

A simple method for the determination of the optical constants n , k and the thickness of a weakly absorbing thin film

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La cavité II peut s'ouvrir, contrairement à la cavité I, ce qui permet de la retirer d'un ensemble expérimental sans avoir à la démonter. Par contre son réglage est beaucoup plus délicat et la luminosité maximum plus difficile à obtenir. Dans nos expériences nous avons préféré, pour sa facilité d'utilisation, la cavité I à la cavité II pour les travaux de routine.

4 Conclusion

Les cavités que nous avons conçues ont des performances, tant du point de vue luminosité que domaine de pression, supérieures aux appareils existants. Leur grande efficacité d'excitation permet de les faire travailler dans des conditions expérimentales difficiles. La stabilité de leur décharge en fait des sources très intéressantes de radicaux libres pour des études cinétiques. La cavité I est particulièrement intéressante en ce sens.

Remerciements

Ce travail a été effectué dans le laboratoire de M le Professeur P Goudmand. Nous tenons à le remercier pour tous les conseils qu'il a bien voulu nous donner et pour l'attention bienveillante qu'il a porté à nos efforts.

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A simple method for the determination of the optical constants n , k and the thickness of a weakly absorbing thin film

J C Manificier, J Gasiot and J P Fillard

Université des Sciences et Techniques du Languedoc, Centre d'Etudes d'Electronique des Solides, associé au CNRS, Place Eugene Bataillon, 34060 Montpellier, France

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Abstract We propose a new calculation following traditional methods for deducing optical constants and thickness from the fringe pattern of the transmission spectrum of a thin transparent dielectric film surrounded by non-absorbing media. The particular interest of this method, apart from its easiness, is that it makes a directly programmable calculation possible; the accuracy is of the same order as for the iteration method.

1 Introduction

The measurement of the transmission T of light through a parallel-faced dielectric film in the region of transparency is sufficient to determine the real and imaginary parts of the complex refractive index $\eta = n - ik$, as well as the thickness t . Both Hall and Ferguson (1955) and Lyashenko and Miloslavskii (1964) developed a method using successive approximations and interpolations to calculate these three quantities. We propose a similar method of analysis in the same range of applicability and precision. Our method can be distinguished from that of Lyashenko and Miloslavski (1964) in two ways: firstly data handling, calculation and computation are easier, and secondly it gives an explicit expression for n , k , t . The accuracy for n and t will be emphasized.

2 Theory

Figure 1 represents a thin film with a complex refractive index $\eta = n - ik$, bounded by two transparent media with refractive

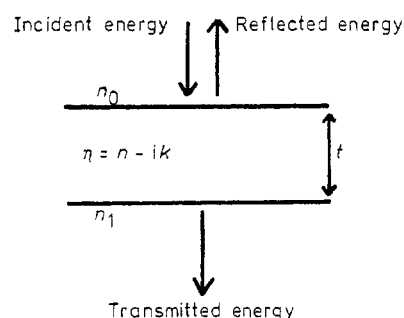


Figure 1 Reflection and transmission of light by a single film

indices, n_0 and n_1 . Considering a unit amplitude for the incident light, in the case of normal incidence the amplitude of the transmitted wave is given by

$$A = \frac{t_1 t_2 \exp(-2\pi i \eta t / \lambda)}{1 + r_1 r_2 \exp(-4\pi i \eta t / \lambda)} \quad (1)$$

in which t_1, t_2, r_1, r_2 are the transmission and reflection coefficients at the front and rear faces (Heavens 1965). The transmission of the layer is given by

$$T = \frac{n_1}{n_0} |A|^2. \quad (2)$$

The exact expression is given in the appendix. In the case of weak absorption (see appendix) with $k^2 \ll (n - n_0)^2$ and $k^2 \ll (n - n_1)^2$,

$$T = \frac{16n_0 n_1 n^2 \alpha}{C_1^2 + C_2^2 \alpha^2 + 2C_1 C_2 \alpha \cos(4\pi n t / \lambda)} \quad (3)$$

where $C_1 = (n + n_0)(n_1 + n)$, $C_2 = (n - n_0)(n_1 - n)$ and

$$\alpha = \exp(-4\pi k t / \lambda) = \exp(-K t). \quad (4)$$

K is the absorption coefficient of the thin film.

Generally, outside the region of fundamental absorption ($h\nu > EG$: thin film gap) or of the free-carrier absorption (for higher wavelengths), the dispersion of n and k is not very large. The maxima and minima of T in equation (3) occur for

$$4\pi n t / \lambda = m\pi \quad (5)$$

where m is the order number. In the usual case ($n > n_1$, corresponding to a semiconducting film on a transparent non-absorbing substrate, $C_2 < 0$), the extreme values of the transmission are given by the formulae

$$T_{\max} = 16n_0 n_1 n^2 \alpha / (C_1 + C_2 \alpha)^2 \quad (6)$$

$$T_{\min} = 16n_0 n_1 n^2 \alpha / (C_1 - C_2 \alpha)^2. \quad (7)$$

By combining equations (6) and (7), Lyashenko and Miloslavskii (1964) developed an iterative method allowing the determination of n and α and, using (4) and (5), k and t .

We propose an important simplification of this method: we consider T_{\min} and T_{\max} as continuous functions of λ through $n(\lambda)$ and $\alpha(\lambda)$. These functions which are the envelopes of the maxima $T_{\max}(\lambda)$ and the minima $T_{\min}(\lambda)$ in the transmission spectrum are shown in figure 2. The ratio of equations (6) and (7) gives

$$\alpha = \frac{C_1[1 - (T_{\max}/T_{\min})^{1/2}]}{C_2[1 + (T_{\max}/T_{\min})^{1/2}]} \quad (8)$$

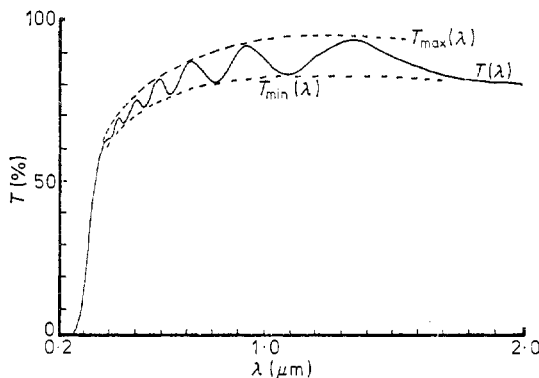


Figure 2 Typical transmission spectrum for a thin SnO_2 film of uniform thickness, $t = 0.9 \pm 0.04 \mu\text{m}$

Then, from equation (6),

$$n = [N + (N^2 - n_0^2 n_1^2)^{1/2}]^{1/2} \quad (9)$$

where

$$N = \frac{n_0^2 + n_1^2}{2} + 2n_0 n_1 \frac{T_{\max} - T_{\min}}{T_{\max} T_{\min}}.$$

Equation (9) shows that n is explicitly determined from T_{\max} , T_{\min} , n_1 and n_0 at the same wavelength.

Knowing n , we can determine α from equation (8). The thickness t of the layer can be calculated from two maxima or minima using equation (5):

$$t = \frac{M \lambda_1 \lambda_2}{2(n(\lambda_1) \lambda_2 - n(\lambda_2) \lambda_1)} \quad (10)$$

where M is the number of oscillations between the two extrema ($M = 1$ between two consecutive maxima or minima); $\lambda_1, n(\lambda_1)$ and $\lambda_2, n(\lambda_2)$ are the corresponding wavelengths and indices of refraction. Knowing t and α we are able to calculate the extinction coefficient k from equation (4). It is worthwhile noting that expressions (8), (9) and (10) can be easily calculated using a programmable pocket calculator.

3 Precision of the method and experimental precautions

The relative error was determined using equation (11) obtained by combining equations (8) and (6):

$$\frac{n^2}{C_1 C_2} = \frac{T_{\min} T_{\max}}{4n_0 n_1 (T_{\min} - T_{\max})} \quad (11)$$

giving

$$f(n, n_0, n_1) \frac{dn}{n} = \left(\frac{dT_{\min}}{T_{\min}} \right) \left(\frac{T_{\max}}{T_{\max} - T_{\min}} \right) - \left(\frac{dT_{\max}}{T_{\max}} \right) \left(\frac{T_{\min}}{T_{\max} - T_{\min}} \right) \quad (12)$$

and

$$f(n, n_0, n_1) = - \frac{2(n^2 - n_0 n_1)(n^2 + n_0 n_1)}{(n^2 - n_0^2)(n^2 - n_1^2)}.$$

We assumed: (i) that we could neglect the comparatively insignificant error in n_1 , which is usually the case for a glass substrate of known index of refraction, and (ii) that the errors for the two envelopes $T_{\max}(\lambda)$ and $T_{\min}(\lambda)$ are non-correlated. We then obtained

$$\frac{\Delta n}{n} = \frac{\Delta T}{T} \left(\frac{T_{\max} + T_{\min}}{T_{\max} - T_{\min}} \right) \frac{1}{|f(n, n_0, n_1)|} \quad (13)$$

with $\Delta T/T$ being the relative precision of measurements ($\Delta T/T = \Delta T_{\min}/T_{\min} = \Delta T_{\max}/T_{\max}$).

The function $f(n, n_0, n_1)$ has been plotted on figure 3 for $n_0 = 1$ and for two particular values of n_1 , $n_1 = 1.51$ and $n_1 = 1.6$, corresponding respectively to the two extreme values for a conventional glass substrate. Equation (12) shows that due to the presence of the ratio $(T_{\max} + T_{\min})/(T_{\max} - T_{\min})$ the accuracy is strongly affected when the amplitude of oscillations is weak. A necessary condition for a good fringe pattern is that the difference between n and n_1 should be as great as possible. Similarly, from equation (10), we obtain

$$\frac{dt}{t} = d\lambda \frac{n(\lambda_1) \lambda_2^2 - n(\lambda_2) \lambda_1^2}{\lambda_1 \lambda_2 (n(\lambda_1) \lambda_2 - n(\lambda_2) \lambda_1)} + dn(\lambda_2) \frac{\lambda_1}{n(\lambda_1) \lambda_2 - n(\lambda_2) \lambda_1} - dn(\lambda_1) \frac{\lambda_2}{n(\lambda_1) \lambda_2 - n(\lambda_2) \lambda_1}. \quad (14)$$

This can usually be simplified in the case of a weak dispersion of n ($n(\lambda_1) \simeq n(\lambda_2)$, $dn(\lambda_1) \simeq dn(\lambda_2)$) and leads to

$$\Delta t/t \simeq \Delta \lambda (\lambda_1 + \lambda_2) / \lambda_1 \lambda_2 + \Delta n/n. \quad (15)$$

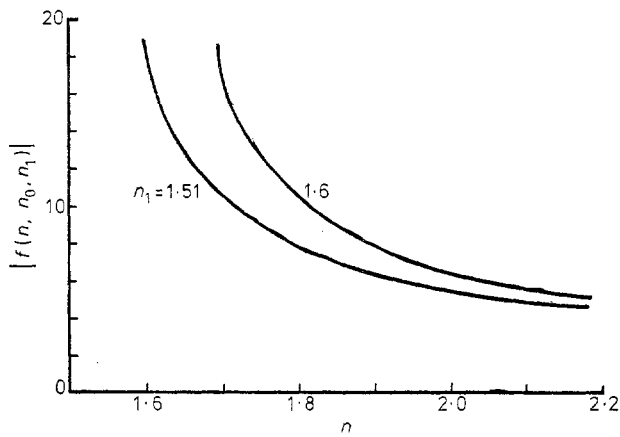


Figure 3 Variation of $|f(n, n_0, n_1)|$ with n for two values of n_1 . $n_1 = 1.51$ and $n_1 = 1.6$; $n_0 = 1$ in both cases

On the other hand, if $dn(\lambda_1)/n(\lambda_1) \neq dn(\lambda_2)/n(\lambda_2)$, which is the case if the number of minima T_{\min} and maxima T_{\max} is low, we obtain

$$\frac{\Delta t}{t} \simeq \Delta \lambda \frac{\lambda_1 + \lambda_2}{\lambda_1 \lambda_2} + \frac{\Delta n}{n} |(\lambda_2 + \lambda_1)/(\lambda_2 - \lambda_1)|. \quad (16)$$

It can then be seen that the error in t can be very important due to the coefficient $(\lambda_2 + \lambda_1)/(\lambda_2 - \lambda_1)$.

Moreover, some experimental care should be taken in the application of the above method: (i) the effective bandwidth of the spectrophotometer should be kept smaller than the half-width of the interference maximum when using a $0.2\text{--}2\text{ }\mu\text{m}$ spectrophotometer: this leads to an upper limit for the thickness of the film of the order of $10\text{ }\mu\text{m}$; (ii) the sample must be homogeneous and parallel-faced; (iii) the variation of n and k with the wavelength should be small; this condition fails in the vicinity of the fundamental absorption short-wavelength region.

This method was applied to SnO_2 films deposited on a glass substrate by a spray (or vacuum evaporation) technique (Manificier *et al* 1975, 1976). It is shown from this work that our method is much more convenient and useful and leads to the same results as the iterative method of Lyashenko and Miloslavskii (1964). Typically, using an Aminco DW-Z uv/vis and a Beckman DK2A spectrophotometer, with $\Delta T/T \simeq 1\%$ we obtained $\Delta n/n = 2\text{--}5\%$ (n between 1.8 and 2.2). The accuracy in t is a critical function of the definition of the maxima and minima. In the best cases the precision is of the order of 4%.

In conclusion, we can say that this method provides a very simple way of calculating n , k and t with a precision in the same range as that of the iterative method of Lyashenko and Miloslavskii (1964), in the case of a weakly absorbing film surrounded by non-absorbing media.

Appendix

The exact expression is given by the following equation:

$$T = \frac{16n_0n_1(n^2 + k^2)\alpha}{A + B\alpha^2 + 2\alpha[C \cos(4\pi nt/\lambda) + D \sin(4\pi nt/\lambda)]} \quad (17)$$

with

$$\begin{aligned} A &= [(n + n_0)^2 + k^2][(n + n_1)^2 + k^2] \\ B &= [(n - n_0)^2 + k^2][(n - n_1)^2 + k^2] \\ C &= -(n^2 - n_0^2 + k^2)(n^2 - n_1^2 + k^2) + 4k^2n_0n_1 \\ D &= 2kn_1(n^2 - n_0^2 + k^2) + 2kn_0(n^2 - n_1^2 + k^2). \end{aligned}$$

Equation (17) is the same as equation (1) in the paper by Hall and Ferguson (1955). With the conditions

$$k^2 \ll (n - n_0)^2 \quad (18)$$

$$k^2 \ll (n - n_1)^2 \quad (19)$$

which are satisfied if the absorption is weak enough, we obtain

$$\begin{aligned} A &= (n + n_0)^2(n + n_1)^2 \\ B &= (n - n_0)^2(n - n_1)^2 \\ C &= -(n^2 - n_0^2)(n^2 - n_1^2) + 4k^2n_0n_1 \\ D &= 2kn_1(n^2 - n_0^2) + 2kn_0(n^2 - n_1^2). \end{aligned}$$

It is easy to show that $D \ll C$, so, near a maximum or minimum for T (cf equation (5)) we can neglect $D \sin(4\pi nt/\lambda)$. Moreover, in most practical cases $n > n_0$ and $n > n_1$, leading to

$$4n_0n_1/(n + n_0)(n + n_1) < 1$$

and so to

$$4k^2n_0n_1 \ll (n^2 - n_0^2)(n^2 - n_1^2).$$

If

$$k^2 \ll (n - n_0)(n - n_1) \quad (20)$$

then we obtain equation (3) in the text.

For example, in the case of SnO_2 on a glass substrate (Manificier *et al* 1976): $n = 2$; $n_0 = 1$; $n_1 = 1.5$; then (19) is the more restrictive condition and leads to $k^2 \ll 0.25$. In practice, $k < 0.1$ would be a reasonable limit.

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