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REGULAR ARTICLE

# Ellipsometric and Maxwell-Garnett Model Studies of Silicon Nitride-based Bilayer Structures Elaborated by LPCVD

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In this work, we study the geometrical and optical properties of thin layers' silicon oxynitride ($SiO_xN_y$) prepared by low pressure chemical vapor deposition technique from the $SiH_2Cl_2$, $N_2O$ and $NH_3$ at 850 °C, using spectroscopic ellipsometry measurements. For this purpose, Maxwell-Garnett (MG) model was used. It is applied to silicon oxynitride, considering the material as a heterogeneous medium formed by silicon oxide ($SiO_2$) and the silicon nitride ($Si_3N_4$). The thickness of $SiO_xN_y$ films was computed based on the gas flow rate of $NH_3$ and with the ratio $R = NH_3/N_2O$. Moreover, its refractive index was calculated at wavelength 830 nm as a function of both gas flow rate of $NH_3$ and $N_2O$. In addition, the volume fraction of $SiO_2$ and $Si_3N_4$ was evaluated versus this flow rates. It was observed that the thickness increases with increasing gas flow rate of $NH_3$ and with the ratio R. The results also show that the refractive index decreases with gas flow rate of $N_2O$. However, it increases with the rate of $NH_3$, ranging from 1.458 to 1.597. Furthermore, the results proved that knowing the volume fraction of $SiO_2$ and $Si_3N_4$ allows us to quantify the evolution of the refractive index of $SiO_xN_y$. Finally, this paper makes it possible to choose the experimental parameters of precursors deposition based on the application to which it is dedicated.

**Keywords**: Ellipsometry, Silicon oxynitride, Refractive index, Maxwell-Garnett

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## 1. INTRODUCTION

The silicon oxide $SiO_2$ and silicon nitride $Si_3N_4$ have complementary properties. Indeed, on the one hand $SiO_2$ with a refractive index of 1.46, present an excellent dielectric property, a good transparency in the visible range, and low mechanical stress. But, it constitutes a weak barrier to the diffusion of ions and dopants [1-3]. On the other hand, $Si_3N_4$ which has a refractive index of 2.02, is denser and thus offers a better barrier to diffusion. However, it is a mediocre dielectric and has low resistance to cracking due to significant mechanical stress [4-8]. Thereby, it has become appealing to find a compromise between the properties of $SiO_2$ and $Si_3N_4$ by manufacturing intermediate materials [9, 10].

The silicon oxynitride $SiO_xN_y$ combines the dielectric and mechanical qualities of silica, along with the advantage of serving as a diffusion barrier to impurities, distinguished by the nitrides. It is a good candidate for the production of antireflective coatings and is also used in waveguides, integrated optics, and microelectronics, as intermetallic insulators and passivation insulators [11-13]. Investigating the characteristics of these materials is essential for defining their optical, physical, and chemical properties. Consequently, numerous studies have been conducted in this area. The X-ray photoelectron spectroscopy characterizations of $SiO_xN_y$ stoichiometries was presented by Temple [14]. The same authors have indicated the effect of the gas mixture $SiH_4/N_2O/NH_3/N_2$ on the properties of $SiO_xN_y$ thin films deposited by plasma enhanced chemical vapor deposition (PECVD) [15]. Afterwards, Soman [16] studied several samples of $Si_xN_y$ and $SiO_xN_y$ layers elaborated by PECVD with different compositional and optical properties by changing the gas mixing ratio during deposition, resulting in materials with variable refractive index. Recently, Mortreuil [17] have studied the dielectric layers of $SiO_xN_y$ where deposited by plasma. They analyzed the influence of the dielectric films thickness on charging phenomena in such films using Kelvin probe force microscopy and conductive atomic force microscopy. More recently, Evtukh [18] have investigated the peculiarities of the structure and electrical conductivity of nanocomposite $SiO_xN_y$(Si) and $SiAl_zO_xN_y$(Si) films in the temperature range of 95 – 340 K. In his work Radoi [11] focuses on the analysis of laser-ablated silicon oxynitride layers, found with laser reactive ablation technique. Their approach is based to use the Bruggeman model

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(BEMA) in wave-length range (320-700 nm) to characterize the $SiO_xN_y$ layers elaborated by LRA and LPCVD. However, in our previous work [19] we obtained that the Maxwell-Garnett model is more accurate than that of Bruggeman, and that the latter is not suitable for nanocomposites and optical materials [20]. Furthermore, the wavelength ($\lambda$ = 830 nm) which the extinction coefficient being zero is not included in the wavelength range used in Radoi's research paper.

For this purpose, this work seeks to study the geo-metrical and optical properties of silicon oxynitride. So, several samples were elaborated using the low pressure chemical vapor deposition (LPCVD) technique from $SiH_2Cl_2$, $N_2O$, and $NH_3$ gaseous precursors. The spectroscopic ellipsometry spectra were fitted in the wave-length range (450-900 nm) using the Maxwell-Garnett (MG) model. This model was carried out by assuming the silicon oxynitride material as a heterogeneous medium formed of silicon oxide $SiO_2$ and silicon nitride $Si_3N_4$ [17].

## 2. EXPERIMENTAL PROCESS

The silicon oxynitride thin films were deposited on a silicon substrate using the LPCVD technique, employing a mixture of dichlorosilane, nitrous oxide, and ammonia as precursors. The $SiH_2Cl_2$ and $[N_2O + NH_3]$ flow rates were kept constant, respectively at 200 sccm and 160 sccm throughout the elaboration process, while the gas flow ratio $R = NH_3/N_2O$ was varied from 0.3 to 0.23. The series of samples was deposited at a temperature of 850 °C and a pressure of 450 mtorr for a duration of 50 min. Measurements of the fabricated films using spectroscopic ellipsometry were performed with a rotating analyzer ellipsometer, model SpecEL2000-VIA. These measurements correspond to the ellipsometric angles Ψ and Δ, which were independently extracted as a function of wavelength across a measurement range of 450 to 900 nm, with an incidence angle of 70°, as shown in Fig. 1.

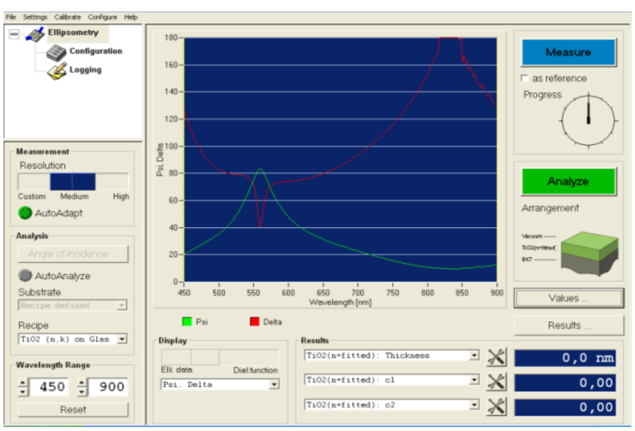



**Fig. 1** – Characterization of the sample where $R = 0.1$ by spectroscopic ellipsometry

## 3. THEORY AND MODELING

The This study utilized the ellipsometric technique, which is described by the following fundamental equation [1, 13]:

$$\rho = \frac{R_p}{R_S} = \tan \psi e^{j\Delta} \tag{1}$$

Where Ψ and Δ represent the ellipsometric angles. The refractive index of the mixture, calculated using the Maxwell-Garnett model, is expressed as follows [20].

$$\frac{n^2_{S_iO_xN_y} - n^2_{S_iO_2}}{n^2_{S_iO_xN_y} + 2n^2_{S_iO_2}} = \left(1 - f_{S_iO_2}\right)\frac{n^2_{S_3N_4} - n^2_{S_iO_2}}{n^2_{S_3N_4} + 2n^2_{S_iO_2}} \tag{2}$$

This equation simplifies to the following form:

$$\begin{aligned}&\left(n^2_{S_3N_4} + 2n^2_{S_iO_2} + f_{S_iO_2}(n^2_{S_iO_2} - n^2_{S_3N_4})\right)n^2_{S_iO_xN_y} - \\ &-(2f_{S_iO_2}+1)n^2_{S_iO_2}n^2_{S_3N_4} - (2-2f_{S_iO_2})n^4_{S_iO_2} = 0\end{aligned} \tag{3}$$

With:

$$f_{S_iO_2} + f_{S_3N_4} = 1 \tag{4}$$

Where $n_{SiOxNy}$ is the refractive index of the layer under study, $f_{SiO2}$ and $f_{Si3N4}$ are the volume fractions of the $SiO_2$ and $Si_3N_4$ respectively. The $n_{SiO2}$ and $n_{Si3N4}$ represent their refractive index, its values of were taken from the literature [1]. A Simplex technique was used to minimize the error function (5) [11]. This approach allowed us to align the theoretical curves of the angles $\Psi_{th}{}^i$ and $\Delta_{th}{}^i$ calculated by our model, with the experimental data ($\Psi_{exp}{}^i$ and $\Delta_{exp}{}^i$). This was accomplished by adjusting the thickness and the volume fraction of one of the two constituents.

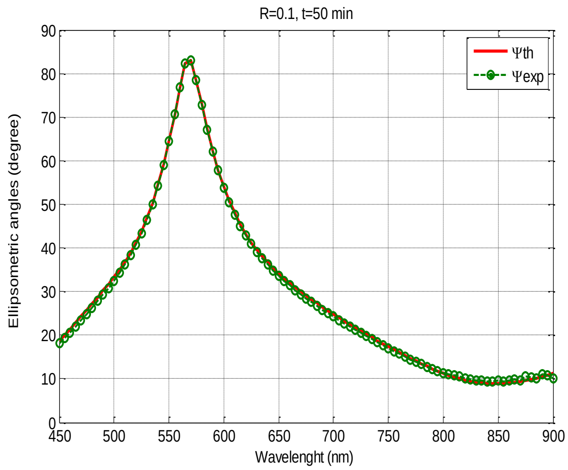
$$\chi^2 = \frac{1}{N}\sum_{i=1}^{N}(\psi^i_{\exp} - \psi^i_{th})^2 + (\Delta^i_{\exp} - \Delta^i_{th})^2 \tag{5}$$

Where $N$ is the number of measurements.

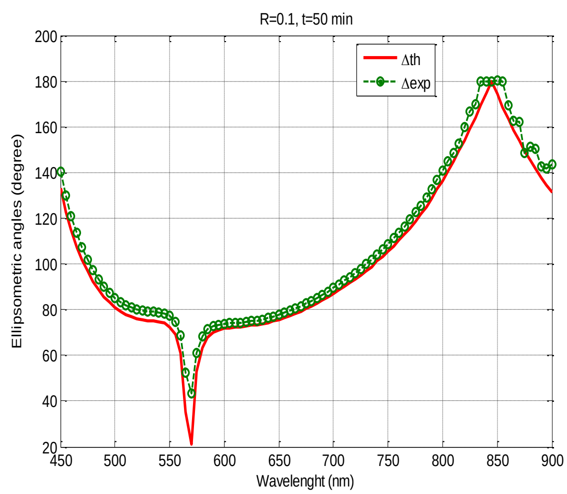
## 4. RESULTS AND ANALYSIS

### 4.1 Validation of the Used Model

To validate the Maxwell-Garnett model, we com-pared its results with experimental data. To do this, we focused on the sample deposited with an ammonia flow rate of 20 sccm and a nitrous oxide flow rate of 140 sccm. By applying the minimization procedure, the theoretical curves were successfully fitted to the experimental data, yielding an error of $2.38 \times 10^{-5}$. This indicates a high level of accuracy in matching the theoretical curves to the experimental spectra, as shown in Fig. 2 (a and b). These results validate that the minimization technique used in the proposed model aligns with the experimental findings, highlighting the model's effectiveness.




a




b

**Fig. 2** – Experimental and theoretical curves of the ellipsometric angles for $SiO_xN_y$ layer: (a) Ψ and (b) Δ

### 4.2 Effect of Precursors on Thickness

To highlight the evolution of the geometrical properties of the elaborated films, their thickness is analyzed with a varied ammonia gas flow rate and the ratio $R$. Fig. 3 shown the variation of thickness as a function of the $NH_3$ flow. Based on the results given in Fig. 3, we can notice that the thickness is an increasing function with the ammonia flow rate. Furthermore, Fig. 4 illustrates the variation of the thickness of the deposited $SiO_xN_y$ films for different values of $R$. As illustrated, as the gas flow ratio $R$ increases, the thickness of the deposited silicon oxynitride layers increases. So, the introduction of more reactive gases, therefore the quantity of nitrous oxide and ammonia multiply, we will then have increasingly thick silicon oxynitride films, varying between 287 and 398 nm.

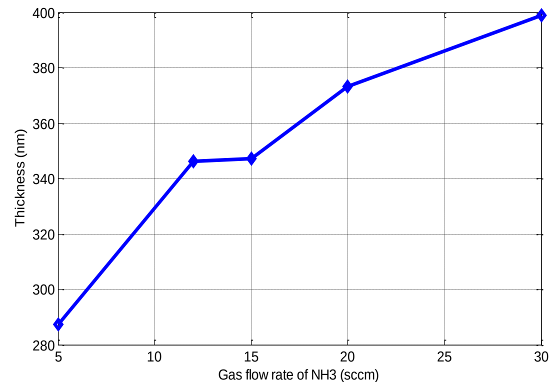



**Fig. 3** – Thickness variation with gas flow rate of $NH_3$

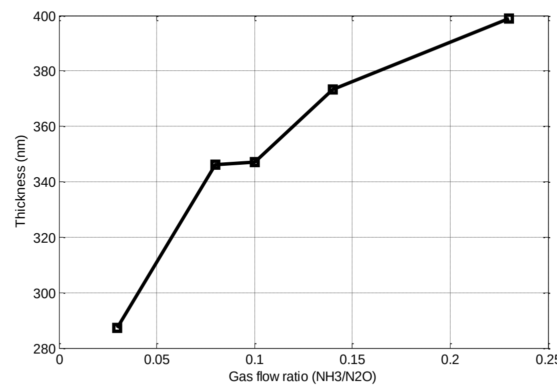



**Fig. 4** – Variation of the thickness vs. gas flow ratio $NH_3/N_2O$

### 4.3 Effect of Precursors on Refractive Index

In this section, the variation of the refractive index of $SiO_xN_y$ at the wavelength $\lambda = 830$ nm, where the extinction coefficient was zero, was carried out as a function of the gas flow rate of $NH_3$ and $N_2O$. Based on the results shown in Fig. 5, we can see that the refractive index of $SiO_xN_y$ is an increasing function with the flow gas controlling the nitrogen $NH_3$, ranging from 1.458 to 1.597. Nevertheless, the profile illustrated in Fig. 6 shows that the refractive index decreases as the flow rate of $N_2O$ governing oxygen increases.

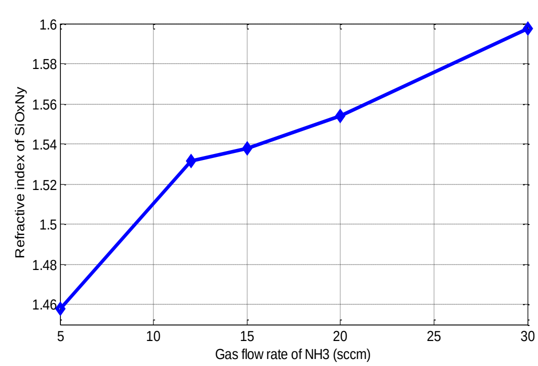



**Fig. 5** – Refractive index as a function of $NH_3$ flow at 830 nm

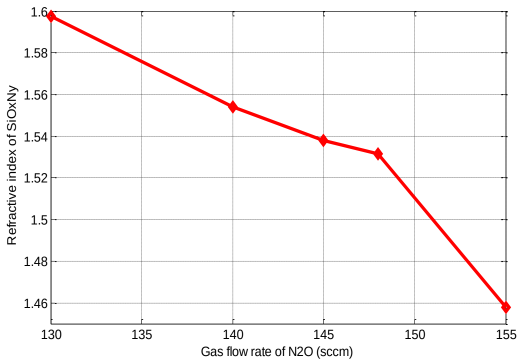



**Fig. 6** – Refractive index as a function of $N_2O$ flow at 830 nm

### 4.4 Effect of Precursors on Volume Fraction of Its Constituents

As mentioned earlier, part of this study is devoted to the optical properties of the $SiO_xN_y$ films. The volume fraction of $SiO_2$ and that of $Si_3N_4$ define the values of its refractive index. The knowledge of their variations with $NH_3$ and $N_2O$ gaseous precursors is used to quantify the evolution in the refractive index of $SiO_xN_y$. According to Fig. 7, the volume fraction of $SiO_2$ decreases as the $NH_3$ rate increases. In this case, the lowest value of the silica percentage (99 %) is obtained for an oxynitride film with a refractive index equal to 1.458.

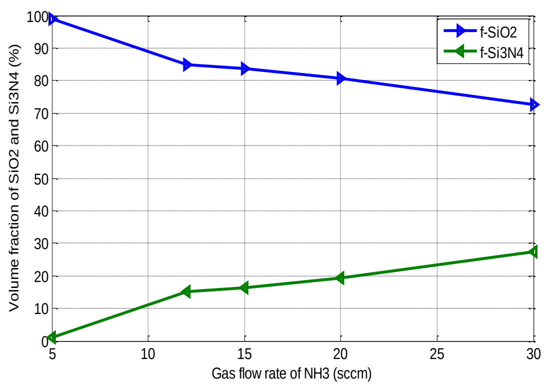



**Fig. 7** – Volume fraction of $SiO_2$ and of $Si_3N_4$ variation with gas flow rate of $NH_3$

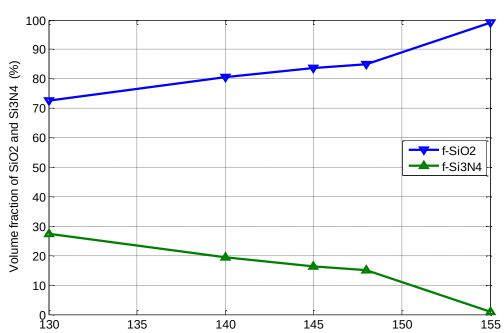



**Fig. 8** – Volume fraction of $SiO_2$ and of $Si_3N_4$ variation with gas flow rate of $N_2O$

Moreover, the volume fraction of $Si_3N_4$ rises with the flow of ammonia. We can observe that, the predominant influence of the ammonia flow on the incorporation of the silicon nitride phase. As a result, the refractive index of $SiO_xN_y$ is increasingly approaching that of $Si_3N_4$. In the contrary, we can see in previous figure (Fig. 8) the influence of $N_2O$ flow on the incorporation of the silicon oxide $SiO_2$ phase. Consequently, the refractive index of $SiO_xN_y$ decreases.

## 5. CONCLUSIONS

In conclusion, this work contributes to the study of the geometrical and optical parameters of silicon oxynitride ($SiO_xN_y$) thin films prepared by the LPCVD process, using spectroscopic ellipsometry spectra and the Maxwell-Garnett model.

Comparing the curves predicted by this model with those obtained experimentally allows us to validate its good accuracy. Thus, the results show that the thicknesses of $SiO_xN_y$ films increases from 287 to 398 nm with both $NH_3$ and $N_2O$ flow. Then, at the wavelength ($\lambda = 830$ nm) where the extinction coefficient being zero, the refractive index of the deposited films varies from 1.458 to 1.597 as the gas flow rate of $NH_3$ increases. This can be explained by the incorporation of nitrogen into the produced films. In contrast, the refractive index decreases when the $N_2O$ flow rises. This is due to the incorporation of oxygen into the deposited films.

In addition, the results proved that the variation of the volume fraction of $SiO_2$ and of $Si_3N_4$ with the gas flow rate of $NH_3$ and $N_2O$ allows us to quantify the evolution of the refractive index of $SiO_xN_y$. Thereby, the results showed also that the refractive index is inversely proportional to the percentage of $SiO_2$. Nevertheless, it is proportional to the volume fraction of the $Si_3N_4$. Moreover, the obtained results showed that, for a good optical characteristic of these films, the $SiO_xN_y$ must be exibit very high refractive index intermediate be-tween that of $SiO_2$ and $Si_3N_4$.

To sum up, this paper permits to choose the experimental parameters of the precursor deposition according to the application to which it is destined.

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## Еліпсометричні дослідження та моделювання за Максвеллом–Гарнеттом двошарових структур на основі нітриду кремнію, отриманих методом LPCVD


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У даній роботі досліджено геометричні та оптичні властивості тонких плівок оксинітриду кремнію ($SiO_xN_y$), отриманих методом низькотемпературного хімічного осадження з газової фази при зниженому тиску (LPCVD) із використанням прекурсорів $SiH_2Cl_2$, $N_2O$ та $NH_3$ при температурі 850 °C. Для аналізу використано спектроскопічну еліпсометрію. Для моделювання було застосовано модель Максвелла–Гарнетта (MG), яка розглядає $SiO_xN_y$ як гетерогенне середовище, що складається з фаз оксиду кремнію ($SiO_2$) та нітриду кремнію ($Si_3N_4$). Основні результати: Товщина плівки залежить від швидкості подачі аміаку ($NH_3$) та співвідношення $R = NH_3 / N_2O$ – з її збільшенням товщина $SiO_xN_y$ зростає. Показник заломлення при довжині хвилі 830 нм зростає зі збільшенням $NH_3$ та знижується зі зростанням $N_2O$, змінюючись у діапазоні від 1.458 до 1.597. Об'ємна частка компонентів ($SiO_2$ та $Si_3N_4$) була розрахована залежно від швидкостей подачі газів, що дозволяє кількісно описати зміну оптичних властивостей плівки. Таким чином, наведені результати дозволяють визначити оптимальні параметри осадження, виходячи з цільового застосування $SiO_xN_y$-покриттів в електроніці та фотоніці.



**Ключові слова**: Еліпсометрія, Оксинітрид кремнію, Показник заломлення, Модель Максвелла–Гарнетта.