DETERMINATION OF OPTICAL PARAMETERS OF THIN FILMS FROM TRANSMITTANCE SPECTRA

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Received 30 April 2011; accepted 15 May 2011.

1. Introduction

The knowledge of the wavelength dependent complex refractive index of thin films and other parameters is very important not only for their characterization, but also for the design and modelling of optical components, optical coatings or thin film solar cells. There are many different techniques for determining optical parameters (refractive index, extinction coefficient, optical band gap) and film thickness from spectrometric data. For example: (i) Measuring reflection and transmission at the same location [1]. (ii) Matrix methods (suitable for multilayer films). (iii) Methods using only a single transmission spectrum where belongs the envelope methods [5] and methods based on the "fitting" of dispersion relations that are the subject of this paper.

Method introduced in this paper was developed to analyse TCO films (Transparent Conductive Oxides), particularly samples of scandium doped zinc oxide (ZnO:Sc) thin films, which is a promising material for application in thin film photovoltaic cells and various optoelectronic devices. Samples were prepared by the rf magnetron sputtering using a BOC Edwards TF 600 electron beam and sputtering system. Transmittances spectra were measured by using UV/Vis double-beam spectrophotometer SPECORD 210 in its maximal wave range of 190 – 1100 nm.

2. Optical parameter identification

In presented analysis the Swanepoel's model [1] for optical transmission spectra is applied. Additionally, it is assumed that the refractive index follows the Sellmeier equation

(1a) and the extinction coefficient is modelled by the Cauchy phenomenological formula (1b).

$$n^2(\lambda) = A_n + B_n\lambda^2 / \left(\lambda^2 - C_n^2\right),\ k(\lambda) = K_1 + K_2\lambda^{-2} + K_3\lambda^{-4} \qquad \text{(1a), (1b)}$$

Phase mapping

Recently we have developed a complex method for optical parameter identification based on the knowledge of the phase difference defined by eq. 2

$$\Delta\varphi = \varphi_1 - \varphi_2 = 4\pi d\left(\frac{n(\lambda_1)}{\lambda_1} - \frac{n(\lambda_2)}{\lambda_2}\right), \qquad (2)$$

where the phase angle φ_i is $4\pi d\, n(\lambda_i)/\lambda_i$ (from [5]) and wave lengths λ_i (i=1,2) denote locations of two different local extremes of the transmission spectra (see Fig. 1) that appear for the sake of interference. For easier understanding, subscripts M or E are later used to denote quantities based on the model or from the experiment, respectively.

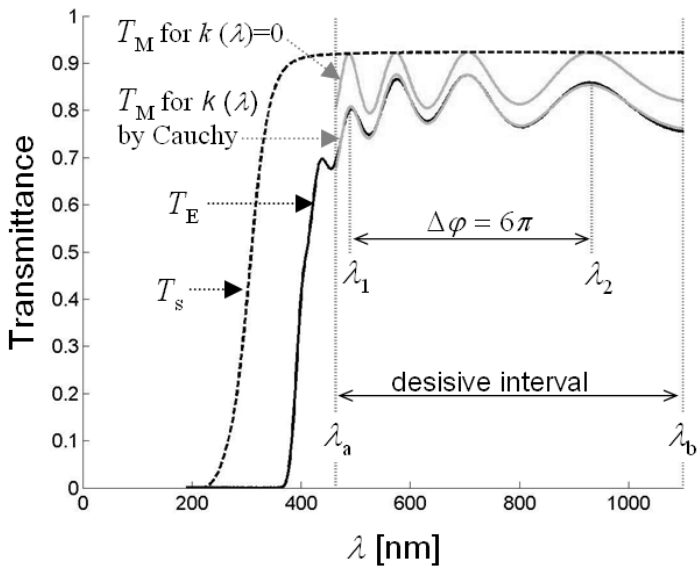

Fig.1: *Transmittance spectra. Symbol* T_s *denotes the substrate transmittance;* T_E *and* T_M *are the experimentally measured and the modelled spectra, respectively. Interval* (λ_a, λ_b) *marks the valid area significant for optimization and the fitting process. Wavelengths* λ_1, λ_2 *demonstrate an example of the phase difference* $\Delta\varphi$ *estimation.*

According to the structure of Swanepoel's model, the phase difference $\Delta\varphi$ between wave lengths λ_1 and λ_2 is an integer multiple of π, therefore can be easily estimated from the measured transmission spectra. After substitution for the refraction index n from eq. (1a) into eq. (2) we obtain formula for the phase shift predicted by the model

$$\Delta\varphi_{\mathrm{M}} = 4\pi d\left[\left(\frac{A_{\mathrm{n}}}{\lambda_1^2} + \frac{B_{\mathrm{n}}}{\lambda_1^2 - C_{\mathrm{n}}^2}\right)^{1/2} - \left(\frac{A_{\mathrm{n}}}{\lambda_2^2} + \frac{B_{\mathrm{n}}}{\lambda_2^2 - C_{\mathrm{n}}^2}\right)^{1/2}\right]. \tag{3}$$

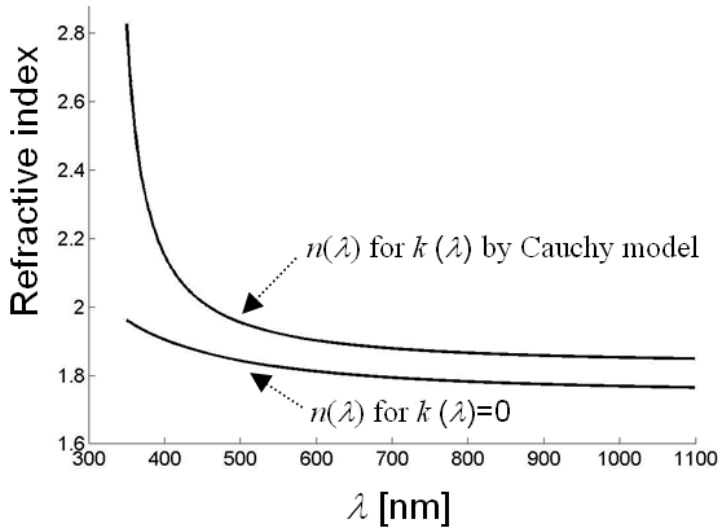

Fig.2: *Refractive index resulting from the identification process. Figure compares the refractive index estimated with the initial assumption of the extinction coefficient equal to zero with the final results adopting the Cauchy model.*

Comparing the experimental value of $\Delta\varphi_{\mathrm{E}}$ with the model prediction of $\Delta\varphi_{\mathrm{M}}$ allow us to effectively select convenient parameter values that are later used as an initial point for consequent fitting process. In the first approximation is assumed a zero extinction coefficient $k = 0$, thus there are four parameters left to be identified, namely A_{n}, B_{n} and C_{n} from the Sellmeier model and the layer thickness d. By performing extensive numerical experiments we have observed low sensitivity to the parameter C in relatively broad band of wave length values. Therefore fixing the value of C came out to be a convenient step for mapping the space of all possible parameter values in selected range by evaluating residual function $R_{\Delta\varphi} = \Delta\varphi_{\mathrm{E}} - \Delta\varphi_{\mathrm{M}}$.

Further optimization is necessary to correct the initial choice of C or to include identification of parameters characterizing absorption. This leads to a nonlinear multi-parametric optimization process where gradient methods can be effectively applied. A residual function in the form of a definite integral (4) with bounds λ_{a} and λ_{b} specifying decisive interval was used.

$$R_{\mathrm{T}} = \frac{1}{\lambda_{\mathrm{b}} - \lambda_{\mathrm{a}}}\left[\int_{\lambda_{\mathrm{a}}}^{\lambda_{\mathrm{b}}} \left(T_{\mathrm{E}}(\lambda) - T_{\mathrm{M}}(\lambda)\right)^2 d\lambda\right]^{1/2}. \tag{4}$$

3. Results

Table 1 presents parameter set identified at the initial approximation with the extinction coefficient is fixed to be zero, as well as the final result of the identification process. The refraction index for both cases is presented at Fig. 2 and for the comparison of the measured transmission spectra with the model see Fig. 1. Note that resulting curve T_E is nearly perfectly following the experimental spectra T_E.

Tab. 1: *Resulting parameters.*

	A_n [1]	B_n [1]	C_n [m]	K_1 [1]	K_2 [m^2]	K_3 [m^4]	d [nm]
Initial approx.	-0.406	3.453	1.52E-7	0	0	0	790
Final result	2.754	0.605	3.29E-7	0.00544	-1.9E-16	3.27E-35	748

4. Discussion

The comparison of T_E and T_M curves at Fig. 1 shows that the model is approximating the experiment very well within the selected decisive interval. Additionally, by our preliminary comparison with other methods are especially reliable the identified values of the film thickness *d*. Presented concept allows very general modification of the extinction coefficient model and some limited modifications of the refraction index model. The main advantage of the presented method, from the computational point of view, is the possibility to apply gradient methods for the final optimization process. The main limitation method is then the possibility to determine the phase difference value from the experiment and low absorption within the indicated decisive interval.

Acknowledgement

The result was developed within the CENTEM project, reg. no. CZ.1.05/2.1.00/03.0088 that is co-funded from the ERDF within the OP RDI program of the Ministry of Education, Youth and Sports.

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