A METHOD FOR DETERMINING OPTICAL CONSTANTS OF THIN FILMS

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A new method for determining optical constants of films on a transparent base is proposed on the basis of measured energetical coefficients of reflectance and transmittance and also of the films thickness. Optical constants are calculated by using an iteration process. The initial approximation is found either graphically or from simplified formulae.

The problem of determining optical constants (coefficients of refraction n_1 and absorption k) plays an important role in physical as well as in technical investigations of thin films optics. In the spectrophotometric method, refraction and absorption coefficients are determined basing on measured coefficients of reflectance R (from the side of the air), transmittance T and the film thickness d.

Energetic coefficients of reflectance and transmittance of the absorbing film on a transparent base for the case of normal light illumination are given by well-known

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formulae [1–3]

$$R = \left|\frac{(n_0-n_1+ik)(n_1-ik+n_2)+(n_1-ik-n_2)(n_0+n_1-ik)e^{-2i\delta_1}}{(n_0+n_1-ik)(n_1-ik+n_2)+(n_0-n_1+ik)(n_1-ik-n_2)e^{-2i\delta_1}}\right|^2 \tag{1}$$

$$T = \frac{16n_0n_2(n_1^2+k^2)e^{-4\pi kd/\lambda}}{|(n_0+n_1-ik)(n_1-ik+n_2)+(n_0-n_1+ik)(n_1-ik-n_2)e^{-2i\delta_1}|^2} \tag{2}$$

where $\delta_1 = \frac{2\pi}{\lambda}(n_1-ik)\,d$, n_0 — refraction coefficient of the air, n_2 — refraction coefficient of the base, d — thickness of the film, λ — wavelength. As the determination of the optical constants on the basis of Eqs (1) and (2) is in general difficult, graphical methods [4–6] are employed. They are, however, of limited accuracy, and require preparing a large number of plots. Optical constants can be also determined by means of approximate methods [7–10]. The accuracy of the values obtained for n_1 and k depends on satisfying the assumptions made in a particular approach. The method we propose in this paper enables calculations to a high degree of accuracy of the optical constants of the absorbing film on a transparent base, by making succesive approximations.

The conditions for the existence of the solutions of the set of Eqs (1), (2) in the region of physically relevant values of n_1, k, R, T, d have not been generally investigated. Finding these conditions and their discussion will be the subject of a subsequent paper. Here, we shall restrict ourselves to presenting the method, giving some examples of its applications and to some remarks about error propagation.

We determine the optical constants in two steps. First, we evaluate graphically or on the basis of approximate formulae the zeroth-order approximation of n_1, k, and later, using an iteration process, we obtain the optical constants to a high degree of accuracy.

To find the zeroth-order approximation we rewrite Eqs (1), (2) in the following form

$$n_1 = f_1(n_1, k) \tag{3}$$

$$k = f_2(b_1, k) \tag{4}$$

where

$$f_1(n_1, k) = \frac{R|t|^2 - W - \overline{W} - |u|^2 - (n_1^2+n_2^2+k^2)[(n_0-n_1)^2+k^2]}{2n_2[(n_0-n_1)^2+k^2]} \tag{5}$$

$$f_2(n_1, k) = \sqrt{\frac{T|t|^2 e^{4\pi kd/\lambda}}{16n_0n_2} - n_1^2} \tag{6}$$

and

$$t = (n_0+n_1-ik)(n_1-ik+n_2)+(n_1-ik-n_2)(n_0-n_1+ik)e^{-\frac{4\pi id(n_1-ik)}{\lambda}} \tag{7}$$

$$U = (n_1-ik-n_2)(n_0+n_1-ik)e^{-4\pi id/\lambda(n_1-ik)} \tag{8}$$

$$W = (n_0-n_1+ik)(n_1+n_2-ik)\,\overline{U}. \tag{9}$$

By $g_{1k}(n_1)$ we denote the function $f_1(n_1, k)$ for fixed k. Next, we make plots of the function $y = g_{1k}(n_1)$ for different values of k.

The intersection points of these curves with the straight line $y = n_1$ give pairs of values satisfying Eq. (3). The resulting plots are presented in Fig. 1.

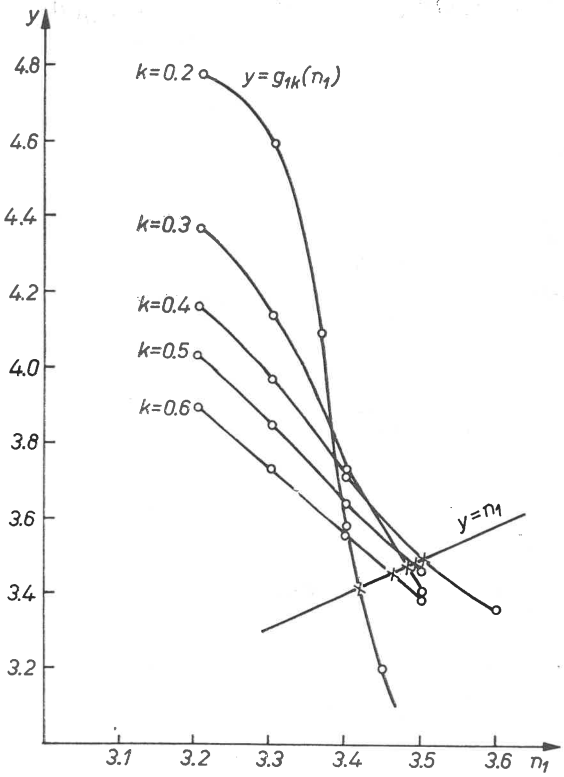

Fig. 1

Analogously, we find pairs of values satisfying Eq. (4). Let us denote by $g_{2n_1}(k)$ the function $f_2(n_1, k)$ for a fixed n_1, and make the plots of the function $y = g_{2n_1}(k)$ for different values of n_1. The intersection points of these curves with the straight line $y = k$ give the requested pairs of values (Fig. 2).

Among all pairs of values n_1 and k satisfying Eq. (3) and Eq. (4) we look for pairs of values satisfying Eqs (3) and (4) simultaneously. To find them, we construct two plots in the system (n_1, k) using the previously obtained two sets of pairs of values n_1 and k. The intersection point of the curves determines the zeroth-order approximation of the optical constants (Fig. 3).

To obtain more exact values of the optical constants, the iteration method proposed by Brown [11, 12] is used. Eqs (1) and (2) are written in the form

$$\varphi_1(n_1, k) = 0 \tag{10}$$

$$\varphi_2(n_1, k) = 0 \tag{11}$$

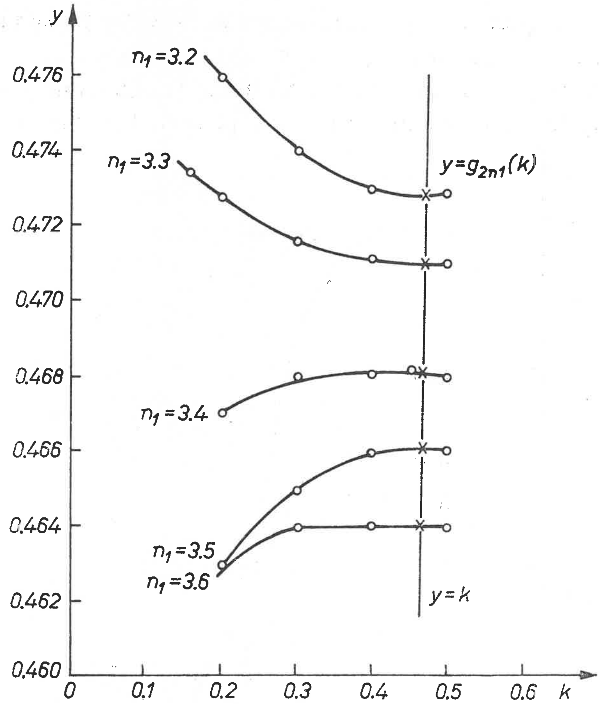

Fig. 2

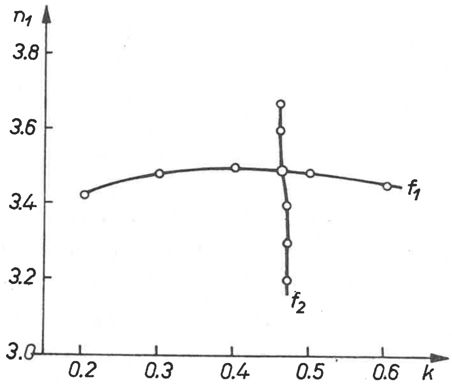

Fig. 3

where

$$\varphi_1(n_1, k) = R|t|^2 - W - \overline{W} - |U|^2 - [(n_1+n_2)^2 + \\ + k^2]\,[(n_0-n_1)^2+k^2]$$

$$\varphi_2(n_1, k) = -T|t|^2 + 16 n_0 n_2 (n_1^2+k^2) e^{-4\pi k d/\lambda}.$$

The function $\varphi_1(n_1, k)$ is expanded in Taylor's series in the neighbourhood of the zeroth-order point (n_1^0, k^0), keeping only linear terms of the series. On equating the expression obtained to zero, we express n_1 as a linear function on k:

$$n_1 = l(k). \tag{12}$$

After inserting (12) into (11) we obtain an equation with one unknown which, solved by Newton's method, yields the first-order approximation k^1.

This value is then inserted into (12) giving the first-order approximation n_1^1. Next, we expand the function φ_1 in the neighbourhood of the point (n_1^1, k^1) rejecting all non-linear terms in Taylor's series, *etc.*

This procedure is carried out until two subsequent approximations $n_1^{(j-1)}, k^{(j-1)}$ and $n_1^{(j)}, k^{(j)}$ differ (in absolute value) by less than a given $\varepsilon > 0$, *i. e.*

$$|n_1^{(j)} - n_1^{(j-1)}| < \varepsilon \quad |k^{(j)} - k^{(j-1)}| < \varepsilon.$$

In the cases considered by us the method was rapidly convergent, and the zeroth-order approximation sometimes differed markedly from the exact value.

Calculations were performed on ODRA-1204 computer at the Computing Centre of the University of Wrocław.

The method presented above was used for the first time to determine the optical constants of the PbO films [13]. Yellow and red films of PbO exhibit an absorption band in ultraviolet from 300 nm to 400 nm [8, 14]. In this range interferential extrema do not appear on the curves of reflectance and transmittance coefficients as functions of the wavelength. The convergence of the method for different values of the zeroth-order approximations was investigated in [13]. The graphically determined zeroth-order approximation for the red-film of PbO of the thickness $d = 3250$ Å, with $\lambda = 3500$ Å, equals respectively

$$n_1^0 = 3.45, \; k^0 = 0.469.$$

The results of subsequent approximations are presented in Table I.

TABLE I

j	$n_{(j)}^1$	$k^{(i)}$
1	3.498043	0.466376
2	3.497834	0.466420
3	3.497834	0.466420

As it is seen from Table I the method is rapidly convergent. The graphically determined zeroth-order approximation of the refraction and absorption coefficients for the yellow film of PbO of the thickness $d = 6005$ Å and for the wavelength $\lambda = 3500$ Å equals respectively

$$n_1^0 = 2.9, \quad k^0 = 0.27. \tag{13}$$

After three iteration steps results remain fixed, up to the fifth digit after the decimal point:

$$n_1^{(3)} = 2.90591, \quad k^{(3)} = 0.2700. \tag{14}$$

It is easily seen, that the zeroth-order approximation values differ but slightly from the values (14). When taken as a zeroth-order approximation values differ from (14) more than values (13), *e. g.* if we take $n_1^0 = 2.4$ and $k^0 = 0.3$, then the sequence of approximations is also convergent yielding the values (14). In general, however, the zeroth-order approximation range for which the above method is convergent depends on the parameters R, T, d, λ, n_2.

The question arises to what degree of accuracy are we able to determine optical constants, owing to the accuracy in measuring energetical coefficients of reflectance and transmittance and the thickness of the film. As the dependence of n_1 and k on R, T, d, λ, n_2 is of a complex form, it is not clear whether maximal errors $\Delta n_1, \Delta k$ correspond to maximal errors $\Delta R, \Delta T, \Delta d$.

Some information can be gained from the discussion of errors for certain sets of parameters R, T, d, λ, n_2. Their values are presented in Table II. The values of the last columns of Table II were obtained by a systematic investigation of the behaviour of n_1 and k when the parameters were changed in the range $R \pm \Delta R, T \pm \Delta T, d \pm \Delta d$.

It turns out that the parameter values with maximal errors in general do not correspond to extreme values of the optical constants. The values of n_1 and k presented in Table II correspond to the maximal errors.

Using the method presented above we have determined the optical constants for Al films taking advantage of the energetic coeffitients for different thickness and wavelengths, as given by Hass and Waylonis [15], who determined n_1 and k for Al films using Hadley's graphical method.

TABLE II

λ (Å)	n_2	R	ΔR	T	ΔT	d (Å)	Δd (Å)	n_1	k	Δn_1	Δk
3500	1.477	0.241	0.005	0.002	0.0002	6005	300	2.899	0.271	0.045	0.018
3000	1.488	0.217	0.004	0.003	0.0003	6005	300	2.725	0.215	0.042	0.012
3000	1.70	0.291	0.006	0.002	0.0002	3250	162	3.281	0.419	0.063	0.029
3500	1.65	0.317	0.006	0.0026	0.0002	3250	162	3.514	0.467	0.026	0.023

TABLE III

λ (nm)	d (Å)	R	T	Results of paper [15]		Numerical results	
				n_1	k	n_1	k
220	200	0.763	0.152	0.14	2.35	0.140	2.353
220	400	0.906	0.011	0.14	2.35	0.141	2.352
300	160	0.740	0.16	0.25	3.35	0.254	3.313
300	400	0.914	0.005	0.25	3.35	0.259	3.383
400	200	0.849	0.059	0.40	4.45	0.403	4.456
400	400	0.921	0.004	0.40	4.45	0.385	4.377
546	200	0.856	0.035	0.80	5.92	0.840	6.029
546	400	0.912	0.002	0.80	5.92	0.844	6.069

Besides, it was found [15] that for Al films of thickness exceeding 100 Å the optical constants do not depend on the thickness, and energetic coeffitients R and T calculated from optical constants fit experimental data.

The results obtained by us for Al films and those from [15] are presented in Table III. Optical constants determined in [15] graphically and calculated by the above-presented method in general coincide. Discrepancies appear for small values of the transmittance coefficients. Energetical coefficients calculated from optical constants determined numerically are exactly equal to initial data.

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