



Optical characterization of inhomogeneous thin films with randomly rough boundaries

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Abstract: An inhomogeneous polymer-like thin film was deposited by the plasma enhanced chemical vapor deposition onto silicon single-crystal substrate whose surface was roughened by anodic oxidation. The inhomogeneous thin film with randomly rough boundaries was created as a result. This sample was studied using the variable-angle spectroscopic ellipsometry and spectroscopic reflectometry. The structural model including the inhomogeneous thin film, transition layer, and identically rough boundaries was used to process the experimental data. The scalar diffraction theory was used to describe the influence of roughness. The influence of the scattered light registered by the spectrophotometer due to its finite acceptance angle was also taken into account. The thicknesses and optical constants of the inhomogeneous thin film and the transition layer were determined in the optical characterization together with the roughness parameters. The determined rms value of the heights of roughness was found to be in good agreement with values obtained using AFM. The results of the optical characterization of the studied inhomogeneous thin film with rough boundaries were also verified by comparing them with the results of the optical characterization of the inhomogeneous thin film prepared using the same deposition conditions but onto the substrate with a smooth surface.

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1. Introduction

Homogeneous and inhomogeneous thin films are often encountered in many branches of fundamental research, applied research, and industrial innovations. Therefore, methods for studying their physical and chemical properties are needed. This implies that the optical properties of thin films are studied intensively. An enormous effort has been devoted to the development of methods of the optical characterization of homogeneous thin films so far (see e.g. [1–18]). Less attention has been devoted to the optical characterization of inhomogeneous thin films. Despite of this reality, several papers dealing with the optical characterization of these films have been published (see e.g. [19–32]). The homogeneous and inhomogeneous thin films often exhibit miscellaneous defects. Transition layers, overlayers, non-uniformity in thickness and optical constants, random boundary roughness, and artificial optical anisotropy belong to the defects occurring most often in practice.

Considerable attention has been devoted to the optical characterization of homogeneous thin films exhibiting these defects. For example, silicon single crystal substrates covered by thermally grown silicon oxide films containing transition layers were studied using ellipsometry in paper [33]. The influence of overlayers of ZnTe epitaxial thin films deposited onto gallium arsenide single crystal substrates on the optical characterization of these films was investigated in paper [34]. The optical characterization of homogeneous thin films exhibiting the thickness non-uniformity was performed, for example, in papers [35–42]. The influence of an artificial anisotropy on determining optical parameters of transparent homogeneous thin films was investigated, for

instance, in paper [43]. The homogeneous thin films exhibiting random roughness of boundaries were optically characterized, for example, in papers [43–52]. The Rayleigh-Rice theory (RRT) was utilized for expressing the optical quantities of the rough thin films exhibiting the rms values of heights of irregularities substantially smaller than wavelengths of incident light (see papers [44–48]). The scalar diffraction theory (SDT) was used to express optical quantities of the rough thin films with the larger rms values of the heights of the irregularities (see papers [45–47,49–52]).

Substantially less attention has been devoted to the optical characterization of inhomogeneous thin films exhibiting defects in comparison with homogeneous thin films. The influence of transition layers on the results of the optical characterization of inhomogeneous thin films was investigated in [53,54]. The optical characterization of the inhomogeneous thin films with thickness non-uniformity and transition layers was performed in papers [55,56]. To our knowledge, only one paper has been devoted to the optical characterization of inhomogeneous thin films exhibiting random roughness of their boundaries so far (see [57]). In this paper, the values the optical and roughness parameters of the inhomogeneous thin films originated by thermal oxidation of gallium arsenide single crystal substrates were determined by processing the experimental data corresponding to spectroscopic ellipsometry and spectroscopic reflectometry. For including random roughness into formulae for ellipsometric parameters and reflectance the RRT was utilized.

Note that it is often necessary to confirm the results obtained at the optical characterization of thin films exhibiting random roughness of boundaries by means of a separate determination of the parameters describing roughness of individual boundaries. There are many methods for determining these boundary parameters. Optical methods belong to the most efficient for this purpose. A detailed review of the papers dealing with the optical characterization of randomly rough surfaces is presented in paper [58]. Briefly speaking, the main optical techniques used to characterize various randomly rough surfaces are as follows: interferometry and interferometric microscopy (see e.g. [59–62]), spectroscopic photometry (see e.g. [63,64]), monochromatic and spectroscopic ellipsometry (see e.g. [64–68]), confocal laser scanning microscopy (see e.g. [69,70]), laser speckle techniques (see e.g. [71–73]) and techniques based on measuring scattered light (see e.g. [58,69,74–78]). The checking of the results obtained for the boundaries of the rough thin films within the optical characterization of these films can also be carried out using non-optical techniques. The most important of them is atomic force microscopy (AFM) (see e.g. [79,80]).

In this paper, the results achieved in the optical characterization of the inhomogeneous polymer-like thin film with randomly rough boundaries are presented. The optical characterization is based on the combined method of the variable-angle spectroscopic ellipsometry and spectroscopic reflectometry. It is shown that the description of the roughness using the SDT and the model with identically rough boundaries provides a suitable approach for interpreting the experimental data. In order to interpret the experimental data, it was also necessary to take into account the transition layer between the inhomogeneous polymer-like film and the substrate. The optical constants of the polymer-like thin film, the optical constants of the transition layer, the mean thicknesses of the thin film and the transition layer, and the roughness parameters of the boundaries are determined in the optical characterization. The values of the roughness parameters are verified by AFM. The results concerning the polymer-like thin film prepared under the same deposition condition but deposited on the substrate with a smooth surface are presented for comparison.

2. Experiment

The results concerning two samples of polymer-like films are presented in this paper. Both polymer-like films were prepared under the same deposition conditions but one was deposited onto the substrate with a rough surface and the other onto the substrate with a smooth surface.

Therefore, the boundaries of the first film exhibit random roughness while the boundaries of the other film are smooth.

2.1. Sample preparation

The inhomogeneous $\text{SiO}_x\text{C}_y\text{H}_z$ thin films were deposited by the plasma enhanced chemical vapor deposition (PECVD) using the parallel-plate PECVD reactor with capacitively coupled glow discharge at working frequency 13.56 MHz. The reaction chamber was made of a glass cylinder closed by two stainless steel flanges and the parallel electrodes were made of graphite. The film was deposited using the mixture of methane (CH_4) and hexamethyldisiloxane ($\text{C}_6\text{H}_{18}\text{Si}_2\text{O}$ - HMDSO). In order to create the inhomogeneity of the films, the methane flow rate was gradually reduced from 5.5 sccm to 0 sccm for 4 minutes. The supplied RF power was 50 W and it was applied to the lower electrode.

Two samples with $\text{SiO}_x\text{C}_y\text{H}_z$ inhomogeneous thin films were prepared during the same deposition. The first film was deposited onto a double side polished silicon-single crystal substrate. The second film was deposited onto a silicon-single crystal substrate with a randomly rough surface. The randomly rough surface was prepared by anodic oxidation of the substrate at a constant voltage followed by the dissolution of the grown oxide film in a mixture of water and hydrofluoric acid.

In order to increase the adhesion of the films, the silicon substrates were pretreated in argon (Ar) discharge for 5 minutes prior to the deposition of $\text{SiO}_x\text{C}_y\text{H}_z$ thin films. The applied power was 50 W and the flow rate of argon was 5 sccm.

2.2. Experimental data

The ellipsometric quantities were measured using the Horiba Jobin Yvon UVISSEL phase modulated ellipsometer for five incidence angles in the interval $55\text{--}75^\circ$ within the spectral range 0.6–6.5 eV (190–2066 nm). The experimental ellipsometric data are represented by the quantities I_s , I_c and I_n representing the independent components of the normalized Mueller matrix of isotropic systems [81]. The reflectance at near-normal incidence angle of 6° was measured using the Perkin Elmer Lambda 1050 spectrophotometer within the spectral range 0.69–6.5 eV (190–1800 nm).

The Bruker Dimension Icon atomic force microscope was used to investigate the topography of the surfaces of the samples.

3. Theoretical background

3.1. Structural model

The difference between the thin polymer-like films deposited onto the rough and smooth surfaces is that the boundaries of the first film are rough while the boundaries of the second film are smooth. Otherwise, both the films are assumed to have the same structure. The inhomogeneity of the polymer-like films corresponds to the profile of the optical constants along the coordinate perpendicular to the mean planes of the boundaries. Thin transition layers between the substrates and the inhomogeneous thin films are considered in the structural model. These transition layers are modeled as thin homogeneous layers.

In the structural model of the thin inhomogeneous film deposited onto the substrate with the rough surface, it was assumed that all boundaries exhibit identical roughness.

The substrate with smooth boundaries had both sides polished, therefore, it was necessary to take into account the reflections on the back side of the substrate, which influence the measured optical quantities in the spectral region where the substrate becomes transparent. The method described in [81] was used to include this effect in the calculation of reflectance and ellipsometric

quantities. The substrate with the randomly rough surface was polished only from the front side, therefore, reflections on the back side of the substrate were not considered in this case.

3.2. Inhomogeneous thin film with smooth boundaries

At each point of the inhomogeneous film, the optical constants are calculated on the basis of a Kramers–Kronig consistent dispersion model, which depends on several dispersion parameters. Therefore, the profile of the optical constants is given by specifying the dependencies of these dispersion parameters on the coordinate z , which determines the depth inside the inhomogeneous film. The profile of the optical constants of the inhomogeneous thin film is modeled by assuming a linear profile of the dispersion parameters

$$p_\alpha(z) = p_\alpha^U + (p_\alpha^L - p_\alpha^U) \frac{z}{d}, \quad (1)$$

where the index α is used to distinguish the individual dispersion parameters, $p_\alpha(z)$ is the value of the dispersion parameter at depth z inside the inhomogeneous film, p_α^U are the values of the dispersion parameters at the upper boundary of the film and p_α^L are the values of the dispersion parameters at the lower boundary.

The approximate method described in [25] is used to calculate the optical quantities of the inhomogeneous film. This approximate method combines the Richardson extrapolation with the method based on dividing the inhomogeneous film into a large number of thin homogeneous films with the optical constants chosen such that they approximate the continuous profile of the optical constants. The Richardson extrapolation is used to increase the rate of convergence to the exact results. It was found that dividing the inhomogeneous film into 32 thin homogeneous films provided sufficient accuracy.

Of course, there are also other methods that can be used to calculate the Fresnel coefficients of inhomogeneous thin films with arbitrary refractive index profiles (see e.g. [21,82]). All methods must give the same results provided the optical quantities of the inhomogeneous films are calculated with sufficient accuracy. For example, the method presented in [21] is based on the perturbation series which starts with the Fresnel coefficients corresponding to the Wentzel–Kramers–Brillouin–Jeffreys (WKBJ) approximation and then adds corrections corresponding to internal reflections in the inhomogeneous layer. In this case, the corrections of at least the first order, which account for single reflections inside the inhomogeneous film, must be included in order to obtain accurate results.

3.3. Inhomogeneous thin film with randomly rough boundaries

The theoretical approach based on the SDT is used to calculate the optical quantities of the inhomogeneous polymer-like thin film deposited onto the substrate with a rough surface. The approach using the SDT is valid if the roughness can be viewed as locally smooth. In theory, this means that the lateral dimensions of roughness should be much larger than the wavelength of light, however, in practice, the SDT works well even if the lateral dimensions of roughness are comparable to the wavelength of light. Within the SDT the reflection coefficients of thin films with identically rough boundaries are expressed using the following formula [22,65,67]

$$r_{\text{SDT},q} = \int_{-\infty}^{\infty} \int_{-\cot \theta_0}^{\cot \theta_0} \int_{-\infty}^{\infty} r_q(\eta, \eta_x, \eta_y) w(\eta, \eta_x, \eta_y) e^{ivz\eta} d\eta d\eta_x d\eta_y, \quad (2)$$

where the index $q = p, s$ is used to distinguish between the p and s polarized waves. The symbol $\eta = \eta(x, y)$ represents the values of the function describing the deviations from the mean plane, $\eta_x = \partial_x \eta(x, y)$ and $\eta_y = \partial_y \eta(x, y)$ are the derivatives of $\eta(x, y)$ with respect to coordinates x and y . The symbol $r_q(\eta, \eta_x, \eta_y)$ denotes the local reflection coefficient, which, in general, depends on the deviations from the mean plane η at the given point and on the slope of the tangent plane, i.e.

on the derivatives η_x and η_y . The symbol $w(\eta, \eta_x, \eta_y)$ denotes the probability density function for the distribution of η , η_x and η_y . The symbol v_z represents the z -component of the vector given as the difference of the wavevector of the incident wave and the wavevector of the reflected wave. It can be calculated as

$$v_z = \frac{4\pi}{\lambda} n_0 \cos \theta_0, \quad (3)$$

where λ is the wavelength of the incident light, θ_0 is the incidence angle and n_0 is the refractive index of the ambient. For surfaces and thin films with identically rough boundaries, the local reflection coefficients do not depend on the deviation from the mean plane η . Moreover, if the slopes of the roughness are small, we can neglect the dependence of the local reflection coefficient on η_x and η_y . The formula (2) is then simplified into the following form

$$r_{\text{SDT},q} = r_{0,q} \int_{-\infty}^{\infty} w_1(\eta) e^{iv_z \eta} d\eta = r_{0,q} \chi_1(v_z), \quad (4)$$

where $r_{0,q}$ is the reflection coefficient corresponding to the thin film with perfectly smooth boundaries, $w_1(\eta)$ is the probability density function for the distribution of the heights of the roughness and $\chi_1(v_z)$ is the corresponding characteristic function. The reflection coefficients $r_{0,q}$ are calculated in the manner described in section 3.2, i.e. in the same way as for the inhomogeneous polymer-like thin film deposited onto the substrate with a smooth surface.

In this work, the one-dimensional distribution of the heights of the roughness is modeled using the normal distribution. The probability density function and the characteristic function are then given as

$$w_1(\eta) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{\eta^2}{2\sigma^2}\right), \quad \chi_1(v_z) = \exp\left(-\frac{1}{2}\sigma^2 v_z^2\right). \quad (5)$$

When expressing the reflectance, it is necessary to take into account not only the coherent contribution, which corresponds to the specular reflectance, but also the incoherent contribution, which corresponds to the light scattered on the rough boundaries and registered by the spectrophotometer due to its finite acceptance angle. The total reflectance R_T is, therefore, expressed as [83–85]

$$R_T = R_c + R_i, \quad (6)$$

where R_c represents the coherent component and R_i represents the incoherent component. The coherent component can be calculated from the reflection coefficient as

$$R_c = |r_{\text{SDT}}|^2 = |r_0|^2 |\chi_1(v_z)|^2 = R_0 \exp\left(-\frac{16\pi^2 n_0^2 \sigma^2}{\lambda^2}\right), \quad (7)$$

where $R_0 = |r_0|^2$ is the reflectance that would be measured if the boundaries were perfectly smooth. Note that since we assume near-normal incidence of light, it is not necessary to use the index q distinguishing the p and s polarized waves in the above formula.

In order to calculate the incoherent component, it is necessary to specify the two-dimensional probability density function for distribution of heights of roughness at two (possibly distinct) points. This function can be chosen as follows [83–85]

$$w_2(\eta_1, \eta_2; t) = w_1(\eta_1) \delta(\eta_1 - \eta_2) C(t) + w_1(\eta_1) w_1(\eta_2) [1 - C(t)], \quad (8)$$

where η_1 and η_2 are the heights at two (possibly different) points, t is distance between these two points in the mean plane and the symbol $C(t)$ denotes the autocorrelation coefficient. The

autocorrelation coefficient will be modeled using the Gaussian function

$$C(t) = \exp(-t^2/\tau^2), \quad (9)$$

where τ is the autocorrelation length. The incoherent part of the reflectance is then given by the following formula [83]

$$R_i = R_0 \left[1 - \exp\left(-\frac{16\pi^2 n_0^2 \sigma^2}{\lambda^2}\right) \right] \left[1 - \exp\left(-\frac{\pi^2 n_0^2 (\alpha\tau)^2}{\lambda^2}\right) \right], \quad (10)$$

where the symbol α is the semivertex angle of the cone of acceptance of the detector.

It should be noted that because the slopes of the roughness are neglected in (4), the ellipsometric ratio $r_{\text{SDT,p}}/r_{\text{SDT,s}} = r_{0,p}/r_{0,s}$ is independent on the roughness parameters. Therefore, the method presented in this section predicts that the ellipsometric quantities are not affected by the random roughness of the boundaries.

3.4. Dispersion models

The Campi–Coriasso dispersion model is used to describe both the inhomogeneous film and the transition layer between this film and the substrate. The imaginary part of the dielectric function is given by the following formula [86,87]

$$\varepsilon_i(E) = \frac{2N_{\text{vc}}}{\pi E} \frac{B(E - E_g)^2 \Theta(E - E_g)}{[(E_c - E_g)^2 - (E - E_g)^2]^2 + B^2(E - E_g)^2}, \quad (11)$$

where E denotes photon energy, N_{vc} is the strength of the interband electronic transitions, $\theta(\cdot)$ represents the Heaviside function, E_g is the band gap energy and E_c and B are the parameters ($E_c > E_g$). The real part of the dielectric function is calculated from the imaginary part using the Kramers–Kronig relations.

While the optical constants of the transition layer are determined by four parameters N_{vc} , E_g , E_c , B of the Campi–Coriasso dispersion model, it is necessary to specify two sets of these parameters for the inhomogeneous thin film. The first set N_{vc}^{U} , E_g^{U} , E_c^{U} , B^{U} determines the optical constants at the upper boundary of the inhomogeneous film and the second set N_{vc}^{L} , E_g^{L} , E_c^{L} , B^{L} determines the optical constants at the lower boundary.

It was found that seeking the values of the parameters B^{U} and B^{L} in the Campi–Coriasso model (11) independently at the upper and lower boundaries of the inhomogeneous thin film did not improve the quality of the fit of the experimental data and only increased correlation among the sought parameters. For this reason, the same value was used at the upper and lower boundaries (i.e. $B^{\text{U}} = B^{\text{L}}$).

The optical constants of the silicon single-crystal substrate were fixed in values determined in [88].

3.5. Data processing

The values of the structural and dispersion parameters were determined using the least-squares method. Within the least-squares method, the ellipsometric and spectrophotometric data for each sample were processed simultaneously.

Two values of the thickness of the inhomogeneous thin film were determined within the processing of the experimental data. One of the thicknesses was determined on the basis of the ellipsometric experimental data and the other was determined on the basis of measured spectral dependencies of reflectance. It was necessary to assume that these thicknesses are not the same, since each instrument exhibits different systematic errors. Moreover, the films could exhibit slight non-uniformity in thicknesses and we could not ensure that the ellipsometric and spectrophotometric measurements were performed on the exactly same spot on the sample.

3.6. Results

The topography of the sample with substrate roughened by the anodic oxidation was measured by AFM before the polymer-like thin film was deposited onto it and after the deposition of the film was finished. In this way it was possible to obtain information concerning the roughness of the upper as well as the lower boundaries of the thin film. The AFM scans of the topography of the silicon substrate with rough surface and the upper boundary of the inhomogeneous film deposited onto it are shown in Fig. 1. No other filtering apart from the plane leveling was performed for the AFM scans. The rms values of the heights of the rough silicon surface and the upper boundary determined from the AFM scans are 14.7 nm and 15.2 nm, respectively. The fact that these values are very similar supports the assumption that the roughness of the thin film can be described using the model with identically rough boundaries. The rms values of the heights of roughness were determined from the variance of the z values calculated for the set consisting of all pixels of the AFM scans. In the case of the inhomogeneous thin film deposited onto the substrate with a smooth surface, it was found that the roughness of the upper boundary is below the resolving power of AFM.

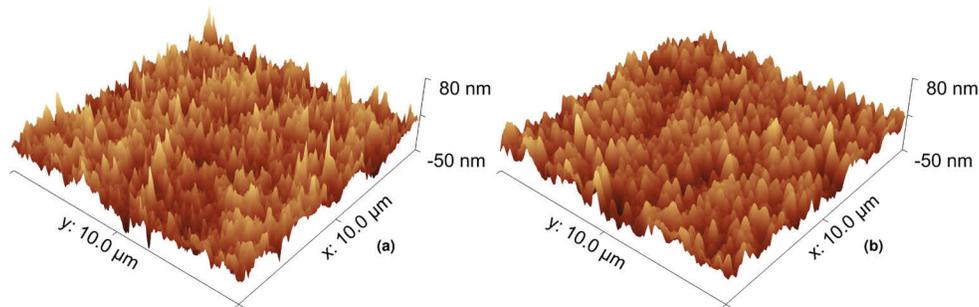


Fig. 1. Topography of (a) the rough silicon surface and (b) the upper boundary of the inhomogeneous thin film.

The distributions of the heights of the rough surface and the upper boundary of the inhomogeneous thin film determined from the AFM scans are shown in Fig. 2. The fits by the Gaussian functions, which are also shown in this figure, correspond to the rms values of the heights 14.26 ± 0.03 nm for the rough surface and 15.11 ± 0.04 nm for the upper surface of the inhomogeneous thin film. It is evident that the fits by the Gaussian functions are very good, which means that the assumption of normally distributed heights of roughness used in the theoretical part is correct.

The measured spectral dependencies of ellipsometric quantities and reflectance together with fits by theoretically calculated curves are shown in Figs. 3 and 4. The values of the structural and dispersion parameters giving to the best fit of experimental data are shown in Tables 1–3. The uncertainties of the parameters shown in these tables correspond to those determined by the least-squares method. The rms value of the heights of the roughness $\sigma = 14.7$ nm agrees well with the values 14.7 nm and 15.2 nm determined by AFM.

It should be noted that only the product $\alpha\tau$ of the semivertex acceptance angle α and the autocorrelation length τ can be determined from the optical measurements. We should be careful when interpreting the value of $\alpha\tau$. Firstly, the equation (11) was derived using the assumption that the spectrophotometer accepts light falling within the ideal cone with semivertex angle α while the real behavior of the spectrophotometer is certainly more complex. Secondly, only the part of the roughness with lateral dimensions larger than $\lambda/(2\pi \sin \alpha)$ contributes to the scattered light falling within the cone of acceptance. Therefore, the autocorrelation length τ is relevant only for the description of roughness in this limited interval of spatial frequencies. For these

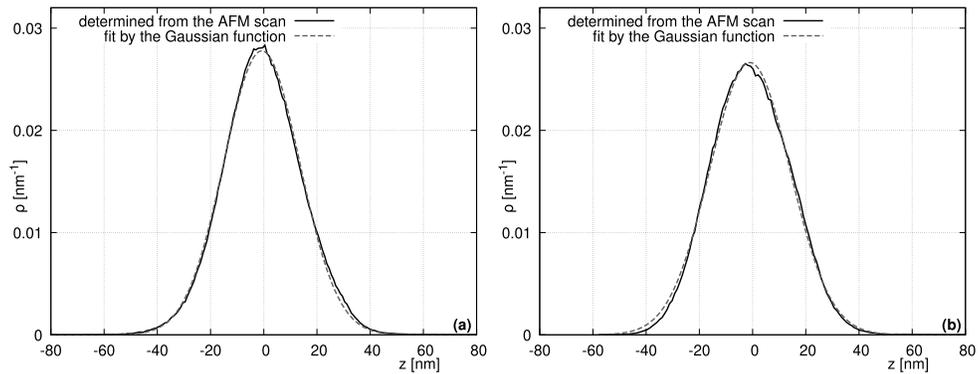


Fig. 2. The distribution of heights determined from the AFM scans of (a) the rough silicon surface and (b) the upper boundary of the inhomogeneous thin film.

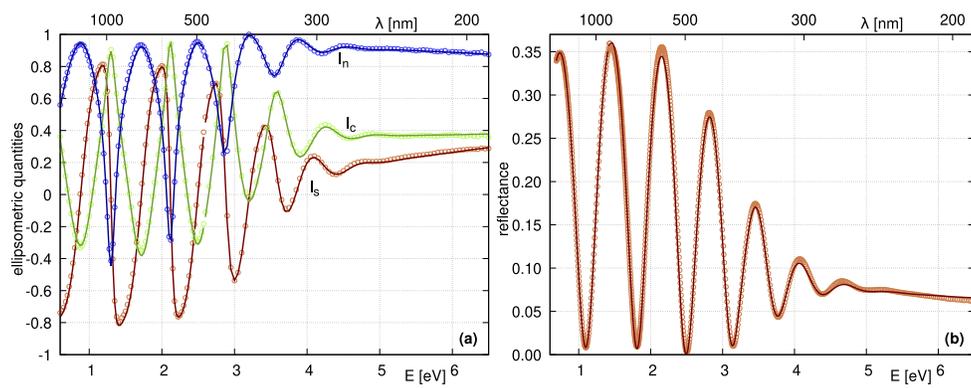


Fig. 3. Spectral dependencies of (a) the ellipsometric quantities at incidence angle 70° and (b) reflectance for inhomogeneous thin film with rough boundaries. Points represent experimental values, curves represent the fits by theoretically calculated values.

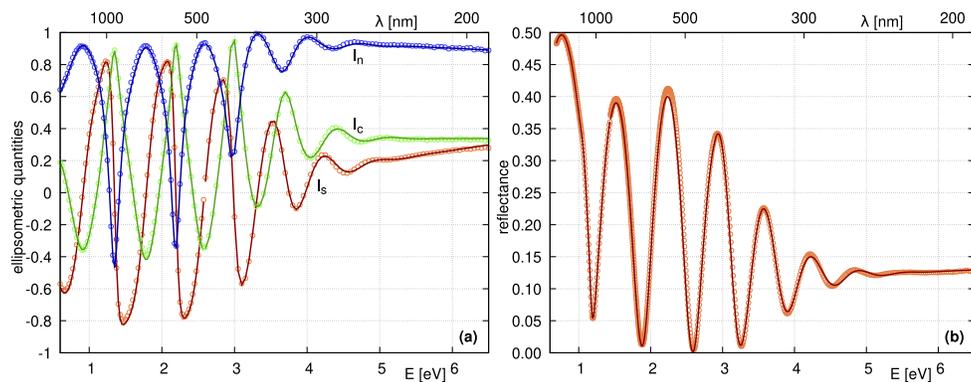


Fig. 4. Spectral dependencies of (a) the ellipsometric quantities at incidence angle 70° and (b) reflectance for inhomogeneous thin film with smooth boundaries. Points represent experimental values, curves represent the fits by theoretically calculated values.

Table 1. Values of the structural parameters.

parameter		rough	smooth
thickness of inhomogeneous film (ellipsometry)	d_e [nm]	503.6 ± 0.2	482.2 ± 0.4
thickness of inhomogeneous film (reflectance)	d_r [nm]	498.1 ± 0.1	477.3 ± 0.3
thickness of the transition layer	d_{tl} [nm]	21.4 ± 0.4	20.5 ± 0.6
rms value of the heights of roughness	σ [nm]	14.7 ± 0.2	
product of autocorrelation length and acceptance half-angle	$\alpha\tau$ [nm]	33 ± 2	

Table 2. Values of the dispersion parameters – thin film with rough boundaries.

parameter		inhomogeneous film		transition
		upper	lower	layer
Total strength of interband transitions	N_{vc} [eV ²]	959 ± 39	989 ± 35	1095 ± 165
Band gap energy	E_g [eV]	1.37 ± 0.01	3.32 ± 0.06	2.62 ± 0.01
Peak position	E_c [eV]	13.9 ± 0.2	16.8 ± 0.2	5.5 ± 0.2
Peak width	B [eV]		23 ± 1	8 ± 2

Table 3. Values of the dispersion parameters – thin film with smooth boundaries.

parameter		inhomogeneous film		transition
		upper	lower	layer
Total strength of interband transitions	N_{vc} [eV ²]	754 ± 20	944 ± 25	1268 ± 222
Band gap energy	E_g [eV]	1.46 ± 0.02	2.77 ± 0.05	2.87 ± 0.01
Peak position	E_c [eV]	12.3 ± 0.1	17.4 ± 0.2	4.9 ± 0.2
Peak width	B [eV]		16.2 ± 0.5	10 ± 2

reasons, it is quite difficult to interpret the individual terms in the product $\alpha\tau$. Despite these limitations, it was shown in paper [66] that this model is successful in interpreting the reflectance spectra of samples with rough surfaces in the ultraviolet region.

The spectral dependencies of the optical constants of the inhomogeneous thin films at the upper and the lower boundaries and the profiles of the optical constants at selected photon energy are shown in Fig. 5. It is evident that the optical constants corresponding to the inhomogeneous thin films deposited onto substrates with rough and smooth surfaces are very similar. They are almost identical for photon energies below $E \lesssim 3$ eV, however, somewhat larger differences are visible in the ultraviolet region. This should not be too surprising since there is no reason to assume that the polymer-like thin films grow identically when they are deposited onto the rough and smooth surfaces. That the films are not completely identical is also obvious from the fact that they have slightly different thicknesses (see Table 1). It should be noted that the accuracy of the determined optical constants in the ultraviolet region may be affected by the use of a very simple model, in which the roughness does not have any influence on the ellipsometric quantities. On the other hand, the good fits of the experimental data by theoretically calculated curves and the fact that the rms value of the heights of the roughness determined by the optical method agrees with the values determined by AFM indicate that the model works well.

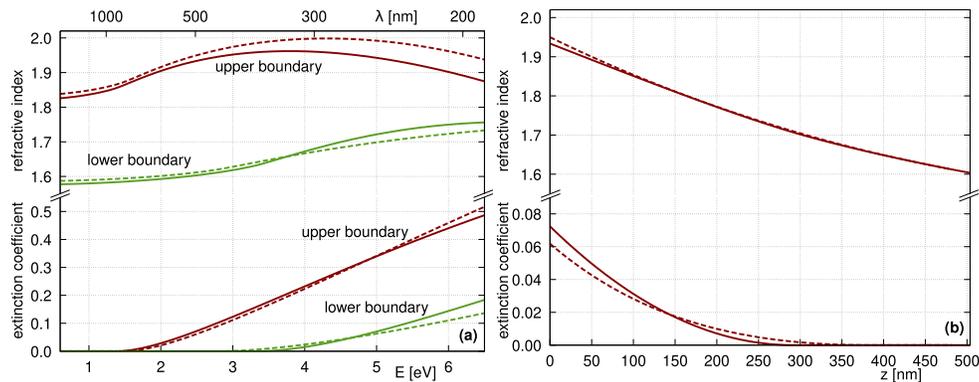


Fig. 5. (a) Spectral dependencies of the optical constants of the inhomogeneous thin films. (b) Profiles of the optical constants calculated for the photon energy $E = 2.5$ eV. The solid curves correspond to the thin film with rough boundaries while the dashed curves correspond to the thin film with smooth boundaries.

A more complex description of the random roughness which takes into account the influence of the slopes on the ellipsometric quantities would require a model with more parameters. Since the description of inhomogeneity already requires a relatively large number of parameters, adding more parameters describing roughness leads to overparameterization. If we wanted to use such a model in the optical characterization, then some of the parameters would have to be fixed in values determined by some other independent method. For example, we could fix the values of the roughness parameters in values determined by AFM, and seek only the optical constants and thicknesses of the inhomogeneous thin film and transition layer within the optical characterization. However, the sensitivity to components of the roughness with different spatial frequencies is different for AFM and the optical methods, therefore, extracting the roughness parameters relevant for the optical characterization from the AFM scans would be a difficult task.

The spectral dependencies of the optical constants of the transition layers are shown in Fig. 6. The most likely explanation for the presence of the transition layers is that they represent the upper layers of the silicon substrates with structure perturbed by the cleaning in the argon plasma. This is supported by the fact that their optical constants are close to those of the crystalline silicon

but with less pronounced structures. From Table 1 and Fig. 6 it is evident that the thicknesses and optical constants of the transition layers are almost the same for the films deposited onto the substrates with rough and smooth surfaces.

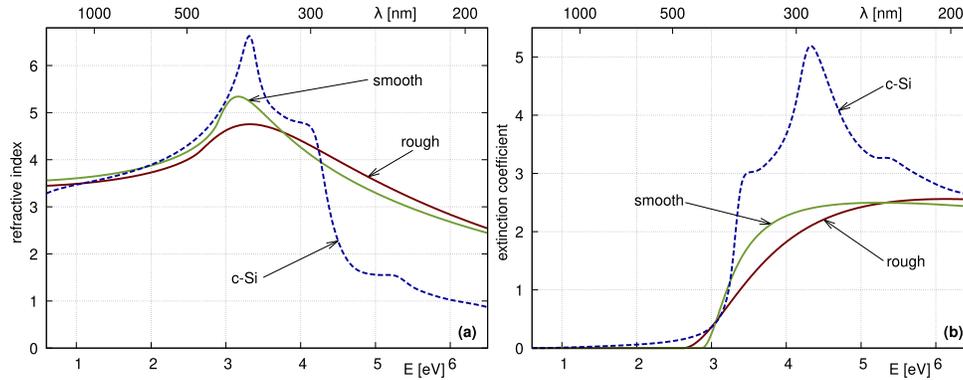


Fig. 6. Spectral dependencies of (a) the refractive index and (b) the extinction coefficient of the transition layers. The dashed curves represent the optical constants of crystalline silicon.

It should be noted that the degree of polarization $P = (I_s^2 + I_c^2 + I_n^2)^{1/2}$ calculated from the experimentally measured ellipsometric quantities is very close to unity. The small differences from unity can be explained by considering a finite spectral resolution of the ellipsometer. If this effect is taken into account, then the agreement between the experimental and theoretically calculated values is within the experimental errors. Therefore the roughness does not introduce depolarization. This means that either the contribution of the scattered light is negligible or the measured ellipsometric quantities are not influenced because its polarization state is identical with that of the specularly reflected beam.

4. Conclusion

The sample of the inhomogeneous polymer-like thin film with randomly rough boundaries was studied using the variable-angle spectroscopic ellipsometry and spectroscopic reflectometry. The boundaries of the polymer-like thin film are rough because it was deposited on the substrate with a rough surface.

The scalar diffraction theory and a simple model assuming identically rough boundaries were used for describing the influence of roughness on the measured optical quantities. In order to correctly interpret the measured values of reflectance, it was also necessary to take into account that part of the scattered light is registered by the spectrophotometer due to its finite acceptance angle. A transition layer between the inhomogeneous thin film and the silicon single-crystal substrate was also considered in the structural model. The Campi–Coriasso dispersion model was used to model the optical constants of both the inhomogeneous thin film and the transition layer. The thicknesses and optical constants of the inhomogeneous thin film and the transition layer were determined in the optical characterization together with the roughness parameters. The rms values of the heights of the roughness of the upper and lower boundaries of the inhomogeneous thin film were also determined using AFM and it was found that their values agree well with the value determined in the optical characterization. The optical characterization of the inhomogeneous polymer-like thin film with smooth boundaries prepared under the same deposition conditions as the film with rough boundaries was also performed in order to verify the results concerning the optical constants and thicknesses of the inhomogeneous thin film and transition layer.

The results of this work show that it is possible to use the optical method to investigate the properties of complex systems that combine optical inhomogeneity, transition layers and random

roughness of boundaries. However, it should be emphasized that some compromises must be made when choosing the structural and dispersion models. Even if the boundaries are smooth, the model assuming the inhomogeneous thin film and transition layer requires a relatively large number of parameters. Therefore, describing the randomly rough boundaries using a model that is too complex and requires many parameters could lead to overparameterization.

The heights of roughness influence the reflectance in a relatively straightforward way, and, moreover, the influence of the heights roughness is relatively uncorrelated with the effects introduced due to the inhomogeneity of the film and the presence of the transition layer. This is probably one of the reasons, why the presented method allowed to determine the rms value of the heights of roughness relatively precisely for the studied sample, and why we can expect the method to succeed also for samples exhibiting roughness with similar properties.

The property that the roughness of the lower boundary is almost identical with that of the upper boundary as well as the presence of the transition layer are relatively general properties of polymer-like films prepared using the employed deposition technique (i.e. the plasma enhanced chemical vapor deposition). Therefore, it is reasonable to expect that the approach presented in this work will be usable for a wide range of films prepared using this technology differing in the rms values of the heights of the roughness, film thicknesses, and profiles of the optical constants of the inhomogeneous thin films. Of course, the model should work also for thin films prepared using other technologies as long as the relevant conditions are fulfilled.

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