\lambda} \pi K V_m \sum_j \frac{\alpha_j}{\lambda^{2j}}, \quad (2a)$$

where  $\lambda$  is the wavelength of light and  $K$  is a constant relating  $V_m$  to the maximum value of the oscillating strain of the optical element. The oscillating optical element is characterized by its thickness  $d$ , its strain-optic coefficient,  $Q(\lambda)$ , and its refractive index  $n(\lambda)$ . The wavelength-dependent parameters  $Q$  and  $n$  can be parameterized using a standard Cauchy expansion, as shown in the second part of Eq. 2a.

It is common, though not universal, to set  $V_m$  to a voltage such that  $A = 2.4048$ . Eq. 2a can be re-arranged to give

$$V_m = \frac{A\lambda}{d\pi K \sum_j \frac{\alpha_j}{\lambda^{2j}}}. \quad (2b)$$

The modulator voltage required to keep  $A$  constant is nearly linear with wavelength with a small dispersive correction.

Similarly, the static strain of the PEM can be expressed as

$$\delta_o = \frac{d}{\lambda} \pi P_o n^3(\lambda) Q(\lambda) = \frac{d}{\lambda} \pi P_o \sum_j \frac{\alpha_j}{\lambda^{2j}} \quad (2c)$$

where  $P_o$  is the static strain of the modulators. If Eqs. 2b and 2c are multiplied, then

$$V_m \delta_o = \frac{AP_o}{K}. \quad (2d)$$

For any particular modulator, the product of the voltage required to keep  $A$  constant and the static strain will be a constant.

By employing a polarizer before the PEM, dynamically elliptically polarized light is generated, with the ellipticity changing at the frequency of the PEM. A convenient way of representing the polarization state of the

light beam after it has passed through the PEM is by its Stokes vector. The Stokes vector is a 4-vector, defined by<sup>11,12</sup>

$$S = \begin{pmatrix} I_o \\ Q \\ U \\ V \end{pmatrix} = \begin{pmatrix} I_o \\ I_o - I_{90} \\ I_{45} - I_{-45} \\ I_{rc} - I_{lc} \end{pmatrix}, \quad (3)$$

where  $I_o$  is the intensity of the light beam, and  $I_o$ ,  $I_{45}$ ,  $I_{90}$ , and  $I_{-45}$  are the light intensities for linearly polarized light at  $0^\circ$ ,  $45^\circ$ ,  $90^\circ$ , and  $-45^\circ$  respectively with respect to the plane of incidence.  $I_{rc}$  and  $I_{lc}$  are the intensities of right- and left-circularly polarized light, respectively. All elements of the Stokes vector are intensities and therefore are real. The total light intensity

$$I_o \geq (Q^2 + U^2 + V^2)^{1/2}, \quad (4)$$

where the equality holds only if the light beam is totally polarized.

The Stokes vector for unpolarized light passing through a polarizer-PEM pair is given by

$$S_{P-PEM} = \begin{pmatrix} 1 \\ C_m C_b + S_m S_b Y_\delta \\ S_m C_b - C_m S_b Y_\delta \\ S_b X_\delta \end{pmatrix} \quad (5)$$

where

$$C_m = \cos(2\theta_m); S_m = \sin(2\theta_m) \quad (6a)$$

$$C_b = \cos(2\theta_b); S_b = \sin(2\theta_b) \quad (6b)$$

The angle  $\theta_m$  is the azimuthal angle of the modulator with respect to the plane of incidence and the angle  $\theta_b$  is the azimuthal angle of the polarizer with respect to the PEM. The quantities  $X_\delta$  and  $Y_\delta$  are the time-dependent basis functions, given by

$$X_\delta = \sin(\delta) = \sin(A \sin(\omega t + \phi) + \delta_o), \quad (7a)$$

$$= \sin(A \sin(\omega t + \phi)) + \delta_o \cos(A \sin(\omega t + \phi)) = X + \delta_o Y$$

$$Y_\delta = \cos(\delta) = \cos(A \sin(\omega t + \phi) + \delta_o). \quad (7b)$$

$$= \cos(A \sin(\omega t + \phi)) - \delta_o \sin(A \sin(\omega t + \phi)) = Y + \delta_o X.$$

The last expansion that separates out the static strain-induced retardation assumes that the static strain is small with respect to 1.

The characterization of any polarizer-PEM pair requires four separate calibrations. The azimuthal orientation of the optical elements is described by two angles  $\theta_b$  and  $\theta_m$  (see Eqs. 5 and 6). The PEM is described by two wavelength-dependent parameters  $A(\lambda)$  and  $\delta_o(\lambda)$ .

### 3. TWO-MODULATOR GENERALIZED ELLIPSOMETER

The intensity of the light beam through the 2-MGE is given by

$$Intensity = S_1^T M S_0, \quad (8)$$

where  $S_0$  is the Stokes vector for the polarization state generator (PSG), given by Eq. 5 and  $S_1^T$  is the transposed Stokes vector for the polarization state detector (PSD). The Mueller matrix  $M$  is a 4X4 real matrix and represents the light interaction with all elements between the PSG and the PSD. The light intensity can be expressed as a dc term plus 8 constant terms multiplied by 8 basis functions<sup>1</sup>

$$Intensity = I_{dc} + I_{X0} X_{0\delta} + I_{Y0} Y_{0\delta} + I_{X1} X_{1\delta} + I_{Y1} Y_{1\delta} + I_{X0X1} X_{0\delta} X_{1\delta} + I_{X0Y1} X_{0\delta} Y_{1\delta} + I_{Y0X1} Y_{0\delta} X_{1\delta} + I_{Y0Y1} Y_{0\delta} Y_{1\delta} \quad (9a)$$

The 0 and 1 subscripts of X and Y refer to the PSG and the PSD, respectively. The strain-induced retardation can be separated out, giving

$$Intensity = \begin{pmatrix} 1 & X_0 & Y_0 & X_1 & Y_1 & X_0X_1 & X_0Y_1 & Y_0X_1 & Y_0Y_1 \end{pmatrix} \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & \delta_0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -\delta_0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & \delta_1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -\delta_1 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & \delta_1 & \delta_0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -\delta_1 & 1 & 0 & \delta_0 \\ 0 & 0 & 0 & 0 & 0 & -\delta_0 & 0 & 1 & \delta_1 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\delta_0 & -\delta_1 & 1 \end{pmatrix} \begin{pmatrix} I_{dc} \\ I_{X0} \\ I_{Y0} \\ I_{X1} \\ I_{Y1} \\ I_{X0X1} \\ I_{X0Y1} \\ I_{Y0X1} \\ I_{Y0Y1} \end{pmatrix} \quad (9b)$$

The 8 constant terms  $I_{X0}$ ,  $I_{Y0}$ , etc. are usually normalized by the  $I_{dc}$  term (to eliminate fluctuations of the incident light intensity, and to eliminate the dependence of the sample reflectivity), so the time-dependence of the intensity is determined by 8 parameters.

The basis functions for any polarization modulation ellipsometer, including the 2-MGE, are the functions X and Y, shown in Eq. 7. These basis functions are related to the common Fourier basis functions using an infinite series including integer Bessel functions:

$$X = \sin(A \sin(\omega t + \phi)) = \sum_{j=1} J_{2j-1}(A) \sin((2j-1)(\omega t + \phi)) \quad (10a)$$

$$Y = \cos(A \sin(\omega t + \phi)) = J_0(A) + 2 \sum_{j=1} J_{2j}(A) \cos(2j(\omega t + \phi)) \quad (10b)$$

For many ellipsometric applications, A is chosen to be 2.4048 radians, where  $J_0(A) = 0$ ,  $J_1(A) = 0.5192$ ,  $J_2(A) = 0.4318$ ,  $J_3(A) = 0.1990$ ,  $J_4(A) = 0.0647$ ,  $J_5(A) = 0.0164$ ,  $J_6(A) = 0.0034$ , etc. At this value for A, the Fourier expansion of the X and Y basis functions have no dc terms and the series converges very rapidly. The  $J_1(A)$  and  $J_2(A)$  are also within 15% of their maximum values for  $A = 2.4048$ .

Because the basis functions of PEM's are so closely related to Fourier basis functions, lock-in amplifiers have often been used to measure the coefficients  $I_X$ , and  $I_Y$  if only a single PEM is used, where  $I_X$  is proportional to the 1f signal, and  $I_Y$  is proportional to the 2f signal. However, this solution becomes untractable when more than one PEM

is used, since one would have to have at least eight lock-in amplifiers to analyze the time-dependent intensity given in Eq. 9. Furthermore, the PEM's are resonant devices, and their frequency and phase are set by the physical dimensions and temperature of the optical element. Therefore, any data analysis will require a precise knowledge of the instantaneous phase of both of the PEM's. The technique that we have used to deconvolute the time-dependent intensity incorporates a trigger circuit that initializes a waveform digitization when the phase of each PEM is at a known value.<sup>1</sup>

The first four basis functions of the 2-MGE are easily determined from the expressions given in Eqs. 10, but the last four are product functions, and therefore include sum and difference Fourier functions in the Bessel function expansions. The result is that many Fourier components are created with a significant amplitude (see Table I of ref. 1); for two PEM's operating at 50 and 60 kHz, the time-dependent intensity includes 31 Fourier components at frequencies less than 240 kHz, all with a significant amplitude.

Information concerning the values of the individual elements of the sample Mueller matrix is included in the measured values of the eight coefficients of the time-dependent basis functions. The elements of the sample Mueller matrix can, in turn, be related to the ellipsometric parameters of the sample. The particular elements of the sample Mueller matrix will depend upon the azimuthal orientations of the PSG and the PSD. Schematically, the measured elements of the sample Mueller matrix can be represented by:

$$\mathbf{M}(\theta_{m0}=0,90;\theta_{m1}=0,90)=\begin{pmatrix} 1 & \bullet & I_{Y0} & I_{X0} \\ \bullet & \bullet & \bullet & \bullet \\ I_{Y1} & \bullet & I_{Y0Y1} & I_{X0Y1} \\ I_{X1} & \bullet & I_{Y0X1} & I_{X0X1} \end{pmatrix}; \quad \mathbf{M}(\theta_{m0}=0,90;\theta_{m1}=\pm 45)=\begin{pmatrix} 1 & \bullet & I_{Y0} & I_{X0} \\ I_{Y1} & \bullet & I_{Y0Y1} & I_{X0Y1} \\ \bullet & \bullet & \bullet & \bullet \\ I_{X1} & \bullet & I_{Y0X1} & I_{X0X1} \end{pmatrix} \quad (11a,b)$$

$$\mathbf{M}(\theta_{m0}=\pm 45;\theta_{m1}=0,90)=\begin{pmatrix} 1 & I_{Y0} & \bullet & I_{X0} \\ \bullet & \bullet & \bullet & \bullet \\ I_{Y1} & I_{Y0Y1} & \bullet & I_{X0Y1} \\ I_{X1} & I_{Y0X1} & \bullet & I_{X0X1} \end{pmatrix}; \quad \mathbf{M}(\theta_{m0}=\pm 45;\theta_{m1}=\pm 45)=\begin{pmatrix} 1 & I_{Y0} & \bullet & I_{X0} \\ I_{Y1} & I_{Y0Y1} & \bullet & I_{X0Y1} \\ \bullet & \bullet & \bullet & \bullet \\ I_{X1} & I_{Y0X1} & \bullet & I_{X0X1} \end{pmatrix} \quad (11c,d)$$

where the sign is not included in the representation. If an element of the sample Mueller matrix has a filled-in dot (•) then that particular element cannot be measured in the given configuration. If an element of the sample Mueller matrix can be measured, then the appropriate normalized constant term is shown.

For a simple isotropic sample where there are no windows, the sample Mueller matrix is given by

$$\mathbf{M}=\begin{pmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{pmatrix}, \quad (12a)$$

where the  $N$ ,  $S$ , and  $C$  parameters are given by

$$N = \cos(2\psi) \quad (12b)$$

$$S = \sin(2\psi) \sin \Delta, \quad (12c)$$

$$C = \sin(2\psi) \cos \Delta. \quad (12d)$$

These parameters are related to the traditional ellipsometry parameters  $\psi$  and  $\Delta$  by

$$\rho = \frac{r_{pp}}{r_{ss}} = \tan(\psi) e^{i\Delta} = \frac{C + iS}{1 + N}, \quad (12e)$$

where  $r_{pp}$  and  $r_{ss}$  are the complex reflection coefficients for light polarized in the plane of incidence or perpendicular to the plane of incidence, respectively. Therefore, the sample Mueller matrix for an isotropic sample can be completely characterized if  $\theta_{m0} = \pm 45^\circ$ ;  $\theta_{m1} = 0^\circ, 90^\circ$  or if  $\theta_{m0} = 0^\circ, 90^\circ$ ;  $\theta_{m1} = \pm 45^\circ$ , where several of the measured parameters will be zero.

### 3.1 Straight-through configuration

When the 2-MGE is placed in the straight-through configuration, the sample is just free space, where the Mueller matrix is the identity matrix ( $N = S = 0$ ;  $C = 1$ ). If the azimuthal angles of each of the polarizers are set close to  $\pm 45^\circ$ , the eight constant prefactors of Eqs. 9 are given by

$$I_{X0} = 0, \quad (13a)$$

$$I_{Y0} = P_{01} [J_0(A_1) \cos(2\theta_m) - 2 \sin(2\theta_m) \varepsilon_{b1}], \quad (13b)$$

$$I_{X1} = 0, \quad (13c)$$

$$I_{Y1} = P_{01} [J_0(A_0) \cos(2\theta_m) + 2 \sin(2\theta_m) \varepsilon_{b0}], \quad (13d)$$

$$I_{X0X1} = -P_{01}, \quad (13e)$$

$$I_{X0Y1} = -P_{01} [\delta_I + \delta_0 \cos(2\theta_m)], \quad (13f)$$

$$I_{Y0X1} = -P_{01} [\delta_0 + \delta_I \cos(2\theta_m)], \quad (13g)$$

$$I_{Y0Y1} = P_{01} \cos(2\theta_m), \quad (13h)$$

where  $P_{01} = \sin(2\theta_{b0}) \sin(2\theta_{b1}) = \pm 1$  and the static strain-induced retardation has been incorporated into the 8 coefficients. The angle  $\theta_m$  is the angle of the PSG with respect to the PSD, and  $\varepsilon_{b0}$  and  $\varepsilon_{b1}$  are the errors of  $\theta_{b0}$  and  $\theta_{b1}$  from  $\pm 45^\circ$ . The quantities  $J_0(A_0)$  and  $J_0(A_1)$  are the 0<sup>th</sup> order integer Bessel functions at angles  $A_0$  and  $A_1$ , where it is assumed that  $A_0$  and  $A_1$  are set near 2.4048 radians, where  $J_0(A_0)$  and  $J_0(A_1)$  are small. Note that all elements except  $I_{X0X1}$  and  $I_{Y0Y1}$  are close to zero.

When the 2-MGE is set such that the PEM's are aligned or perpendicular to each other, then  $\theta_m = 0^\circ$  or  $90^\circ$  and  $\sin(2\theta_m) = 0$  and  $\cos(2\theta_m) = \pm 1$ . In this situation,  $I_{Y0}$  and  $I_{Y1}$  are proportional to  $J_0(A_1)$  and  $J_0(A_0)$ , respectively. Since

$$J_0(A_i) = -0.5196 (2.4048 - A_i), \quad (14)$$

the measurement of  $I_{Y0}$  and  $I_{Y1}$  [and therefore  $J_0(A_0)$  and  $J_0(A_1)$ ] is a direct and very sensitive method for measuring the deviation of  $A_i$  from 2.4048. Such sensitivity is not available from single PEM ellipsometers.

Similarly, if the two PEM's are aligned at  $\pm 45^\circ$  with respect to each other, then  $\sin(2\theta_m) = \pm 1$  and  $\cos(2\theta_m) = 0$ . In this case,  $I_{Y0}$  and  $I_{Y1}$  are proportional to  $\varepsilon_{b1}$  and  $\varepsilon_{b0}$ , respectively, which are the errors of  $\theta_{b1}$  and  $\theta_{b0}$  from  $\pm 45^\circ$ .

Therefore, the  $\pm 45^\circ$  alignment can be used to set  $\theta_{b0}$  and  $\theta_{b1}$  to precisely  $\pm 45^\circ$ . Again, this sensitivity is not available with one-modulator systems, where one has to first establish the  $0^\circ$  and/or the  $90^\circ$  positions of the polarizer with respect to the PEM and then rely on the precision of the rotator to get the  $\theta_b$  to  $\pm 45^\circ$ . In addition;  $I_{XOYI} = \pm \delta_I$  and  $I_{YOXI} = \delta_O$ , which allows for the precise measurement of the static strain-induced retardation as a function of wavelength.

All of these calibrations do not depend upon getting the PEM's precisely aligned or set at  $\pm 45^\circ$  with respect to each other, since errors in  $\theta_m$  do not enter in first order. Residual errors in  $\theta_m$  are always multiplied by another small number, so the product is always small to second order.

### 3.2 Ellipsometry Configuration

In the ellipsometry configuration with an isotropic sample and without windows

$$I_{Y0} = - [N \sin (2 \theta_{m0}) - C J_0(A_1)] , \quad (15a)$$

$$I_{Y1} = - [N \sin (2 \theta_{m1}) - C J_0(A_0)] , \quad (15b)$$

where it is assumed that  $\theta_{b0} = \theta_{b1} = 45^\circ$ . If  $J_0(A_0) = J_0(A_1) = 0$ , then these two parameters can be used to set the azimuthal angles of the PEMs  $\theta_{m0}$  and  $\theta_{m1}$  precisely with respect to the plane of incidence. This calibration is not possible if the sample is anisotropic, since the anisotropy may also contribute to  $I_{Y0}$  and  $I_{Y1}$ . Clearly, the most accurate calibrations of  $\theta_{m0}$  and  $\theta_{m1}$  occur when  $N$  is large.

During any ellipsometry configuration measurement of a 2-MGE, it is possible to monitor the value of the Bessel angle  $A$  by monitoring the higher harmonics. For example, if the coefficient  $I_{Y0}$  were large, then the value of  $A_0$  could be measured by determining both the  $2\omega_0$  and the  $4\omega_0$  components. If the intensities of these two components are  $I(2\omega_0)$  and  $I(4\omega_0)$  respectively, then

$$R = \frac{I(4\omega_0)}{I(2\omega_0)} = \frac{f_{4\omega_0} J_4(A_0 + \delta A)}{f_{2\omega_0} J_2(A_0 + \delta A)} = \frac{f_{4\omega_0}}{f_{2\omega_0}} \frac{J_4(A_0)}{J_2(A_0)} \left( 1 + \delta A \left( \frac{1}{J_4(A_0)} \frac{dJ_4(A_0)}{dA} - \frac{1}{J_2(A_0)} \frac{dJ_2(A_0)}{dA} \right) \right) \quad (16a)$$

or

$$A = A_0 + \delta A = A_0 + (6.416 R \frac{f_{2\omega_0}}{f_{4\omega_0}} - 0.9609) \cdot \quad (16b)$$

In Eqs. 16, the quantities  $f_{2\omega_0}$  and  $f_{4\omega_0}$  are the electronic gains at the frequencies  $2\omega_0$  and the  $4\omega_0$  components, respectively. Similar expressions can be developed for any of the frequency components that are reasonably large. While this technique can always be used, care must be taken to also monitor the errors in both the measured parameters and in the quantities  $f_{2\omega_0}$  and  $f_{4\omega_0}$ .

Obviously, this technique could also be used with the 1<sup>st</sup> and 3<sup>rd</sup> harmonics (to measure the  $I_{X0}$  and the  $I_{X1}$  components), but this requires that the associated element of the sample Mueller matrix be large. With one-modulator ellipsometers, these harmonics can be used to also measure  $A$  whenever  $|S|$  is large, but two-modulator ellipsometers require the  $m_{14}$  and the  $m_{41}$  components to be large, which rarely happens.

### 3.3 Windows

If there are windows or lenses between the sample and the PSG and/or the PSD, then the retardation from these elements must be taken into account for very accurate measurements. If it can be assumed that the window strain is small (so that the linear approximation can be used), the sample-windows Mueller matrix becomes<sup>7</sup>

$$\mathbf{M}_G = \mathbf{M}_{w1} \mathbf{M} \mathbf{M}_{w0} = \begin{pmatrix} 1 & -N & 0 & S_0 N \\ -N & 1 & S_1 S & -S_0 - S_1 C \\ 0 & S_0 S & C - WS & S + WC \\ -S_1 N & S_1 + S_0 C & -(S + WC) & C - WS \end{pmatrix}. \quad (17a)$$

Each window is described by a static retardation  $\delta_{w0}$  and  $\delta_{w1}$  (which will depend upon wavelength in a similar manner as the PEM static retardation, given in Eq. 2c) and by a fast axis direction  $\theta_{w0}$  and  $\theta_{w1}$ . The window between the sample and the PSG is labeled  $w0$  and the window between the sample and the PSD is labeled  $w1$ . In Eq. 11,

$$S_0 = \delta_{w0} \sin(2\theta_{w0}), \quad (17b)$$

$$S_1 = \delta_{w1} \sin(2\theta_{w1}), \quad (17c)$$

$$W = \delta_{w0} \cos(2\theta_{w0}) + \delta_{w1} \cos(2\theta_{w1}). \quad (17d)$$

The cosine terms are not independent, and always enter to first order as the term  $W$ , so only three parameters must be specified to characterize the windows. However, the  $W$  parameter is difficult to measure in the ellipsometry configuration, since it is impossible to separate it from the measured values of  $S$  and  $C$ . One possible solution is to place the 2-MGE in the straight-through configuration, where it is known that  $N = S = 0$  and  $C = 1$ . In this case, configurations where either  $\theta_{m0}$  and/or  $\theta_{m1}$  are  $0^\circ$  or  $90^\circ$  can be used to measure  $W$ . The parameters  $S_0$  and  $S_1$  can be measured in the ellipsometry configuration if the sample is isotropic. This is particularly useful in that these parameters can be measured *in-situ* at the same time and configuration that the actual ellipsometric parameters  $N$ ,  $S$ , and  $C$  are measured.

## 4. CALIBRATIONS

The most helpful configuration for the calibration of the 2-MGE is the straight-through configuration, where the polarizer azimuthal angle  $\theta_p$ , the modulator static strain  $\delta_0$  and the modulator voltage  $V_m$  required to give a modulation amplitude of 2.4048 can all be easily determined. Aside from the calibration of the azimuthal angles of the PEM's with respect to the plane of incidence, this is the only required calibration for the instrument. Moreover, the PEM's are remarkably stable, so this calibration need only be done once unless extremely accurate measurements are required. Small drifts can be observed in the modulator voltage of  $<0.3\%$  and in the static strain-induced birefringence of  $<0.002$ , but these too can be eliminated with more frequent calibration.

The straight-through calibration is performed using four zones:  $\theta_{m0} = 0^\circ$  and  $\theta_{m1} = -45^\circ, 0^\circ, 45^\circ, 90^\circ$ . The  $\pm 45^\circ$  zones are used to measure the static retardation, while the  $0^\circ$  and the  $90^\circ$  zones are used to determine the  $J_0(A_i)$  for each modulator, which is then used to determine the required voltage to give  $A_i = 2.4048$ ,  $i = 0, 1$ .

Figure 1 shows an example of the values of  $J_0(A_i)$  obtained from the  $\theta_{m1} = 0^\circ$  and the  $\theta_{m1} = 90^\circ$  configurations. These values indicate how far  $A_i$  is off from 2.4048 (see Eq. 14) and the are then used to calculate the correct voltage to be applied to each of the PEM's to give a modulator amplitude  $A_i$  of 2.4048. This corrected voltage is shown in Fig. 2 for each of the modulators. The data in Fig. 2 does not extrapolate to zero, nor is it quite a straight line. Generally, the error in the set voltage is 0.001-0.002 volts, which results in an error in  $A_i$  of  $\pm 0.002$ -0.003. A 3-term Cauchy fit is usually sufficient to fit this data with a reduced  $\chi^2 \sim 1$ .

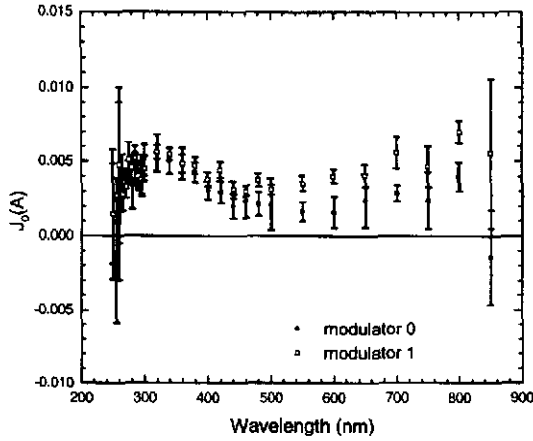


Fig. 1. The values of  $J_0(A)$  obtained from a straight-through configuration measurement where the azimuthal angular difference between the PSG and the PSD is  $0^\circ$  or  $90^\circ$ .

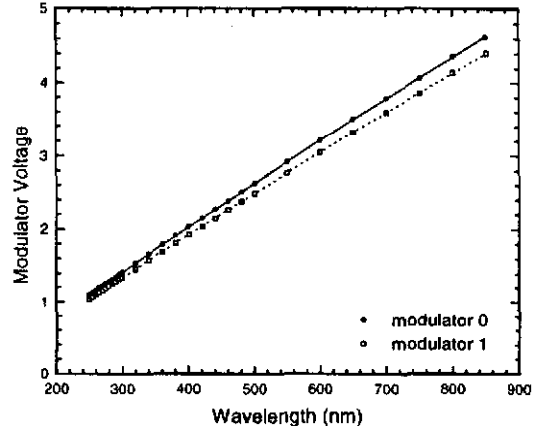


Fig. 2. The values of the applied voltage to each modulator required to obtain a modulator amplitude  $A$  of 2.4048, determined from the experimentally determined values of  $J_0(A)$  shown in Fig. 1.

In the  $\theta_{m1} = \pm 45^\circ$  configurations, the errors in the polarizer azimuthal angles are measured, as is the static strain-induced retardation. The errors in the polarizer azimuthal angles  $\theta_{bi}$  do not depend upon wavelength, so the results can be averaged over all data points taken. This results in a measurement of the error in  $\theta_{bi}$  that is accurate to  $\pm 0.01$ - $0.02^\circ$ . The strain-induced retardation is also measured in this configuration and is shown in Fig. 3 for both modulators, where the fitted line requires three Cauchy coefficients to get a reduced  $\chi^2 \sim 1$ .

Figure 4 shows a plot of typical windows parameters obtained for two fused silica lenses. The  $W$  parameter is obtained in the straight-through configuration, while the  $S_0$  and the  $S_1$  parameters are obtained in the ellipsometry configuration where the sample is an isotropic material such as crystalline silicon. Although the corrections are small, they are easily measurable, and very accurate measurements require that these corrections be applied.

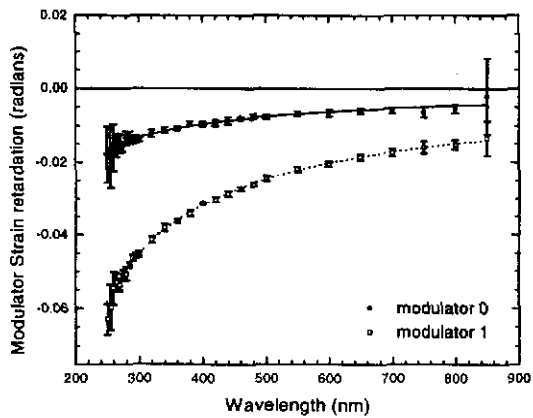


Fig. 3. The static strain-induced retardation for the two modulators used in the 2-MGE.

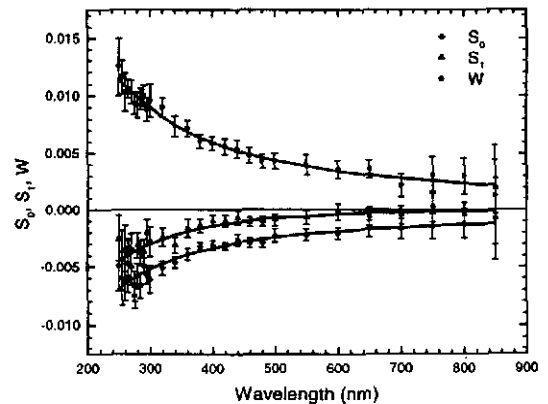


Fig. 4. Typical strain-induced retardation parameters for lenses used in 2-MGE measurements

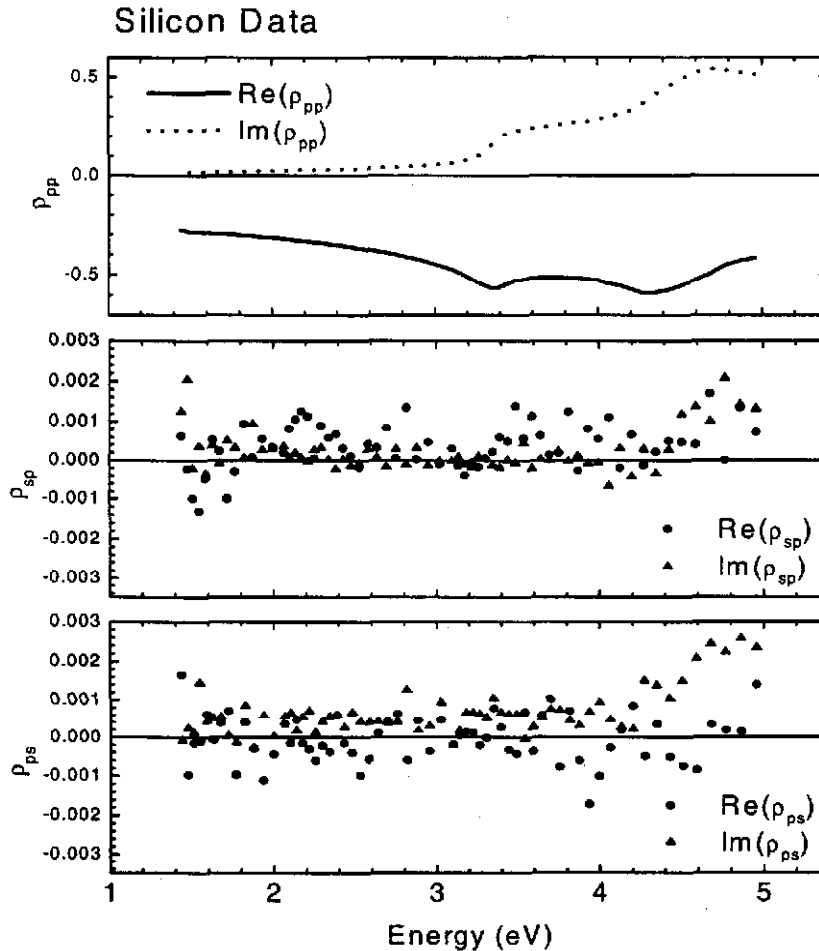


Fig. 5 Sample Rho data for silicon taken at an angle of incidence of 65.26°.

Sample data for silicon is shown in Figure 5. Any residual systematic errors in these calibrations tend to show up in the cross polarization elements, so a good test is to perform the measurements on an isotropic material such as silicon, where it is known that the cross-polarization reflection coefficients are zero. As can be seen, the cross-polarization terms are extremely small, and are less than 0.001 over the central part of the spectrum. (Reduced light in the UV and IR tends to increase the error in these parts of the spectrum.). Therefore, this measurement shows that the 2-MGE is capable of measuring the cross-polarization reflection coefficients very accurately and that calibration errors can essentially be eliminated from the measurement.

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## REFERENCES

1. G. E. Jellison, Jr. and F. A. Modine, "Two-modulator generalized ellipsometry: experiment and calibration," *Appl. Opt.* **36**, 8184-8189 (1997); "Two-modulator generalized ellipsometry: theory", *Appl. Opt.* **36**, 8190-8198 (1997).
2. G. E. Jellison, Jr. F. A. Modine, and L. A. Boatner, "Measurement of the optical functions of uniaxial materials by two-modulator generalized ellipsometry: rutile ( $\text{TiO}_2$ )," *Opt. Lett.* **22**, 1808-1810 (1997).
3. G. E. Jellison, Jr. and L. A. Boatner, "Optical functions of uniaxial  $\text{ZnO}$  determined by generalized ellipsometry," *Phys. Rev. B* **58**, 3586-3589 (1998).
4. G. E. Jellison, Jr., J. O. Ramey, and L. A. Boatner, "Optical functions of  $\text{BiI}_3$  as measured by generalized ellipsometry," *Phys. Rev. B* **59**, 9718-9721 (1999).
5. G. E. Jellison, Jr., F. A. Modine, and L. A. Boatner, "Measurements of the optical functions of uniaxial crystals using 2-modulator generalized ellipsometry (2-MGE), SPIE **3425**, 232-238 (1998).
6. Chi Chen, G. E. Jellison, Jr., and L. A. Boatner, "Optical functions of several tetragonal rare-earth phosphates determined from two-modulator generalized ellipsometry," manuscript in preparation.
7. G. E. Jellison, Jr., "Windows in ellipsometry experiments," *Appl. Opt.* (in press).
8. G. E. Jellison, Jr. and F. A. Modine, "Accurate calibration of a photo-elastic modulator in polarization modulation ellipsometry," SPIE **1166**, 231-241 (1989).
9. F. A. Modine and G. E. Jellison, Jr. "Errors in polarization measurements due to static retardation in photoelastic modulators," *Appl. Phys. Comm.* **12**, 121-139 (1993).
10. J. Badoz, M. P. Silverman, and J. C. Canit, *J. Opt. Soc. Am. A* **7**, 672 (1990).
11. D. S. Kliger, J. W. Lewis, and C. E. Randall, *Polarized Light in Optics and Spectroscopy*, (Academic Press, New York) (1990).
12. C. Brosseau, *Fundamentals of Polarized Light: A Statistical Optics Approach*, Wiley, New York, 1998.

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