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A three-intensity technique for polarizer–sample-analyser photometric ellipsometry and polarimetry

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Abstract. This work presents a novel three-intensity-measurement technique to determine the ellipsometric parameters ψ and Δ in a polarizer–sample–analyser photometric ellipsometer. This technique can be employed to correct the azimuthal misalignment of the analyser with respect to the plane of incidence. By performing two sets of measurements with this technique with the polarizer's azimuth at +45° and −45°, respectively, we can simultaneously determine the azimuthal deviation of the polarizer and further improve the ellipsometric measurements. Applying this technique in the transmission mode allows us to obtain the phase retardation and the optical axis of a waveplate at the same time.

1. Introduction

A measurement technique known as ellipsometry [1, 2] can be used to deduce the optical properties of materials by measuring the changes in polarization of the reflected light. The measured ellipsometric parameters ψ and Δ are used to determine the optical parameters, such as the complex refractive index [3] and film thickness [4] of the sample. The rotating-element ellipsometer [5–7] is now known to be more competitive than the conventional null ellipsometer for automation. Owing to the intrinsic dispersion property of a compensator, rotating-analyser ellipsometry (RAE) has been used for spectroscopic ellipsometry [8]. Two methods to determine the ellipsometric parameters in the RAE from the measured intensity have been proposed: the least-squares method [9] and Fourier transform [5] technique. Both techniques utilize the over determination technique to enhance the accuracy of measurement, even though it is known [10, 11] that only three intensity measurements are needed for determining the ellipsometric parameters in this polarizer–sample–analyser (PSA) arrangement. However, a three-intensity-measurement method has been applied to video polarimetry [12] for mapping the 2D polarization state of a natural environment. Their three intensity measurements were obtained with the azimuthal angles of the analyser differing from each other by 60°. Our previous work applied a similar technique [13] to determine the optical axis of an anisotropic medium, indicating that the determination was independent of the polarizer's quality (for example, the degree of polarization). Meyer *et al* [9] proposed an equation to represent the intensity distribution of the elliptically polarized light in a PSA set-up. By applying the least-squares method to 36 measurements in a cycle, they obtained the intensity distribution and deduced the ellipsometric parameters in a PSA system. In this work, we demonstrate that the three intensity measurements are sufficient to obtain the equation of the elliptically distributed intensity, thereby reducing the number of measurements needed for obtaining the ellipsometric parameters. These three intensity measurements are evenly distributed over half a cycle, allowing the determination to de-couple two primary errors; namely the imperfection and misalignment of the analyser. The system error attributed to the misalignment of the polarizer can be further reduced and is explained in the following section.

By performing the ellipsometric measurements at two azimuths of the polarizer that differ by 90°, Kawabata [14] determined the orientation of the optical components relative to the incidence plane and the ellipsometric parameters of the specimen simultaneously. This two-zone method can also be applied in this three-intensity-measurement technique. Two sets of three intensity measurements can be performed by setting the polarizer's azimuthal angle at 45° and −45° to the incident plane, respectively. The ellipsometric parameters obtained by the three-intensity-measurement technique are insensitive to the azimuthal misalignment of the polarizer; moreover, the concurrently obtained azimuth deviation of the polarizer provides a simple procedure for azimuthal adjustment of the polarizer.

This three-intensity-measurement method can not only reduce the number of measurements needed for calculating the ellipsometric parameters without losing its accuracy but also allow us to measure the azimuthal deviation of the polarizer with respect to the incident plane. This method's versatility is confirmed by measuring the thickness of a thin film (SiO_2) and the refractive index of a bulk medium (Pt). The azimuthal deviations determined by this technique closely correspond to those obtained in our previous studies [15].

This three-intensity-measurement method can be easily converted into transmission made to measure the anisotropic medium. Via this technique, the optical axis and phase retardation of a quarter-wave plate can be determined simultaneously. The fact that the azimuthal position of the polarizer can be easily determined before subjecting the waveplate to the transmission mode ensures that this method can be used to confirm whether the waveplate is only a linear birefringence material without any optical activity. In this study, we apply this technique to measure the optical axis and phase retardation of quartz and mica quarter-wave plates. Measuring the quartz waveplate allows us to observe the existence of the optical activity clearly.

2. Theoretical background

2.1. Reflection ellipsometry

The ellipsometric parameters ψ and Δ defined as

$$\tan\psi\, \mathrm{e}^{\mathrm{i}\Delta} = \frac{r_p}{r_s}$$

where r_p and r_s denote the reflection coefficients in the planes parallel (p) and perpendicular (s) to the incident plane, respectively [16].

$$I(P, A) = I_0[\sin^2 P \sin^2 A + \tan^2\psi \cos^2 P \cos^2 A + 1/2 \tan\psi \cos\Delta \sin(2P)\sin(2A)] \quad (1)$$

where the azimuths P and A are the transmission axes of the polarizer and analyser, respectively. Aligning the azimuth of the polarizer at 45° with respect to the incidence plane allows us to simplify the intensity distribution further to

$$J(A) = 0.5I_0[\sin^2 A + \tan^2\psi \cos^2 A + \tan\psi \cos\Delta \sin(2A)]. \quad (2)$$

Using the analyser's azimuth A versus the intensity, the reflected light can be expressed in a polar coordinate, thereby forming an elliptical distribution. Meyer *et al* [9] proposed that the intensity distribution of the reflected light can also be written as

$$J(A) = L\cos^2(A-\beta) + T\sin^2(A-\beta). \quad (3)$$

By comparing equations (2) and (3), the following relationships can be obtained:

$$\tan(2\beta) = -\cos\Delta \tan(2\psi) \quad (4)$$

$$\tan^2\psi = \frac{1 - R\cos(2\beta)}{1 + R\cos(2\beta)} \quad (5)$$

where $R = (L-T)/(T+L)$. Since L, T and β are the three quantities needed for determining the intensity distribution of equation (3), these three quantities can be obtained simply by measuring the intensity at three different azimuths of the analyser to obtain the three quantities. In this PSA ellipsometry, the errors are primarily caused by the imperfection and misalignment of the polarizers (of the polarizer and analyser, for example). These two effects can be de-coupled by properly selecting three azimuthal angles of the analyser for normalization of the intensity. Let A, $A+P$ and $A+G$ be the azimuthal settings of the analyser relative to the incidence plane. Then the total intensity $(J(A) + J(A+P) + J(A+G))$ for normalization should be independent of the azimuthal angle of the analyser. Thus $[1 + \cos(2P) + \cos(2G)]$ and $[\sin(2P) + \sin(2G)]$ must be zero. Under these conditions and with these trigonometric identities, it is easy to prove that the only choices of azimuthal angles are 60° and 120°, respectively. By using the intensity distribution of equation (3), the following equation can be obtained:

$$\tan[2(\beta - A)] = \frac{\sqrt{3}[J(A+60) - J(A+120)]}{2J(A) - J(A+60) - J(A+120)} \quad (6)$$

which is independent of R. Therefore, the determination of β is independent of the degree of polarization of the polarizer [13]. Hence, in this study, we measure the three radiances through three analysers evenly spaced by 60°. The azimuths of the polarizer and analyser can be well aligned relative to the incidence plane by the intensity ratio technique [15]. According to equation (6), one can show that

$$\tan(2\beta) = \frac{\sqrt{3}[J(60) - J(120)]}{2J(0) - J(60) - J(120)} \quad (7)$$

$$R\cos(2\beta) = \frac{2J(0) - J(60) - J(120)}{J(0) + J(60) + J(120)}. \quad (8)$$

The ellipsometric parameters can be obtained by substituting equations (7) and (8) into equations (4) and (5). The sign of $\cos\Delta$ cannot be determined in the PSA system. Hence, the handedness becomes the intrinsic problem of the system [7].

If the azimuth of the polarizer deviates by a small angle α, then, because $P = \pm 45° + \alpha$, equation (1) can be rewritten as

$$J(A) = 0.5I_0\{\sin^2 A + \tan^2\psi \cos^2 A \pm[(\sin^2 A - \tan^2\psi \cos^2 A)\sin(2\alpha) + \tan\psi \cos\Delta \sin(2A)\cos(2\alpha)]\}. \quad (9)$$

Similarly, equation (5) is modified to

$$\tan^2\psi = \frac{[1 - \sin(2\alpha)][1 - R\cos(2\beta)]}{[1 + \sin(2\alpha)][1 + R\cos(2\beta)]} \quad (10a)$$

for $P = 45° + \alpha$; and

$$\tan^2\psi = \frac{[1 + \sin(2\alpha)][1 - R'\cos(2\beta')]}{[1 - \sin(2\alpha)][1 + R'\cos(2\beta')]} \quad (10b)$$

for $P = -45° + \alpha$, where R' and β' are the corresponding parameters of equation (10a).

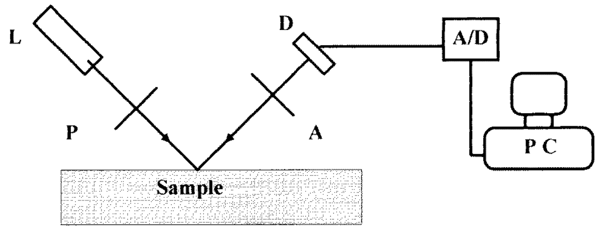

Figure 1. The schematic set-up of the PSA ellipsometer: L, light source (He–Ne laser); P, polarizer; A, analyser; and D, detector.

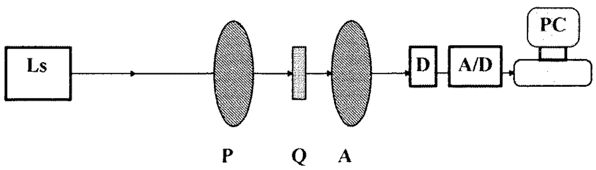

Figure 2. A schematic diagram of the polarimeter set-up. Ls, light source; P, linear polarizer; Q, quarter-wave plate; A, linear analyser; D, detector; and PC, computer.

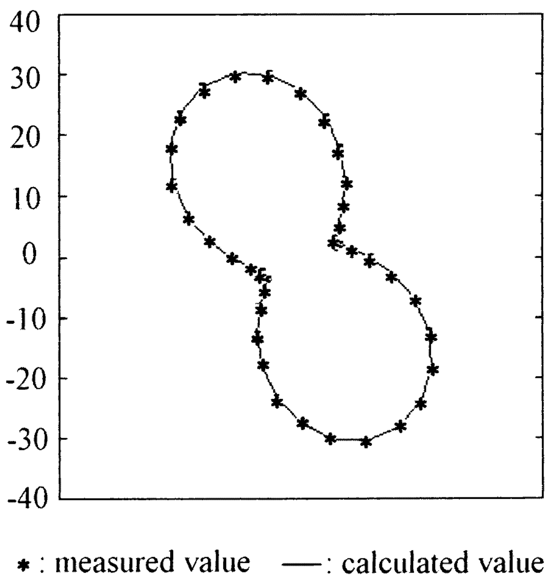

Figure 3. The intensity distribution in the polar coordinate of Pt at angle of incidence 70° and P = 45°: (∗) are the measured intensities and the full line is the calculated intensity when ψ = 32.77° and Δ = 125.5°.

The value of $\tan\psi$ is made independent of the azimuthal deviation (α) of the polarizer by taking the product of equations (10a) and (10b). The azimuthal deviation α can also be obtained by substituting equation (10a) into equation (10b); thus, the accuracy of Δ can be improved by taking $\beta_{ave} = (180 - \beta' + \beta)/2$.

2.2. Transmission ellipsometry

The Stokes vector $\boldsymbol{S}_i$ for linearly polarized light transmitted through a waveplate can be measured by a linear analyser. The measured Stokes vector $\boldsymbol{S}_f$ can be written as

$$\boldsymbol{S}_f = \mathbf{M}_A \mathbf{M}(C, \Delta) \boldsymbol{S}_i$$

where $\mathbf{M}(C, \Delta)$ denotes the Mueller matrix of a waveplate with a phase shift Δ and azimuth C of its fast axis. It is given [17] as

$$\mathbf{M}(C, \Delta) = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos^2(2C) + \cos\Delta \sin^2(2C) & \cos(2C)\sin(2C)(1 - \cos\Delta) & -\sin\Delta \sin(2C) \\ 0 & \cos(2C)\sin(2C) & \sin^2(2C) + \cos\Delta\cos^2(2C) & \sin\Delta\cos(2C) \\ 0 & \sin\Delta\sin(2C) & -\sin\Delta\cos(2C) & 0 \end{bmatrix}.$$

The Mueller matrix of an analyser $\mathbf{M}_A$ is given as

$$\mathbf{M}_A = \frac{1}{2}\begin{bmatrix} 1 & \cos(2A) & \sin(2A) & 0 \\ \cos(2A) & \cos^2(2A) & \cos(2A)\sin(2A) & 0 \\ \sin(2A) & \cos(2A)\sin(2A) & \sin^2(2A) & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}$$

where A represents the azimuthal position of the analyser measured from the axis of reference. The transmitted radiance at A can be calculated from $\boldsymbol{S}_f$ and written as

$$I_q(A) = \frac{I_0}{2}\{1 + \cos[2(A - C)]\cos[2(P - C)] + \cos\Delta \sin[2(A - C)]\sin[2(P - C)]\}. \quad (11)$$

If one removes the compensator, that is $\Delta = 0$, the transmitted radiance of equation (11) can be expressed as

$$I_p(A) = I_0 \cos^2(P - A) \quad (12)$$

which is Malus' law for the transmitted radiance of two polarizers. Let

$$I = \tfrac{2}{3}[I_p(0) + I_p(60) + I_p(120)]$$

$$I_0 = \tfrac{2}{3}[I_q(0) + I_q(60) + I_q(120)]$$

be the normalization intensities; then one can easily prove that

$$\tan(2P) = \frac{I_b}{I_a} \qquad \tan(2C) = \frac{\cos(2P) - I_a''}{I_b'' - \sin(2P)} \quad (13)$$

$$\cos\Delta = \frac{I_b'' - I_a''\tan(2C)}{\sin(2P) - \cos(2P)\tan(2C)} \quad (14)$$

where

$$I_a = [2I_p(0) - I_p(60) - I_p(120)]/I$$
$$I_a'' = [2I_q(0) - I_q(60) - I_q(120)]/I_0$$
$$I_b = \sqrt{3}[I_p(60) - I_p(120)]/I$$
$$I_b'' = \sqrt{3}[I_q(60) - I_q(120)]/I_0.$$

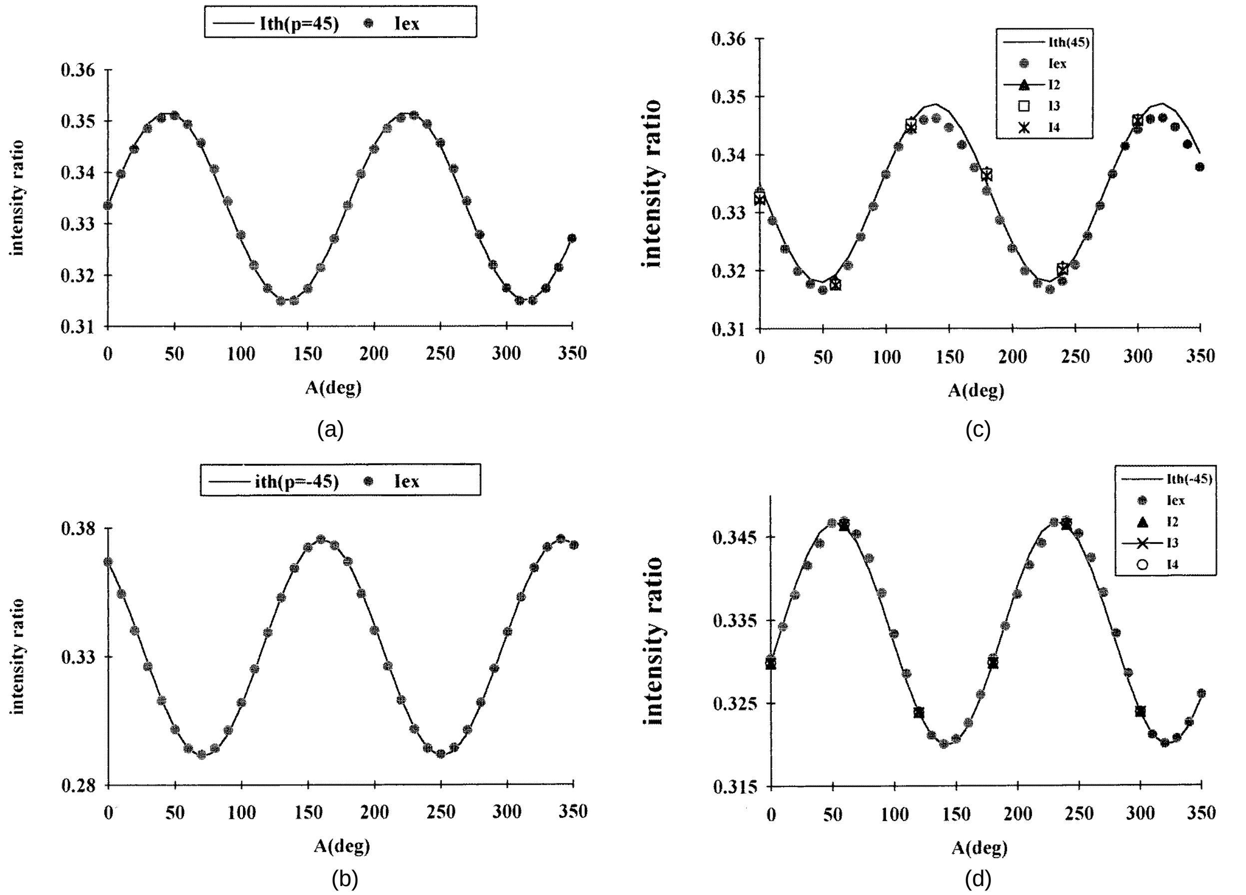

Figure 4. Intensity distributions of the output from the waveplates for various azimuthal angles (A) of the analyser; showing the azimuthal angle versus the intensity ratio: full lines are theoretical distributions and symbols are experimental values. For the quartz waveplate (a) $P = 45.07°$, $C = 0.07°$ and $\Delta = 86.76°$; and (b) $P = -44.95°$, $C = -2.98°$ and $\Delta = 86.07°$. For the mica waveplate (c) $P = 45.15°$, $C = 0.26°$, and $\Delta = 92.63°$; and (d) $P = -45.12°$, $C = 0.27°$ and $\Delta = 92.25°$.

3. Experimental procedures

Optical properties of a thermally grown SiO_2 thin film and a Pt film of over 500 Å thickness deposited on a silicon substrate are investigated by using the PSA photometric ellipsometer. Figure 1 depicts the experimental set-up. The light (L, from a He–Ne laser) passes through a polarizer (P) whose azimuthal angle is set to be $\pm 45°$ with respect to the incidence plane of the sample. The angle of incidence (θ_i) is taken to be $70 \pm 0.02°$ so that the results can be compared with those obtained by using the null ellipsometer (Rudolph AutoEL II). The analyser (A) is mounted on a rotator controlled by a stepping motor. One set of experiments consisting of 36 measurements is performed in one cycle for comparing with the theoretical deduction. Other sets of experiments consist of three measurements only, that is, of the intensities at the azimuth of the analyser at 0°, 60° and 120°, respectively. All intensities are measured using a power meter (D) (Newport 818-SL), digitized by a multimeter (Keithley 195A) and stored in a PC for calculating the ellipsometric parameters. After offsetting the azimuth of the polarizer by 1°, the same experimental procedures are repeated for examining the azimuthal deviation and its effects on the ellipsometric parameters.

Similar experiments are performed in transmission ellipsometry. Figure 2 depicts its set-up. Two compound quarter-wave plates (Melles Griot 02 WRQ 007 and Melles Griot 02 WRM 011) are measured. The fact that the azimuthal position of the polarizer can be determined independently in the transmission ellipsometry accounts for why, in this study, we measure the phase retardation and azimuth C of the waveplate by setting the azimuth P of the polarizer at around $\pm 45°$ to the horizontal axis; the azimuth P is measured before insertion of a waveplate.

4. Results

The values of $\tan\psi$ and $\cos\Delta$ were deduced from equations (3)–(6) by using three intensity measurements. By substituting the deduced values into equation (2), we plotted the intensity distribution in a polar coordinate, as shown in figure 3. This finding confirms that the measured intensities closely corresponded to the calculated intensity distribution, further implying that three intensity measurements are sufficient to deduce the ellipsometric parameters. Following Kawabata's idea [14], we further improved the results by performing the three intensity measurements at two azimuthal angles of the polarizer differing by 90° (namely at $\pm 45°$). Our results are

Table 1. Results of ellipsometric measurements of PSA photometric ellipsometry for a Pt film on a Si substrate. Standard deviations are in parentheses. The null method employed a Rudolph AutoEL II.

Azimuthal of P (degrees)	ψ (degrees)	ψ (degrees)	Δ (degrees)	Δ (degrees)	ψ (degrees)	Δ (degrees)	α (degrees)
	+45	−45	+45	−45	Improved	Improved	
1	32.839	34.348	125.584	120.702	33.589	123.116	0.093
2	32.845	34.346	125.571	120.728	33.591	123.123	0.095
3	32.839	34.347	125.577	120.699	33.588	123.110	0.093
4	32.839	34.344	125.582	120.714	33.587	123.121	0.094
5	32.845	34.348	125.570	120.702	33.592	123.109	0.092
Mean	32.840 (0.003)	34.346 (0.002)	125.579 (0.005)	120.711 (0.011)	33.588 (0.002)	123.118 (0.005)	0.094 (0.002)
Vary P by 1°	31.856 (0.007)	35.237 (0.005)	125.476 (0.03)	120.891 (0.02)	33.526 (0.003)	123.47 (0.02)	1.163 (0.003)
Null					33.58 (0.02)	123.34 (0.04)	

Table 2. The same as table 1 but for a thin film of SiO_2 on a Si substrate.

Azimuthal of P (degrees)	ψ (degrees)	ψ (degrees)	Δ (degrees)	Δ (degrees)	ψ (degrees)	Δ (degrees)	α (degrees)
	+45	−45	+45	−45	Improved	Improved	
1	51.342	50.578	80.232	81.378	50.960	80.80	−0.025
2	51.344	50.577	80.240	81.379	50.960	80.81	−0.029
3	51.334	50.578	80.249	81.389	50.956	80.82	−0.022
4	51.333	50.578	80.239	81.394	50.956	80.82	−0.017
5	51.344	50.578	80.240	81.378	50.961	80.81	−0.028
Mean	51.334 (0.005)	50.575 (0.006)	80.243 (0.009)	81.385 (0.02)	50.957 (0.004)	80.82 (0.02)	−0.022 (0.005)
Null					50.96 (0.01)	81.23 (0.02)	

comparable with those measured by using a conventional null ellipsometer, such as those shown in tables 1 and 2. These tables indicate that our method, while requiring no compensator, can still achieve less than 0.1% errors for both parameters. Herein, we repeat the measurements after offsetting the azimuthal angle of the polarizer by 1°; the obtained ellipsometric parameters deviate by 0.2% only and the initial azimuthal deviation of the polarizer is also revealed.

The phase retardation Δ and optical axis C of the waveplate are obtained by substituting three intensity measurements into equations (13) and (14). Table 3 summarizes those results. The theoretical intensity distribution can be obtained by substituting the measured Δ and C into equation (11). The 36 normalized radiances are compared with the theoretical intensity distribution as illustrated in figures 4(a)–(d). Again, the three intensity measurements are sufficient to determine the phase retardation and optical axis of the wave plates.

5. Concluding remarks

The complex refractive index of Pt film can be deduced from the ellipsometric parameters by considering it as a bulk medium. Those parameters are 2.065 − i3.924 from the three-intensity PSA ellipsometer and 2.089 − i3.955 from the null ellipsometer, respectively. We also measured the ellipsometric parameter ψ for the angles of incidence 45° and 70°, which are 40.96 and 33.59°, respectively. In addition, the intersection of two iso-tan ψ [18] curves is used to obtain the refractive index of the Pt film as 2.075 − i3.937. All these refractive indices deduced from various methods were within 1% of one another. The thickness of the thin film of SiO_2 deduced from the ellipsometric parameters is 1133 Å by the PSA and 1136 Å by the null method. The standard azimuthal deviation (α) closely resembles that which we obtained previously [15]. This three-intensity PSA photometric ellipsometry can achieve the same level of accuracy as the two-zone average null ellipsometry. The data deemed necessary for obtaining the ellipsometric parameters in the three-intensity

Table 3. The phase retardation and azimuthal angles of a quarter-wave plate under various azimuthal angles of the polarizer.

Quartz	P = 45.07(0.006)	−44.95
C	0.07(0.01)	−2.98(0.05)
Δ	86.76(0.04)	86.07(0.01)
Mica	P = 45.15	−44.98
C	0.26(0.04)	0.27
Δ	92.63(0.07)	92.25

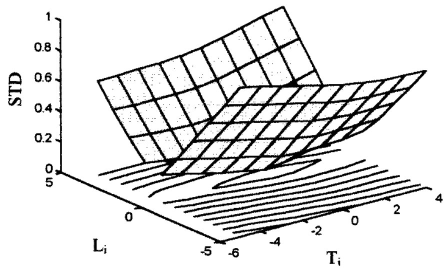

Figure 5. The standard deviation at various L and T, where the contour line is on the L–T plane.

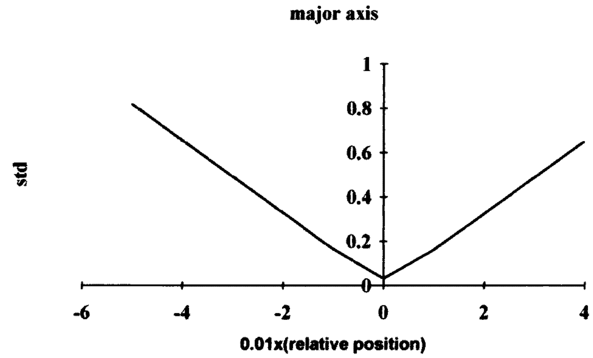

Figure 6. The standard deviation when $T = T_t$.

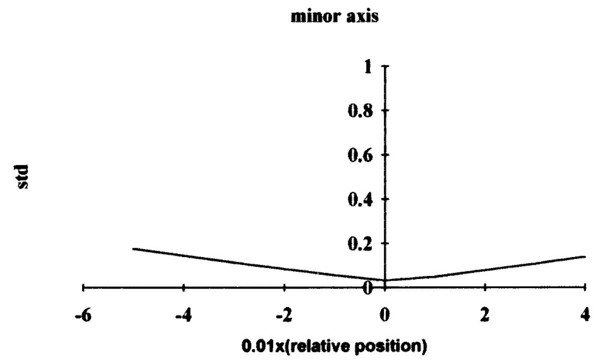

Figure 7. The standard deviation at $L = L_t$.

PSA photometric ellipsometry are less than those in the rotating-analyser ellipsometry (RAE). Therefore, it is more feasible to develop it for 2D measurements. Although we do not have a RAE to measure the same sample, our standard deviations of both ellipsometric parameters for Pt are comparable to those obtained by Kawabata [14] for gold. By using a set of 36 intensity measurements for a least-squares fit (appendix), we also found that our result has the least standard deviation, approximately the limit of the intensity fluctuation (8×10^{-4}) of the system. Moreover, the system can be improved if we improve the dynamic range of the detector and reduce the intensity fluctuation of the light source.

For an anisotropic medium, such as a waveplate, its phase retardation and azimuthal angle can be simultaneously measured by this three-intensity PSA technique. In theory, the phase retardation and azimuth C of the waveplate should be independent of the azimuthal setting of the polarizer. Herein, measuring the quartz-wave plate, both its phase retardation and its optical axis depend on the azimuthal setting of the polarizer. This discrepancy is much smaller for the mica waveplate. Although the quartz crystal is an optically active birefringence material [19], its optically active property is much smaller [20] than its linear birefringence property when it is used as a waveplate. Therefore, the optical activity has been neglected in most waveplate-related experiments. With this technique, the optical activity is clearly observable when one measures the optical axis of the first-order quartz quarter-wave plate. This enhanced optical activity should be further investigated.

Acknowledgments

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Appendix A. The least-squares fit for a set of Pt measurements

In the text, we have already proved that β deduced from the three-intensity technique is insensitive to the quality of the polarizer. By deviating L and T from the three-intensity-measured values, L_t and T_t, we take

$$L_i = L_t[1 + 0.01(5 - \mathrm{i})] \qquad T_i = T_t[1 + 0.01(5 - \mathrm{i})]$$

and substitute the values into

$$J_i(A) = L_i \cos^2(A - \beta) + T \sin^2(A - \beta).$$

The standard deviations between the theoretical values J_i and the 30 intensity measurements are calculated and plotted in figures 5–7. We can conclude that L_t and T_t are the least standard deviations of all.

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