International Journal of New Horizons in Physics

http://dx.doi.org/10.18576/ijnhp/030102

Spectroellipsometric Characterization of Sputtered Silicon Nitride Films Using Two Different Dispersion Relations

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Received: 21 Nov. 2015, Revised: 22 Dec. 2015, Accepted: 24 Dec. 2015.
Published online: 1 Jan. 2016.

Abstract: Silicon nitride thin films were prepared on polished silicon wafers by reactive radio frequency sputtering. The optical constants and the thicknesses of the deposited layers were determined by spectroscopic ellipsometry in the wavelength range from 250 nm to 1690 nm. The dielectric function of the silicon nitride thin films was modeled by two different ways: on the one hand the Tauc-Lorentz dispersion relation was applied; on the other hand the combination of the Cauchy dispersion relation and the Urbach absorption tail was used. The evaluations of the measured ellipsometric spectra based on the two different modeling yielded similar results concerning the thickness of the surface roughness layer, as well as the thickness and the refractive index of the silicon nitride layers.

Keywords: Silicon nitride, optical properties, refractive index, sputtering, ellipsometry.

1 Introduction

Silicon nitride thin films find a considerable number of uses in technology, for example in semiconductor industry, photovoltaic applications and in optoelectronics because of their attractive properties. These films have been deposited by various techniques, such as pyrolysis of a mixture of silane and ammonia [1], plasma-enhanced chemical vapor deposition [2, 3], hot-wire chemical vapor deposition [4], or radio frequency (RF) sputtering [5]. The precise knowledge of the thickness and the optical properties of the deposited films are important in the design of multilayer optical systems. The aim of the present communication is to report the optical constants, layer thicknesses and growth rates of silicon nitride thin films deposited at room temperature by radio frequency sputtering.

2 Experimental

The silicon nitride layers were deposited by RF sputtering at room temperature onto single crystalline Si (100) substrates. RF sputter deposition was carried out in a Leybold Z400 apparatus pumped by a combination of a mechanical and a turbomolecular pump to a base pressure better than $5*10^{-5}$ Pa. Sputtering has been performed in high purity nitrogen gas with an applied wall potential in the range of 1.3 – 1.6 kV DC. The target was coupled to the RF generator operating at 13.56 MHz via a network for an impedance match between the generator and its load. A silicon wafer served as target for the deposition process. Table 1 shows the preparation parameters (the applied wall potential and the duration of deposition) for the different samples.

Table 1: Preparation parameters for the different silicon nitride samples.

Sample number	Voltage [kV]	Duration [min]
no-332-b	1.3	80
no-331-b	1.4	70
no-330-b	1.5	60
no-333-b	1.6	55

Characteristic optical properties of thin film structures can be derived from spectroscopic ellipsometry (SE) measurement, which is known to be a high-precision optical characterization technique [6-9]. Ellipsometry determines the change in polarized light upon light reflection on a sample. SE measures two quantities, Ψ and Δ. These are the amplitude ratio Ψ and phase difference Δ between light waves known as p- and s-polarized light waves. The method of Variable Angle Spectroscopic Ellipsometry (VASE) makes possible measurements at multiple angles of incidence. An additional angle will change the length of the light penetrating through the materials. Multiple angles are helpful to improve the confidence limits of the results yielded by the evaluation of the measured spectra.

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SE measurements have been done using a rotating compensator spectroscopic ellipsometer (Model M-2000DI, produced by the J.A. Woollam Co., Inc.) in the wavelength range of 191-1690 nm with angles of incidence of 64º, 67º and 70º; the measured data were analyzed using the computer code of WVASE32 [10].

3 Results and Discussion

The optical constants of the deposited layers have been evaluated from SE measurements using a three layer optical model consisting of a surface roughness layer, a bulk-like layer, a silicon dioxide layer and the substrate. The layer adjacent to the single crystalline silicon substrate represents the thin native silicon oxide film. The bulk-like layer represents the amorphous silicon nitride film, whereas its refractive index and extinction coefficient in the first case was modeled using the Tauc-Lorentz dispersion relation [11-13]. The roughness layer was taken into account on the basis of effective medium approximation [14], the roughness layer consists of 50% of the underlying material and 50% of void. The wavelength dependence of the refractive index and the extinction coefficient for the single crystalline silicon substrate and for the silicon dioxide was taken from the database of the computer code of WVASE32 [10]. During the evaluation of the measured SE data, eight free parameters were involved in the computation: the thickness of the surface roughness layer, the thickness of the bulk-like silicon nitride layer, the thickness of silicon oxide layer, and the other five ones are the parameters belonging to the Tauc-Lorentz dispersion relation. The calculated spectra were fitted to the measured ones using a regression algorithm. The measure of the fit quality is the mean squared error (MSE). The unknown parameters are allowed to vary until the minimum of MSE is obtained. In order to avoid the "local" minimum in the regression algorithm, a careful global search procedure has been performed (involving a wide range of starting parameter values).

Figs. 1 and 2 illustrate the measured and generated values of the ellipsometric parameters Ψ and Δ for sample no-333-b, respectively. The agreement between the measured and generated spectra is acceptable. Fig. 3 shows the refractive index n and extinction coefficient k as function of wavelength for the sample no-333-b. Table 2 shows the thickness of the surface roughness layers, the thickness of the bulk-like silicon nitride layers, the total layer thickness (the total layer thickness is equal to the thickness of the bulk-like silicon nitride layer plus the half of the thickness of the surface roughness layer), the parameters of the Tauc-Lorentz dispersion relation yielded by the evaluation, the growth rate data and the refractive index n and extinction coefficient k at the wavelength of 350 nm together with the mean squared error MSE characterizing the quality of the fit. We found that the refractive index n of the bulk-like silicon nitride layer varies in a relatively narrow range from 1.872 to 1.948 at the wavelength of 350 nm for different sample preparation parameters. These values are systematically smaller than refractive index of the

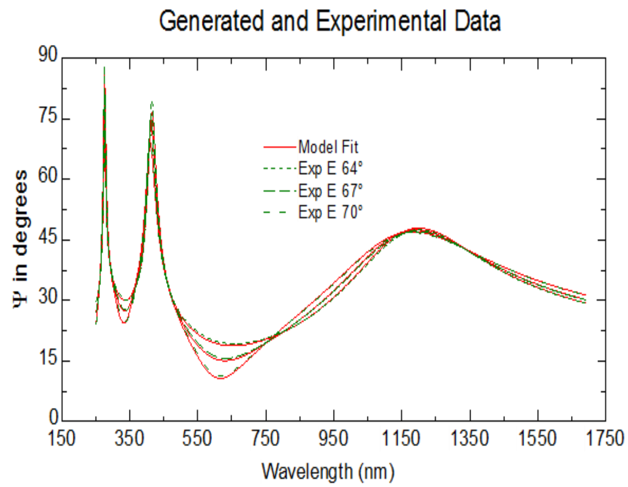

Fig. 1: Measured and fitted values of the ellipsometric parameter Ψ for sample no-333-b using a three-layer optical model based on the Tauc-Lorentz dispersion relation.

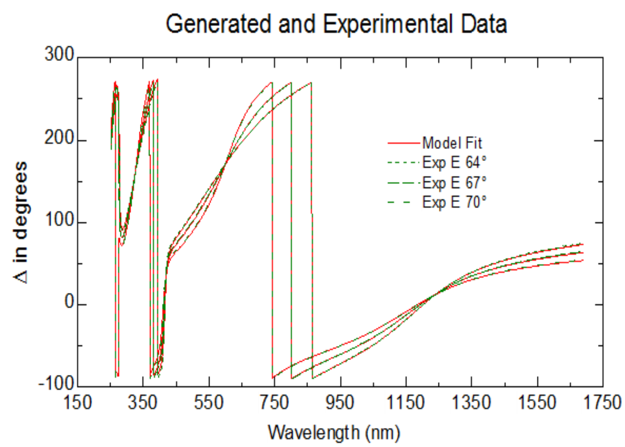

Fig. 2: Measured and fitted values of the ellipsometric parameter Δ for sample no-333-b using a three-layer optical model.

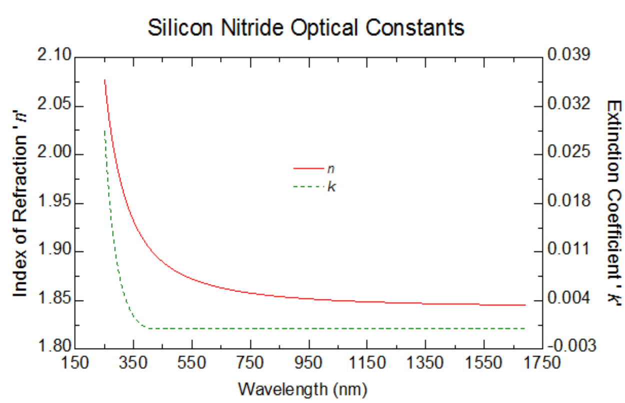

Fig. 3: Refractive index (n) and extinction coefficient (k) as a function of the wavelength for the bulk-like silicon nitride layer of sample no-333-b.

Table 2. Results of the evaluation based on the Tauc-Lorentz dispersion relation. Rows 2-5 show the thickness values, the forthcoming rows display the parameters of the Tauc-Lorentz dispersion relation, namely Ampl (amplitude), En (peak transition energy), C (broadening term), Eg (optical band gap), E1-Offset (additional fitting parameter). Row 11 displays the DC voltage applied during the RF sputtering, row 12 shows the values for the growth rate, rows 13 and 14 show the refractive indices and extinction coefficients at wavelength of 350 nm, row 15 displays the values of the mean squared error (MSE).

1	Sample number	no-332-b	no331-b	no-330-b	no-333-b
2	Thickness of the surface roughness layer [nm]	5.09±0.06	3.5±0.1	3.5±0.2	6.25±0.08
3	Thickness of the bulk-like silicon nitride layer [nm]	169.7±0.1	176.9±0.2	180.9±0.3	182.8±0.2
4	Total layer thickness [nm]	172.3±0.2	178.7±0.3	182.7±0.5	185.9±0.3
5	Thickness of the SiO_2 layer [nm]	0.79±0.09	1.2±0.2	6.4±0.3	0.8±0.1
6	Ampl [eV]	60.0±0.6	34.4±0.9	28.3±0.9	23.6±0.4
7	En [eV]	10.53±0.05	8.49±0.08	8.0±0.1	7.37±0.03
8	C [eV]	2.34±0.02	1.66±0.05	1.93±0.07	0.81±0.02
9	Eg [eV]	3.299±0.007	3.23±0.01	3.10±0.01	2.99±0.02
10	E1-Offset	0.89±0.03	1.91±0.03	2.09±0.04	2.34±0.01
11	DC Voltage [kV]	1.3	1.4	1.5	1.6
12	Growth rate [nm/min]	2.154	2.553	3.015	3.380
13	n (350 nm)	1.9016	1.9002	1.8847	1.9321
14	k (350 nm)	0.0007	0.0010	0.0024	0.0018
15	MSE	37.92	67.41	104.6	60.09

Table 3: Results of the evaluation based on the combination of the Cauchy dispersion relation with the Urbach absorption tail. The deposition parameters (columns 2-3) and parameters of the Cauchy dispersion ($n = A + B/\lambda^2 + C/\lambda^4$; columns 4-6) with Urbach absorption tail ($k = G*e^{H[E-F]}$ with F= 4.959 eV (250 nm); columns 7-8) together with the uncertainties yielded by the evaluation.

1	2	3	4	5	6	7	8
Sample number	Voltage [kV]	Duration [min]	A	B	C	G	H
no-332-b	1.3	80	1.8056± 0.0003	0.00945± 0.00008	0.000259± 0.000005	0.0302± 0.0004	2.22± 0.06
no-331-b	1.4	70	1.8135± 0.0004	0.0068± 0.0001	0.000413± 0.000008	0.037± 0.001	2.3± 0.1
no-330-b	1.5	60	1.7986± 0.0005	0.0063± 0.0002	0.00047± 0.00001	0.054± 0.002	2.03± 0.07
no-333-b	1.6	55	1.8438± 0.0004	0.00642± 0.00008	0.000517± 0.000006	0.0350± 0.0009	2.23± 0.08

stoichiometric silicon nitride (2.05 [5]) suggesting that the layers contain some oxygen from the residual gas of the chamber.

The DC voltage influences the growth rate markedly; the increase of the DC voltage from 1.3 kV to 1.6 kV caused an increase of the growth rate from 2.15 nm/min to 3.38 nm/min.

Table 4: Columns 2-5 contain the layer thickness values obtained during evaluation using the combination of the Cauchy dispersion relation and the Urbach absorption tail (the total layer thickness is equal to the thickness of the bulk-like silicon nitride layer plus the half of the thickness of the surface roughness layer). Columns 6-7 show refractive index n and the extinction coefficient k at the wavelength of 350 nm for the bulk-like silicon nitride layers. Column 8 displays the mean squared error MSE characterizing the fit quality.

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1	2	3	4	5	6	7	8
Sample number	Thickness of the surface roughness layer [nm]	Thickness of the bulk-like silicon nitride layer [nm]	Total layer thickness [nm]	Thickness of the SiO_2 layer [nm]	Refractive index n at the wavelength of 350 nm	Extinction coefficient k at the wavelength of 350 nm	Mean squared error MSE
no-332-b	5.01±0.07	170.5±0.1	173.0±0.2	0.0±0.2	1.8999	0.0013	42.41
no-331-b	3.6±0.1	178.1±0.2	179.9±0.3	0.0±0.2	1.8966	0.0014	69.59
no-330-b	3.8±0.2	181.4±0.2	183.3±0.5	5.8±0.2	1.8809	0.0030	109.7
no-333-b	6.38±0.09	183.4±0.2	186.6±0.3	0.2±0.1	1.9306	0.0015	60.13

In the second case the evaluation method based on the combination of the Cauchy dispersion relation and the Urbach absorption tail was selected. During the evaluation of the measured SE data, eight free parameters were involved in the computation: the thickness of the surface roughness layer, the thickness of the bulk-like silicon nitride layer, the thickness of silicon oxide layer, three parameters of the Cauchy dispersion formula and the other two ones are the parameters belonging to the expression describing the Urbach absorption tail. Table 3 list the deposition parameters (columns 2-3) and parameters of the Cauchy dispersion (columns 4-6) with Urbach absorption tail (columns 7-8) together with the uncertainties yielded by the evaluation. Table 4 shows the layer thickness values and the refractive index n and the extinction coefficient k at the wavelength of 350 nm for the bulk-like silicon nitride layers. The last column displays the mean squared error MSE characterizing the fit quality.

4 Conclusion

Silicon nitride thin films were deposited on single-crystalline silicon by radio frequency sputtering at room temperature. The measured ellipsometric spectra were evaluated using two different dispersion relations for modeling the wavelength dependence of the refractive index and extinction coefficient of the bulk like silicon nitride layer. The as-deposited silicon nitride films showed refractive index at the wavelength of 350 nm in the range of 1.881 - 1.931 according to the evaluation based on the combination of the Cauchy dispersion relation with the Urbach absorption tail. The evaluation based on the Tauc-Lorentz dispersion relation yielded a range of 1.885 – 1.932 for the refractive index at the wavelength of 350 nm. It is worthwhile to note that the combination of the Cauchy dispersion relation with the Urbach absorption tail does not satisfy the Kramers–Kronig relations. The thicknesses of the surface roughness layer and the bulk-like silicon nitride layer were determined with high precision. The increase in the DC voltage increases the layer growth rate markedly.

Acknowledgements

Authors are thankful to Ms. Amanda Wai for her assistance in the sample preparation. Support from OTKA K115852, M-ERA-NET „Watersafe” 117847, FP7 E450EDL and SEA4KET as well as the TÉT_12_DE-1-2013-0002 projects are greatly acknowledged.

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