EFFECT OF NON-UNIFORM LAYER THICKNESS ON THE INTERFERENCE STRUCTURE OF OPTICAL TRANSMITTANCE

By

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The optical transmittance of thin films is calculated assuming slowly varying thickness. The approximate expression, deduced here is compared with computer simulated spectra. It is shown that with this expression the error due to uneven layer thickness can be estimated. A new method is suggested for the determination of the optical constants, extending previous procedures, wich are only valid for films of even thickness.

Introduction

Several authors have proposed the determination of optical parameters from transmission measurements only [1—6]. The procedure is based on the interference structure of the spectra, *i.e.* on a resolved series of maxima and minima of transmittance. In general, three equations of transmittance, more specifically two for the envelop curves and the third for the extrema condition are sufficient to determine three unknown parameters: the real and immaginary parts of the refractive index and the layer thickness.

This method supposes that (i) the optical constants are slowly varying functions of the wavelength and (ii) the film thickness is exactly constant. These conditions are seldom fully satisfied, for example in the case of slabs split from special laminated crystals [4] or vacuum deposited films of certain materials [5]. With most of the current technologies, *e.g.* with the CVD method one cannot obtain a constant layer thickness with the accuracy required by the method. At slowly varying thickness the deformation of spectra can be minimized by reducing the diameter of the light beam, however, this possibility is limited due to the signal-to-noise ratio. Thus, we determined the error caused by thickness uneveness and developed an improved method to obtain the optical constants of such real layers.

Theory

i) *The model.* The transmittance of a homogeneous film of varying thickness is given by the following expression,

$$T_{\mathrm{meas}} = \frac{\int J(x,y)\,T(x,y)\,dx\,dy}{\int J(x,y)\,dx\,dy},$$

where $T(x, y)$, and $J(x, y)$ are the local transmittance and the distribution of light density, respectively. The coordinates are perpendicular to the incident light. The dependence of the local transmittance on the coordinates x and y is caused by the variation in thickness wich can often be described in case of real films by rather complicated functions.

As the number of unknown parameters increases in the expression of the thickness, the evaluation of the measurement becomes contestable. By reducing the diameter of the light beam, the change in thickness of the measuring area becomes small enough and the bounding surfaces of the films can be approximated by two planes in the interesting range. For the sake of simplicity it is assumed that the light density is constant inside the rectangular measuring area and the thickness varies linearly along the edges. The measured transmittance can be approximated by an average function

$$T_{\mathrm{av}} = \frac{1}{2h}\int_{-h}^{h} T(d_0+\xi)\,d\xi, \tag{1}$$

where h is the largest deviation from the mean thickness d_0. It can be shown that this approximation is valid for less rigorous conditions, too.

In the following discussion the films with $\varepsilon \equiv h/d_0 > 0$ will be referred to as real samples, whereas those of $\varepsilon = 0$ as perfect samples.

Let us consider a weakly absorbing thin film deposited onto a thick transparent substrate. Under typical experimental conditions the interference takes place only within the film and the interference between the incident light beam and the reflected beam from the backside of the substrate can be disregarded [1]. The compex refractive index of the film is denoted by $\hat{n} = n_2(1+i\varkappa)$ and generally the indices 1, 2 and 3 stand for air, film and substrate, respectively. No special attention will be paid for free films (thin crystalline sheets, without substrate) since all the expressions below are also valid for those by substituing $n_3 = n_1 = 1$.

The transmittance of the sample can be given by the expression [1]:

$$T = \frac{T_f \cdot T_s}{1 - R_f \cdot R_s}, \tag{2}$$

where T_f, T_s and R_f, R_s denote the transmission and reflexion coefficients of the film and the substrate, respectively. The corresponding expressions are derived in many monographs, for example in [7—9]. Our treatment is based mainly on [7].

Substituting the transmission and reflexion coefficients into Eq. (2) we obtain

$$T = \frac{P}{Q_1 e^{\alpha_0 \varkappa} + Q_2 e^{-\alpha_0 \varkappa} + M \cos(\alpha + \delta)}, \tag{3}$$

where the following abbreviations are introduced:

$$P = (1-\varrho_{12}^2)(1-\varrho_{23}^2)(1+\varkappa^2)T_s, \tag{4a}$$

$$Q_1 = (1-R_s)\cdot \varrho_{12}^2, \tag{4b}$$

$$Q_2 = (\varrho_{12}^2-R_s)\cdot \varrho_{23}^2, \tag{4c}$$

$$M = 2|\varrho_{12}\cdot \varrho_{23}|\cdot(1+R_s^2-2R_s\cos\varphi_{12})^{1/2}\cdot \operatorname{sgn}(n_3-n_2), \tag{4d}$$

$$\operatorname{tg}\delta = \frac{R_s\sin 2\varphi_{12}}{1-R_s\cos 2\varphi_{12}}, \tag{4e}$$

$$\alpha = \alpha_0+\varphi_{12}+\varphi_{23} = \frac{4\pi n_2}{\lambda}d+\varphi_{12}+\varphi_{23} = \gamma d+\varphi_{12}+\varphi_{23}, \tag{4f}$$

$$R_s = 1-T_s = \left(\frac{n_3-1}{n_3+1}\right)^2. \tag{4g}$$

All the other notations are the same as in *Ref.* [7]:

$$\varrho_i^2 = \frac{(n_i-n_j)^2+n_2^2\varkappa^2}{(n_i+n_j)^2+n_2^2\varkappa^2}, \tag{5a}$$

$$\operatorname{tg}\varphi_{ij} = \frac{2n_in_j\varkappa}{n_2^2(2+\varkappa^2)-(n_i^2+n_j^2)}. \tag{5b}$$

We note in passing that only the denominator of Eq. (3) is thickness dependent.

The average value of the transmission can be obtained by inserting Eq. (3) into Eq. (5). For practical purposes an expression is needed which is as simple as possible, with an accurancy somewhat better than that of the measurements. It would be useful if the transmittance of the corresponding perfect layer having the same thickness as the average thickness of the real film could be calculated from the measured values. In this case the method, wich has been elaborated for the determination of the optical constants of perfect layers should remain applicable with a slight modification for real layers, too. Such an expression can be obtained by expanding the integrand of Eq. (1) with regard to ξ. The result takes the following form:

$$T_{av} = T_0[1+K(n_2, \varkappa, n_3, \varepsilon)] = T_0\left[1+N\cdot\sum_n \frac{h^{2n}}{(2n+1)!}\,\partial_\xi^{2n} N(\xi)\Big|_{\xi=0}\right], \tag{6}$$

where T_0 is the transmission of the film of thickness d_0, and $N(\xi)$ denotes the denominator of Eq. (3) with $d=d_0+\xi$. The correction function $K(n_2, \varkappa, n_3, \varepsilon)$ is expected to be small, ($K\ll 1$). Utility of Eq. (6) depends on the fact, how many terms of the series have to be considered to obtain an acceptable approximation.

(ii) Mathematical considerations. Let us consider the following derivate:

$$\partial N(\xi)|_{\xi=0} = \gamma^k\varkappa^k Q_1 e^{\alpha_0\varkappa}+(-\gamma\varkappa)^k e^{-\alpha_0\varkappa}+(-1)^{\mathrm{INT}\left(\frac{k+1}{2}\right)}\gamma^k\cdot M\cdot\begin{Bmatrix}\cos\\ \sin\end{Bmatrix}(\alpha+\sigma).$$

By combining this expression with Eq. (6) it can be seen that the quantity γ occurs in each case in the product $h\gamma=\alpha_0\varepsilon$. Taking into account that $\alpha_0=m\pi$, with $m=1, 2, \ldots$, at the wavelengths corresponding to the extrema of the transmittance, we conclude that even for $\varepsilon\ll1$ the convergence of the expansion (6) may be rather slow. For weakly absorbing materials $\varkappa\ll1$ therefore the derivates of the exponential functions become soon negligible but the terms, which ordiginate from the trigonometric part of denominator decrease tiresome-slowly. In order to get a practicable expression from Eq. (6), we need to sum the trigonometric terms in closed form.

To get an convenient overlook of the terms in expansion (6), let us arrange the derivates of $N^{-1}(\xi)$ as follows:

$$(N^{-1})' = -\frac{N'}{N^2},$$

$$(N^{-1})'' = -\frac{N''}{N^2}+\frac{2N'^2}{N^3},$$

$$(N^{-1})''' = -\frac{N'''}{N^2}+\frac{6N''\cdot N'}{N^3}-\frac{3N'^3}{N^4},$$

$$(N^{-1})'''' = -\frac{N''''}{N^2}+\frac{8N'''\cdot N'+6N''^2}{N^3}-\frac{36N''\cdot N'^2}{N^4}+\frac{24N'^4}{N^5}. \tag{7}$$

During our subsequent considerations we shall use indices referring to the rows and columns of this table.

Terms of the q-th row and r-th column of the table wich are not multiplied by powers of $\varkappa$ can be summed as

$$M^r N^{-(r+1)}\gamma^q \sum_p C_p^r(q)\cdot\cos p\cdot(\alpha+\delta), \quad \text{(for even } q\text{'s)} \tag{8a}$$

or

$$M^r N^{-(r+1)}\gamma^q \sum_p S_p^r(q)\cdot\sin p\cdot(\alpha+\delta), \quad \text{(for odd } q\text{'s).} \tag{8b}$$

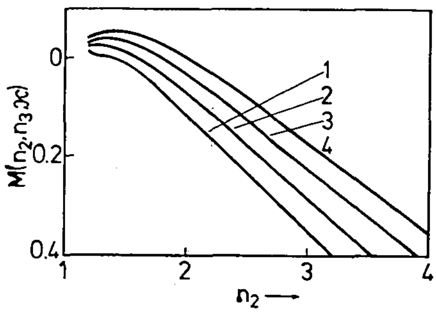

Fig. 1. The function $M(n_2, n_3, \varkappa)$ (at $\varkappa=0$ and at $n_3=1.4$, 1.6, 1.8 and 2 for curve 1, 2, 3 and 4, respectively) for estimation of the order of terms in Eq. (11)

The sum runs over $p=r-2k\geqq0$, with $k=0, 1, 2, \ldots$. The subsequent columns in the table are multiplied by increasing power of η, where $\eta=M/N$, and $\eta\ll1$, therefore they give a decreasing contribution to the correction function. It will be useful to rearrange the terms of the expansion according to the powers of η and $\varkappa$. For weakly absorbing materials $N\approx1$, therefore knowing the value of $M(n_2, \varkappa, n_3)$ the contributions of different terms can be estimated. The values of M are plotted against n_2 for $\varkappa=0$ and at different n_3 in Fig. 1.

The terms of each column can be

summed by deducing a generalised fomula for each coefficient of Eq. (8a) and (8b). It can easily be seen that

$$S_1^1(2n+1) = (-1)^n \quad \text{and} \quad C_1^1(2n) = (-1)^{n+1},$$

and so the term of the highest value in the correction function can be expressed as:

$$\eta \cos(\alpha+\delta) \cdot \sum_{n=1}^{\infty} \frac{(-1)^{n+1}(\alpha_0 \varepsilon)^{2n}}{(2n+1)!} = \eta \varphi(\alpha_0 \varepsilon) \cdot \cos \alpha^*,$$

where $\alpha^* = \alpha + \delta$ and the function $\varphi(x)$ is introduced

$$\varphi(x) = \frac{x - \sin x}{x}. \tag{9}$$

Any coefficient of the expression standing in any row of the second column can be calculated from the coefficients of the first and second column standing in the previous row. After some algebra we obtain:

$$C_0^2(2n+2) = S_1^1(2n+1),$$
$$C_2^2(2n+2) = 2 \cdot S_2^2(2n+1) - S_1^1(2n+1),$$
$$S_2^2(2n+1) = 2 \cdot C_2^2(2n) + C_1^1(2n).$$

This set of equations leeds to recursion formulae and finally we get:

$$C_0^2(2n) = (-1)^{n-1},$$
$$C_2^2(2n) = (-1)^n \cdot (2^{2n-1} - 1),$$
$$S_2^2(2n+1) = (-1)^{n+1}(2^{2n} - 1).$$

The summation over all the terms in the second column yields the following contribution to the correction function:

$$\eta^2 \left\{ \left[\varphi(\alpha_0 \varepsilon) - \frac{1}{2} \varphi(2\alpha_0 \varepsilon) \right] \cdot \cos 2\alpha + \varphi(\alpha_0 \varepsilon) \right\}.$$

Coefficients of the trigonometric functions of the 3rd and 4th rows were determined by similar procedure as the previous one and the corresponding expressions are listed below:

$$C_3^3(2n) = (-1)^{n+1} \cdot 4^{-1}(9^n - 3 \cdot 4^n + 3),$$
$$C_1^3(2n) = (-1)^n \cdot 3 \cdot 4^{-1} \cdot (4^n - 4),$$
$$S_3^3(2n+1) = (-1)^n \cdot 3 \cdot 4^{-1} \cdot (9^n - 2^{2n+1} + 1),$$
$$S_1^3(2n+1) = (-1)^{n+1} \cdot 3 \cdot 2^{-1}(4^n - 1),$$
$$C_4^4(2n) = (-1)^n \cdot 8^{-1} \cdot (16^n - 4 \cdot 9^n + 6 \cdot 4^n - 4),$$
$$C_2^4(2n) = (-1)^{n+1} \cdot (9^n - 4 \cdot 4^n + 7),$$
$$C_0^4(2n) = (-1)^n 3 \cdot 4^{-1} \cdot (4^n - 4).$$

The result of the summation over the terms in these columns is expressed in Eq. (11) as a function of $\varphi(x)$, again. The graph of $\varphi(x)$ can be seen in Fig. 2.

The mixed terms in the expansion wich depend on both η and $\varkappa$ are also collected as increasing power of η. The $(\eta \cdot \varkappa)$ type terms originate from the second column, where the numerator can be expressed as

$$\sum_{k=1}^{q-1} \binom{q}{k} \cdot (\partial^k N) \cdot (\partial^{q-k} N). \tag{10}$$

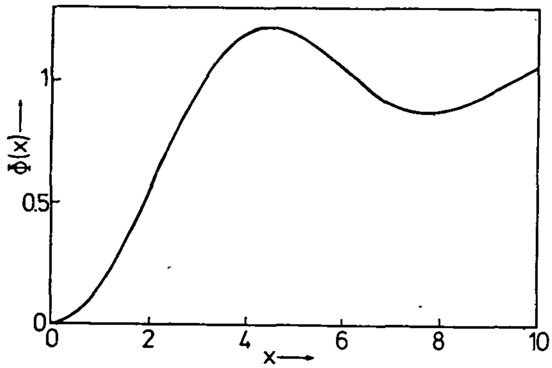

Fig. 2. The function $\varphi(x)$ for the estimation the error according to Eq. (12)

The index q refers to the row of the table, again. Since only the terms of the first derivate of $N(\xi)$ appear in the sum the summation can readily be performed. The terms multiplied by $\eta^2\varkappa$ originate from the third row. The previously calculated values of $C_p^r(q)$ and $S_p^r(q)$ were utilized for facilitation of the collection. Finally, the correction function defined by Eq. (6) can be written as:

$$\begin{aligned}
K(n_2, \varkappa, n_3, \varepsilon) &= \eta\varphi(\beta)\cdot\cos\alpha^* + \eta^2\{\varphi(\beta) + [\varphi(\beta) + 0.5\cdot\varphi(2\beta)]\cdot\cos 2\alpha^*\} + \\
&+ \eta^3\{[3\varphi(\beta) - 0.75\varphi(2\beta)]\cos\alpha^* + [0.75\varphi(\beta) - 0.75\varphi(2\beta) + 0.25\varphi(3\beta)]\cos 3\alpha^*\} + \\
&+ \eta^4\{3\varphi(\beta) - 0.75\varphi(2\beta)] + 0.5[7\varphi(\beta) - 4\varphi(2\beta) + \varphi(3\beta)]\cdot\cos 2\alpha^* + \\
&+ 0.125[4\varphi(\beta) - 6\varphi(2\beta) + 4\varphi(3\beta) - 4\varphi(4\beta)]\cdot\cos 4\alpha^*\} + \\
&+ 2\eta\varkappa N^{-1}[\varphi(\beta) + \cos\beta - 1]\cdot(Q_1 e^{\alpha_0\varkappa} - Q_2 e^{-\alpha_0\varkappa})\cdot\sin\alpha^* - \\
&+ 0.75\eta^2\varkappa N^{-1}[4\varphi(\beta) + 4\cos\beta - 3 - \cos 2\beta - \varphi(2\beta)]\cdot(Q_1 e^{\alpha_0\varkappa} - Q_2 e^{-\alpha_0\varkappa})\sin 2\alpha^* + \\
&+ \varkappa^2\beta^2 N^{-2}[(2Q_1^2 + \varkappa^2\beta^2)e^{2\alpha_0\varkappa} - NQ_1 e^{\alpha_0\varkappa} - 4Q_1Q_2 + NQ_2 e^{-\alpha_0\varkappa}],
\end{aligned} \tag{11}$$

where the abbreviation $\beta = \alpha_0\varepsilon$ is used. With the assumption that $\max \varkappa \leqq \eta^2$, as well as Q_2 has the same order as η^2, the neglected terms in Eq. (11) are in the order of η^5. For practical calculations many terms of this expression can be neglected depending on the values of n_2, n_3, $\varkappa$ and ε. At the extrema of the transmittance further simplification is possible since $\alpha^* \approx \pi$, and the mixed terms cancel.

It is interesting to note that the first term in the correction function has a simple physical meaning stating that the thickness uneveness should fade the interference pattern. Restricting the approximation only to the first term and writting M instead of η we have:

$$T_{av} = P \cdot \frac{1 + M\varphi(\beta)\cos\alpha^*}{e^{\alpha_0 x} + M\cos\alpha^*}. \tag{12}$$

This approximation showes that the difference between the maxima and the minima of the fraction (12) decreases with increasing β (increasing ε or decreasing wavelength). From Eq. (12) it can be concluded that the difference in transmittance of real and perfect films tends to zero as β^2, since for small arguments $\varphi(\beta) \approx \beta^2/6$.

The requirement of the layer eveness *i.e.* perfect samples can be quantitated on basis of this simplified approximation (12). The largest deviation from the average thickness must be less than $(\sigma/M)^{1/2} \cdot \lambda/4\pi n$, where σ is the permitted error of transmittance. For a sample of $n_2=2$, $n_3=1{,}5$ and an expected value of $\sigma \sim 10^{-3}$ we obtain $h < \lambda/200$.

Comparison with computer generated spectra

The validity of the approximation developed here was checked by computer simulation. Transmission spectra of uneven films were calculated by numeric integration of Eq. (3). Typical results are illustrated in Fig. 3. The parameter values

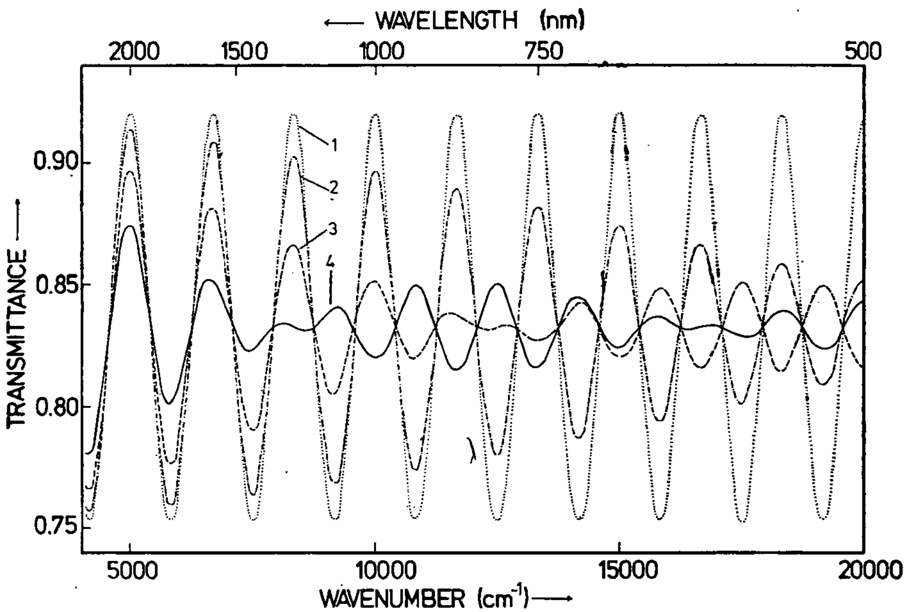

Fig. 3. Computer simulated spectra for films of an average thickness of 1500 nm (see in text). The highest deviation of the thickness from its average value are 0 for curve 1, 50 nm for curve 2, 100 nm for curve 3 and 150 nm for curve 4

$n_2=2$, $n_3=1.5$ and $\varkappa=0$, as well as the average layer thickness are the same for each spectrum; the displayed set of curves was generated by varying the layer uneveness parameter ε. The first curve (curve 1 in Fig. 3) displays the transmission pattern of an even film, whenever the other curves are characterized by a thickness deviation of 50nm, 100nm, and 150nm, respectively. Similar spectra were generated for $\varkappa=0.01$ and for the wavelength dependence of $\varkappa$, too. The simulated spectra well agree with the calculated curves using the correction function given in Eq. (11). Using only two terms from Eq. (11) the error was found to be less than $3 \cdot 10^{-3}$ and if the third term is included the error is further reduced below 10^{-4}.

The computer simulated spectra of uneven films were evaluated by the method, described in Ref. [4, 5], wich is valid for perfect films only. The obtained values of the refractive index differ considerably from those, used for simulation. The calculated thickness of the film displayed an apparent wavelength dependence. As seen from Eq. (12) the error in thickness and the refractive index increases with decreasing wavelength.

Fig. 3 illustrates an interesting phenomenon, what we call "*pattern inversion*". At the wavelengths, where the transmittance of the appropriate perfect film reaches its maxima, the real film shows transmittance minima in the short wavelength part of the spectra. In the long wavelength side of the spectra, on the other hand, the position of the maxima coincidences. Whenever pattern inversion occure the calculated interference order should be altered by 1, *e.g.* it changes from even into odd or vice versa. It is surprising that this phenomenon appeares already at relatively small values of $\varepsilon(\sim 0.07)$.

On the evaluation of the spectra

In the case of the perfect films one can obtain two equations from Eq. (3) with $\cos\alpha^*=1$ and $\cos\alpha^*=-1$, respectively, wich are valid for the envelop curves of the extrema of the transmittance. Utilising the extrema condition of the interference, we have three equations for 3 unknow parameters n_2, $\varkappa$, d. The problem is overdetermined since the thickness of the film can be obtained from each extremum. This condition permits the determination of the fourth unknown parameter ε, in the case of real films. We have found a value of ε wich minimizes the apparent wavelength dependence of the thickness.

The determination of the optical constants of real films can be done as follows: with any arbitrarily chosen ε and estimated n_2 the value of the correction function can be obtained in the zeroth order approximation. Now, Eq. (6) yields the approximate transmittance of the corresponding perfect film and the procedure outlined in [1—5] can be used for the determination of the optical constants. The results of the first calculation yield a better approximation of the correction function and we have to iterate until the change of the optical constants remain less than a preset value. The method is self consistent for every ε, but the apparent wavelength dependence of the calculated thickness is a good criteria for choosing the true ε value. After 2 or 3 arbitrarily choosen ε an appropriate value of ε can be found by interpolation.

Concluding remarks

The thickness uneveness of the film has a drastic effect on the interference structure of the transmission spectrum. The derived expression describes well the properties of the model, however the model involves a strong assumption concerning the thickness variation in the measuring area of the sample. Decreasing the diameter of the measuring light beam the conditions of the model are better fulfiled. At the same time the thickness uneveness decreases and the measured transmittance better approaches the spectra of the perfect film and, therefore the succesive approximation described above converges more rapidly.

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ВЛИЯНИЕ НЕОДНОРОДНОСТИ ТОЛЬЩИНЫ ПЛЕНОК НА ИНТЕРФЕРЕНЦИОННУЮ СТРУКТУРУ ОПТИЧЕСКОГО ТРАНСМИССИОННОГО СПЕКТРА ТОНКИХ ПЛЕНОК

М. И. Терек

Получено аналитическое выражение для трансмиссионного спектра тонких пленок с медленно меняющейся толщиной (ТПМТ). Проведено сравнение спектров полученных моделированием на вычислительной машине с исползованием аналитический формулоы- Выведенное выражение позволяет оценить ошибки от неоднородности толщины пленки. Действие распространено метода на определение оптически хконстант на основе интерферен. ционной структуры спектров на ТПМТ.