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Design of a real-time spectroscopic rotating compensator ellipsometer without systematic errors

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ABSTRACT

We describe a spectroscopic ellipsometer in the visible domain (400–800 nm) based on a rotating compensator technology using two detectors. The classical analyzer is replaced by a fixed Rochon birefringent beamsplitter which splits the incidence light wave into two perpendicularly polarized waves, one oriented at +45° and the other one at −45° according to the plane of incidence. Both emergent optical signals are analyzed by two identical CCD detectors which are synchronized by an optical encoder fixed on the shaft of the step-by-step motor of the compensator. The final spectrum is the result of the two averaged Ψ and Δ spectra acquired by both detectors. We show that Ψ and Δ spectra are acquired without systematic errors on a spectral range fixed from 400 to 800 nm. The acquisition time can be adjusted down to 25 ms. The setup was validated by monitoring the first steps of bismuth telluride film electrocrystallization. The results exhibit that induced experimental growth parameters, such as film thickness and volumic fraction of deposited material can be extracted with a better trueness.

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1. Introduction

Nowadays, in situ monitoring of optical interfaces is a growing challenge both for fundamental studies in material science and for industrial purposes in terms of real time quality diagnostic of thin film processes. Then spectroscopic ellipsometry appears to be a powerful tool as a non-invasive, contactless and sensitive method. However, dynamic ellipsometric measurements are marred by random and systematic errors. For a rotating compensator ellipsometer, these errors have been studied [1–6]. In order to reduce random errors, a tracking method has been developed [5] and a common way to reduce systematic errors of ellipsometric angles is to calculate the mean of two measurements at two analyzer positions with the incidence plane −45° and +45° [2]. This method is called double-zone measurements. Generally, the analyzer is mounted on a motorized rotation stage. The rotation of the optical component from −45° to +45° requires few seconds and then it is not suitable for dynamic measurements. Recently Marsillac et al. [7] proposed a broadband in situ real time spectroscopic ellipsometry analysis of sputtered Ag nanoparticle films over a wide range (0.75–6.5 eV) with a rotating-compensator multichannel ellipsometer at an angle of incidence of 65° (with pairs of Ψ, Δ spectra collected each 3 s). Strong dependences of the particle size and film thickness were observed from the nucleation regime throughout coalescence and analyzed in separate modes evidenced in the dielectric function: intraband, particle plasmon polariton, and interband transitions. Using the same setup, Begou et al. reported first co-evaporated growth dynamics and accurate dielectric functions of $CuInSe_2$ [8] and recently of (Ag, Cu)InSe2 thin films [9]. Rodenhausen et al. [10] combined spectroscopic ellipsometry method with an acoustical one (Quartz Crystal Microbalance sensor). The authors determine the thickness and the porosity of transparent organic layers for thicknesses smaller than the wavelength of the probing light with bio-applications such as self-assembled monolayer chemisorption or DNA detection. Here, we propose an experimental setup taking the systematic errors into account and being compatible to in situ and real time measurements. In situ experiments of electrochemical growth of film were conducted on electrodes in order to study the interest of the proposed setup. Indeed, electrodeposition, as a cost-effective and high throughput fabrication process, is the method of choice to synthesize nanoengineered thermoelectric materials [11,12]. The transport properties of electroplated films are mainly due to their crystallinity and morphology and then to their related electrocrystallisation process [13–16]. The electrodeposition of bismuth telluride, which is widely used as thermoelectric material at room temperature [17,18], has been monitored by the setup presented in this paper.

2. Ellipsometer configuration

This spectroscopic ellipsometer is based on a Rotating Compensator Ellipsometer (RCE) [19]. The detected signal is the dot-product of the first row of the Mueller matrices of the optical configuration with the input Stokes vector. The detected irradiance, *I*, has the form:

$$I = I_0\left[a_0 + \sum_n \left(a_{2n}\cos 2nC + b_{2n}\sin 2nC\right)\right], \tag{1}$$

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Table 1
Systematic errors in RCE with $A = 45°$ or $A = -45°$. $sgnA = 1$ for $A > 0$ and $sgnA = -1$ for $A < 0$.

Origin	$\begin{bmatrix}\delta\Psi\\\delta\Delta\end{bmatrix}$	Origin	$\begin{bmatrix}\delta\Psi\\\delta\Delta\end{bmatrix}$
δA	$\begin{bmatrix}-sgnA\sin2\Psi\\0\end{bmatrix}$	γ_A	$2\begin{bmatrix}0\\sgnA\end{bmatrix}$
δP	$\begin{bmatrix}-sgnA\cos\Delta\\2sgnA\sin\Delta\cot2\Psi\end{bmatrix}$	γ_P	$\begin{bmatrix}sgnA\cos2P\sin\Delta\cos^2 2\Psi-\sin2P\cos\Delta\cos2\Psi\sin2\Psi\\2(-\sin2P\cos^2\Delta+sgnA\cos2P\cos\Delta\cot2\Psi)\end{bmatrix}$
δC	$2\begin{bmatrix}sgnA\cos\Delta\\-2sgnA\sin\Delta\cot2\Psi\end{bmatrix}$	γ_C	$\begin{bmatrix}0\\0\end{bmatrix}$

where a_{2n} and b_{2n} are the Fourier coefficients of the irradiance ($n = 1, 2$), I_0a_0 is the average incident-light irradiance and $C = \omega t$ is the angle of the fast axe of the compensator at time t.

The ellipsometric parameters of the sample are defined by:

$$\tan\Delta = \frac{\alpha_2\sin2P-\beta_2\cos2P}{\alpha_4\sin2P-\beta_4\cos2P}\left(\frac{1-\cos\delta_c}{2\sin\delta_c}\right)$$
$$\tan\Psi = \frac{K\cdot sgnA\cdot\sin2A\left(1\pm\pm\sqrt{1+1/K^2}\right)}{(1+\cos2A)}, \tag{2}$$

where

$$\frac{1}{K} = \sqrt{\frac{\left(\alpha_2^2+\beta_2^2\right)(1-\cos\delta_c)^2/\sin^2\delta_c+4(\beta_4\cos2P-\alpha_4\sin2P)^2}{2(\alpha_4\cos2P+\beta_4\sin2P)}},$$

where A and P are the position of the analyzer and the polarizer respectively, $sgnA = 1$ for $A > 0$ and $sgnA = -1$ for $A < 0$ and β_{2n} and α_{2n} are the Fourier coefficients of the detected irradiance with $\alpha_{2n} = G.a_{2n}$ and $\beta_{2n} = G.b_{2n}$ and G is considered to be the overall gain and is assumed to be real and frequency independent. δ_c is the phase retardation of the compensator.

For $A = +45°$ and $A = -45°$, the ellipsometric parameters are given by:

$$\tan\Delta_{A=+45°} = \tan\Delta_{A=-45°} = \frac{\alpha_2\sin2P-\beta_2\cos2P}{\alpha_4\sin2P-\beta_4\cos2P}\left(\frac{1-\cos\delta_c}{2\sin\delta_c}\right) \tag{3}$$

and

$$\tan\Psi_{A=+45°} = \tan\Psi_{A=-45°} = K\left(1\pm\pm\sqrt{1+1/K^2}\right). \tag{4}$$

All errors independent of their origins, propagate through the Fourier coefficients a_0, a_{2n} and b_{2n}. The systematic errors due to the positioning of the optical elements (i.e. δA, δP and δC) can be reduced by a suitable calibration procedure, but other errors due to the imperfection of the components (i.e. γ_A, γ_P, γ_C) affect the measurements. They are evaluated by calculating the Jacobian matrix relating $\delta\Delta$ and $\delta\Psi$ to the errors δa_0, δa_{2n} and δb_{2n} [2]. When the analyzer is set to +45° or −45° the systematic errors on Ψ and Δ for RCE are given in Table 1. We can see that the double-zone measurement is useful for eliminating errors due to a mispositioning of an optical element and errors due to the imperfection of the analyzer and the compensator. However, positioning the analyzer from +45° or −45° is too long (i.e. approximately 1 or 2 s) to be compatible with real-time measurements. In our experimental configuration the classical analyzer is replaced by a fixed Rochon birefringent beamsplitter which splits the incidence light wave into two perpendicularly polarized waves, one oriented at +45° (i.e. the undeflected beam $\mathbf{S}_{S+45°}$) and the other at −45° (i.e. the deflected beam $\mathbf{S}_{S-45°}$) according the plane of incidence (Fig. 1). Each light beam is analyzed by a Larry 2048 pixel CCD detector mounted on a Jobin-Yvon CP140 spectrograph. They acquire simultaneously light and are synchronized by an optical encoder fixed on the shaft of the step-by-step motor of the compensator. The minimum integration time (11 ms) and readout time (10 ms) of the 2048 pixels of each detector limit the acquisition of ellipsometric spectra to 25 ms (4 ms is reserved for calculating Ψ and Δ versus wavelength).

For each detector the ellipsometric spectra are determined and we proceed to an arithmetic average of Ψ and Δ spectra. Thus, no displacement of the Rochon birefringent beamsplitter is required. Table 2 shows that all errors except γ_p (i.e. polarizer imperfections) are canceled. For the latter, the particular position of the polarizer $P = 0°$ or $P = 90°$ vanishes the corresponding systematic errors on Ψ and Δ as shown in the following equations:

$$\delta\Psi = -\sin2P\cos\Delta\cos2\Psi\sin2\Psi\gamma_P, \tag{5}$$

$$\delta\Delta = -2\sin2P\cos^2\Delta\gamma_P. \tag{6}$$

By this method all systematic errors can be eliminated.

3. In situ electrodeposition monitoring: results and discussion

A homemade electrochemical cell was developed, where the working electrode acts as an Electromechanical Quartz Crystal Microbalance

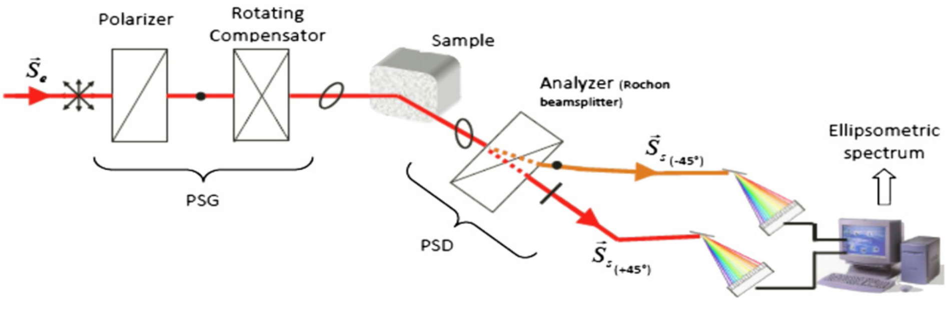

Fig. 1. Diagram of the real-time spectroscopic rotating compensator ellipsometer (RTSRCE).

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