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Silicon nitride/silicon oxynitrid/silicon dioxide thin film multilayer characterized by variable angle spectroscopic ellipsometry

Yi-Ming Xiong
University of Nebraska-Lincoln

Paul G. Snyder
University of Nebraska-Lincoln, psnyder1@unl.edu

John A. Woollam
University of Nebraska-Lincoln, jwoollam1@unl.edu

Graded refractive index silicon oxynitride thin film characterized by spectroscopic ellipsometry

Paul G. Snyder, Yi-Ming Xiong, and John A. Woollam
Center for Microelectric and Optical Materials Research, Department of Electrical Engineering, University of Nebraska, Lincoln, Nebraska 68588-0511

Ghanim A. Al-Jumaily
M/S 179-111, Jet Propulsion Laboratory, Pasadena, California 91109

F. J. Gagliardi
Barr Associates, Inc., Westford, Massachusetts 01886

A graded refractive index silicon oxynitride (SiO_xN_y) thin film was prepared on a silicon substrate by ion assisted deposition. Spectroscopic ellipsometry (SE) was used to optically analyze the film. The measured SE spectra (2500–8200 Å) were analyzed with several fitting models, whose construction was based on an Auger depth profile of the film. In each model, the optical response of SiO_xN_y was described using the Bruggeman effective medium approximation, by modeling it as a physical mixture of two distinct phases: silicon dioxide and silicon nitride. Grading was modeled by varying the silicon nitride volume fraction with depth below the surface, according to an assumed profile. Fitting results were very sensitive to the profile chosen, which was different for each model. Experimentation with the profile led to a model which produced a remarkably good fit, over the entire spectral range. As a result, the film thickness and its refractive index profile were determined. The index profile determined by SE analysis was found to be consistent with the Auger profile.

I. INTRODUCTION

Graded refractive index optical thin films, such as rugate filters, are being increasingly recognized and considered as an attractive alternative to traditional multilayer dielectric stack devices for spectral filters and broad band antireflection coatings.[1,2] Several methods are used to fabricate the films,[3] including ion assisted deposition (IAD).[4,5] In this method, the film compositional variation is realized by reactive ion beams through control of the relative gas fluxes. The dependencies of refractive index in the composition can, in turn, be employed for fabrication of desired optical films.

An effective and nondestructive means of accurately determining the film thickness and its refractive index profile is needed to facilitate film analysis. In this work, spectroscopic ellipsometry (SE),[6] a nondestructive optical technique, was used to study an IAD deposited graded refractive index silicon oxynitride (SiO_xN_y) thin film. The ellipsometrically measured spectra were analyzed with several fitting models, constructed based on an Auger depth profile of the film. The model calculations were carried out by assuming SiO_xN_y to be a physical mixture[7–9] of two distinct phases, silicon dioxide (SiO_2) and silicon nitride (Si_3N_4), using the Bruggeman effective medium approximation (EMA).[10] As a result, the film thickness and its refractive index profile were determined.

II. EXPERIMENTAL

A. Sample preparation

The SiO_xN_y film was deposited onto a 50 mm diam Si(100) substrate wafer in a Balzers 760 system. The substrate was heated to about 300 °C with a quartz halogen heater. The deposition system was cryogenically pumped to a base pressure of 1.50×10^{-6} Torr. High purity (99.999%) Si was evaporated using one of two electron evaporation sources, where the evaporation rate was controlled by a crystal rate controller. An 8 cm Kaufman ion source with beam expanding grids was used for precleaning and ion bombardment during deposition, and was located 35 cm away from the substrate. The bombarding species were nitrogen ions, with beam energy and current set at 500 eV and 100 mA, respectively. The relative arrival ratio of ions to Si atoms was about 1:1. The film chemistry (composition versus thickness) was controlled by linearly varying the ratio of nitrogen flowing into the ion source and oxygen in the backfill. The film compositional variation was measured by sputter depth profiling by Auger electron spectroscopy (AES).

B. Ellipsometry

SE was used to optically characterize the deposited SiO_xN_y film. Details about ellipsometry can be found in Ref. 11, and only a brief description is given here. In ellipsometry, linearly polarized light is incident at an oblique angle onto the sample. The reflected light from the sample surface is, in general, elliptically polarized. The new polarization state of the reflected light, containing information about sample's properties, is determined using a polarizer prism. The ellipsometrically measured data are tan ψ and cos Δ, defined by tan $\psi \cdot \exp^{i\Delta} = R_p/R_s$, where R_p and R_s are the complex reflection coefficients for light polarized parallel (p) and perpendicular (s) to the plane of incidence, respectively. For a substrate covered with a thin
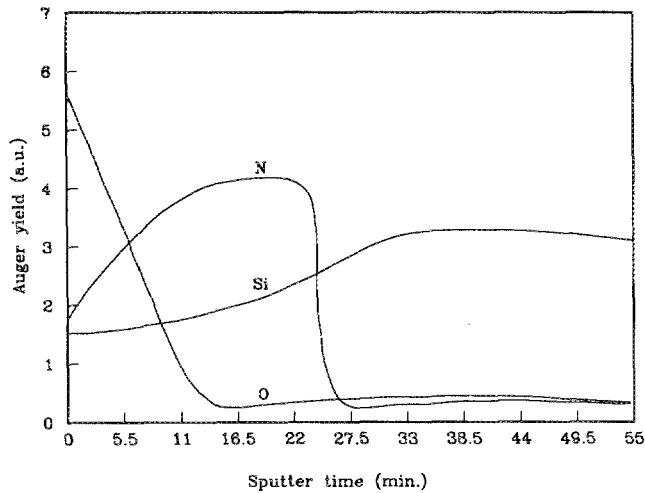
FIG. 1. Auger in-depth profile of the graded refractive index SiO_xN_y deposited on Si substrate.

film, the measured data are numerically fitted with an appropriate model, which should best represent the sample under study. In this model, the unknown parameters, such as film thickness, optical constants, and even constituent volume fractions (if the film is a composite material) can be chosen as fitting parameters to be determined from standard ellipsometric equations.[11] Using a regression algorithm,[12] the fitting parameters are varied, until the squared differences between the measured and calculated data at the corresponding wavelengths and angles of incidence are minimized. The quality of the fit can be judged quantitatively by the biased estimator (σ), defined by

$$\sigma = \frac{1}{N} \sum_{i=1}^{N} \left(\frac{(\psi_i^m - \psi_i^c)^2}{\delta \psi_i^2} + \frac{(\Delta_i^m - \Delta_i^c)^2}{\delta \Delta_i^2} \right), \qquad (1)$$

where N is the total number of measurements, and superscripts m and c stand for the measured and calculated data, respectively. The experimental errors in ψ_i and Δ_i are given by $\delta \psi_i$ and $\delta \Delta_i$, which are estimated values.

All SE measurements were performed at room atmosphere with an automatic rotating analyzer ellipsometer (similar to that described in Ref. 13, with the added capability of variable angle of incidence), at 75° angle of incidence and in the 2500–8200 Å spectral range at 50 Å intervals. The value of angle chosen here is not critical, because of the large variations in the spectra due to optical interference in the transparent film. Prior to and during the measurements, no chemical treatment was made on the sample surface. The average uncertainties in the ψ and Δ measurements were within ±0.01° and ±0.03°, respectively. The angle of incidence was accurate to within ±0.01°. The beam diameter was about 1 mm.

III. MODELING

The optical properties of SiO_xN_y have been studied before (see Refs. 7–9 and 14–15). As indicated in Refs. 7 and 9, the optical behavior of homogeneous SiO_xN_y can be successfully described by modeling it to be a simple dielectric
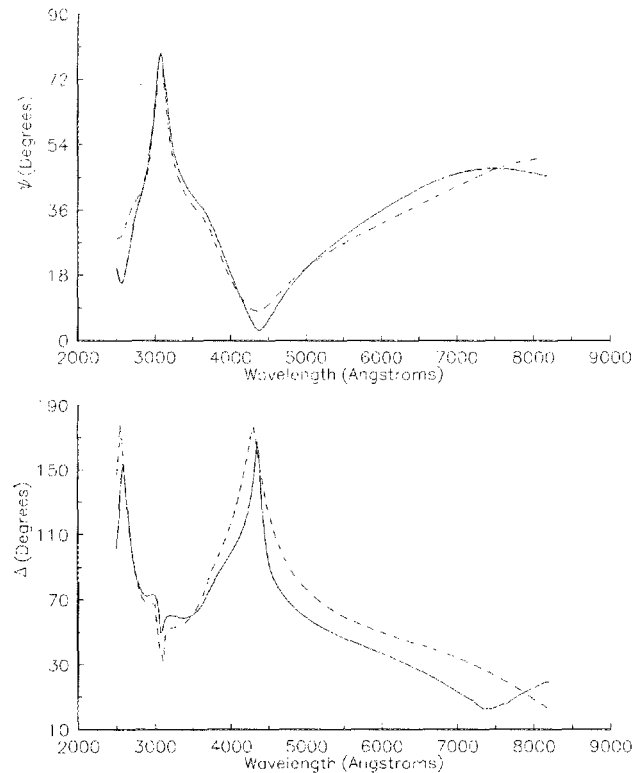

FIG. 2. Measured (solid lines) and calculated (dashed lines) ψ and Δ spectra at 75° angle of incidence, obtained using the first fitting model described in the text.

physical mixture of two distinct phases, SiO_2 and Si_3N_4, using the Bruggeman EMA. According to the EMA, the effective refractive index n of SiO_xN_y can be obtained from the following equations:

$$f_{SiO_2} \frac{n_{SiO_2}^2 - n^2}{n_{SiO_2}^2 + 2n^2} + f_{Si_3N_4} \frac{n_{Si_3N_4}^2 - n^2}{n_{Si_3N_4}^2 + 2n^2} = 0, \qquad (2)$$

and

$$f_{SiO_2} + f_{Si_3N_4} = 1, \qquad (3)$$

where f_{SiO_2} and $f_{Si_3N_4}$ represent the relative volume fractions of SiO_2 and Si_3N_4 in the mixture, and n_{SiO_2} and $n_{Si_3N_4}$ are the refractive indices of SiO_2 and Si_3N_4, respectively. Furthermore, at each wavelength (within the given spectral range), the refractive index of SiO_xN_y has been found to be a linear function of its constituent volume fraction (i.e., f_{SiO_2} or $f_{Si_3N_4}$).[7,9]

The SiO_xN_y film studied in this work is inhomogeneous, i.e., its refractive index profile varies with depth. AES depth profile of the sample in Fig. 1 clearly shows the inhomogeneity of film composition versus its depth (corresponding roughly with the sputter time scale). Thus, it would be unphysical to model this film as a homogeneous EMA mixture. From Fig. 1, it can be seen that the N/O ratio is large and fairly constant, starting from the SiO_xN_y film/Si substrate interface to the point corresponding to about 15 min into the sputtering, and that the ratio de-
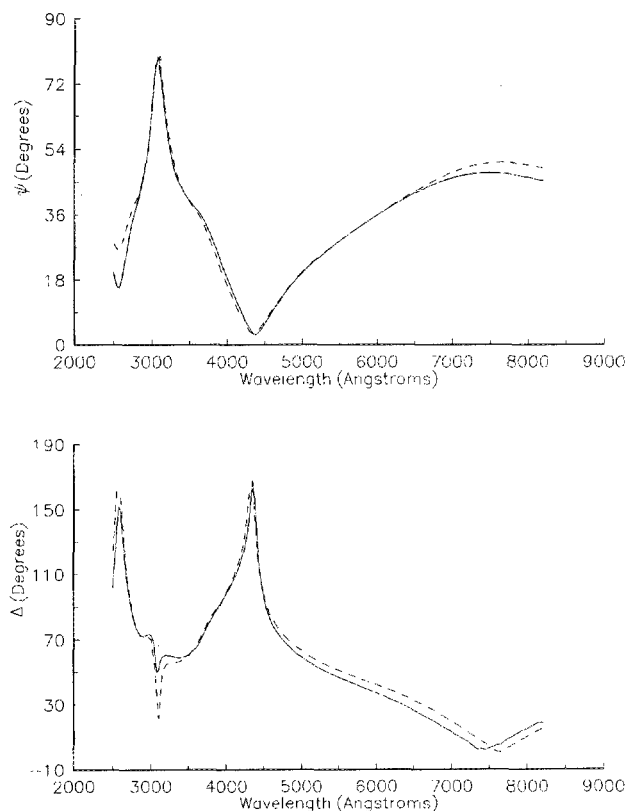
TABLE I. SE determined results, obtained using the three different fitting models described in the text.

	First fitting model
Total layer thickness (uniform)	1360 (Å)
Layer composition	36% SiO_2 + 64% Si_3N_4
σ	7752
	Second fitting model
Total layer thickness (linearly graded)	1377 (Å)
Starting layer composition	0% SiO_2 + 100% Si_3N_4
Ending layer composition	73% SiO_2 + 27% Si_3N_4
σ	2005
	Third fitting model
First sublayer total thickness (linearly graded)	400 (Å)
Starting layer composition	0% SiO_2 + 100% Si_3N_4
Ending layer composition	8% SiO_2 + 92% Si_3N_4
Second sublayer total thickness (linearly graded)	943 (Å)
Starting layer composition	11% SiO_2 + 89% Si_3N_4
Ending layer composition	80% SiO_2 + 20% Si_3N_4
σ	1330

FIG. 3. Measured (solid lines) and calculated (dashed lines) ψ and Δ spectra at 75° angle of incidence, obtained using the second fitting model described in the text.

creases continuously, in a nearly linear fashion, from that point (15 min) on to the sample surface (0 min).

Based on these observations, three different fitting models were constructed for SE data analysis. The main differences are in the assumed profile shapes. The simplest model, assuming a uniform layer of SiO_xN_y with constant composition, was used for comparison with the graded index models, to illustrate the gross effects of a graded versus an ungraded layer. The second model assumed a simple linear variation in the relative volume fraction of Si_3N_4, starting from 100% at the film/substrate interface, and decreasing towards the surface (where the film was oxygen rich). The third model, a refinement of the second, added another linearly graded interfacial layer near the substrate, which was less steeply graded than the layer above it.

To approximate the effects of grading, each graded layer was divided into many sublayers of equal thickness. Each sublayer had a uniform composition, and the composition from one sublayer to the next was varied in a linear staircase fashion (see Figs. 4 and 6). In practice, division into ten sublayers produced a very good approximation to continuous grading.

In carrying out the above multilayer model calculations, the layer's total thickness, and its starting and ending layer volume fractions were chosen as the fitting parameters. The refractive indices of SiO_2, Si_3N_4, and Si substrate, were taken as the bulk values given in the literature.[16] It is known that very thin films often have different optical con-

stants from the bulk. However, the validity of using bulk values in this study was verified, by accurately modeling measurements on pure SiO_2 and Si_3N_4 films of roughly the same thickness.

IV. RESULTS AND DISCUSSION

The film thickness and its refractive index profile were determined by numerically fitting the SE data, using the above-described fitting models. Figs. 2 and 3 show the measured and calculated (best-fit) ψ and Δ spectra obtained using the first and second models, respectively. The σ values in Table I also provide a quantitative comparison
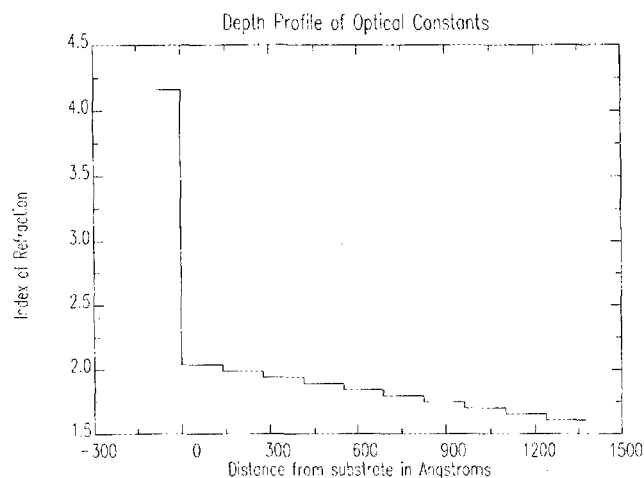

FIG. 4. SE determined refractive index profile of the film at 5350 Å wavelength, obtained using the second fitting model described in the text.
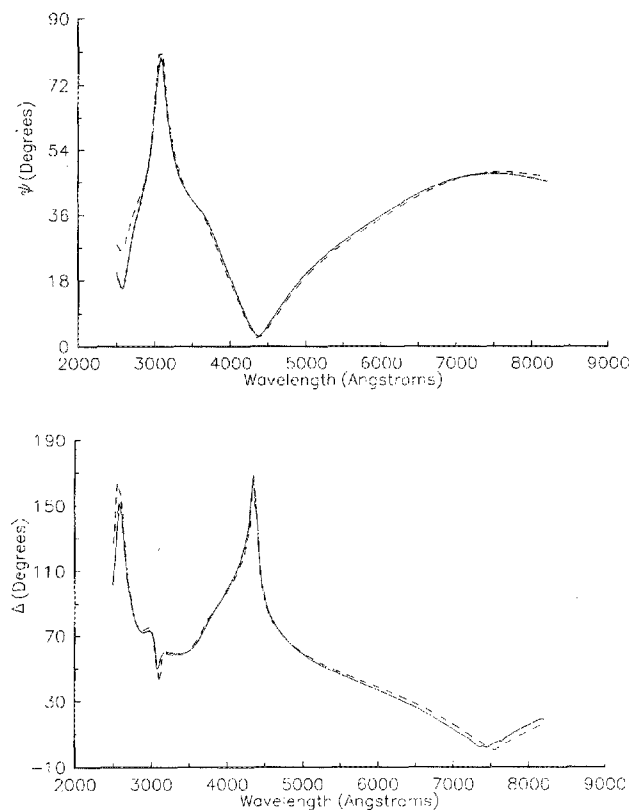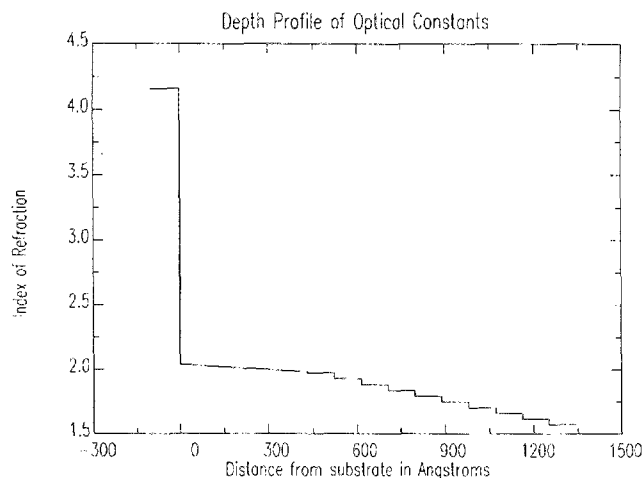
FIG. 6. SE determined refractive index profile of the film at 5350 Å wavelength, obtained using the third fitting model described in the text.

FIG. 5. Measured (solid lines) and calculated (dashed lines) ψ and Δ spectra at 75° angle of incidence, obtained using the third fitting model described in the text.

of the results. These figures graphically illustrate how much the fits are improved in going from a uniform to a graded profile. Peaks and valleys in the ψ and Δ spectra are due to the optical interference within the film. Their positions, shapes, and amplitudes are very sensitive to the layer thickness and assumed profile. Clearly, the first fitting model only yields a qualitative fit. With the second model, the fit is much improved, indicating that the assumed profile is now much closer to the real profile. The associated refractive index profile (based on the best-fit results) is shown in Fig. 4. The best results were obtained from the third fitting model. Fitting results for this model, shown in Fig. 5, are visibly better than for the other two models. Figure 6 illustrates the associated refractive index profile. This index profile is also very consistent with the Auger profile (Fig. 1). It clearly demonstrates that the refractive index profile of the SiO_xN_y film is indeed graded nonuniformly, as suggested by the Auger profile. The final fitting results (i.e., thicknesses, volume fractions, and the σ values) are given in Table I.

The large values of σ in Table I indicate (a) the estimates for $\delta\psi$ and $\delta\Delta$ were too low (which is likely), and/or (b) the model used did not perfectly reflect the true structure and refractive index profile in the sample. Considering that a complex profile was modeled simply, with only a few fitting parameters, and that strong optical interference structure was present in the data, the σ values obtained are not unreasonable.

Another model was constructed to determine whether an interfacial layer existed at the SiO_xN_y film/Si substrate boundary. The model was similar to the third model described above, but added a thin interfacial layer, which consisted of a mixture of equal volume fractions of Si and Si_3N_4. The mixture's optical constants were modeled with the Bruggeman EMA. The fit improved significantly (by 30%) when the interfacial layer thickness was set to 30 Å, without changing the parameters of the film above it (except to adjust its total thickness). However, because of the complexity of the model and the possibility of hidden parameter correlations, we consider the results for the interface to be only qualitative.

V. CONCLUSION

It has been shown that an inhomogeneous SiO_xN_y thin film structure with nonuniformly graded index profile can be successfully characterized by SE. For the sample studied, both the film thickness and its refractive index profile were determined. The SE data are extremely sensitive to the refractive index profile, as well as the layer thickness. We therefore expect that SE will prove to be valuable for fast, quantitative, nondestructive, and possibly *in situ* analysis of more complicated rugate structures.

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