

Phase thickness approach for determination of thin film refractive index dispersion from transmittance spectra

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Abstract. A novel approach for determination of refractive index dispersion $n(\lambda)$ and thickness d of thin films of negligible absorption and weak dispersion is proposed. The calculation procedure is based on determination of the phase thickness of the film in the spectral region of measured transmittance data. All points of measured spectra are included in the calculations. Barium titanate and titanium oxide thin films are investigated and their $n(\lambda)$ and d are calculated. The approach is validated using Swanepoel's method and it is found to be applicable for relatively thinner films when measured transmittance spectra have one minimum and one maximum only.

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1 Introduction

For many optical applications the physical thickness d of a thin film, its refractive index n and optical dispersion $n = n(\lambda)$ are very important. In some thin films technologies it isn't possible to determine film optical parameters in situ, during the deposition process. It is not so easy to determine these film characteristics and after film deposition too. The reason of this situation is in the fact that even for bulk materials, direct measurements of the refractive index n are quite complex. Most precise information for a thin film may be obtained from transmission spectra measurements. However they are strongly dependent on the optical parameters of the film ($n(\lambda)$, d) and the substrate, absorption and smoothness of the substrate front and back side.

Due to interference of light, a ripple (an oscillating) structure with maxima and minima is typical for thin films optical transmittance/reflectance spectra. Optical parameters of thin films can be extracted from optical spectra using several conventional methods [1–15].

In envelope methods [1–9] spectral extrema and their positions are applied for determination of $n(\lambda)$ and d . These methods are applicable for films of thickness in a limited range, for example $d > 0.2 \mu\text{m}$ for most oxide films [2]. For thinner films, the number of extrema becomes insufficient and it is very difficult to locate them [2]. Application of relatively simple equations is an advantage of the envelope method, this method do not require specific software for determination of thin film refractive index dispersion. However, the envelope methods use only

extrema points and their positions, i.e. only a small number of all measurements are taken into account. Thin films of thickness higher than $0.5 \mu\text{m}$ and negligible absorption in the visible spectral region usually have transmittance spectra with number of extrema high enough for accurate determination of the refractive index dispersion. Thin films of thickness in the range $0.2\text{--}0.4 \mu\text{m}$ may have spectra with low number of extrema points (3–5) which may increase the degree of uncertainty of the final results. Another disadvantage of the envelope methods is the usage of interpolated points (not really measured) between two successive measured maxima or minima [1,2].

Curve fitting methods apply numerical optimisation to fit a theoretical model to measured data in order to find dispersion $n(\lambda)$ and thickness d [9–15]. These methods need very specialised and flexible software environment for the calculations because of the variety of dispersion equations in literature [9]. Special care should be taken for the theoretical model of dispersion and for determination of the initial values of the optimised parameters. Curve fitting methods include all measured points of transmittance spectra, but these methods require very specific software and very careful selection of the initial data [9–15].

The main purpose of this paper is to propose a new approach for determination of refractive index dispersion $n(\lambda)$ and thickness d of thin films of negligible absorption and weak dispersion. The main idea is to combine the advantages of both upper mentioned groups of methods: application of relatively simple mathematical support (envelope methods) and processing of all points of measured transmittance spectra (curve fitting methods). The presented procedure is important for experimental

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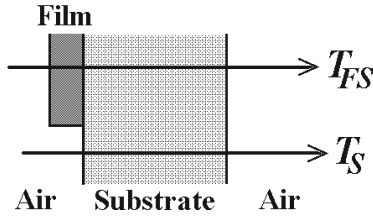


Fig. 1. Thin film – substrate system surrounded by air; T_{FS} – transmittance of coated substrate; T_S – transmittance of uncoated substrate.

investigations of thin films with negligible absorption in the spectral region of interest.

2 Analysis of thin film transmittance spectra

Let us describe the optical system which transmittance will be analysed. It is nonabsorbing glass substrate of refractive index n_S (with parallel boundaries), which is covered with thin film of refractive index n , see Figure 1. The substrate is considered to be optically thick and no interference effects are assumed. The film is also considered to be nonabsorbing but optically thin and all interference patterns in transmittance spectra are supposed to be caused by interference of light in the film. The system is surrounded by air of refractive index $n_{AIR} = 1$. The case of normal incidence of light is investigated.

Transmittance of the optical system, described above, for the case of weakly absorbing thin film of complex refractive index n^* ($n^* = n - ik$, where n is the real refractive index, $i = \sqrt{-1}$, k is the extinction coefficient, $k \ll n$) is given by [2]:

$$T_{FS} = \frac{Ax}{B - Cx \cos \varphi + Dx^2},$$

where

$$\begin{aligned} A &= 16n^2n_S; \quad B = (n+1)^3(n+n_S^2); \\ C &= 2(n^2-1)(n^2-n_S^2); \quad D = (n-1)^3(n-n_S^2); \\ \varphi &= 2\delta = 4\pi nd/\lambda; \quad \alpha = 4\pi k/\lambda; \quad x = \exp(-\alpha d). \end{aligned}$$

In the last equations $\delta = \frac{\varphi}{2} = \frac{2\pi nd}{\lambda}$ is the film phase thickness and α is the film absorption coefficient.

For many optical materials there are spectral regions where optical absorption is negligible and for these materials and regions it is reasonable to suppose $k = 0$, $\alpha = 0$ and $x = 1$. It leads to the following simplified expression for transmittance of the investigated system:

$$T_{FS} = \frac{A}{B - C \cos \varphi + D}. \quad (1)$$

After some transformations, the following expression can be derived:

$$\frac{1}{T_{FS}} - \frac{V}{2} = \frac{1}{8}(U - V)(1 - \cos \varphi) = \frac{1}{4}(U - V)\sin^2 \delta, \quad (2)$$

where

$$U = \frac{n^4 + n_S^2}{n^2 n_S} = \frac{n^2}{n_S} + \frac{n_S}{n^2}, \quad V = \frac{n^2(n_S^2 + 1)}{n^2 n_S} = n_S + \frac{1}{n_S}. \quad (3)$$

Transmittance T_S of uncoated substrate is given by the equation [2]:

$$T_S = \frac{2n_S}{n_S^2 + 1},$$

and therefore the reciprocal value of T_S will be:

$$\frac{1}{T_S} = \frac{n_S^2 + 1}{2n_S} = \frac{1}{2} \left(n_S + \frac{1}{n_S} \right) = \frac{V}{2}. \quad (4)$$

The substitution of $\frac{V}{2}$ from (4) into (2) leads to:

$$\frac{1}{T_{FS}} - \frac{1}{T_S} = \frac{1}{4}[U - V]\sin^2 \delta. \quad (5)$$

It can be seen that in a case of a film-substrate system with no refractive index dispersion ($n(\lambda) = \text{const.}$, $n_S(\lambda) = \text{const.}$) $\frac{1}{T_{FS}} - \frac{1}{T_S}$ is a periodic function of δ (or $1/\lambda$) with constant amplitude and extrema points defined from the following conditions:

1. If $\delta = 0, \pi, 2\pi, \dots, m\pi$, then $\sin^2 \delta = 0$ and $\frac{1}{T_{FS}} - \frac{1}{T_S} = 0$, see (5). In this case the phase thickness δ is equal to π multiplied by an integer number. These minimal values of $\frac{1}{T_{FS}} - \frac{1}{T_S}$ correspond to transmittance maxima T_{MAX} which are equal to substrate transmittance T_S . For these values of the phase thickness the optical thickness nd is an integer number of $\lambda/2$: $nd = m\frac{\lambda}{2}$.
2. If $\delta = \frac{\pi}{2}, \frac{3\pi}{2}, \frac{5\pi}{2}, \dots, \frac{2L+1}{2}\pi$ (L is an integer number), then $\sin^2 \delta = 1$ and $\frac{1}{T_{FS}} - \frac{1}{T_S} = \frac{1}{4}[U - V]$. Now the phase thickness δ is equal to π multiplied by a semi-integer number. These maximal values of $\frac{1}{T_{FS}} - \frac{1}{T_S}$ correspond to transmittance minima T_{MIN} which are smaller than substrate transmittance T_S . For these values of the phase thickness δ the optical thickness nd is a semi-integer number of $\lambda/2$: $nd = \frac{(2L+1)}{2}\frac{\lambda}{2} = m\frac{\lambda}{2}$. Here L is an integer number.

The integer and semi-integer numbers m corresponding to transmittance maxima or minima are also known as interference orders [2]. Their values can be determined from the extrema positions in the thin film transmittance spectra. If the numbers of m are known, then it will be possible to find the phase thickness δ at all extrema points and to make some assumptions for δ at each point of the measured spectra.

3 Description of the approach

Let us analyse equation (5) from the point of view of determination of the film refractive index dispersion $n(\lambda)$. As it was mentioned above, transmittance spectra of coated and uncoated substrate are measured usually at discrete

wavelengths λ_j in a spectral region of the substrate transparency (not at a single wavelength). At each measurement wavelength λ_j the term $V_j = n_{sj} + \frac{1}{n_{sj}} = \frac{2}{T_{sj}}$ can be easily calculated from the measured transmittance of uncoated substrate T_{sj} . To find the film refractive index n_j for a given wavelength λ_j , the corresponding numerical value of the term U_j is necessary. It can be found from the equation:

$$U_j = V_j + \frac{4 \left(\frac{1}{T_{FSj}} - \frac{1}{T_{sj}} \right)}{\sin^2 \delta_j}, \quad (6)$$

but only in the case if the phase thickness of the film δ_j for the same wavelength λ_j is known. *Note, that if $\sin^2 \delta \rightarrow 0$ (for $\delta = m\pi$), small uncertainty of δ will cause high uncertainty in the term U .*

If U_j is known, then the corresponding refractive index value n_j can be calculated from the following biquadratic equation:

$$n_j^4 - n_{sj}U_jn_j^2 + n_{sj}^2 = 0. \quad (7)$$

There are two positive roots of this equation – one higher than the substrate index n_{sj} and another one smaller than n_{sj} . In our experiment, materials of refractive index higher than that of the substrate were used and the corresponding root was chosen.

The substrate refractive index n_{sj} at λ_j can be found from the equation [2]:

$$n_{sj} = \frac{1}{T_{sj}} + \left(\frac{1}{T_{sj}^2} - 1 \right)^{\frac{1}{2}}. \quad (8)$$

Equations (3, 4, 6, 7, 8) show that it is possible to calculate thin film refractive index n_j if the corresponding phase thickness δ_j is known.

In brief, our approach consists of the following main steps:

1. Determination of interference orders m corresponding to the extrema wavelengths in the measured transmittance spectrum.
2. Determination of the thin film phase thickness δ at each measurement wavelength using interference orders m and Cauchy expression for the refractive index dispersion.
3. Calculation of the initial values of the refractive index at each wavelength from analytically derived equation (7) using the phase thickness δ , measured film-substrate transmittance $T_{FS}(\lambda)$ and measured uncoated substrate transmittance $T_S(\lambda)$.
4. Determination of the film thickness d from the phase thickness δ and the initial values of the refractive index.
5. Calculation of the final values of the refractive index at each wavelength from the phase thickness δ and film thickness.

In more details, the proposed approach is explained in Section 5 and it is illustrated using transmittance spectra of some vacuum deposited thin films.

4 Experimental details

To test the capabilities of the phase thickness approach, barium titanate and titanium oxide films were investigated.

Barium titanate films were deposited by RF magnetron sputtering of a 76 mm diameter water-cooled BaTiO₃ ceramic disk at sputtering pressure about 1 Pa. Depositions were carried out using high frequency power 350 W and a target-substrate distance 8 cm. The films were deposited in sputtering atmosphere 100% Ar. During the deposition, the substrate temperature was kept $\sim 10^\circ\text{C}$ by means of water circulation. The physical thickness of all films was in the range 0.1–0.3 μm . One half of each substrate was left uncoated during the film deposition.

Titanium oxide films were deposited by magnetron reactive sputtering of a 76 mm diameter water-cooled titanium target at total sputtering pressure of ~ 1 Pa (Ar + O₂) and oxygen concentration in the range 25–35%. Preliminary gas pressure into the recipient was $\sim 8 \times 10^{-3}$ Pa. Deposition rates ~ 6.4 nm/min were obtained at a power-supply voltage 500 V, discharge current 0.5 A, and a target-substrate distance 8 cm. The glass substrates were mounted on a stainless-steel substrate holder, which was maintained at a temperature $\sim 200^\circ\text{C}$. One half of each substrate was left uncoated during film deposition.

The optical transmittance of coated and uncoated substrates were measured in the wavelength region $\lambda = 0.33\text{--}0.80$ μm using automatic spectrophotometric recorder “Specord UV – VIS” after film deposition. Accuracy in measurements was $\sim 0.5\%$. Typical measured transmittance spectrum of uncoated glass substrate $T_S(\lambda)$ and interference spectrum of a substrate with film $T_{FS}(\lambda)$ are shown in Figures 2a (for barium titanate films) and 2b (for titanium oxide films).

The spectra given in Figures 2a and 2b allow us to make some important assumptions for the substrates and films. Transmittance of the substrates $T_S(\lambda)$ is relatively constant ($\sim 92\%$) in the range 0.37–0.80 μm and in this range the substrate absorption may be neglected [2].

Transmittance maxima in the barium titanate thin film spectrum $T_{FS}(\lambda)$ in Figure 2a are very close to the substrate transmittance, which means very low values of extinction coefficient k , i.e. $k \sim 0$ [2]. The depth of the minima weakly depends on the wavelength, which means weak dispersion of the refractive index $n(\lambda)$ [2]. Barium titanate films are studied in the spectral region $\lambda = 0.37\text{--}0.80$ μm .

The spectra shown in Figure 2b can be analysed in a similar way to make some assumptions for the titanium oxide thin films. Transmittance maxima in the thin film interference spectrum $T_{FS}(\lambda)$ are very close to the substrate transmittance in the spectral region $\lambda > 0.45$ μm , which means $k \sim 0$ [2]. In the spectral region $\lambda < 0.45$ μm there are no interference maxima close to the substrate transmittance, i.e. the absorption of the film can not be neglected. This conclusion is in agreement with the well known absorption of titanium oxide thin films in the blue zone of the visible spectrum and in the near ultraviolet region [16]. The depth of the minima in the spectral region

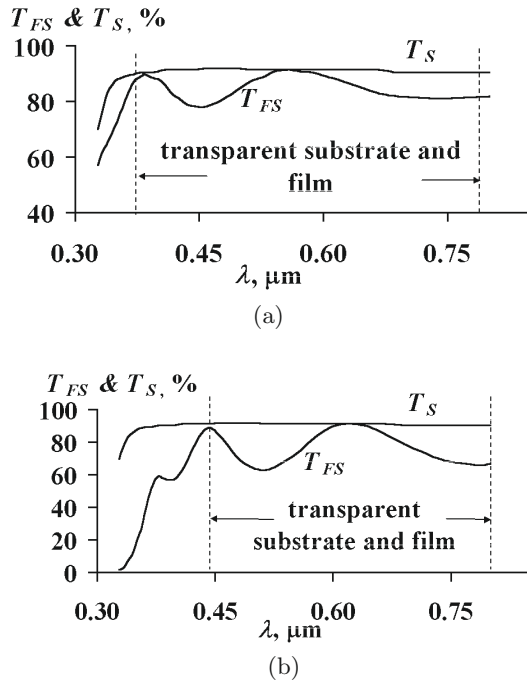


Fig. 2. Measured transmittance spectra of uncoated glass substrates $T_S(\lambda)$ and interference spectra of coated substrates $T_{FS}(\lambda)$: (a) with barium titanate thin film; (b) with titanium oxide thin film.

$\lambda = 0.45\text{--}0.80\ \mu\text{m}$ also weakly depends on the wavelength, and again weak dispersion of the refractive index $n(\lambda)$ can be assumed for this spectral region [2].

5 Results and discussion

5.1 Films with several extrema in their transmittance spectra

The calculation procedure is divided into five different steps, which are described in details below.

I. The first step in our approach is determination of interference orders m corresponding to all extrema points of the transmittance spectra. As the phase thickness $\delta = 2\pi n d \frac{1}{\lambda}$ increases when $1/\lambda$ increases, the numbers m also become higher. At least one interference order should be found for determination of all values of m .

The numbers m corresponding to two successive transmittance minima can be determined in the following way. If these minima are located at wavelengths λ_1 and λ_2 ($\lambda_1 > \lambda_2$) then the optical thickness of the film is related with these wavelengths as follows:

$$n_1 d = (2L + 1) \frac{\lambda_1}{4}, \quad n_2 d = (2L + 3) \frac{\lambda_2}{4}, \quad (9)$$

where n_1 and n_2 are refractive index values at wavelengths λ_1 and λ_2 . For these minima $\sin^2 \delta = 1$ and the following relations can be written:

$$\frac{1}{T_{FS}} - \frac{1}{T_S} = \frac{1}{4} [U - V] \Rightarrow U = V + 4 \left(\frac{1}{T_{FS}} - \frac{1}{T_S} \right),$$

Table 1. Calculated values of m for the spectra given in Figure 2a (barium titanate).

$\lambda_{EXT}, \mu\text{m}$	0.741	0.562	0.455	0.385
$m = \delta_{EXT}/\pi$	1.5	2.0	2.5	3.0
δ_{EXT}, rad	4.712	6.283	7.854	9.425

Table 2. Calculated values of m for the spectra given in Figure 2b (titanium oxide).

$\lambda_{EXT}, \mu\text{m}$	0.787	0.617	0.513	0.444
$m = \delta_{EXT}/\pi$	1.5	2.0	2.5	3.0
δ_{EXT}, rad	4.712	6.283	7.854	9.425

and

$$n^4 - n_S U n^2 + n_S^2 = 0. \quad (10)$$

As V and n_S can be calculated from T_S (see Eqs. (4, 8)), equation (10) allows determination of n_1 and n_2 . The value of L can be calculated from:

$$\frac{n_1 \lambda_2}{\lambda_1 n_2} = \frac{2L + 1}{2L + 3}$$

and it should be rounded to the nearest integer number. When L is known, at $\lambda_1 m = \frac{2L+1}{2}$.

The results from the first step for determination of the interference orders m are presented in Tables 1 (barium titanate film) and 2 (titanium oxide film). After that, the values of m were used for determination of the phase thickness δ_{EXT} corresponding to the extrema wavelengths λ_{EXT} of the investigated spectra.

II. The second step is to find an appropriate expression for the phase thickness δ as a function of λ taking into account refractive index dispersion of the film.

In many practical situations Cauchy expression of the following type

$$n(\lambda) = a + \frac{b}{\lambda^2} \quad (11)$$

is used to describe refractive index dispersion. The last equation for $n(\lambda)$ leads to the following equation for δ :

$$\delta = 2\pi n(\lambda) d \frac{1}{\lambda} = 2\pi \left(a + \frac{b}{\lambda^2} \right) d \frac{1}{\lambda} = \left(A + \frac{B}{\lambda^2} \right) \frac{1}{\lambda}, \quad (12)$$

where $A = 2\pi a d$ and $B = 2\pi b d$. Equation (12) shows that if A and B are known, then it will be easy to calculate the phase thickness δ_j at all measurement wavelengths λ_j , not only at extrema positions.

To propose one possible way for determination of A and B let us transform the last equation into

$$\delta \lambda = A + B \frac{1}{\lambda^2}. \quad (13)$$

This equation shows that in the case of Cauchy dispersion equation (11) the product $(\delta \lambda)$ is a linear function of $\frac{1}{\lambda^2}$ with slope B and intercept A . A and B can be determined from δ_{EXT} and λ_{EXT} values using a graphical presentation of the values of $(\delta_{EXT} \lambda_{EXT})$ as a function of $\frac{1}{\lambda_{EXT}^2}$.

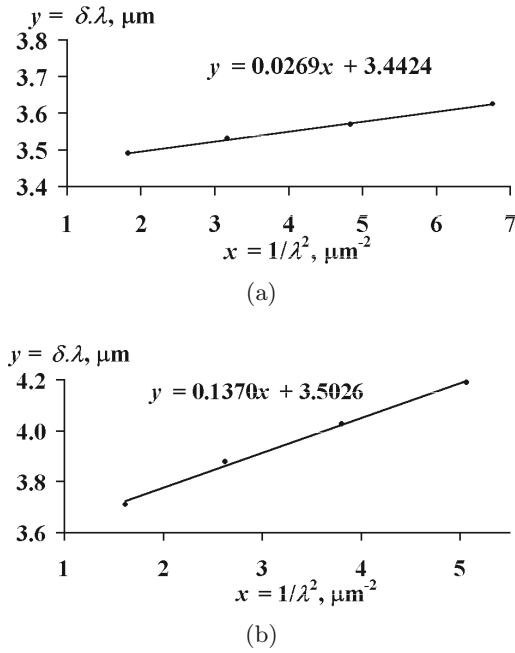


Fig. 3. Graphical presentation of the product $\delta\lambda$ (denoted as y) as a function of $1/\lambda^2$ (denoted as x) for the spectra given in Figure 1; circles (•) correspond to the extrema points in Figure 1; the solid lines are linear approximations of the extrema points with equations given on the charts; (a) barium titanate thin film; (b) titanium oxide thin film.

It is illustrated in Figure 3. The values given in Tables 1 and 2 are used to obtain these charts.

From the approximation of the transmittance extrema points in Figures 2a and 2b (shown with • symbols) with linear functions (the solid lines) the following equations for the phase thickness δ were derived:

$$\delta = \left(3.4424 + \frac{0.0269}{\lambda^2} \right) \frac{1}{\lambda} \quad (14)$$

for the barium titanate thin film and

$$\delta = \left(3.5026 + \frac{0.1370}{\lambda^2} \right) \frac{1}{\lambda} \quad (15)$$

for the titanium oxide thin film. In these equations λ is in μm .

III. The third step is to find initial values of the refractive index n . It is carried out by means of equations (6) and (7).

Analysis of the initial values of n shows that special care should be taken for the points near transmittance maxima, where $\sin^2 \delta \rightarrow 0$. Note again that near transmittance maxima ($\delta_j \rightarrow m\pi$), there is high level of uncertainty in the term U and therefore in the values of n due to division by very small numbers. This is illustrated in Figure 4 where the values of U and the initial values of refractive index n_{INITIAL} are shown as a function of λ .

IV. The next step in our study is to find the film thickness d . It can be calculated at each wavelength λ_j using

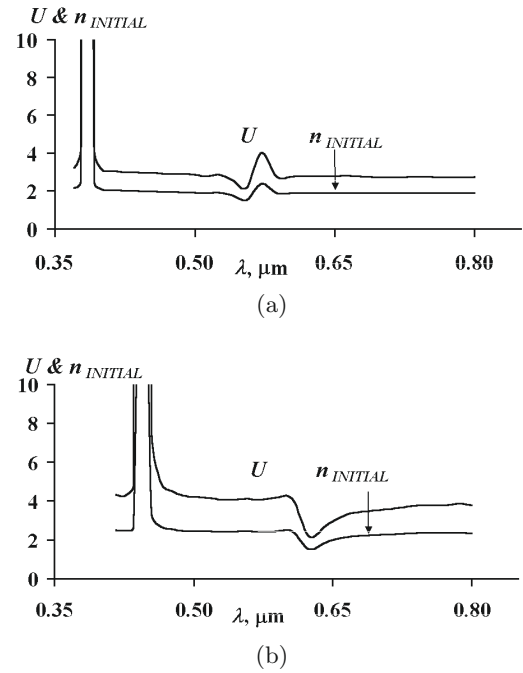


Fig. 4. Values of U and the initial values of refractive index n_{INITIAL} as functions of λ ; (a) barium titanate thin film; (b) titanium oxide thin film.

corresponding values of the phase thickness δ_j and the initial values of refractive index n_j according to the following equation:

$$d_j = \frac{\delta_j \lambda_j}{2\pi n_j}. \quad (16)$$

Figure 5 shows the calculated values of d as functions of the wavelength for both presented samples. The dashed horizontal lines represent the graphical estimation of the film thicknesses – $d = 0.29 \mu\text{m}$ for barium titanate thin film and $d = 0.26 \mu\text{m}$ for titanium oxide thin film. As it can be seen, the deviations from the horizontal lines are high near the wavelengths where $\sin^2 \delta \rightarrow 0$.

During the calculation of the film thickness d , there are two possible ways to minimise uncertainties near the points where $\sin^2 \delta \rightarrow 0$. The first one is to exclude the points where the refractive index values are unacceptable and afterwards to determine the average of all d_j :

$$d_A = \frac{1}{N_1} \sum_{j=1}^{N_1} d_j, \quad (17)$$

where N_1 is the number of all points taken into account in calculation of the film thickness. As it was not easy to find reasonable criteria for exclusion of any points from the measured spectra, all points were included in the calculation of d_A . For the spectra presented in Figure 2, equation (17) leads to $d_A = 0.278 \mu\text{m}$ for the barium titanate thin film and $d_A = 0.256 \mu\text{m}$ for the titanium oxide thin film.

The second way for reduction of uncertainties near the points where $\sin^2 \delta \rightarrow 0$ is to weight the values of d_j with

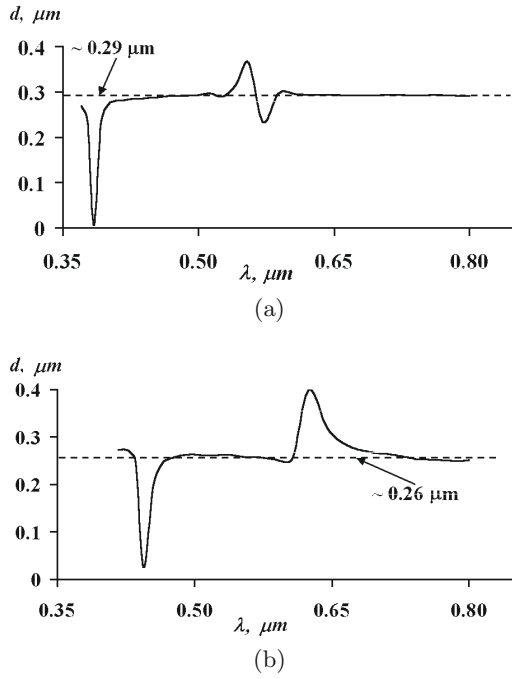


Fig. 5. Thin film thickness d as a function of λ ; (a) barium titanate thin film; (b) titanium oxide thin film.

values of some appropriate weight function. The main idea is to take these points with minimal weight. An asymptotic study of equations (5) and (6) leads to the conclusion that the functions $\sqrt{\frac{1}{T_{FS}} - \frac{1}{T_S}}$ and $\sqrt{U - V}|\sin \delta|$ could be useful ones to weight the values of d_j . The function of $\sin^2 \delta$ also seems to be an appropriate weight. All these weight functions (WF) are taken into account during the calculation of the weighted average thickness value according to the equation:

$$d_W = \frac{\sum_{j=1}^{N_2} d_j WF_j}{\sum_{j=1}^{N_2} WF_j}. \quad (18)$$

In the last equation N_2 is the number of all points of transmittance spectra and WF_j denotes the corresponding weight function value. For the spectra presented above the film thicknesses calculated according to equations (17, 18) are presented in the upper half of Table 3 for all WF .

It can be seen that the estimated maximal relative difference ε between d_A and d_W ($\varepsilon = 2|d_A - d_W|/(d_A + d_W)$) is higher for the barium titanate film ($\sim 4.1\%$) compared to the titanium oxide film ($\sim 1.9\%$).

V. The last step is to find the final values of the refractive index n as a function of λ . When the film thickness d is known the final values of the refractive index $n(\lambda_j)$ can be determined at each wavelength λ_j from the phase thickness δ_j using the equation:

$$n(\lambda_j) = \frac{\delta_j \lambda_j}{2\pi d}. \quad (19)$$

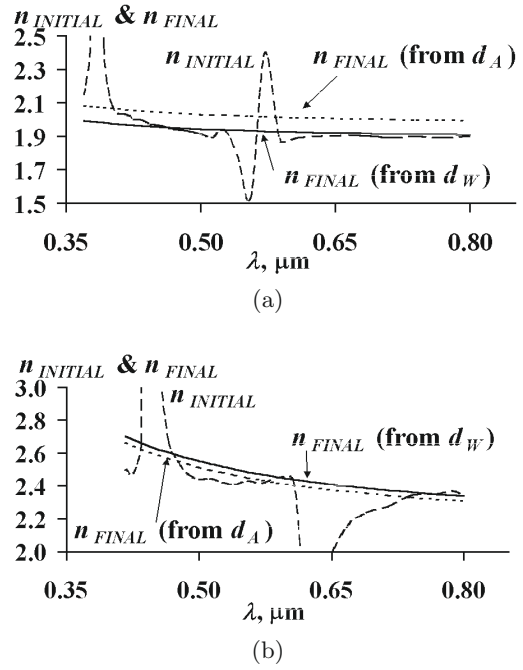


Fig. 6. Initial values ($n_{INITIAL}$) and final values (n_{FINAL} calculated from d_A and d_W) of the refractive index for: (a) barium titanate thin film; (b) titanium oxide thin film.

The final values of the refractive index were calculated according to equation (19) using both values of the film thickness d_A and d_W . The initial and final values of the refractive index ($n_{INITIAL}$ and n_{FINAL}) are presented in Figures 6a (barium titanate film) and 6b (titanium oxide film).

Figure 6 shows that there is a certain disagreement between final refractive index values calculated from d_A and d_W (Eqs. (17, 18)). The relative difference is $\sim 1.9\%$ for the titanium oxide thin film and $\sim 4.1\%$ for the barium titanate thin film. These numbers can be explained very well with the observed differences between d_A and d_W and equation (19) used to obtain the final refractive index values. Therefore, some additional criterion is necessary to find which expression for calculation of the film thickness d (Eq. (17) or (18); d_A or d_W) is more correct. As at this stage of the investigation the refractive index dispersion and the film thickness are known, the calculation of thin film transmittance spectrum (according to Eq. (1)) is already possible. Therefore, the comparison between measured and calculated spectra can be used as a criterion to find which is the better way to calculate the film thickness d .

Table 3 (the lower half) shows the root mean square difference δT_{RMS} between measured and calculated spectra using all proposed WF . It is calculated by means of the following equation:

$$\delta T_{RMS} = \sqrt{\frac{1}{N_2} \sum_{j=1}^{N_2} (T_{FSj} - T_{CALCj})^2}. \quad (20)$$

Table 3. Thin film thicknesses d (in μm) using different weight functions and root mean square difference between measured and calculated thin film transmittance spectra δT_{RMS} (in %).

	Weight function	none	$\sqrt{\frac{1}{T_{FS}} - \frac{1}{T_S}}$	$\sqrt{U - V} \sin \delta $	$\sin^2 \delta$
$d, \mu\text{m}$	barium titanate	0.278	0.285	0.285	0.290
	titanium oxide	0.256	0.255	0.255	0.260
$\delta T_{RMS}, \%$	barium titanate	1.97	1.04	1.05	0.62
	titanium oxide	1.85	1.90	1.90	1.70

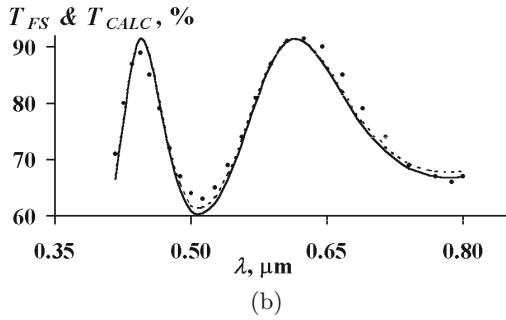
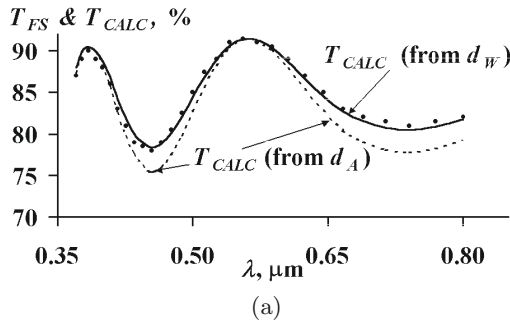
**Fig. 7.** Comparison between measured T_{FS} (●) and calculated T_{CALC} spectra using the value of d_W (solid line) or the value of d_A (dashed line) for: (a) barium titanate thin film; (b) titanium oxide thin film.

Table 3 (the lower half) shows that the values of δT_{RMS} are lower, if $\sin^2 \delta$ is taken as WF . The difference between all WF is more clearly seen for the barium titanate thin film, where δT_{RMS} decreases from 1.97% (for d_A) to 0.62% (for d_W with $WF \sin^2 \delta$). Accordingly, $\sin^2 \delta$ is used as WF in the next part of our study.

The comparison between measured and calculated spectra is presented in Figure 7. Figure 7a shows that for the barium titanate thin film the measured spectrum (● symbols) is much better reproduced with the solid line calculated from d_W . The dashed line (calculated from d_A) is not so close to the measurements near transmittance minima (near 0.45 and 0.75 μm) and the differences are of order of 2–3%. These differences can be explained with the difference between d_A and d_W . It can be seen that the spectrum calculated from d_W is much more close to the

Table 4. Comparison between refractive index values calculated using Swanepoel's method – $n^*(\lambda)$ and according to the presented approach – $n^{**}(\lambda)$ for the barium titanate thin film; λ_{EXT} – the extrema wavelengths; T_S – uncoated substrate transmittance; T_{MAX} and T_{MIN} – transmittance maxima and minima envelopes and m – interference orders (barium titanate).

$\lambda_{EXT}, \mu\text{m}$	T_S	T_{MAX}	T_{MIN}	m	$n^*(\lambda)$	$n^{**}(\lambda)$
0.385	0.905	0.905	0.740	3.0	2.014	1.987
0.455	0.920	0.920	0.780	2.5	1.987	1.959
0.555	0.915	0.915	0.795	2.0	1.965	1.936
0.747	0.905	0.905	0.810	1.5	1.943	1.915
Film thickness		$d^* = 0.286 \mu\text{m}$			$d^{**} = 0.290 \mu\text{m}$	

measured one in comparison with the spectrum calculated from d_A .

Figure 7b shows that for the titanium oxide thin film both calculated spectra (from d_W and d_A) describe very well the measurements and these both spectra are practically identical. However, the small difference between d_W and d_A ($\sim 1\%$) and the small differences between the corresponding calculated transmittance spectra can be regarded mostly as a case of coincidence rather than as a case of a rule.

Figure 7 shows that the spectra calculated from the weighted average thickness d_W describe very well measured transmittance spectra. It is a reason to conclude that the determination of the final refractive index n_{FINAL} using d_W gives more realistic results in comparison with the case of using d_A .

Tables 4 and 5 present comparison between refractive index values calculated using Swanepoel's method $n^*(\lambda)$ [2] and according to the presented approach – $n^{**}(\lambda)$ (calculated from the phase thickness using d_W). In both tables λ_{EXT} are the extrema wavelengths; T_S denote uncoated substrate transmittance; T_{MAX} and T_{MIN} correspond to transmittance maxima and minima envelopes and values of m are interference orders.

In brief the calculations according to Swanepoel's method are performed using the following steps:

1. Determination of the initial refractive index values $n_{INITIAL}$ at the extrema wavelengths using the equation:

$$n_{INITIAL} = \left[N + (N^2 - n_S^2)^{1/2} \right]^{1/2},$$

Table 5. Comparison between refractive index values calculated using Swanepoel's method $n^*(\lambda)$ and according to the presented approach – $n^{**}(\lambda)$ for the barium titanate thin film; λ_{EXT} – the extrema wavelengths; T_S – uncoated substrate transmittance; T_{MAX} and T_{MIN} – transmittance maxima and minima envelopes and m interference orders (titanium oxide).

λ_{EXT} μm	T_S	T_{MAX}	T_{MIN}	m	$n^*(\lambda)$	$n^{**}(\lambda)$
0.444	0.915	0.890	0.60	3.0	2.569	2.566
0.513	0.915	0.905	0.63	2.5	2.468	2.460
0.625	0.915	0.915	0.65	2.0	2.369	2.356
0.787	0.905	0.905	0.66	1.5	2.294	2.277
Film thickness		$d^* = 0.260 \mu\text{m}$		$d^{**} = 0.260 \mu\text{m}$		

where $N = 2n_S \frac{T_{MAX} - T_{MIN}}{T_{MAX} T_{MIN}} + \frac{n_S^2 + 1}{2}$ [2].

- Calculation of the film thickness by means of two adjacent transmittance maxima/minima (using the extrema wavelengths λ_1 and λ_2 and the corresponding initial refractive indices n_1 and n_2), according to the equation

$$d = \frac{\lambda_1 \lambda_2}{2(\lambda_1 n_2 - \lambda_2 n_1)} (\text{averaged}) [2].$$

- Determination of the interference orders m using $m = \frac{2n_{INITIAL}d}{\lambda}$ (rounded to integer or semi-integer number) [2].
- Calculation of the final refractive index values using the initial ones, the interference orders, the film thickness and Cauchy expression (11) [2].

It can be seen from the last two columns of Tables 4 and 5 that the maximal difference between n^* and n^{**} is not higher than 1.5%, which is in the limits of the accuracy of the Swanepoel's method.

The agreement between n^* and n^{**} and the agreement between measured and calculated transmittance is a base to verify the results and to validate the presented phase thickness approach for determination of refractive index dispersion and thickness of nonabsorbing thin films.

5.2 Films with two extrema in their transmittance spectra

It is well-known that when the film thickness decreases the number of extrema in corresponding transmittance spectra also decreases [2]. This leads to difficulties in construction of envelopes. Figure 8 shows two spectra of films of thickness less than $0.2 \mu\text{m}$.

The spectrum in Figure 8a corresponds to barium titanate thin film. Two extrema in the spectral region of substrate transparency ($\lambda > 0.37 \mu\text{m}$) can be seen – maximum at $\lambda = 0.570 \mu\text{m}$ and minimum at $\lambda = 0.385 \mu\text{m}$. This minimum is the only one in the whole measured spectrum and it is not possible to construct the minima envelope (and to use Swanepoel's method). On the other hand both extrema could be used for determination of phase thickness and refractive index dispersion.

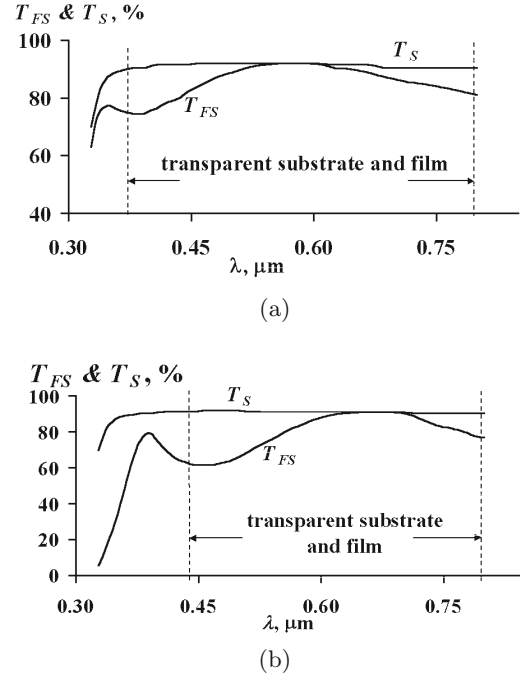


Fig. 8. Transmittance spectra with insufficient number of extrema for construction of envelopes; (a) barium titanate thin film; (b) titanium oxide thin film.

Figure 8b shows transmittance spectrum of titanium oxide film deposited at the same conditions like the sample presented above (but for two times shorter deposition time). There are also two extrema in the spectral region of negligible thin film absorption $\lambda = 0.45\text{--}0.80 \mu\text{m}$: maximum at $\lambda = 0.670 \mu\text{m}$ and minimum at $\lambda = 0.455 \mu\text{m}$. For this sample the minimum at $\lambda = 0.455 \mu\text{m}$ is also the only one in the whole measured spectrum and it is not possible to construct the minima envelope too. However, both extrema could be used for determination of phase thickness and refractive index dispersion.

At low film thicknesses the interference orders m could be estimated if the refractive index dispersion $n(\lambda)$ is supposed to be low. In our cases with transmittance spectra with one minimum at λ_1 and one maximum at λ_2 , ($\lambda_1 < \lambda_2$) it can be written:

$$n_1 d = (m + 0.5) \frac{\lambda_1}{2}, \quad n_2 d = m \frac{\lambda_2}{2}. \quad (21)$$

As for all transparent materials with negligible absorption the refractive index dispersion is low, it is reasonable to suppose that $n_1 \approx n_2$. Then from equation (21) it follows $(m + 0.5) \frac{\lambda_1}{2} \approx m \frac{\lambda_2}{2}$. The number m should be rounded to the nearest integer number. For our spectra in both cases $m = 1$.

Figure 9 shows plots of the product $\delta\lambda$ versus $1/\lambda^2$ for the spectra given in Figure 8. The extrema points were interpolated with straight lines for determination of the phase thickness dispersion.

Determination of the film thickness leads to the following values: $d_A = 0.148 \mu\text{m}$ and $d_W = 0.142 \mu\text{m}$ for the barium titanate film and $d_A = 0.132 \mu\text{m}$ and $d_W = 0.137 \mu\text{m}$

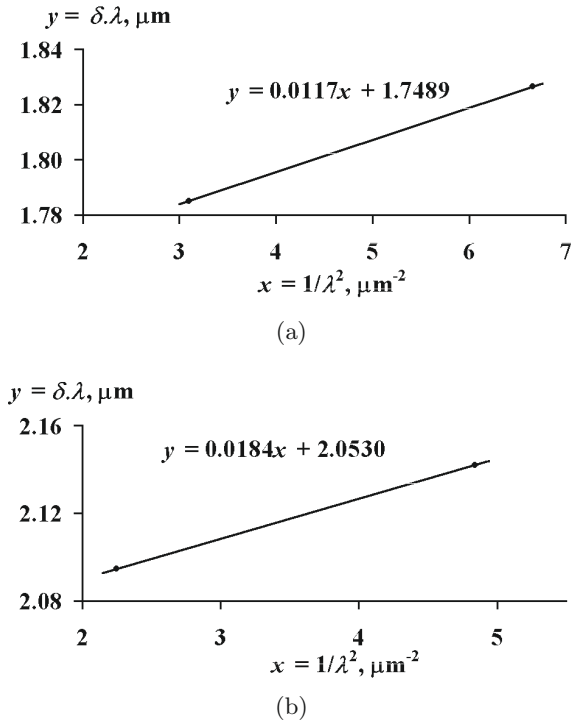


Fig. 9. Plots of the product $\delta\lambda$ versus $1/\lambda^2$ for the spectra given in Figure 7: (a) barium titanate thin film; (b) titanium oxide thin film.

for the titanium oxide film. The corresponding final values of the refractive index are presented in Figure 10. For both samples there are $\sim 5\%$ relative differences between values of d_A and d_W and between the corresponding refractive index values.

Figures 11a and 11b show comparison between measured transmittance T_{FS} (\bullet symbols) and calculated transmittance T_{CALC} according to equation (1) using averaged thickness d_A (dashed lines) and weighted average thickness d_W (solid lines). In the spectral regions near measured transmittance maxima ($\sim 0.57 \mu\text{m}$ for barium titanate thin film and $\sim 0.67 \mu\text{m}$ for titanium oxide thin film) both calculated spectra describe very well measured data. However, in the spectral regions near measured transmittance minima ($\sim 0.39 \mu\text{m}$ for barium titanate thin film and $\sim 0.45 \mu\text{m}$ for titanium oxide thin film) the solid lines are closer to the measurements. It is a reason to conclude again that the final refractive index values calculated from weighted average thickness d_W are more correct. Agreement between measurements and calculations is a base to conclude that the phase thickness approach is applicable for determination of thin film optical parameters ($n(\lambda)$ and d) in cases when measured transmittance spectra have one minimum and one maximum only.

In concluding remarks, it should be noted that even if the thin film optical absorption can not be fully neglected, the calculated refractive index values and thickness still can be used as initial ones for further optimization in order to find the complex refractive index.

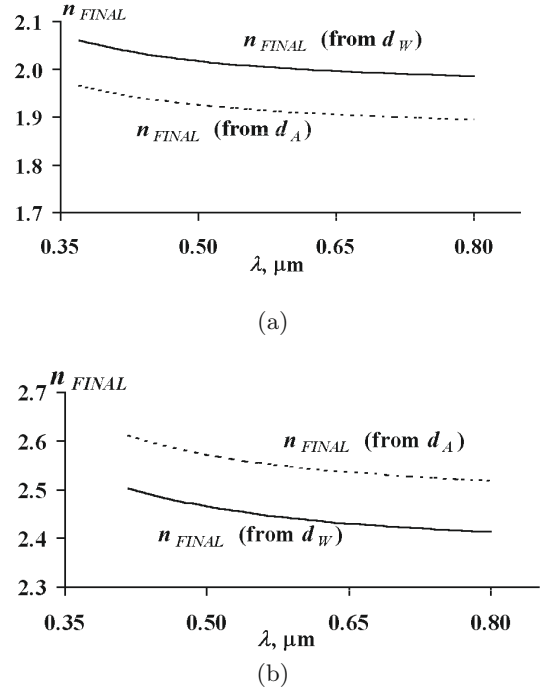


Fig. 10. Final values of refractive index calculated from d_A and from d_W ; (a) barium titanate thin film; (b) titanium oxide thin film

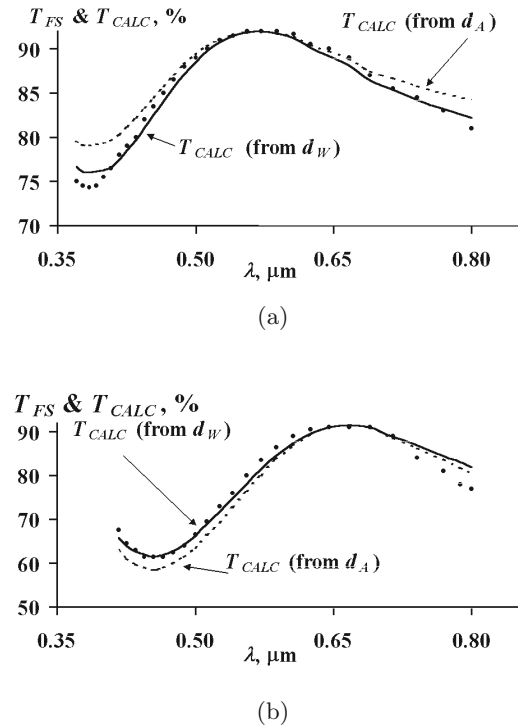


Fig. 11. Comparison between measured T_{FS} (\bullet) and calculated T_{CALC} spectra using the value of d_W (solid lines) or the value of d_A (dashed lines): (a) barium titanate thin film; (b) titanium oxide thin film.

6 Conclusions

A new approach for determination of refractive index dispersion $n(\lambda)$ and thickness d of thin films of negligible absorption and weak dispersion is proposed. The calculation procedure is based on application of relatively simple mathematical support, which is important for experimental investigations. All points of measured transmittance spectra are included in data processing.

The approach is demonstrated with determination of $n(\lambda)$ and d of barium titanate and titanium oxide thin films in the spectral regions 0.37–0.80 μm and 0.45–0.80 μm correspondingly.

The observed agreement between refractive index values calculated using Swanepoel's method and using presented procedure as well and the agreement between measured and calculated transmittance is a base to verify the results and to validate the presented phase thickness approach for determination of refractive index dispersion and thickness of nonabsorbing thin films.

The phase thickness approach is also applied for determination of refractive index dispersion and thickness of films of thickness less than 0.2 μm , which have transmittance spectra with number of extrema not enough for application of Swanepoel's method. Very good agreement between measured and calculated transmittance from the weighted average thickness is observed. It is a base to conclude that the phase thickness approach is applicable for determination of thin film optical parameters ($n(\lambda)$ and d) in cases when measured transmittance spectra have one minimum and one maximum only.

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References

1. J.C. Manifacier, J. Gasiot, J.P. Fillard, J. Phys. E: Sci. Instrum. **9**, 1002 (1976)
2. R. Swanepoel, J. Phys. E: Sci. Instrum. **16**, 1214 (1983)
3. D. Minkov, R. Swanepoel, Opt. Eng. **32**, 3333 (1993)
4. J.M. Gonzalez-Leal, R. Prieto-Alcon, J.A. Angel, D. Minkov, E. Marquez, Appl. Opt. **41**, 7300 (2002)
5. Q. Ren, C.B. Ma, J. Mater. Sci. Lett. **19**, 7 (2000)
6. D.J. Won, C.H. Wang, H.K. Jang, D.J. Choi, Appl. Phys. A **73**, 595 (2001)
7. K. Ayadi, N. Haddaoui, J. Mater. Sci. - Mater. El. **11**, 163 (2000)
8. L. Soliman, A. Ibrahim, Fizika A **6** 4, 181 (1997)
9. D. Poelman, P.F. Smet, J. Phys. D: Appl. Phys. **36**, 1850 (2003)
10. Sh.C. Chiao, B.G. Bovard, H.A. Macleod, Appl. Opt. **34**, 7355 (1995)
11. J.A. Dobrowolski, F.C. Ho, A. Waldorf, Appl. Opt. **22**, 3191 (1983)
12. T. Pencheva, M. Nenkov, J. Mod. Opt. **43**, 2449 (1996)
13. M. Nenkov, T. Pencheva, J. Opt. Soc. Am. A **15**, 1852 (1998)
14. E.G. Birgin, I. Chambouleyron, J.M. Martínez, J. Comput. Phys. **151**, 862 (1999)
15. S. Ventura, E.G. Birgin, J.M. Martínez, I. Chambouleyron, J. Appl. Phys. **97**, 043512 (2005)
16. H.A. Macleod, *Thin film optical filters*, 2nd edn. (Adam Hilger Ltd., Bristol, 1986)