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PAPERS

Approximate methods for the optical characterization of inhomogeneous thin films: Applications to silicon nitride films

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In this paper the overview of the most important approximate methods for the optical characterization of inhomogeneous thin films is presented. The following approximate methods are introduced: Wentzel–Kramers–Brillouin–Jeffreys approximation, method based on substituting inhomogeneous thin films by multilayer systems, method based on modifying recursive approach and method utilizing multiple-beam interference model. Principles and mathematical formulations of these methods are described. A comparison of these methods is carried out from the practical point of view, *ie* advantages and disadvantages of individual methods are discussed. Examples of the optical characterization of three inhomogeneous thin films consisting of non-stoichiometric silicon nitride are introduced in order to illustrate efficiency and practical meaning of the presented approximate methods.

Keywords: reflectance, ellipsometric parameters, inhomogeneous thin films, optical characterization

1 Introduction

Optics of thin films belongs to fields of fundamental and applied research exhibiting very extensive scientific activities. This is given by important applications of results achieved within this field in practice. This reality is demonstrated by enormous number of scientific articles and many monographs dealing with optics of thin films. Enormous attention has been devoted to optics of homogeneous thin films in particular (see *eg* [1-13]). A less attention has been devoted to optics of inhomogeneous thin films since from the optical point of view these films are more complicated than homogeneous thin films. This complication consists in fact that inhomogeneous thin films exhibit continuous profiles of the optical constants, *ie* refractive index and extinction coefficient, across these thin films. In other words, functions of single variable describe courses of the refractive index and extinction coefficient from the lower to upper boundaries of these inhomogeneous thin films (single variable corresponds to axis perpendicular to parallel boundaries of these films). As for optics of inhomogeneous thin films one can recommend, for example, these papers [14-29]. Within the last three decades an interest concerning optics of inhomogeneous thin films was intensified. This is caused by better optical behavior of these thin films in comparison with multilayer systems formed by homogeneous thin films with mutually different refractive indices. The inhomogeneous films can substitute these systems in producing various optical devices. For example, inhomogeneous thin films exhibit substantially lower light scattering caused by boundary roughness than the multilayer systems. It is possible to mention rugate filters that consist of single inhomogeneous thin films having relatively complicated profile of refractive index. The rugate filters are practically significant because they can be employed as optical devices with high reflectance within spectral ranges of interest (see *eg* [26, 27]). In some applications they are employed instead of reflectors formed by the multilayer systems exhibiting scattering of light that is not negligible. Antireflection coatings formed by single inhomogeneous thin films with suitable refractive index profiles can serve as other example of a possibility to replace multilayer systems by single inhomogeneous thin films (see *eg* [2]).

The other reason, why the inhomogeneous thin films become more interesting than before, is given by a necessity to perform the optical characterization of thin films consisting of complex materials. The films formed by complex materials are created in modern branches of fundamental and applied research such as microelectronics, nanotechnology, solar energetics *etc.* These films are often inhomogeneous from the optical point of view which implies an effort to develop new and more efficient methods of the optical characterization of these inhomogeneous thin films. This is especially true if inhomogeneous thin films exhibit unusual and complicated refractive index profiles. Representatives of the complex inhomogeneous thin films are silicon nitride films that are rich in silicon (see *eg* [23, 24]). In general it is possible to state that chemical and plasma chemical technologies

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frequently produce the thin films exhibiting more or less degree of optical inhomogeneity.

Optical techniques such ellipsometry and reflectometry are utilized for real-time monitoring and feedback of a growth of homogeneous and inhomogeneous thin films at their preparation in technological arrangements. In this case it is necessary to have in situ methods of their optical characterization with a high speed of calculations of the corresponding optical quantities, which is required by efficient computer algorithms. This fact represents a further reason for developing new efficient methods of the optical characterization of the inhomogeneous thin films and their structures. New approaches are still developed in design of systems formed by inhomogeneous thin films as well.

In this overview paper the approximate methods for the optical characterization of the inhomogeneous thin films mostly utilized in practice will be presented. We will concentrate on the methods corresponding to reflected light from these films. Their illustration will be performed through examples of the optical characterization of inhomogeneous thin films of non-stochiometric silicon nitride.

2 Structural model of inhomogeneous thin films

The structural model of the inhomogeneous thin film used here is given by the following assumptions:

- Ambient is homogeneous, isotropic and non-absorbing.
- Substrate and thin film consist of isotropic, absorbing and non-magnetic materials.
- Boundaries are smooth, flat and mutually parallel.
- Transition layers are not taken into account at the boundaries so that the boundaries are infinitely thin.
- Substrate is optically homogeneous.
- No defects are taken into account in thin film volume.
- Thin film is inhomogeneous, *ie* its refractive index profile is described by a function of single variable corresponding to the axis perpendicular to the parallel boundaries.

3 Description of approximate methods

Theory and formulations of the four approximate methods for optical characterization of the inhomogeneous thin films are described in this section.

3.1 Wentzel–Kramers–Brillouin–Jeffreys (WKBJ) method

This approximate method can be used for the optical characterization of the inhomogeneous thin films if the gradients of their refractive index profiles are very small so that these profiles only influence phase angles of these films. In this case the inhomogeneous thin film can be replaced by a multilayer system containing a large number of sub-layers. The internal reflections from the boundaries of the sub-layers are neglected. Thus, light reflections are only considered from the uppermost and lowest boundaries, *ie* from the boundary between the ambient and adjacent sub-layer and from the boundary between substrate and adjacent sub-layer. After performing the limit for the number of the dividing sub-layers going to infinity one obtains the following formula for the reflection coefficients of this inhomogeneous thin film (for details see *eg* [2])

$$r_0 = \frac{r_\mathrm{U} + r_\mathrm{L}\exp(\mathrm{i}x_d)}{1 + r_\mathrm{U} r_\mathrm{L}\exp(\mathrm{i}x_d)}, \tag{1}$$

$$r_\mathrm{U} = \frac{Y_0 - Y_\mathrm{U}}{Y_0 + Y_\mathrm{U}},\ r_\mathrm{L} = \frac{Y_\mathrm{L} - Y_\mathrm{S}}{Y_\mathrm{L} + Y_\mathrm{S}},$$

$$x_d = \frac{4\pi}{\lambda}\int_0^d \sqrt{n^2(z) - n_0^2\sin^2\varphi_0}\,\mathrm{d}z.$$

Symbols d, n_0, φ_0 and λ represent the thickness of the film, refractive index of the ambient, incidence angle of light onto the upper boundary of the film and wavelength of incident light, respectively. The parameters d and n_0 and variable quantities φ_0 and λ are always real. The other parameters and quantities can be complex in general. Symbols r_0, r_U and r_L denote the reflection coefficients of the film, upper boundary and lower boundary for both the polarizations, *ie* for the p-polarization and s-polarization, respectively. Symbol $n(z)$ represents a function of coordinate z describing the refractive index profile across the film (z is coordinate corresponding to axis perpendicular to the parallel boundaries of the inhomogeneous film). Symbols Y_0, Y_U, Y_L and Y_S denote the optical admittances of the ambient, admittance at the upper boundary, admittance at the lower boundary and admittance of the substrate for both the polarizations, respectively. Note that the following equations are valid for the admittances corresponding to the s-polarization ($Y_{0,\mathrm{s}}$, $Y_{\mathrm{U,s}}$, $Y_{\mathrm{L,s}}$ and $Y_{\mathrm{S,s}}$) and p-polarization ($Y_{0,\mathrm{p}}$, $Y_{\mathrm{U,p}}$, $Y_{\mathrm{L,p}}$ and $Y_{\mathrm{S,p}}$)

$$Y_{i,\mathrm{s}} = n_i\cos\varphi_i,\ \ Y_{i,\mathrm{p}} = \frac{n_i}{\cos\varphi_i},$$

where the index $i = 0, \mathrm{U}, \mathrm{L}, \mathrm{S}$. Symbol x_d denotes the phase angle of the film. Symbols n_U, n_L and n_S represent the refractive indices at the upper boundary, at the lower boundary and the refractive index of the substrate, respectively, while symbols φ_U, φ_L and φ_S are the refraction angles corresponding to the upper boundary, lower boundary and substrate, respectively. It is evident that the following equalities are fulfilled

$$\begin{aligned} n_0\sin\varphi_0 = {} & n_\mathrm{U}\sin\varphi_\mathrm{U} = n(z)\sin\varphi(z) = \\ & n_\mathrm{L}\sin\varphi_\mathrm{L} = n_\mathrm{S}\sin\varphi_\mathrm{S}, \end{aligned}$$

where $\varphi(z)$ is the refraction angle at coordinate z.

The reflection coefficients enable us to calculate reflectances and ellipsometric quantities in reflected light.

By means of these measured optical quantities one can perform the optical characterization of the thin films studied (the formulae for the optical quantities will be presented below). In the case of this approximate method the complete optical characterization of the inhomogeneous thin film is performed if the spectral dependencies of the refractive indices at upper and lower boundaries are determined together with the thickness value (it is usually true that the spectral dependencies of the refractive indices of the ambient and substrates are known or they can be determined in independent ways).

3.2 Approximate method based on replacing inhomogeneous films by multilayer systems

This approximate method is frequently utilized for the optical characterization of the inhomogeneous thin films exhibiting refractive index profiles with relatively large gradients because the WKBJ approximation does not provide satisfactory results in this case. This method is based on replacing the inhomogeneous thin film by a multilayer system containing a large number of sub-layers with very small thicknesses and very small differences in refractive indices of the adjacent sub-layers. The internal reflections from the boundaries of the sub-layers cannot be neglected. This means that the recursive formulae or matrix formalism must be utilized for expressing the reflection coefficients of the multilayer systems substituting the inhomogeneous thin films. Within the recursive approach the formula for these reflection coefficients r of the multilayer system containing N sub-layers is given as follows (see *eg* [1, 2])

$$\begin{aligned} r =& \frac{r_1 + \bar{r}_2 \mathrm{e}^{\mathrm{i}x_1}}{1 + r_1 \bar{r}_2 \mathrm{e}^{\mathrm{i}x_1}},\ \bar{r}_2 = \frac{r_2 + \bar{r}_3 \mathrm{e}^{\mathrm{i}x_2}}{1 + r_2 \bar{r}_3 \mathrm{e}^{\mathrm{i}x_2}},\ \dots \\ &\dots,\ \bar{r}_N = \frac{r_N + r_{N+1} \mathrm{e}^{\mathrm{i}x_N}}{1 + r_N r_{N+1} \mathrm{e}^{\mathrm{i}x_N}}, \end{aligned} \tag{2}$$

where

$$r_1 = \frac{Y_0 - Y_1}{Y_0 + Y_1},\ r_2 = \frac{Y_1 - Y_2}{Y_1 + Y_2},$$

$$\vdots$$

$$r_N = \frac{Y_{N-1} - Y_N}{Y_{N-1} + Y_N},\ r_{N+1} = \frac{Y_N - Y_\mathrm{S}}{Y_N + Y_\mathrm{S}},$$

$$x_w = \frac{4\pi}{\lambda} d_w \sqrt{n_w^2 - n_0^2 \sin^2 \varphi_0}\ ,$$

where symbols d_w, n_w, and Y_w are thickness, refractive index and admitance of the individual sub-layer, respectively ($w = 1, 2, \dots, N$). Note that the over-bar is used to distinguish symbols, it should not be confused with the complex conjugation, which is denoted by symbol *.

Within the matrix formalism reflection coefficients of the multilayer system r are expressed in this way

$$r = \frac{M_{11} - Y_0^{-1} Y_\mathrm{S} M_{22} + Y_\mathrm{S} M_{12} - Y_0^{-1} M_{21}}{M_{11} + Y_0^{-1} Y_\mathrm{S} M_{22} + Y_\mathrm{S} M_{12} + Y_0^{-1} M_{21}}, \tag{3}$$

where M_{mn} $(m, n = 1, 2)$ are elements of the interference matrix of the system replacing the inhomogeneous thin film. The foregoing formula (3) is derived, for example, in monographs [2, 30].

In paper [24] an improvement of the method presented in this section is performed. This improvement consists in improving the convergence of the method using the Richardson extrapolation. If the Richardson extrapolation is used then the error of calculation falls approximately as $N^{-\log_2 N}$, where N is the number of the approximating sub-layers. However, it should be noted that the faster convergence can be achieved using the Richardson extrapolation only if the number of the sub-layers is considerably large (for details see [24]).

It is also possible to use a procedure which automatically determines the number of dividing sub-layers. This automatic procedure contains several steps. In each following step the number of the sub-layers is doubled owing to the preceding step. These steps are repeating until the required precision is reached.

The complete optical characterization of the inhomogeneous thin films with larger gradients is performed if the functions describing the profiles of the complex refractive index are determined together with the spectral dependencies of the boundary refractive indices and thickness. Thus, the thickness values of the individual sub-layers must be sufficiently small in order to obtain a continuous refractive index profile of such the films from the practical point of view.

3.3 Approximate method based on modifying recursive formulae of multilayer systems

Kildemo *et al* [20] published the sophisticated approximate method for deriving the reflection coefficients of the inhomogeneous thin films based on a modification of the recursive formalism for the multilayer systems. They used the recursive formula for the four-layer system for explaining the principle of their derivation. In this derivation systematic trends of reflections were revealed if this four-layer system were subdivided into more and more sub-layers. Using the limit for infinite number of the sub-layers the formula for the reflection coefficients of the inhomogeneous thin films with arbitrary refractive index profiles including those exhibiting large gradients was obtained. In the formulae derived the multiple integrals containing derivatives of the profiles of the refractive index occur. The multiplicity of the integrals is equal to numbers of internal reflections considered inside the film. If two internal reflections are taken into account in maximum then the formula (for details see [20]) for the reflection coefficients is $r =$

$$= \frac{r_\mathrm{U} + B_1 + r_\mathrm{U} r_\mathrm{L} \overline{B}_1 \mathrm{e}^{\mathrm{i}x_d} + r_\mathrm{U} B_2 + r_\mathrm{L} \overline{B}_2 \mathrm{e}^{\mathrm{i}x_d} + r_\mathrm{L} \mathrm{e}^{\mathrm{i}x_d}}{1 + r_\mathrm{U} B_1 + r_\mathrm{L} \overline{B}_1 \mathrm{e}^{\mathrm{i}x_d} + B_2 + r_\mathrm{U} r_\mathrm{L} \overline{B}_2 \mathrm{e}^{\mathrm{i}x_d} + r_\mathrm{U} r_\mathrm{L} \mathrm{e}^{\mathrm{i}x_d}}, \tag{4}$$

where

$$B_1=\int_0^d f(z_1)\mathrm{e}^{\mathrm{i}x(z_1)}\mathrm{d}z_1,\ \ \overline{B}_1=\int_0^d f(z_1)\mathrm{e}^{-\mathrm{i}x(z_1)}\mathrm{d}z_1,$$

$$B_2=\int_0^d\int_0^{z_1} f(z_1)f(z_2)\mathrm{e}^{\mathrm{i}[x(z_1)-x(z_2)]}\mathrm{d}z_1\mathrm{d}z_2,$$

$$\overline{B}_2=\int_0^d\int_0^{z_1} f(z_1)f(z_2)\mathrm{e}^{-\mathrm{i}[x(z_1)-x(z_2)]}\mathrm{d}z_1\mathrm{d}z_2,$$

where

$$f(z)=\frac{1}{2Y(z)}\frac{\mathrm{d}Y(z)}{\mathrm{d}z},$$

$$x(z)=\frac{4\pi}{\lambda}\int_0^z\sqrt{n^2(\nu)-n_0^2\sin^2\varphi_0}\mathrm{d}\nu.$$

In paper [20] the formula corresponding to three internal reflections containing the triple integrals is also introduced explicitly. The triple integrals can be expressed by means of the double integrals (for details see [20]). In their paper Kildemo *et al* [20] presented also the modification of their approximate method in matrix formalism, in which the integrals occur in the elements of the interference matrix of the inhomogeneous film. Note that the theoretical results presented in [20] were applied to processing experimental data measured for inhomogeneous semiconductor films in papers [31, 32].

This method enables us to determine the same quantities of the inhomogeneous films exhibiting the arbitrary refractive index profiles as in the case of the foregoing multilayer system method.

3.4 Approximate method based on multiple beam interference model

This approximate method can also be utilized for the optical characterization of the inhomogeneous thin films exhibiting arbitrary profiles. Thus, it can be employed for characterizing the films with large gradients of the refractive index profiles and complicated profiles. The method is based on calculating corrections to the WKBJ formula corresponding to single, double, triple and multiple internal reflections inside the structure of the inhomogeneous film connected with the profile of the refractive index (see [33]). These corrections contain multiple integrals similar to those obtained in paper [20]. A multiplicity of the integrals is again given by the numbers of the considered internal reflections inside the film. After using the mathematical procedure presented in [33] one then derives the following formula for r

$$r=r_0+\sum_{n=1}^{M}\Delta r_n, \tag{5}$$

where Δr_n denotes the individual correction of the order n and M is the maximum order needed for obtaining a required precision of calculations. The correction of the n-th order is expressed as follows [33]

$$\Delta r_n=\sum_{l=1}^{3^{n-1}}(I_{n,\mathrm{R}}^{(l)}+\overline{I}_{n,\mathrm{R}}^{(l)}). \tag{6}$$

Symbols $I_{n,\mathrm{R}}^{(l)}$ and $\overline{I}_{n,\mathrm{R}}^{(l)}$ represent the partial corrections of the n-th order (see [33]). The partial corrections of the first order are expressed as follows [33]

$$I_{1,\mathrm{R}}^{(1)}=C_{1,\mathrm{R}}^{(1)}j_1^{(1)},\ \ \overline{I}_{1,\mathrm{R}}^{(1)}=\overline{C}_{1,\mathrm{R}}^{(1)}\overline{j}_1^{(1)}, \tag{7}$$

where

$$C_{1,\mathrm{R}}^{(1)}=-\frac{\tau_\mathrm{R}}{(1-\rho)^2},\ \ j_1^{(1)}=\int_0^d f(z_1)\mathrm{e}^{\mathrm{i}x(z_1)}\mathrm{d}z_1,$$

$$\overline{C}_{1,\mathrm{R}}^{(1)}=\frac{\tau_\mathrm{R}Z^2}{(1-\rho)^2},\ \ \overline{j}_1^{(1)}=\int_0^d f(z_1)\mathrm{e}^{-\mathrm{i}x(z_1)}\mathrm{d}z_1,$$

$$Z=r_\mathrm{L}\mathrm{e}^{\mathrm{i}x_d},\ \ \rho=-r_\mathrm{U}r_\mathrm{L}\mathrm{e}^{\mathrm{i}x_d},\ \ \tau_\mathrm{R}=t_\mathrm{U}t'_\mathrm{U}.$$

The symbols t_U and t'_U are calculated as

$$t_\mathrm{U}=\frac{2Y_0}{Y_0+Y_\mathrm{U}},\ \ t'_\mathrm{U}=\frac{2Y_\mathrm{U}}{Y_0+Y_\mathrm{U}}$$

for s-polarization,

$$t_\mathrm{U}=\frac{C_0}{C_\mathrm{U}}\frac{2Y_0}{Y_0+Y_\mathrm{U}},\ \ t'_\mathrm{U}=\frac{C_\mathrm{U}}{C_0}\frac{2Y_\mathrm{U}}{Y_0+Y_\mathrm{U}}$$

for p-polarization with $C_0=\cos\varphi_0$ and $C_\mathrm{U}=\cos\varphi_\mathrm{U}$. The partial corrections of the second order are given in this way [33]

$$I_{2,\mathrm{R}}^{(l)}=C_{2,\mathrm{R}}^{(l)}\,j_2^{(l)},\ \ \overline{I}_{2,\mathrm{R}}^{(l)}=\overline{C}_{2,\mathrm{R}}^{(l)}\,\overline{j}_2^{(l)},\ \ l=1,2,3,$$

$$C_{2,\mathrm{R}}^{(1)}=\frac{r_\mathrm{U}C_{1,\mathrm{R}}^{(1)}}{1-\rho},$$

$$j_2^{(1)}=\int_0^d\int_0^d f(z_1)f(z_2)\mathrm{e}^{\mathrm{i}[x(z_1)+x(z_2)]}dz_2dz_1,$$

$$C_{2,\mathrm{R}}^{(2)}=ZC_{1,\mathrm{R}},$$

$$j_2^{(2)}=\int_0^d\int_0^{z_1} f(z_1)f(z_2)\mathrm{e}^{\mathrm{i}[x(z_1)-x(z_2)]}dz_2dz_1,$$

$$C_{2,\mathrm{R}}^{(3)}=\frac{\rho ZC_{1,\mathrm{R}}^{(1)}}{1-\rho},$$

$$
\begin{gathered}
j_2^{(3)} = \int_0^d \int_0^d f(z_1) f(z_2) \mathrm{e}^{\mathrm{i}[x(z_1)-x(z_2)]} dz_2 dz_1, \\
\overline{C}^{(1)}_{2,\mathrm{R}} = -\frac{1}{Z}\overline{C}^{(1)}_{1,\mathrm{R}}, \\
\overline{j}_2^{(1)} = \int_0^d \int_{z_1}^d f(z_1) f(z_2) \mathrm{e}^{-\mathrm{i}[x(z_1)-x(z_2)]} dz_2 dz_1, \\
\overline{C}^{(2)}_{2,\mathrm{R}} = \frac{r_{\mathrm{U}}\overline{C}^{(1)}_{1,\mathrm{R}}}{1-\rho}, \\
\overline{j}_2^{(2)} = \int_0^d \int_0^d f(z_1) f(z_2) \mathrm{e}^{-\mathrm{i}[x(z_1)-x(z_2)]} dz_2 dz_1, \\
\overline{C}^{(3)}_{2,\mathrm{R}} = \frac{Z\overline{C}^{(1)}_{1,\mathrm{R}}}{1-\rho}, \\
\overline{j}_2^{(3)} = \int_0^d \int_0^d f(z_1) f(z_2) \mathrm{e}^{-\mathrm{i}[x(z_1)+x(z_2)]} dz_2 dz_1.
\end{gathered}
\tag{8}
$$

Note that the following equalities are valid $I_2^{(2)} = \overline{I}_2^{(1)}$, $I_2^{(3)} = \overline{I}_2^{(2)}$, $B_1 = j_1^{(1)}$, $\overline{B}_1 = \overline{j}_1^{(1)}$, $B_2 = j_2^{(2)}$ and $\overline{B}_2 = \overline{j}_2^{(2)}$. By means of the equations for the partial corrections of the second order (see (8)) it is possible to formulate equations enabling us to calculate the partial corrections of the $(n+1)$-th order using those corresponding to the n-th order $(n \geq 1)$. The derivation of these equations is carried out in detail in [33]. This means that one can calculate the corrections Δr_n of arbitrary order which is important for the optical characterization of inhomogeneous thin films exhibiting complex refractive index profiles and profiles with the large gradients.

4 Measurable optical quantities in reflected light

Measurable optical quantities of the inhomogeneous thin films corresponding to reflected light can easily be calculated with using the reflection coefficients r. In practice spectral dependencies of reflectance and ellipsometric quantities measured in specular direction are mostly utilized for characterizing thin films and their systems. At oblique incidence of light reflectances for both the polarizations are calculated as $R_{\mathrm{p}} = r_{\mathrm{p}} r_{\mathrm{p}}^*$ and $R_{\mathrm{s}} = r_{\mathrm{s}} r_{\mathrm{s}}^*$. At near-normal incidence it holds that $\overline{R} = \overline{r}\overline{r}^*$, where $\overline{r}$ denotes the reflection coefficient for $\varphi_0 = 0$ and $\overline{R}$ represents corresponding reflectance. For specular reflection ellipsometric parameters Ψ and Δ are calculated using ellipsometric ratio ρ_{E} by means of equation

$$\rho_{\mathrm{E}} = r_{\mathrm{p}}/r_{\mathrm{s}} = \tan\Psi \exp(\mathrm{i}\Delta).$$

Ellipsometric parameters Ψ and Δ are measured using null ellipsometry. Ellipsometric techniques based on rotating polarizer or analyzer provide measured ellipsometric quantities cos Δ and tan Ψ. Within phase-modulated ellipsometry the associated ellipsometric parameters I_{s}, I_{c}, and I_{n}, are measured. These parameters are given as follows

$$
\begin{gathered}
I_{\mathrm{s}} = -\mathrm{i}\frac{r_{\mathrm{p}} r_{\mathrm{s}}^* - r_{\mathrm{p}}^* r_{\mathrm{s}}}{|r_{\mathrm{s}}|^2 + |r_{\mathrm{p}}|^2}, \quad I_{\mathrm{c}} = \frac{r_{\mathrm{p}} r_{\mathrm{s}}^* + r_{\mathrm{p}}^* r_{\mathrm{s}}}{|r_{\mathrm{s}}|^2 + |r_{\mathrm{p}}|^2}, \\
I_{\mathrm{n}} = \frac{|r_{\mathrm{s}}|^2 - |r_{\mathrm{p}}|^2}{|r_{\mathrm{s}}|^2 + |r_{\mathrm{p}}|^2}.
\end{gathered}
\tag{9}
$$

Note that the associated ellipsometric parameters are the elements of the Mueller matrix for isotropic systems (see *eg* [34]). Of course, other ellipsometric quantities can be used to characterize thin films. Overviews concerning ellipsometry are presented, for example, in [35-37].

5 Comparison of approximate methods

The WKBJ approximation is the simplest approximate method enabling us to perform the optical characterization of the special inhomogeneous thin films, *ie* the films with small gradients of the refractive index profiles. This method always fails at the optical characterization of the inhomogeneous thin films with lager gradients of profiles. Thus, the WKBJ approximation has limited applications in practice. The remaining three approximate methods are evidently more general because they can be usable for the films with arbitrary refractive index profiles. Of course, their comparison is especially interesting if the films with complex or strong gradient profiles are characterized. First, the comparison of the method mostly utilized in practice, *ie* the method based on substituting the inhomogeneous films by the multilayer systems, with the approximate methods based on the modified recursive approach and multiple-beam interference model will be performed. After our numerical analysis concerning the inhomogeneous thin films with complicated refractive index profiles or profiles exhibiting large gradients it was found that the method based on the multilayer system approximation was faster than the remaining two approximate methods utilizing the integrals. This was true even when sophisticated calculations of the integrals are utilized (see paper [33]). This means that the method based on the multilayer system approximation is more advantageous for in situ applications than the methods containing the integrals. However, some disadvantage is connected with this method. It requires very large number of subdividing homogeneous sub-layers of the multilayer system to achieve the desired precision. Thus, the number of points at which the profile of the refractive index must be calculated is much larger than in the methods containing the integrals presented above (for details see [33]). In other words, the speed of the method employing the multilayer approximation can be negatively affected if evaluation of the refractive index profile takes a long time while the impact on the remaining two approximate methods is much lower. This means that the larger speed of the method using the multilayer system approximation need not be unambiguous advantage at the optical characterization of the inhomogeneous films with large gradients of profiles or complex profiles. This statement is true for ex-situ applications in particular.

The speeds of the methods containing the integrals are mutually comparable at calculating the optical quantities of a majority of inhomogeneous films. This is given by the fact that the speed of these calculations is determined by the speed of calculating the mentioned integrals which are mutually similar in both the methods. In detailed numerical analysis it was found that the method of Kildemo *et al* [20] is somewhat faster than the method based on the multi-beam interference model if the films with complex refractive index profiles are characterized. The complex refractive index profiles are exhibited, for example, in the rugate filters. It should be noted that the method utilizing multi-beam interference is more advantageous for including defects of the inhomogeneous films such as roughness of oboundaries or thickness nonuniformity than the method of Kildemo *et al* [20]. For example, at including random boundary roughness into the formulae for the reflection coefficients of the inhomogeneous films it is necessary to calculate statistical mean values of these coefficients and optical quantities calculated using these coefficients (see *eg* [37-45]). These mean values are more easily calculated if the reflection coefficients are expressed by series than by rational functions (it will be discussed elsewhere in details). It is suitable to mention the further disadvantage of the method Kildemo *et al* [20] which consists in difficulties to express corrections of higher orders in the formulae for the reflections coefficients of the inhomogeneous thin films (for details see [20, 33]).

6 Experiment

The illustration of the applications of the methods presented here is performed through the optical characterization of three inhomogeneous non-stoichiometric silicon nitride films deposited by reactive magnetron sputtering of silicon target in argon-nitrogen atmosphere onto silicon single crystal substrates. The profiles of the refractive index of these films were created by changing flow rate of nitrogen during deposition of the films while the other deposition conditions were kept constant. The spectral dependencies of reflectance were measured by Perkin Elmer Lambda 1050 spectrophotometer for the incidence angle of 6° within the spectral range 1.4 – 6.5 eV (190 – 860 nm) while spectral dependencies of the associated ellipsometric parameters were measured using Horiba Jobin Yvon UVISEL phase modulated ellipsometer for several incidence angles from the interval 55° – 75° within the spectral range 0.6 – 6.5 eV (190 – 2066 nm). The optical data were processed by the least-squares method (LSM) taking into account the weights of individual experimental values of measured reflectance and associated ellipsometric parameters.

6.1 Application of the WKBJ method

This non-stoichiometric silicon nitride film exhibited the refractive index profile with very small gradient and therefore the values of the associated ellipsometric parameters used for characterizing this film were calculated using the reflection coefficients expressed by (1). The profile of the complex refractive index of the film was modeled by the linear dependence of the complex dielectric function in this way

$$n^2(z) = n_{\mathrm{U}}^2 - \frac{z}{d}(n_{\mathrm{U}}^2 - n_{\mathrm{L}}^2). \tag{10}$$

Each of the dielectric functions $\varepsilon_{\mathrm{U}}(E) = n_{\mathrm{U}}^2(E)$ and $\varepsilon_{\mathrm{L}}(E) = n_{\mathrm{L}}^2(E)$ was calculated using the dispersion model with the imaginary part of the dielectric function given as

$$\varepsilon_{\mathrm{i},t}(E) = \frac{N_t(E - E_{\mathrm{g},t})^2(E_{\mathrm{h},t} - E)^2}{C_t E^2}\Pi(E_{\mathrm{g},t}, E_{\mathrm{h},t}; E), \tag{11}$$

where the index $t = \mathrm{U}, \mathrm{L}$ is used to distinguish between the dielectric functions at upper and lower boundaries. The parameter N_t determines the strength of the transitions and the parameters $E_{\mathrm{g},t}$ and $E_{\mathrm{h},t}$ are the band gap energy and maximum energy of interband transitions, respectively.

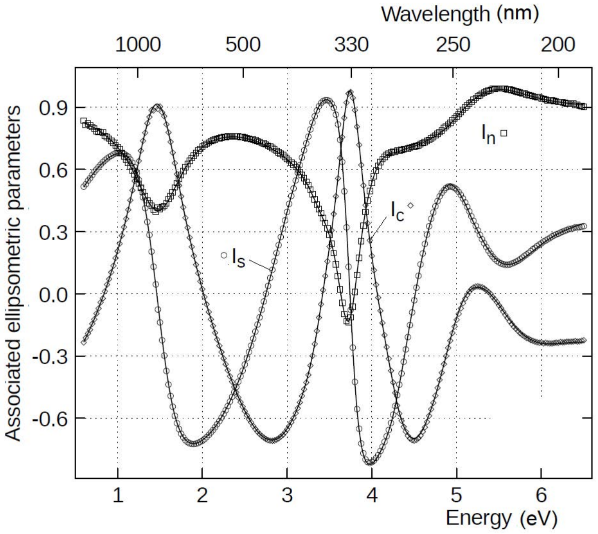

Fig. 1. Spectral dependencies of the measured associated ellipsometric parameters at angle of incidence of 65° and their fits: points denote the experimental values and curves represent the theoretical data

The function $\Pi(a, b; x)$ is defined as

$$\Pi(a, b; x) = \begin{cases} 1, & a \le x \le b \\ 0, & \text{otherwise.} \end{cases}$$

The constant C_t is determined by means of the following equation

$$\int_0^\infty E\varepsilon_{\mathrm{i},t}(E)\mathrm{d}E = N_t.$$

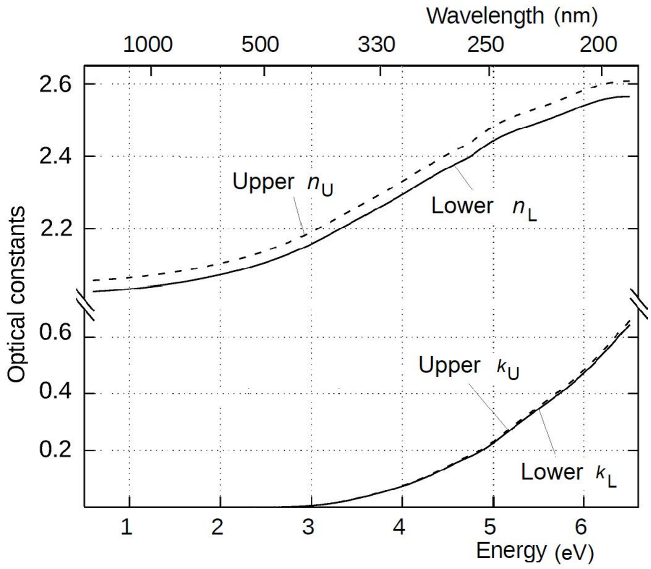

Fig. 2. Spectral dependencies of the optical constants at the upper boundary n_U and k_U and lower boundary n_L and k_L

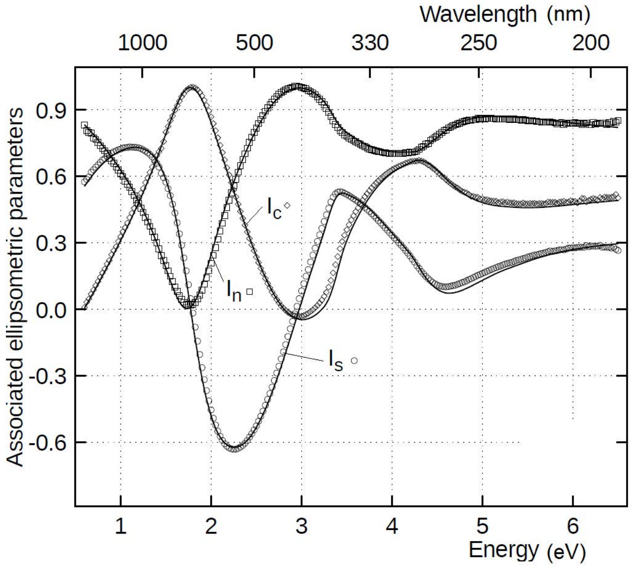

Fig. 3. Spectral dependencies of the measured associated ellipsometric parameters at angle of incidence of 70 ° and their fits: points denote the experimental values and curves represent the theoretical data

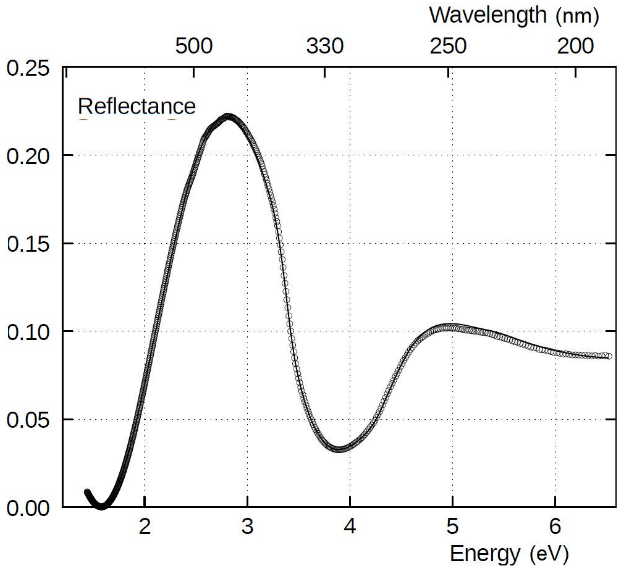

Fig. 4. Spectral dependency of the measured reflectance and its fit: points denote the experimental values and curve represents the theoretical data

The real parts of the dielectric functions $\varepsilon_{r,t}(E)$ were calculated using the Kramers–Kroning relation (see *eg* [46]).

It was found that using the refractive index profile and dispersion model introduced above the very good fits of the experimental data were obtained (see Fig. 1).

In Fig. 2 the spectral dependencies of the boundary refractive indices n_U and n_L together with the spectral dependencies of the corresponding extinction coefficients k_U and k_L are plotted. The thickness of the film was determined in value of 113 nm. This thickness value and the spectral dependencies of n_U and n_L confirm the assumption concerning the very small gradient of the refractive index profile of this film. It should be noted that by means of the three remaining approximate methods the same results and fits of experimental data of this sample were achieved within the experimental accuracy which was expected.

6.2 *Application of the multilayer approximation*

The non-stoichiometric silicon nitride film, whose results of the optical characterization are introduced in this sub-section, exhibited a relatively large gradient of the refractive index profile and therefore the approximation by the multilayer system was utilized (WKBJ approximation completely failed). Within its optical characterization the simultaneous processing of reflectance and ellipsometric data was utilized [24]. The formulae corresponding to the matrix formalism were used to calculate the values of reflectance and associated ellipsometric parameters. This approximate method was employed in the modification including its improvement consisting in using the Richardson extrapolation presented in paper [24]. For this film the relatively complicated function describing the refractive index profile had to be used

$$n^2(z) = n_U^2\left[1 - p\left(\frac{z}{d}\right)\right] + n_L^2 p\left(\frac{z}{d}\right),$$

where $p(x) = 1 - (1 - x)^s$ and s is the parameter of the model. The dielectric functions at the upper and lower boundaries were calculated using the same dispersion model as in the previous example (see (11)).

Using the chosen dispersion model and refractive index profile the excellent fits of the experimental data were obtained (see Fig. 3 and Fig. 4).

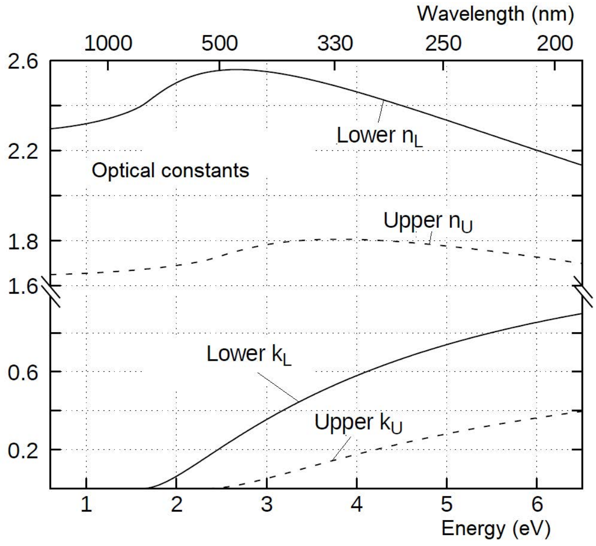

Fig. 5. Spectral dependencies of the boundary refractive indices n_U and n_L and the boundary extinction coefficients k_U and k_L

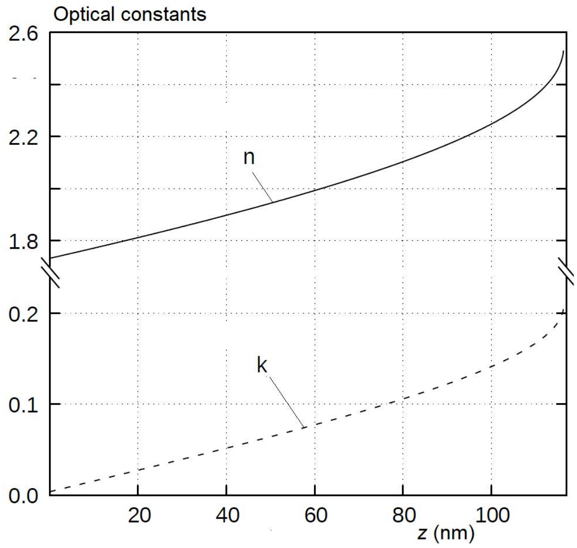

Fig. 6. Profile of refractive index n and extinction coefficient k for $E = 2.5$ eV

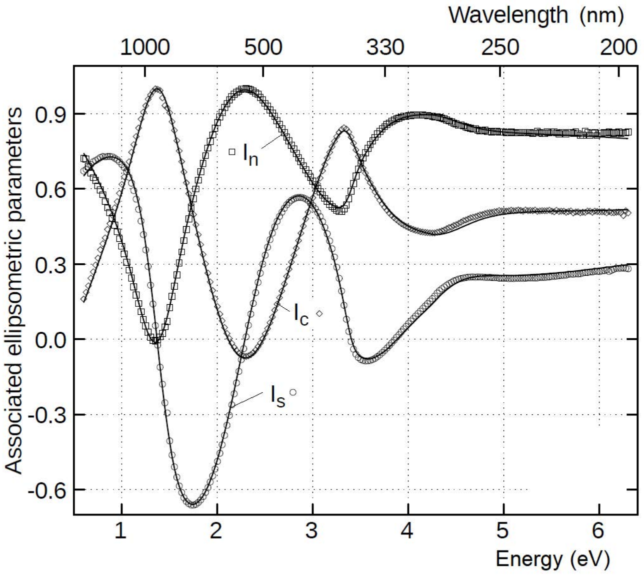

Fig. 7. Spectral dependencies of the measured associated ellipsometric parameters at angle of incidence of 70° and their fits: points denote the experimental values and curves represent the theoretical data

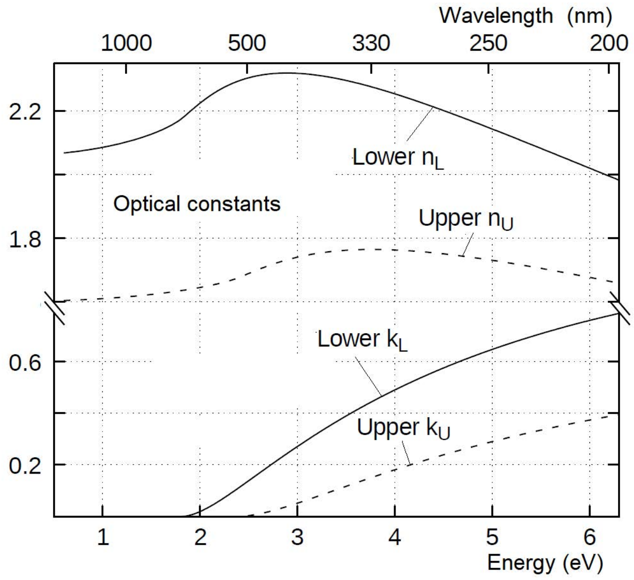

Fig. 8. Spectral dependencies of the optical constants at the upper boundary n_U and k_U and lower boundary n_L and k_L

The spectral dependencies of the refractive indices n_U and n_L and extinction coefficients k_U and k_L calculated by means of the found dispersion parameters are depicted in Fig. 5.

In Fig. 6 the determined profiles of refractive index and extinction coefficient are introduced for the selected wavelength. From Figs. 5 and 6 it is clear that the refractive index and extinction coefficient profiles exhibit considerably large gradients. Note that the thickness of this film was determined in value of 115 nm.

6.3 Application of the method based on multi-beam interference model

In this sub-section the results of the optical characterization of another inhomogeneous non-stoichiometric silicon nitride film with relatively large gradient of the refractive index profile achieved by the multi-beam approximation are presented (third sample). This means that the formula (5) for the reflection coefficients had to be used to calculate the values of the associated ellipsometric parameters (reflectance data again were not utilized for the optical characterization). The existence of a rela-

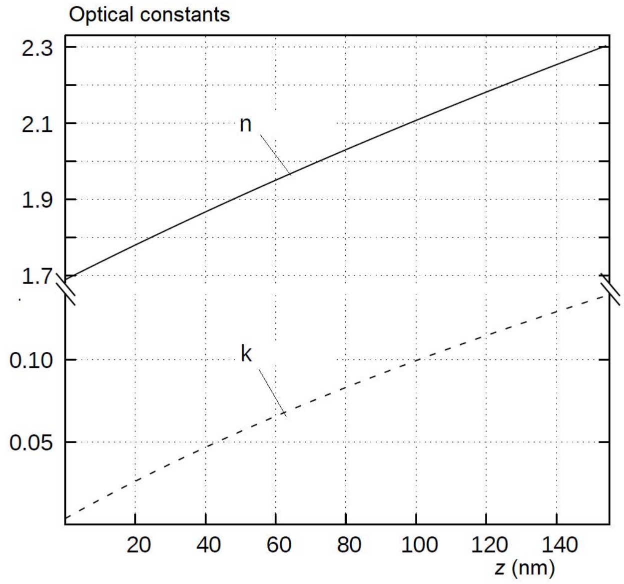

Fig. 9. Profiles of the refractive index n and extinction coefficient k for $E = 2.5$ eV

tively large gradient of the profile in this film was again indicated by an impossibility to fit the experimental data by means of the WKBJ approximation. The model of the refractive index profile as well as the dispersion model are identical with those used in the optical characterization performed by the WKBJ method (see sub-section 6.1, (10) and (11)). The perfect fits of the experimental data was achieved which is seen in Fig. 7.

In Fig. 8 the spectral dependencies of the boundary optical constants are introduced (the Kramers–Kroning relation was again utilized).

From Fig. 8 it can be observed that the spectral dependencies of n_{L} and k_{L} exhibit higher values than the spectral dependencies of n_{U} and k_{U}. Note that the same conclusion is also valid for the other film with the large gradient of the profile discussed in the foregoing sub-section (see sub-section 6.2) in contrast to that whose data are compatible with the WKBJ approximation (see sub-section 6.1). This implies that for the films with the large gradients the concentration of silicon is higher at the lower boundaries than silicon concentration at upper boundaries. The opposite is true for the film with small gradient obeying the WKBJ approximation. The profiles of the refractive index and extinction coefficient are plotted in Fig. 9 for the selected wavelength.

This figure and the spectral dependencies of the boundary refractive indices n_{U} and n_{L} prove the assumption concerning the large gradients of the refractive index. The same statement is fulfilled for the extinction coefficient of this film. The thickness value of this film was found in value of 154 nm. Note that the presented results were achieved with the single, double and triple integrals included into the formula for the reflection coefficients (see [33]).

7 Conclusion

In this overview paper four approximate methods for the optical characterization of the inhomogeneous thin films are described. These methods are useful from the point of view of their practical use and, therefore, they can find a use in the modern branches of research such as microelectronics, nanotechnology, solar energetics *etc.* The principles and mathematical formulations of these methods are presented. It is shown that the inhomogeneous thin films exhibiting very small gradients of their refractive index profiles can be characterized using the simple formulae occurring in the WKBJ approximation. In principle the procedure corresponding to this approximation enables us to determine the spectral dependencies of the boundary optical constants of these films together with the profiles of the optical constants and thicknesses. The WKBJ approximation fails in the optical characterization of the inhomogeneous thin films exhibiting larger gradients of the refractive index profiles. In this case it is possible to use the method based on the approximation of such the inhomogeneous thin films by multilayers systems divided into a large number of very thin sub-layers. The adjacent sub-layers must have very small differences in refractive indices. The efficiency of this approximate method can be improved by using the Richardson extrapolation. Using this multilayer approximation one can determine the same characteristics as at applyying the WKBJ approximation. The same possibilities concerning the characterization of the films with complicated profiles and large gradients of profiles are exhibited by two further methods. One of them is based on the modification of the recursive formalism for multilayer system and the latter of them is based on the mathematical formulation of multi-beam interference of light inside the inhomogeneous films. These two methods also enable us to determine spectral dependencies of the boundary optical constants, profiles of these constants and thicknesses corresponding to the inhomogeneous thin films with arbitrary refractive index profiles including complex profiles exhibiting simultaneously large gradients.

Of course, the good results of the optical characterization of the inhomogeneous thin films mentioned above can be achieved only when the correct dispersion models and correct functions describing the profiles of these films are used. Therefore, one can recommend to use the universal dispersion model together with their special modifications (see *eg* [47]) and multi-sample modification of the LSM in which experimental data of several samples of the same layered system are simultaneously processed (for details see *eg* [48, 49]). The use of the universal dispersion model or its modifications ensures that dispersion models employed are physically correct. The use of the multi-sample method at processing the experimental data enables us to reduce a correlation of the parameters sought by the LSM, which causes an improvement of the results achieved in the optical characterization of the inhomogeneous thin films exhibiting complex refractive index profiles in particular.

In this paper the results of the optical characterization of the three inhomogeneous non-stoichiometric silicon nitride films are presented. These examples show the efficiency of the approximate methods presented here together with a demonstration of their practical meaning. In this conclusion it is, moreover, necessary to point out that by means of the approximate methods based on the multilayer approximation, modification of the recursive formulae and multiple-beam interference model one obtains the same results of characterizing arbitrary inhomogeneous films from the practical point of view. The differences among these methods only consist in the different speeds of achieving the results. The approximation by multilayer systems is the most rapid of them (this is especially true if this approximate method is improved by the Richardson extrapolation). The method based on the multiple-beam interference model is potentially usable in including some defects such as boundary roughness into the formulae for the optical quantities of inhomogeneous thin films (it will be presented elsewhere). The method based on modifying the recursive formulae exhibits a higher speed in the optical characterization of inhomogeneous films than the method based on multibeam interference model if these films have very complicated refractive index profiles (*eg* rugate filters).

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