Journal of New Technology and Materials
JNTM
Vol. 04, N°01 (2014)138-142

OEB Univ. Publish. Co.

Spectroscopic Ellipsometry characterization of thin films deposited on silicon substrate

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Received: 30 April 2014, accepted 26 May 2014

Abstract

In this work we used spectroscopic ellipsometry to study the optical properties of thin films deposited on silicon substrates. Analysis of the ellipsometric spectra allows determining the thickness of the deposited films and the dispersion of their optical indices. The proposed method is to develop a theoretical model to calculate the ellipsometric angles as function of the wavelength of the incident light. The theoretical model is based on the theory of propagation of electromagnetic waves in a stratified medium and effective medium approximation of Bruggman. The theoretical ellipsometric spectrum will be adjusted to the measured spectrum to determine the optical characteristics of the deposited films. In the case of silicon oxynitride thin films this analysis allows among others to determine the chemical composition of the deposited film and to detect any index gradient in the layer.

Keywords: *thin film, ellipsometry, silicon oxynitride, optical properties.*

1. Introduction

Spectroscopic ellipsometry is one of the most accurate and reliable optical techniques to characterize a large variety of materials and thin films on any substrate.Because today's devices such as solar cells[1, 2], light emitting devices[3, 4] and thin film transistors[5] are formed by complex multilayers structures, the correct use of ellipsometry requires appropriate models for data analysis.The Principe of modeling is to adjust the parameters of a theoretical model to minimize the difference between the measured and calculated spectra. Most of the problems encountered in the analysis of ellipsometric spectra are related to inadequate choice of the model rather than the quality of measurements.

Inthiswork, weproposedamathematicalmodelbased on the theory of propagation of electromagnetic waves in stratified media. The optical indices dispersion is described by the Bruggmanapproximationof effective media. This model was used to analyze ellipsometric spectra measured on thin films of silicon oxynitridesdeposited on silicon substrate.

Oxynitrides are particularly interesting compounds, intermediate between silica and nitride, they encompass mechanical and dielectrical qualities of silica, and present the advantage of serving as a diffusion barrier to impurities, like nitrides. These qualities give them opportunities in the field of microelectronics or to achieve antireflection which improves the performance of solar cells[6-15].

2. Theoretical model

Ellipsometry is a method of optical analysis based on the change of polarization state of light upon reflection on a flat surface. We send to the sample a polarized light and the change of polarization introduced by the sample is analyzed. These changes can be represented by two Fresnel coefficients r_s and r_p (Eq.1) [16-18], acting on each of the field components (Fig. 1).The reportof these coefficientsis defined inEq.2:

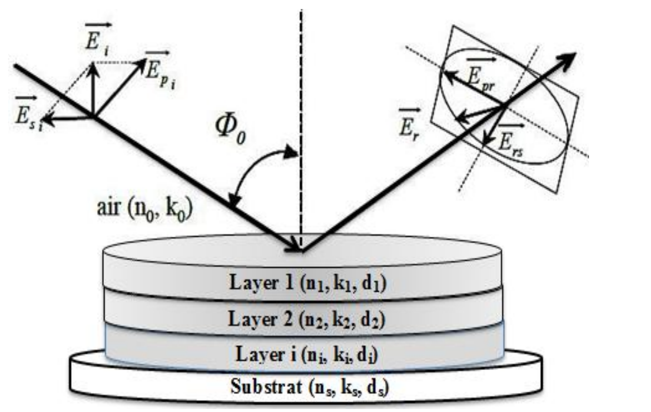

Fig. 1. Schematic view of an ellipsometric measurement, using multilayer optical system.

$$r_p = \frac{E_p^r}{E_p^i} = \frac{\left|E_p^r\right|}{\left|E_p^i\right|} e^{i(\delta_p^r - \delta_p^i)} = \left|r_p\right| e^{i\delta_p}$$

$$r_s = \frac{E_s^r}{E_s^i} = \frac{\left|E_s^r\right|}{\left|E_s^i\right|} e^{i(\delta_s^r - \delta_s^i)} = \left|r_s\right| e^{i\delta_s} \quad \text{(Eq. 1)}$$

$$\rho = \frac{r_p}{r_s} = \tan\psi e^{i\Delta} \quad \text{(Eq. 2)}$$

The measurables of ellipsometry are the phase and the light amplitude changes upon reflection which are denoted as$\psi$and Δ.

$$\tan\psi = \left|\frac{r_p}{r_s}\right| \text{ And } \Delta = \delta_p - \delta_s \quad \text{(Eq. 3)}$$

Ellipsometry is an indirect method; it does not give direct access to the physical parameters of the sample (optical thickness and the optical indices dispersion). It is necessary to use a model for describing the optical response of a sample. i.e. to calculate the reflection coefficients r_p and r_s and then find the Ψ and Δangles. To calculate the optical response of a stratified media, we used the matrix representation of Abeles [19] which represents each stratum by a matrix:

For S mode:

$$\begin{pmatrix} E_0^- \\ E_0^+ \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 & -\frac{1}{N_0 \cos\theta_0} \\ 1 & \frac{1}{N_0 \cos\theta_0} \end{pmatrix} \prod_{j=1}^{L} \begin{pmatrix} \cos\beta_j & \frac{i \sin\beta j}{\tilde{N}_j \cos\theta_j} \\ i\tilde{N}_J \cos\theta_j \sin\beta_j & \cos\beta_j \end{pmatrix} \begin{pmatrix} 1 \\ \tilde{N}_S \cos\theta_S \end{pmatrix}$$

And for P mode:

$$\begin{pmatrix} E_0^- \\ E_0^+ \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 & -\frac{\cos\theta_0}{N_0} \\ 1 & \frac{\cos\theta_0}{N_0} \end{pmatrix} \prod_{j=1}^{L} \begin{pmatrix} \cos\beta_j & \frac{i \sin\beta j \cos\theta_j}{\tilde{N}_j} \\ \frac{i\tilde{N}_J \sin\beta_j}{\cos\theta_j} & \cos\beta_j \end{pmatrix} \begin{pmatrix} 1 \\ \frac{\tilde{N}_S}{\cos\theta_s} \end{pmatrix}$$

Where

$$\beta_j = \frac{2\pi\tilde{N}_j d_j}{\lambda} \cos\theta_j$$

β_j the phase shift due to thejlayer is calculate from θ_j the incidence angle, $\tilde{N}_j$ the complex opticalindex and d_j thelayer thickness.

To model the optical indices of silicon oxynitrideswe used the effective media approximation of Bruggman (BEMA) (Eq.4) [20]. In this approximation, the deposited layers are considered as combinations of homogeneous mixtures of silicon nitride Si_3N_4 and SiO_2 silica. The optical indices are then calculated from the volume fractions $f_n$of SiO_2 and Si_3N_4 and their optical indices published by Palikin his handbook [21].

$$\sum_{n=1}^{L} f_n \frac{\varepsilon_n - \varepsilon}{\varepsilon_n + 2\varepsilon} = 0 \quad \text{(Eq.4)}$$

For minimizingthe difference between measuredandcalculated spectra, quantified by the quadratic error χ^2, we used the simplex method [22].

$$\chi^2 = \frac{1}{2M} \sum_{i=1}^{M} \left(\left(\tan\psi_{ex}^i - \tan\psi_{th}^i \right)^2 + \left(\cos\Delta_{ex}^i - \cos\Delta_{th}^i \right)^2 \right) \quad \text{(Eq.5)}$$

The proposed model allowed highlighting the effect of the thickness and the chemical composition (volume fractions) of the films on the ellipsometric spectra. The confrontation of calculated spectra to ellipsometric measures taken on real structures allowed the validation of the theoretical model.

3. Results and discussions

The proposedmodelisused first to describe the optical indices dispersion ofSiO_xN_yusing the effective media approximation of Bruggman, and then to studythe influence of the chemical composition and the film thicknessontheellipsometricangles. Finally the proposed model was validated by the confrontation between calculated and experimental spectra.

3.1. Optical indices calculated

The effective media approximation of Bruggman permits to have a good description of the optical indices dispersion depending from chemical composition of the deposited films. It thus enables to distinguish between the optical behavior of silica-rich and silica-poorfilms.

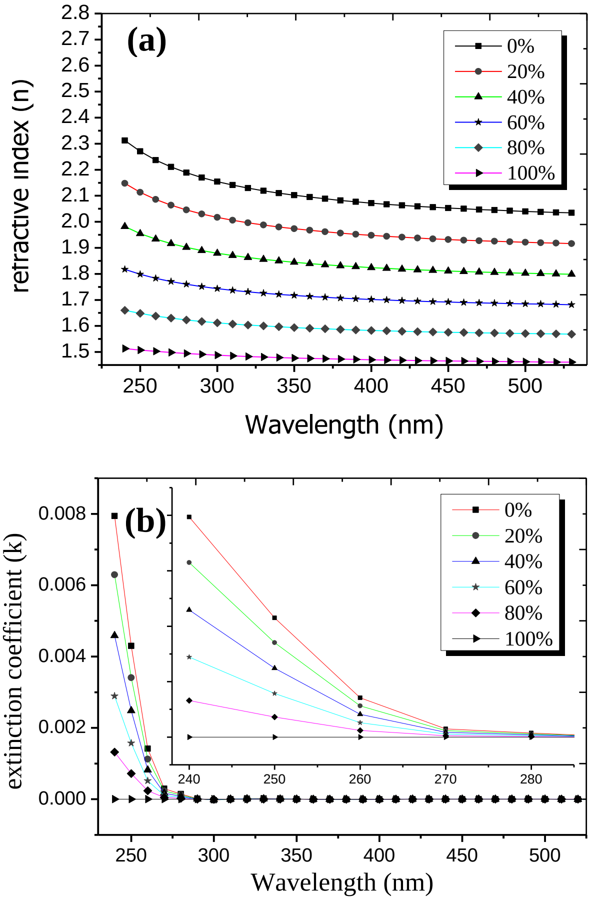

Fig. 2. (a) Calculated refractive index n, and (b) extinction coefficient k of SiO_xN_y thin films as a function of the volume fraction of SiO_2.

In Figure 2 both n and k decrease with increasing the proportion of SiO_2, which explain the dependence between the optical indices and the chemical composition of SiO_xN_y. (b) Shows a strong absorption aboveto6 eV which corresponds to a direct photo-excitation process and a total transparency in the visible and near infrared ranges.

Ellipsometricspectra

Using the proposed theoretical model, ellipsometric spectra were calculated for homogeneous films of oxynitrides containing 20% silica.The thickness variesfrom 150nm to 350nm. The incidence angle was taken equal to 70°in agreement with experiment.

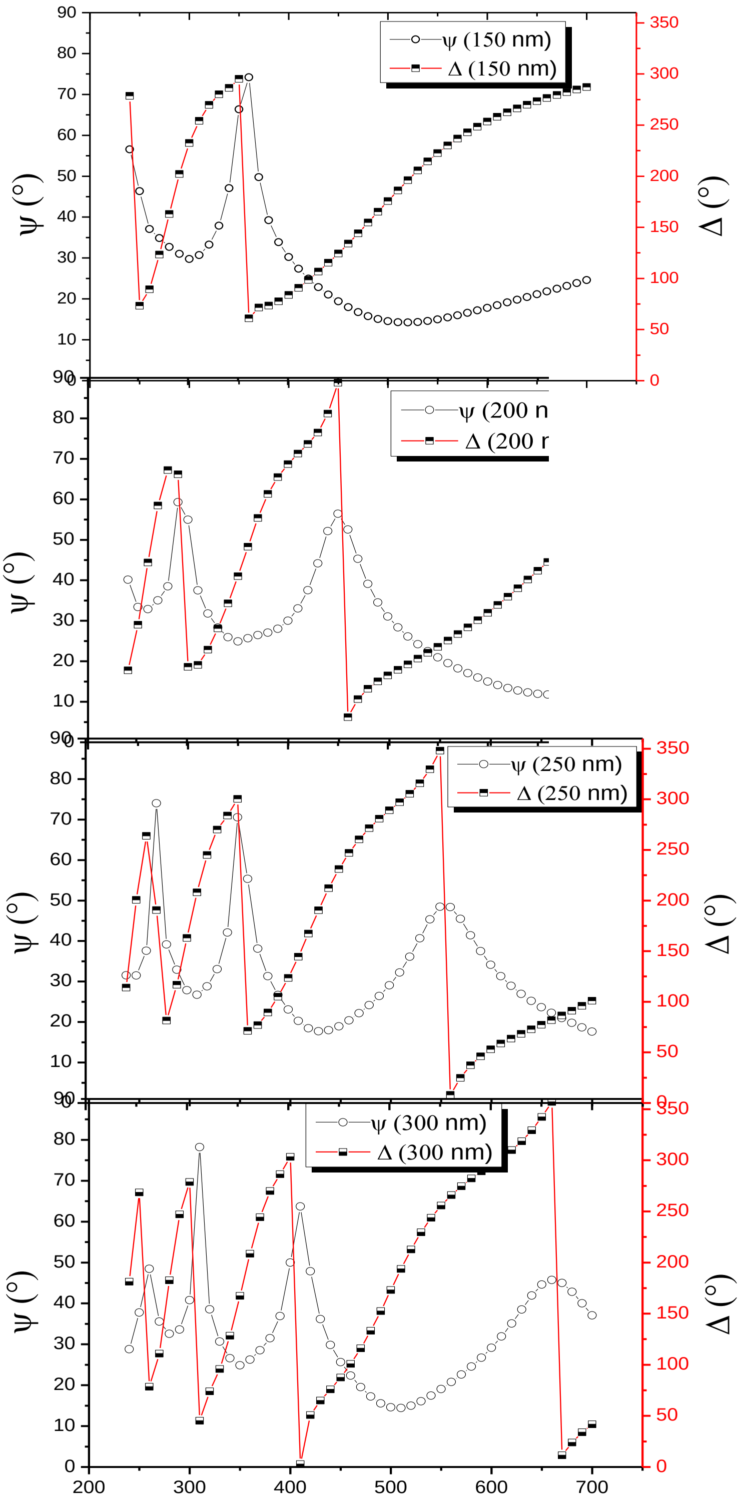

Fig. 3. Ellipsometric spectra calculated forvariable thickness of SiO_xN_y containing 20% silica.

In Figure 3 we observe a pseudo-periodic variation in ellipsometric spectra depending on the wavelength. This variation is due to interferences resulting from reflections at the two interfaces Air / film and film / Substrate. It is noted that when the film thickness increases the number of oscillations increases accordingly.

3.2. Analysis of Measured spectra

To validate the model two samples were used, the first sample presents an oxynitride film deposited using variable precursors flow while the second film is deposited at constant flow.

Ellipsometric spectra recorded between 250nm and 825nm for the two samples were compared with those calculated by the proposed model.The adjustment is done using progressively models with an increasing number of strata (monolayer, bilayer and trilayer models).

The trilayer model gives the best agreement between calculation and measurements, for the two samples**(Fig.4)**. The optical thickness and volume fraction of SiO_2 of each sample are reported in **Table 1**.Both deposited films are silica-rich, and this is probably due to the high reactivity of oxygen

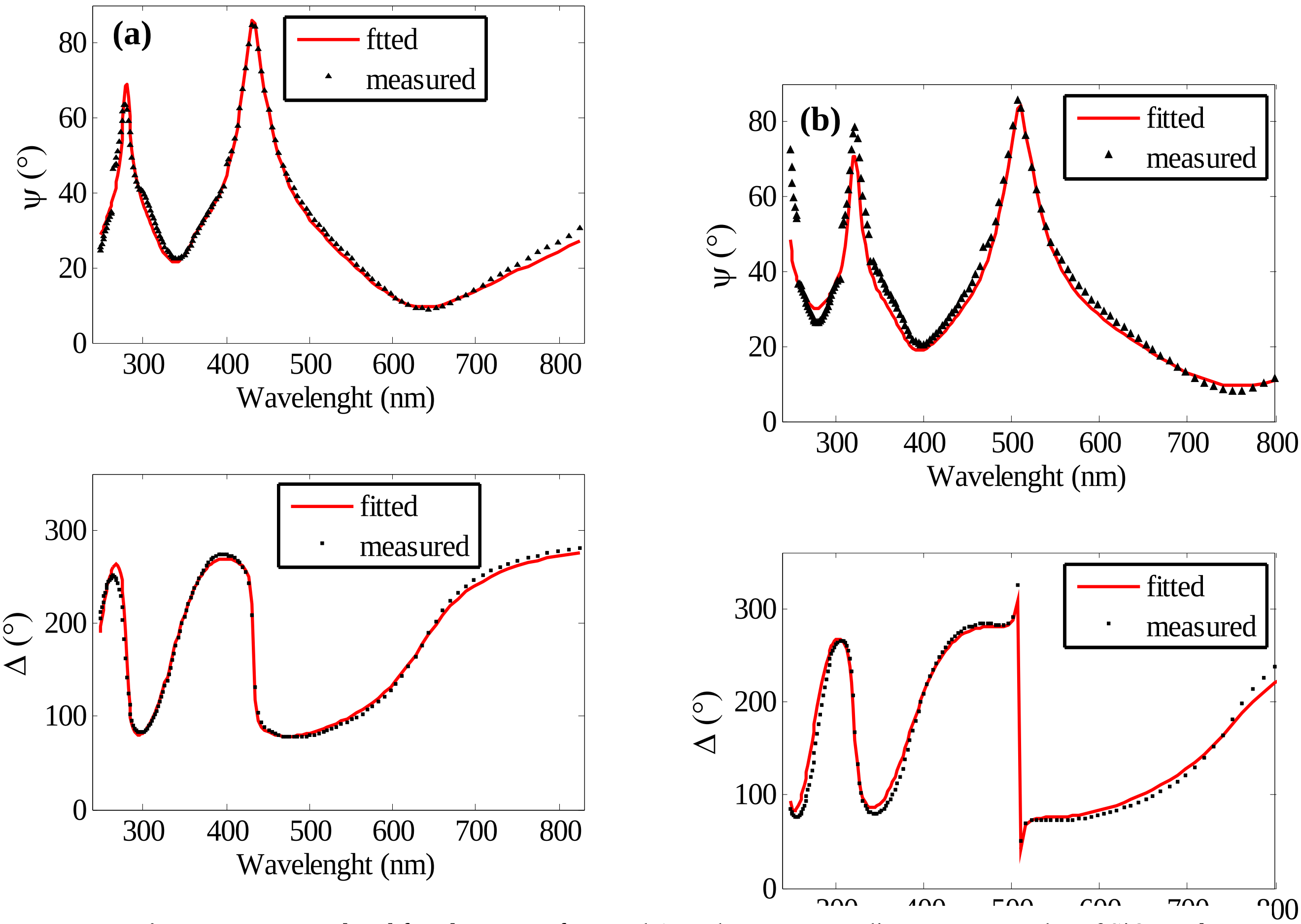

Fig. 4. Experimental and fitted spectra of ψ and Δ angles, corresponding to two samples of SiO_xN_y thin films: (a) 259 nm, (b) 306 nm.

Table 1; Ellepsometric analysis

sample	Sample 1		Sample 2	
	Thickness (nm)	f_{SiO2} (%)	Thickness (nm)	f_{SiO2} (%)
Layer 1	115	98	3	80
Layer 2	104	78	138	85
Layer3	42	50	164	88

The analysisof measured ellipsometric spectra reportedin**table 1** shows that for sample 1 the volume fraction ofSiO_2isgraduallydecreasing fromtheairto the substratewhich means that the deposited film is inhomogeneous in depth.Contrary to this, for sample 2, the volume fraction ofSiO_2isalmostconstant, which indicates thehomogeneityof the deposited thinfilm.These results are in accordance with the sample conditions of preparation.

The quadratic error χ^2shown in figure 5 represents the mean deviation between the experimental and that calculated spectraby different proposed optical systems such as monolayer, bilayer and trilayer. we can clearly see that for the sample 1, more the number of layers increases more the error decreases and the adjustment with multilayer system becomes better.Such comportmentconfirmsthe results of Table1 (inhomogeneousdeposited film). While for the sample 2, the quadratic error is almost constant indicating that the monolayer modelling system is sufficient.This confirms the results of Table 1(comparable volume fraction of SiO_2 in the depth of deposited film).

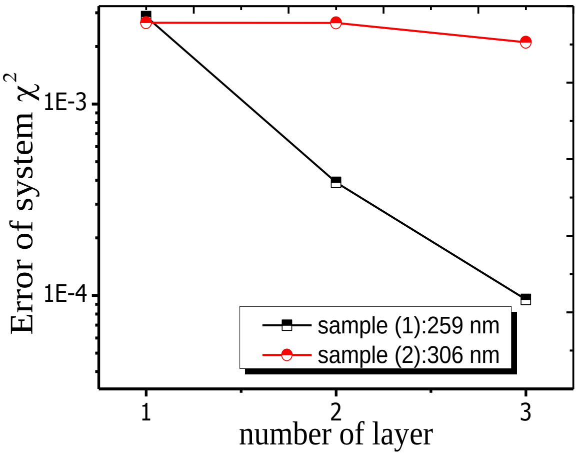

Fig. 5. Quadratic errorχ^2 for three optical systems proposed for analysing ellipsometric spectra of sample S_1 and S_2

4. conclusion

We have investigated the optical properties of thin films of silicon oxynitride by spectral ellipsometry. The good agreement between calculated and measured spectra indicates that the proposed model correctly simulates such optical systems. It is clear that the results obtained by the multilayer system in the case of inhomogeneous samples indicate that more the proposed system is complex, more the adjustment is successful and this appears directly on the values of the error function, which decreases rapidly. For homogeneous film, a monolayer model is sufficient to analyze the ellipsometric spectra. Ellipsometric analysis shows that both deposited films are silica-rich probably because of the high reactivity of oxygen with silicon.

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