# Null ellipsometer with phase modulation

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**Abstract:** A new null ellipsometer is described that uses photoelastic modulator (PEM). The phase modulation adds a good signal-to-noise ratio, high sensitivity, and linearity near null positions to the traditional high-precision nulling system. The ellipsometric angles $\Delta$ and $\psi$ are obtained by azimuth measurement of the analyzer and the polarizer–PEM system, for which the first and second harmonics of modulator frequency cross the zeros. We show that the null system is insensitive to ellipsometer misadjustment and component imperfections and modulator calibration is not needed. In addition, a fast ellipsometer mode for fine changes measurement of ellipsometric angles is proposed.

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## 1. Introduction

Null ellipsometry [1–3] is a classical ellipsometric technique based on measurement of azimuth angles of polarizer, compensator, and analyzer for which the detected intensity is extinguished. The null ellipsometer is highly accurate and almost free of systematic errors. The null method does not need intensity calibration and a polarized light source (or a polarization-sensitive detector) does not bring noticeable systematic errors. Moreover, zone averaging, *i.e.* averaging of obtained ellipsometric angles for different sets of azimuth angles corresponding to nulls, gives measurement insensitive to azimuths misadjustment and polarizing component imperfections. Problems of this method come from a weak signal near nulls and parabolic intensity dependence near null during a polarizer scan due to the Malus' law. This problem can be solved by including of azimuth modulation in null ellipsometry system. A method with Faraday magneto-optical cells was proposed by Winterbottom [4] and realized afterwords [5–7] A reason why the azimuth modulation null ellipsometry has not been widespread and commercialized comes probably from relatively low modulation amplitude of the Faraday cells, interference from stray ac magnetic fields, and spurious reflections in the many-component system.

On the other hand, automatic ellipsometers with high sensitivity and high acquisition rate are required for spectral measurements, *in situ* applications, and fast processes monitoring [8, 9]. Automatic ellipsometers can be divided into two categories: rotating polarizing component ellipsometers and phase-modulation ellipsometers. In the first category, systems with rotating polarizer, analyzer [10–12], and compensator [13, 14] are widely used. In the second category, a photoelastic modulator (PEM) with modulation frequency typically 50 kHz is used as a phase modulator [15–19]. PEM usually consists of a fused silica bar vibrating with natural resonant frequency sustained by a piezoelectric transducer. The periodic stress creates an optical anisotropy in the silica bar showing a photoelastic effect. The modulator, appearing to be the most critical element in the ellipsometer setup, should be carefully calibrated as a function of wavelength and temperature stabilized [16, 20, 21]. The automatic ellipsometers represent a shift from direct measurement of ellipsometric angles to light intensity measurement, *i.e.* a shift from ellipsometry to polarimetry [8]. However, intensity based ellipsometers require precise intensity calibration and precise system adjustment. Consequently, small imperfections in calibration or component adjustment can affect precision of measured data.

In this paper, we propose a new ellipsometric configuration based on the null method combined with the phase modulation. The null ellipsometer with phase modulation takes advantages of both configurations: (i) very high precision, insensitivity to component adjustment, and measurement almost free of systematic errors characteristic for null ellipsometry and (ii) high sensitivity, strong linear signal near nulls, and lock-in detection as advantages of modulation ellipsometric methods. Section 2 deals with theoretical description of the new ellipsometric system and its sensitivity to component imperfections. In Section 3 we propose a fast mode of the ellipsometer for measurement of fine changes of ellipsometric angles.

## 2. New configuration of null ellipsometry with phase modulation

This section deals with description of the new null ellipsometer with phase modulation. Description is based on the Jones matrix formalism. It is shown that the ellipsometric angles $\psi$ and $\Delta$ are directly related to the azimuth angles of polarizer and analyzer, which can be obtained by nulling of signals at second- and first-harmonic frequency of the modulator, respectively. In second part of this section we describe possibility of ellipsometric angles averaging for different null positions (zones) and sensitivity of the system to alignment (azimuth angles errors) and modulator and polarizing component imperfections.

### *2.1. Description of ellipsometer*

Figure 1 schematically shows basic components of the null PMSCA ellipsometric system. The system consists of the polarizer, which is mechanically connected to the modulator (PEM) and rotated by 45° from the modulator axis. Both components can slowly rotate and their azimuth angle $P$ can be precisely monitored during the rotation. Note that $P$ denotes the azimuth of the modulator optical axis. We propose to use the photoelastic modulator (PEM), which enables operation with high amplitude of modulation, appropriate modulation frequency, and spectral range. The retardation angle of PEM is the oscillating function of time

$$\varphi = \varphi_0 + \varphi_A \sin \omega t, \qquad (1)$$

where $\omega = 2\pi f$ is the angular frequency of the PEM phase oscillation, $\varphi_A$ denotes the modulation amplitude, and $\varphi_0$ corresponds to the residual birefringence due to PEM internal stress. The modulator is followed by a sample, which is characterized by the amplitude reflection coefficients $r_{ss}$ and $r_{pp}$ for $s$- and $p$-polarized light, respectively. The ellipsometric angles $\psi$ and $\Delta$ are defined using the ratio $r_{pp}/r_{ss} = \tan\psi \exp(i\Delta)$. Polarization state of reflected light from the sample is measured by a quarter-wave compensator (retardation angle $\phi = 90°$ and azimuth $C = \pm 45°$) and an analyzer with the adjustable azimuth angle $A$. For spectral measurement

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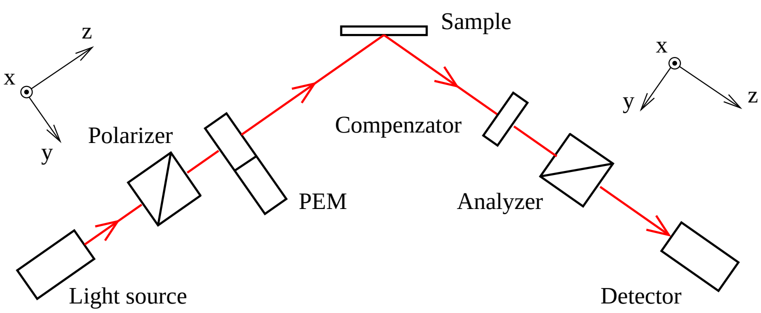


Fig. 1. Schematic description of null PMSCA ellipsometric system consisting of Polarizer–Modulator–Sample–Compensator–Analyzer. Coordinate systems are shown.

the achromatic compensator is needed, we propose to use, for example, the V-shaped Fresnel rhomb, or the zero-order achromatic waveplates [22, 23]. The Jones vector describing polarization of light incident on the detector can be calculated as the matrix product

$$\begin{bmatrix} E_x \\ E_y \end{bmatrix} = \frac{E_0}{\sqrt{2}} \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \overbrace{\begin{bmatrix} \cos A & \sin A \\ -\sin A & \cos A \end{bmatrix}}^{\text{Analyzer at azimuth } A} \overbrace{\begin{bmatrix} \cos(\phi/2) & \pm\sin(\phi/2) \\ \pm\sin(\phi/2) & \cos(\phi/2) \end{bmatrix}}^{\text{Compensator } C=\pm 45^\circ} \times$$

$$\times \overbrace{\begin{bmatrix} r_{ss} & 0 \\ 0 & r_{pp} \end{bmatrix}}^{\text{Sample}} \overbrace{\begin{bmatrix} \cos P & -\sin P \\ \sin P & \cos P \end{bmatrix}}^{\text{Azimuth of PEM}-\text{P}} \overbrace{\begin{bmatrix} \exp(i\varphi/2) & 0 \\ 0 & \exp(-i\varphi/2) \end{bmatrix}}^{\text{Modulator (PEM)}} \overbrace{\begin{bmatrix} 1 \\ 1 \end{bmatrix}}^{\text{Polarizer } 45^\circ}, \quad (2)$$

where the signs $\pm$ correspond to the azimuth angles of the compensator $C = \pm 45^\circ$ and $E_0$ denotes the amplitude of light wave coming from the polarizer. Detected intensity is obtained from Eq. (2) in the form

$$I = E_x E_x^* + E_y E_y^* = \frac{|E_0|^2 |r_{ss}|^2}{2} \left(I_0 + I_S \sin\varphi + I_C \cos\varphi\right), \quad (3)$$

where the asterisk $*$ denotes the complex conjugate and using $r_{pp}/r_{ss} = \tan\psi \exp(i\Delta)$

$$\begin{aligned} I_0 &= \left[1 + \cos 2A \cos\phi + (1 - \cos 2A \cos\phi) \tan^2\psi\right]/2, && (4) \\ I_S &= \tan\psi \left(\sin 2A \sin\Delta \pm \sin\phi \cos 2A \cos\Delta\right), && (5) \\ I_C &= -\sin 2P \left[1 + \cos 2A \cos\phi - (1 - \cos 2A \cos\phi) \tan^2\psi\right]/2 + \\ &\quad + \cos 2P \tan\psi \left(\sin 2A \cos\Delta \mp \sin\phi \cos 2A \cos\Delta\right). && (6) \end{aligned}$$

In terms of the Bessel functions of first kind $J_n$, the terms $\sin\varphi$ and $\cos\varphi$ from Eq. (3) can be expanded using Eq. (1) into the series

$$\begin{aligned} \sin\varphi &= J_0(\varphi_A) \sin\varphi_0 + 2J_1(\varphi_A) \cos\varphi_0 \sin\omega t + 2J_2(\varphi_A) \sin\varphi_0 \cos 2\omega t + \cdots, && (7) \\ \cos\varphi &= J_0(\varphi_A) \cos\varphi_0 - 2J_1(\varphi_A) \sin\varphi_0 \sin\omega t + 2J_2(\varphi_A) \cos\varphi_0 \cos 2\omega t + \cdots. && (8) \end{aligned}$$

The main idea of the proposed ellipsometric method is to search for nulls of the first $I_\omega$ and the second harmonic $I_{2\omega}$ signal. By substitution of Eqs. (7) and (8) into (3) the intensities at the first and second harmonic frequency of the modulator are proportional to $I_\omega \propto (I_S \cos\varphi_0 \,\text{-}\,\text{-}\, I_C \sin\varphi_0)$ and $I_{2\omega} \propto (I_S \sin\varphi_0 + I_C \cos\varphi_0)$. Consequently, the signals $I_\omega$ and $I_{2\omega}$ cross simultaneously the zeros for $I_S = 0$ and $I_C = 0$. In the case of ideal modulators ($\varphi_0 = 0$) $I_\omega \propto I_S$ and $I_{2\omega} \propto I_C$, whereas for real modulator ($\varphi_0 \neq 0$) a coupling between $I_S$ and $I_C$ arises.

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Table 1. Ideal zone positions for null PMSCA ellipsometer ($\phi = 90°$). The azimuth of analyzer $A$ is obtained by nulling of the first harmonic signal. The azimuth angle $P$ of the system polarizer–modulator corresponds to the null of the second harmonic.

| Zone | $C$ | $A\ (I_\omega = 0)$ | $P\ (I_{2\omega} = 0)$ | $\Delta$ | $\psi$ |
|---|---|---|---|---|---|
| 1 | $+45°$ | $A = \Delta/2 + \pi/4$ | $P = \psi$ | $\Delta = 2A - \pi/2$ | $\psi = P$ |
| 2 | $+45°$ | $A = \Delta/2 + \pi/4$ | $P = \psi + \pi/2$ | $\Delta = 2A - \pi/2$ | $\psi = P - \pi/2$ |
| 3 | $+45°$ | $A = \Delta/2 - \pi/4$ | $P = -\psi$ | $\Delta = 2A + \pi/2$ | $\psi = -P$ |
| 4 | $+45°$ | $A = \Delta/2 - \pi/4$ | $P = -\psi + \pi/2$ | $\Delta = 2A + \pi/2$ | $\psi = -P + \pi/2$ |
| 5 | $-45°$ | $A = -\Delta/2 + \pi/4$ | $P = \psi$ | $\Delta = -2A - \pi/2$ | $\psi = P$ |
| 6 | $-45°$ | $A = -\Delta/2 + \pi/4$ | $P = \psi + \pi/2$ | $\Delta = -2A - \pi/2$ | $\psi = P - \pi/2$ |
| 7 | $-45°$ | $A = -\Delta/2 - \pi/4$ | $P = -\psi$ | $\Delta = -2A + \pi/2$ | $\psi = -P$ |
| 8 | $-45°$ | $A = -\Delta/2 - \pi/4$ | $P = -\psi + \pi/2$ | $\Delta = -2A + \pi/2$ | $\psi = -P + \pi/2$ |

Condition $I_S = 0$, which relates mainly to null of $I_\omega$ signal, can be adjusted by rotation of the analyzer [see Eq. (5)]. The analyzer azimuth angle $A$ directly relates to $\Delta$ by the formula

$$\tan\Delta = \mp \frac{\sin\phi}{\tan 2A} = \sin\phi\,\tan\left[\pm(2A \pm \pi/2)\right], \tag{9}$$

where the first sign is related to the azimuth of compensator $C = \pm 45°$ and the second sign in the term $(2A \pm \pi/2)$ corresponds to the periodicity of the function tangents. Similarly, the condition $I_C = 0$, which relates mainly to the null of $I_{2\omega}$ signal, can be adjusted by rotation of the polarizer–PEM system [see Eq. (6)]. The azimuth angle $P$ directly relates to $\psi$ by equations

$$\begin{aligned} &\tan\psi = \pm\alpha_{\mp}\tan P, \qquad \tan\psi = \pm\alpha_{\mp}\tan(P - \pi/2), \qquad \text{for} \quad C = +45° \\ &\tan\psi = \pm\alpha_{\pm}\tan P, \qquad \tan\psi = \pm\alpha_{\pm}\tan(P - \pi/2), \qquad \text{for} \quad C = -45° \end{aligned} \tag{10}$$

$$\alpha_{\pm} = \frac{\sqrt{1 - \cos^2\phi\cos^2\Delta} \pm \cos\phi\,\sin\Delta}{\sin\phi}, \qquad \alpha_+\alpha_- = 1, \qquad \alpha_{\pm}(\phi = 90°) = 1, \tag{11}$$

where the formula $\tan 2P = 2\tan P/(1 - \tan^2 P)$ and Eq. (9) were used. Two solutions and the signs in Eq. (10) correspond to the periodicity of the functions $\tan 2A$ and $\tan 2P$. Different signs in Eqs. (9) and (10) correspond to different zones, *i. e.*, different azimuth angles $P$, $A$, for which the null intensity is obtained. Table 1 summarizes the different zones obtained by the null ellipsometer with phase modulation in the case of ideal compensator ($\phi = 90°$).

### 2.2. *Zone averaging and influence of component imperfections*

This section deals with description of systematic errors coming from component imperfections and ellipsometer misalignment. We show that the proposed ellipsometer is almost insensitive to the systematic errors, which which is a result of nulling-based measurement and zone averaging.

The main advantage of the null ellipsometer proposed is insensitivity to the PEM modulation amplitude $\varphi_A$ according to Eqs. (9)–(11). Consequently, intensity calibration is not needed and $\varphi_A$ can be roughly adjusted to get maximum modulation. Similarly, one can show that the angle between the polarizer and the PEM axes does not affect the positions of nulls. Its adjustment to 45° only maximizes the signal-to-noise ratio and the adjustment is not critical. Moreover, the proposed nulling system shows insensitivity to the PEM residual birefringence $\varphi_0$. Note that for considerable $\varphi_0$, the coupling between $I_S$ and $I_C$ arises and several iterations on $A$ and $P$ azimuth adjustments may be needed in practice to reach ideal null positions.

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As a next source of systematic error, compensator retardation error is discussed, which is important mainly in spectral measurements. According to Eqs. (10) and (11) the effect of imperfect compensator retardation $\phi$ on $\psi$ can be successfully eliminated by averaging of different zones. Influences of the imperfection are opposite in zones 1,2,7,8 and 3,4,5,6 described in Tab. 1. According to Eq. (11) we propose to eliminate completely the compensator imperfection by a geometric averaging of $\tan\psi$ from different zones, which is in correlation to the method for standard null ellipsometry proposed by Yamaguchi [24, 25]. On the other hand, the second order (quadratic) influence of the compensator imperfection to $\Delta$ can be eliminated using Eq. (9) only if spectral dependence of $\phi$ is known.

Moreover, zone averaging eliminates the azimuth misadjustment of the compensator, analyzer, and the polarizer–modulator system. One can show that the signs errors of $\psi$ coming from compensator misadjustment are opposite in the zones 1,2,5,6 and 3,4,7,8. We again propose the geometric averaging of $\tan\psi$ for complete error compensation. The error of $\Delta$ originating from $C$ shows also opposite signs in zones with $C = \pm 45^\circ$. Consequently, the zone averaging makes the measurement almost insensitive to the compensator azimuth error. Similarly, the azimuth errors of $P$ and $A$ are also eliminated by zone averaging. Despite the measurement is insensitive to the initial azimuth misadjustments, we note that the differences of azimuth angles have to be measured precisely. In some cases of practical interest, the sample shows also imperfections coming from its anisotropy (off diagonal elements of the Jones matrix) and depolarization. The proposed zone averaged measurement is insensitive to small parasitic anisotropy. Using the Mueller matrix [1, 3] or the coherence matrix formalism [26] one can show complete insensitivity of the proposed null method to the sample depolarization.

## 3. Measurement of fine ellipsometric angles changes

Fast, or *in-situ* monitoring of fine $\psi$ and $\Delta$ changes can be performed in an ellipsometer mode proposed in this section. The ellipsometric angles are described as the sums of unperturbed values and small perturbations: $\psi = \psi_0 + \delta\psi$ and $\Delta = \Delta_0 + \delta\Delta$ . The measurement can be performed in two steps. (i) the null is adjusted in the same configuration as discussed in previous section by adjusting angles $A$, $P$ corresponding to $\Delta_0$ and $\psi_0$, respectively. (ii) the fine changes of $\delta\psi$ and $\delta\Delta$ can be monitored using measurement of the first and second harmonic intensity of the modulator. The normalized intensities are in the forms:

$$I_S/I_0 = \pm\delta\Delta \sin 2\psi_0, \qquad I_C/I_0 = \pm 2\,\delta\psi. \tag{12}$$

Note that in this mode the system calibration and precise adjustment is needed. For easy calibration we propose to adjust the modulation amplitude $\varphi_A = 137.79^\circ$ for which $J_0 = 0$, $J_1 = 0.51915$, and $J_2 = 0.43175$. Moreover, if the measured process can be repeated, measurements in different zones increases precision and insensitivity to imperfections. This measurement mode can be used, for example, in magneto-optical ellipsometry, where the transverse Kerr effect can be represented as small deviation of $\psi$ and $\Delta$

## 4. Conclusion

New null ellipsometer with PEM has been proposed. Two mode of ellipsometer operation have been discussed: (i) high precision null measurement insensitive to component imperfection and misadjustment and (ii) fast intensity based monitoring of fine $\psi$ and $\Delta$ changes.

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