Determination of surface roughness and optical constants of inhomogeneous amorphous silicon films

1984 J. Phys. E: Sci. Instrum. 17 896

(http://iopscience.iop.org/0022-3735/17/10/023)

Determination of surface roughness and optical constants of inhomogeneous amorphous silicon films

R Swanepoel
Department of Physics, Rand Afrikaans University, Johannesburg, South Africa

Received 16 February 1984

Abstract. Inhomogeneities in thin films have a large influence on the optical transmission spectrum. If not corrected for, this may lead to too large calculated values for the absorption coefficient or the apparent presence of an absorption band tail as well as serious errors in the values of refractive index and thickness. The effects of thickness variation, surface roughness and variation in refractive index on the transmission spectrum are analysed. Analytical expressions are derived from which all the optical constants of an inhomogeneous film can be calculated from the transmission spectrum.

1. Introduction

The suitability of hydrogenated amorphous silicon (α-Si:H) for photovoltaic devices has stimulated great interest in the preparation and characterisation of this material. The optical constants $n(\lambda)$ and $\alpha(\lambda)$ are of particular significance in optoelectronic devices. In a previous article (Swanepoel 1983, from now on referred to as the previous work) it was shown that the assumption of an infinite substrate when analysing the transmission spectrum leads to errors in determining $\alpha(\lambda)$ and methods were given for calculating $n(\lambda)$, the thickness d and $\alpha(\lambda)$ accurately from the transmission spectrum. It was emphasised, however, that the theory applies to uniform films with uniform thickness.

Under certain conditions of preparation α-Si:H films can be in the form of a variation in thickness or a variation in composition and structure that may cause a variation in n. The presence of inhomogeneities drastically changes the transmission spectrum and the formulae used for uniform films are not valid any more.

The optical properties of thin films have been investigated by many workers (Heavens 1964, Clark 1980, Martin *et al* 1983) using the approximation of an infinite substrate. Inhomogeneity in the form of surface roughness was also investigated for InAs (Szczyrbowski and Czapla 1977) and it was found that the effect of surface roughness has a large influence on the calculation of α.

In the present paper the effect of thickness variation on the transmission spectrum is analysed and methods presented to calculate the thickness variation, average thickness, $n(\lambda)$ and $\alpha(\lambda)$ from an experimental transmission spectrum of a non-uniform film of α-Si:H. The effect of variation in n is also discussed. Using these methods it is possible to calculate the optical constants to an accuracy of the order of 1% from a spectrum that otherwise would have been useless.

2. Theory

In the previous work the following case was considered. A thin homogeneous film with uniform thickness d and complex refractive index $\boldsymbol{n} = n - \mathrm{i}k$ or absorption coefficient α. The film is on a transparent substrate with refractive index s. The substrate is considered to be perfectly smooth, but thick enough so that in practice the planes are not perfectly parallel so that all interference effects due to the substrate are destroyed, according to equation (13). The system is surrounded by air with refractive index $n_0 = 1$. Taking all the multiple reflections at the three interfaces into account, it was shown that in the case $k^2 \ll n^2$ the expression for the transmission T for normal incidence is given by

$$T = \frac{Ax}{B - Cx\cos\varphi + Dx^2} \tag{1}$$

where

$$A = 16n^2 s \tag{2a}$$

$$B = (n+1)^3(n+s^2) \tag{2b}$$

$$C = 2(n^2-1)(n^2-s^2) \tag{2c}$$

$$D = (n-1)^3(n-s^2) \tag{2d}$$

$$\varphi = 4\pi nd/\lambda \tag{2e}$$

$$x = \exp(-\alpha d) \tag{2f}$$

$$k = \alpha\lambda/4\pi. \tag{2g}$$

The envelopes around the interference maxima T_{M0} and minima T_{m0} were considered to be continuous functions of λ, where

$$T_{\mathrm{M0}} = \frac{Ax}{B - Cx + Dx^2} \tag{3a}$$

$$T_{\mathrm{m0}} = \frac{Ax}{B + Cx + Dx^2}. \tag{3b}$$

The real part of the refractive index n can now be calculated at any wavelength using the formula

$$n = [N + (N^2 - s^2)^{1/2}]^{1/2} \tag{4}$$

where

$$N = 2s\,\frac{T_{\mathrm{M0}} - T_{\mathrm{m0}}}{T_{\mathrm{M0}}T_{\mathrm{m0}}} + \frac{s^2+1}{2}.$$

It was emphasised that the formulae are valid only for films with uniform thickness and (3) and (4) are *not* valid for films with non-uniform thickness.

The case now considered is shown schematically in figure 1(a). It is assumed that the thickness varies linearly over the illuminated area so that the thickness can be expressed in the following form:

$$d = \bar{d} \pm \Delta d. \tag{5}$$

Δd refers to actual variation in thickness from the average thickness $\bar{d}$ as shown in figure 1(a) and must not be confused with a standard deviation of calculated values or a RMS deviation used by other workers (Szczyrbowski and Czapla 1977). The experimental measurements must thus be done in such a way that the same area is illuminated at all wavelengths.

The theory developed below refers to a linear variation in d on a macroscopic scale of typically a few millimetres as shown in figure 1(a). It is, however, also valid for the case where several irregularities occur periodically over the illuminated area in the form of surface roughness. Three forms of surface roughness where Δd can be defined as in equation (5) are shown in figures 1(b) to 1(d). Figure 1(b) shows a triangular surface roughness and figure 1(c) a rectangular surface roughness. The meaning of

0022-3735/84/100896 + 08 \$02.25

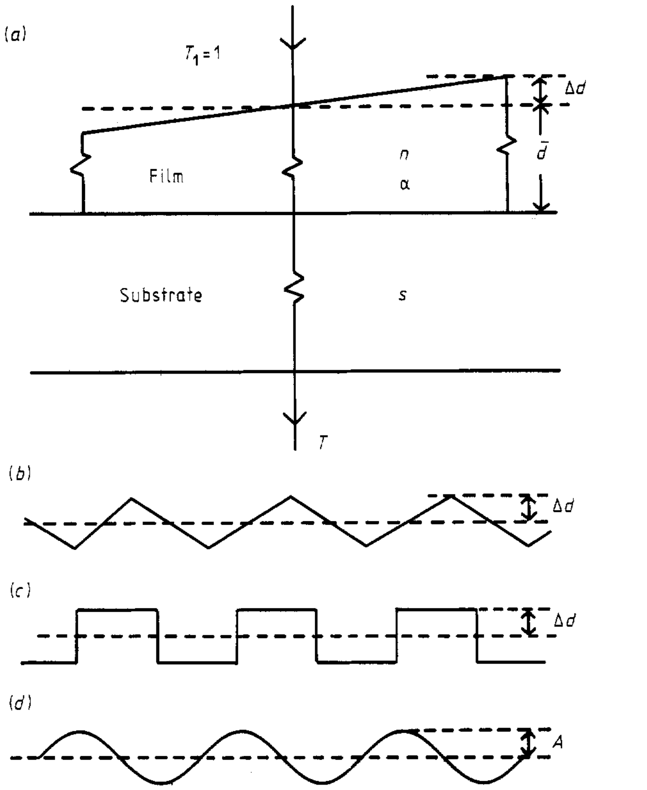

Figure 1. (*a*) System of an absorbing thin film with a variation in thickness on a thick finite transparent substrate; (*b*) triangular surface roughness; (*c*) rectangular surface roughness; (*d*) sinusoidal surface roughness.

Δd is indicated in the figures. Figure 1(*d*) shows a sinusoidal surface roughness with amplitude A and the theory can be applied putting $\Delta d = 4A/\pi$ in equation (5). More random types of surface roughness could be approximated by any of the three forms shown in figure 1, assuming almost normal incidence.

The effect of thickness variation on a typical transmission spectrum is shown in figures 2 and 3. The dotted-curve spectrum is what would have been obtained from a film with uniform thickness while the full-curve spectrum represents that of the same film but with non-uniform thickness. It can be seen that the interference pattern shrinks dramatically if the thickness is not uniform. This effect will now be investigated in detail.

In equation (1) it is assumed that s is known and λ is measured. For non-uniform films the transmission T is then a function of the following form:

$$T = T(\Delta d, n, \bar{d}, x). \tag{6}$$

Another equation is still available for the extremes of the interference fringes

$$2n\bar{d} = m\lambda \tag{7}$$

where m is the order number, integer or half integer.

Thus we have only two equations with five unknown parameters Δd, n, $\bar{d}$, x and m which cannot be solved for equation (6) in the form of equation (1) and further treatment of equation (1) is necessary.

2.1. The transparent region

In the transparent region $\alpha = 0$ or $x = 1$ and equation (1) becomes

$$T = \frac{A}{B - C\cos\varphi + D}. \tag{8}$$

The transmission $T_{\Delta d}$ at a specific wavelength λ for the case of non-uniform thickness according to equation (5) can now be obtained by integrating equation (8):

$$T_{\Delta d} = \frac{1}{\varphi_2 - \varphi_1}\int_{\varphi_1}^{\varphi_2} \frac{A}{B - C\cos\varphi + D}\,\mathrm{d}\varphi \tag{9}$$

where

$$\varphi_1 = 4\pi n(\bar{d} - \Delta d)/\lambda \tag{9a}$$

and

$$\varphi_2 = 4\pi n(\bar{d} + \Delta d)/\lambda. \tag{9b}$$

The integral yields

$$T_{\Delta d} = \frac{\lambda}{4\pi n\,\Delta d}\,\frac{a}{(1-b^2)^{1/2}}$$
$$\times\left[\tan^{-1}\left(\frac{1+b}{(1-b^2)^{1/2}}\tan\frac{\varphi_2}{2}\right) - \tan^{-1}\left(\frac{1+b}{(1-b^2)^{1/2}}\tan\frac{\varphi_1}{2}\right)\right] \tag{10}$$

where

$$a = \frac{A}{B+D} \tag{10a}$$

and

$$b = \frac{C}{B+D}. \tag{10b}$$

The constants in equations (10*a*) and (10*b*) are as defined in equation (2) and will not be simplified further. The integration cannot be carried out from a point on one branch of the tangent curve to a point on another branch. This means that equation (10) cannot be used in the region of the minima of the interference pattern and can only be used in regions around the maxima. This usable region decreases with increasing Δd and equation (10) is thus not a useful expression.

More useful expressions can be obtained by considering the envelopes around the maxima and minima of the spectrum. These envelopes are considered to be *continuous* functions of λ and thus also of $n(\lambda)$. The expression for the maximum envelope can be obtained as follows:

From equation (9*b*) the term $\tan(\varphi_2/2)$ in equation (10) can be written as

$$\tan\frac{\varphi_2}{2} = \frac{\tan(2\pi n\bar{d}/\lambda) + \tan(2\pi n\,\Delta d/\lambda)}{1 + \tan(2\pi n\bar{d}/\lambda)\tan(2\pi n\,\Delta d/\lambda)}.$$

From equations (1) and (2*e*) it follows that at a maximum

$$\frac{2\pi n\bar{d}}{\lambda} = l\pi \qquad l = 0, 1, 2, 3, \ldots$$

and thus at a maximum

$$\tan\left(\frac{2\pi n\bar{d}}{\lambda}\right) = 0$$

and the expression for $\tan(\varphi_2/2)$ reduces to

$$\tan\frac{\varphi_2}{2} = \tan\left(\frac{2\pi n\,\Delta d}{\lambda}\right).$$

Similarly

$$\tan\frac{\varphi_1}{2} = -\tan\left(\frac{2\pi n\,\Delta d}{\lambda}\right).$$

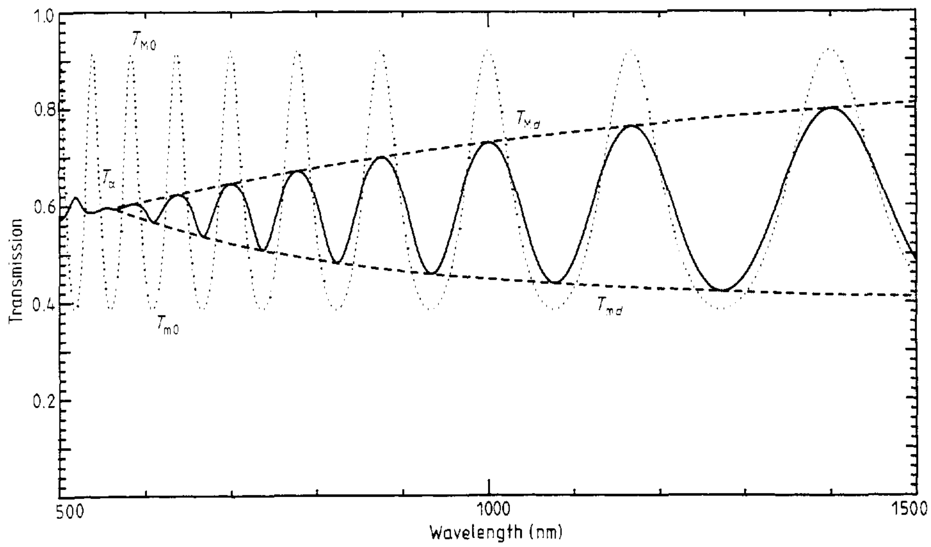

Figure 2. Simulated transmission for a transparent film with uniform thickness of 1 μm (dotted-curve spectrum) compared to that of a film with a thickness variation $\Delta d = 40$ nm (full-curve spectrum).

Substituting the above in equation (10) yields

$$T_{Md} = \frac{\lambda}{2\pi n\,\Delta d}\,\frac{a}{(1-b^2)^{1/2}}\tan^{-1}\left[\frac{1+b}{(1-b^2)^{1/2}}\tan\left(\frac{2\pi n\,\Delta d}{\lambda}\right)\right]. \quad (11)$$

The expression for the envelope around the minima can be obtained by noting that, thinking in terms of continuous envelopes, the concept of maximum changes to minimum if the sign of C is changed in equation (1). Changing the sign of C or b, according to equation (10b), in equation (11) will thus give an expression for the envelope around the minima:

$$T_{md} = \frac{\lambda}{2\pi n\,\Delta d}\,\frac{a}{(1-b^2)^{1/2}}\tan^{-1}\left[\frac{1-b}{(1-b^2)^{1/2}}\tan\left(\frac{2\pi n\,\Delta d}{\lambda}\right)\right]. \quad (12)$$

The range of validity of equations (11) and (12) is

$$0 < \Delta d < \frac{\lambda}{4n}. \quad (13)$$

The behaviour of the equations discussed so far is shown in figure 2. For illustrative purposes a film with the following properties is considered: average thickness $\bar{d} = 1000$ nm; thickness variation $\Delta d = 40$ nm; refractive index $n = 3.5$ (constant) and substrate refractive index $s = 1.51$.

The dotted curve is a plot of equation (8) and thus represents the spectrum of a film with uniform thickness. It was shown in the previous work that the maxima coincide with the transmission of the substrate alone and are given by

$$T_{M0} = T_s = \frac{2s}{s^2+1}. \quad (14)$$

The minima are given by

$$T_{m0} = \frac{4n^2 s}{n^4 + n^2(s^2+1) + s^2}. \quad (15)$$

The solid spectrum is obtained by performing the integration (9) numerically for $\Delta d = 40$ nm. In regions around the maxima where it is valid, equation (10) agrees exactly with the numerical integration.

The upper broken curve T_{Md} is a plot of equation (11) and the lower broken curve T_{md} is a plot of equation (12). At the point $\Delta d = \lambda/4n$ equations (10), (11) and (12) merge to a single point T_α, and the interference pattern is destroyed. From figure 1(a) it can be seen that this means an optical path difference of $\lambda/2$ between the thickest and thinnest parts of the film and bears a similarity to quarter-wavelength layers used for antireflection layers. At this point the transmission represents the interference-free or incoherent transmission which was shown in the previous work to be related to T_{M0} and T_{m0} according to

$$T_\alpha = (T_{M0}T_{m0})^{1/2}. \quad (16)$$

For smaller values of λ such that $\Delta d > \lambda/4n$ a second interference pattern starts to appear but, since it is not of practical importance, it will not be considered further.

The envelopes T_{Md} and T_{md} represented by equations (11) and (12) are continuous functions over the range of practical interest given by equation (13). They are *independent of* $\bar{d}$ and the shrinking of the spectrum is thus only a function of Δd. A thicker film, for example, will produce more and closer-spaced fringes but for the same Δd will always lie between the same envelopes.

Equations (11) and (12) are two independent transcendental equations with only two unknown parameters, n and Δd. They can thus be solved for experimental values using standard computer methods. The equations have one unique solution in the range given by equation (13) and there are no problems with multiple solutions as is often the case in this type of work (Cisneros *et al* 1983).

For a uniform film the departure of T_{M0} from T_s denotes the onset of absorption but in the case of a non-uniform film T_{Md} approaches T_s only in the long-wavelength limit and it decreases consistently with decreasing λ, even for the case where $\alpha = 0$. This decrease in the maxima of an experimental spectrum can mistakenly be attributed to an increase in α or to the existence of an 'absorption band tail' while the effect is actually due to inhomogeneities in the film. Szczyrbowski (Szczyrbowski and Czapla 1977) actually found that the calculated values of α decrease considerably for InAs if the effects of surface roughness are taken into account, thus confirming what is said above. It is impossible to judge from the behaviour of the maxima of the spectrum of a non-uniform film where the onset of absorption starts.

From figure 2 it can be seen that the minima have an upward trend with decreasing λ. This can never happen for a uniform film since it follows from equation (15) that T_{m0} should either stay constant or decrease with decreasing λ. When $\alpha > 0$ it causes T_{m0} to decrease even further. The effect of variation in thickness opposes this trend and any anomalous behaviour of the minima is an indication of inhomogeneities in the film, although slit width may also cause this effect as will be discussed in § 3.

2.2. *The region of weak and medium absorption*

In this region $\alpha > 0$ and the integration (9) should be done over both Δd and x. This is prohibitively difficult analytically and an approximation is to consider x to have some average value over the range of integration with respect to Δd. This approximation is an excellent one provided $\Delta d \ll \bar{d}$. The constants a and b are now redefined as follows:

$$a_x = \frac{Ax}{B + Dx^2} \tag{17a}$$

$$b_x = \frac{Cx}{B + Dx^2} \tag{17b}$$

$$x = \exp(-\alpha\bar{d}). \tag{17c}$$

The equations for the two envelopes now become

$$T_{Mx} = \frac{\lambda}{2\pi n\,\Delta d}\,\frac{a_x}{(1-b_x^2)^{1/2}} \times \tan^{-1}\left[\frac{1+b_x}{(1-b_x^2)^{1/2}}\tan\left(\frac{2\pi n\,\Delta d}{\lambda}\right)\right] \tag{18}$$

$$T_{mx} = \frac{\lambda}{2\pi n\,\Delta d}\,\frac{a_x}{(1-b_x^2)^{1/2}} \times \tan^{-1}\left[\frac{1-b_x}{(1-b_x^2)^{1/2}}\tan\left(\frac{2\pi n\,\Delta d}{\lambda}\right)\right]. \tag{19}$$

(18) and (19) are again two independent equations with two unknown parameters, this time n and x, since Δd is known from solving equations (11) and (12) in the transparent region. Equations (18) and (19) have again one unique solution for n and x in the range

$$0 < x \leqslant 1.$$

The values of n obtained from (18) and (19) can now be fitted to an equation of the form

$$n = \frac{d}{\lambda^2} + c \tag{20}$$

Equation (20) can be used to calculate extrapolated values of n in the region of strong absorption.

The usefulness of constructing continuous envelopes around the extremes of the spectrum lies in that *two* curves are generated from a single one, leading to two independent equations of the form of equation (6). By considering the transparent region and absorption region separately, four equations are actually obtained which together with equation (7) can be used to determine all the parameters of the experimental spectrum, equation (6).

2.3. *Region of strong absorption*

In this region the curves T, T_M and T_m merge into a single curve as shown in figure 3. The effect of Δd is actually beneficial in this region since the interference fringes are smaller and the spectrum approaches the interference-free transmission T_α sooner. The same procedures can thus be used here as in the case of a uniform film. Adding the reciprocals of (3*a*) and (3*b*) and solving for x yields

$$x = \frac{A - (A^2 - 4T_i^2 BD)^{1/2}}{2T_i D} \tag{21}$$

where

$$T_i = \frac{2T_{M0}T_{m0}}{T_{M0} + T_{m0}}. \tag{22}$$

Equation (21) is equivalent to equation (15) of the previous work and the physical meaning of T_i was discussed there. The definitions of curves T_α in equation (16) and T_i in equation (22) are *not* valid in the case of films with non-uniform thickness in the region of weak and medium absorption. In the region of strong absorption all curves merge and equation (21) can be used to calculate x in this region, using equation (20) to obtain n. An empirical way to construct the curve T_i is as follows: draw a smooth curve through the lower third of the interference fringes in the region of medium absorption to join the actual spectrum smoothly in the region of strong absorption.

The treatment in the region of strong absorption for non-uniform films is thus identical to the case of uniform films as discussed in the previous work.

2.4. *Numerical simulation*

To test the accuracy of the theory presented here, a film with the following properties is postulated:

substrate refractive index

$$s = 1.51 \qquad \text{(constant);}$$

average film thickness

$$\bar{d} = 1000\ \text{nm};$$

film refractive index

$$n = \frac{3\times 10^5}{\lambda^2} + 2.6;$$

film absorption coefficient

$$\lg\alpha = \frac{1.5\times 10^6}{\lambda^2} - 8 \qquad (\alpha \text{ in nm}^{-1});$$

thickness variation

$$\Delta d = 30\ \text{nm}.$$

The above values of $n(\lambda)$ and $\alpha(\lambda)$ represent typical values of α-Si:H (Freeman and Paul 1979, Swanepoel *et al* 1984). It is also identical to the case treated in the previous work so that direct comparisons are possible.

The full-curve spectrum shown in figure 3 is a plot obtained using the above film properties and performing the integration (9) numerically with $\Delta d = 30$ nm. The numerical integration was done over both φ and x. Two envelopes T'_M and T'_m are now drawn around the extremes of this spectrum and the values obtained from these 'experimental' envelopes are used in the calculations that follow. For the sake of comparison a plot of equation (1) for $\Delta d = 0$ is shown as the dotted spectrum in figure 3 and the transmission of the substrate alone according to equation (14) is also shown as T_s.

The wavelengths λ and values of T'_M and T'_m at the extremes of the spectrum are given in table 1. The true values of n and x are shown as n_{tr} and α_{tr} for each wavelength in table 1.

In the transparent region equation (7) refers to the wavelength at the points of the spectrum where $dT/d\lambda = 0$. This

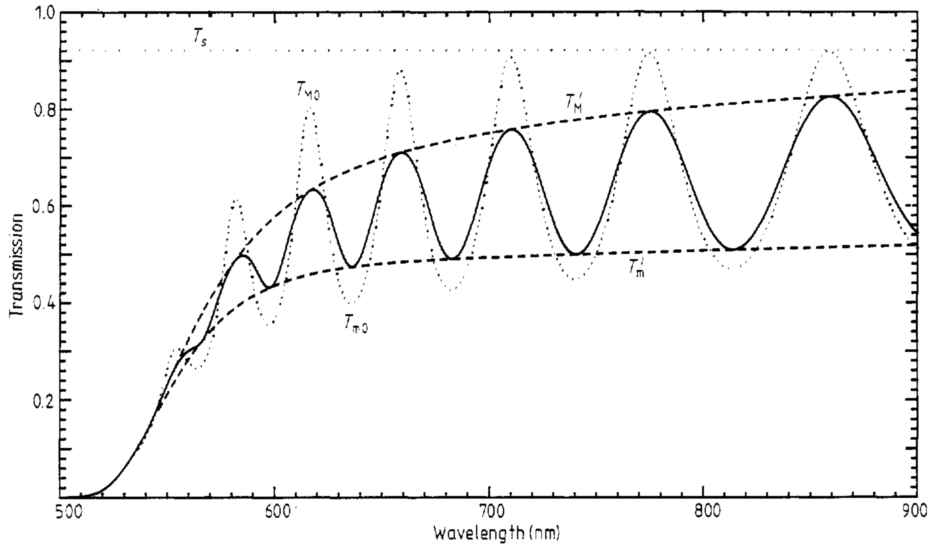

Figure 3. Simulated transmission for an absorbing film with uniform thickness of 1 μm (dotted-curve spectrum) compared to that of a film with a thickness variation $\Delta d = 30$ nm (full-curve spectrum).

is no longer true when $\alpha > 0$. The effect of α on the spectrum is such that the point $dT/d\lambda = 0$ for a maximum occurs at a value of λ that is larger than predicted by equation (7) while that of a minimum occurs at a value of λ that is smaller than that predicted. This effect can be accurately corrected for by reading the values of T and λ at the point where the envelopes are *tangential* to the actual spectrum. When λ is determined in this way equation (7) can be applied right down to the last visible fringe. (This procedure was not given or applied in the previous work.)

The values of n are now calculated using the formula for a uniform film, equation (4), and are shown as n_1 in table 1. It can be seen that the values of n_1 are much lower than the true values n_{tr} and n_1 even decreases with decreasing λ. The error in n_1 ranges from about 10% to 30%. If an attempt is made to calculate the thickness of the film using n_1, a value of about 2000 nm is obtained which represents an error of 100%. Serious errors thus result if a spectrum of a non-uniform film is treated as being that of a uniform film.

The values of T'_M and T'_m are now used in equations (11) and (12) to calculate the values of n and Δd, shown as n_2 and Δd in table 1. Using a fast-converging algorithm, it takes a HP-45B computer only a few seconds to calculate each set of values of n_2 and Δd shown in table 1 to 0.1% precision in T_{Md} and T_{md}, which represents the experimental accuracy.

The values of n_2 agree well with the true values for small α but increase drastically for larger values of α. The values of Δd initially show a small increase from the true value and then decrease drastically for larger values of α. These phenomena are due to the fact that equations (11) and (12) are only valid for $\alpha \equiv 0$ and the computation tries to assign the effect of $\alpha > 0$ to n and Δd. Inspection of the calculated values of Δd gives an indication where the onset of absorption starts. From table 1 it seems that the calculated values of Δd start to decrease when $\alpha \simeq 300\ \mathrm{cm}^{-1}$. The actual value of Δd is now obtained by estimating the asymptotic value for very large λ. The values of table 1 suggest a value for Δd of

$$\Delta d = 30.1 \text{ nm}.$$

This represents an accuracy of less than 1% from the true value. An absolute error of 1% in either T'_M or T'_m leads to a relative error of about 1% in n and a relative error of about 3%

Table 1. Values of λ, T'_M and T'_m for the spectrum of figure 3. Calculation of Δd, n and α (α in units of cm^{-1}).

λ	n_{tr}	α_{tr}	T'_M	T'_m	n_1	n_2	Δd	n_3	x	α
859	3.006	11	0.826	0.514	2.722	3.013	30.17	3.008	0.9988	12
814	3.052	18	0.812	0.509	2.720	3.060	30.12	3.056	0.9989	11
775	3.099	31	0.796	0.505	2.710	3.110	30.25	3.098	0.9969	31
740	3.147	54	0.778	0.500	2.699	3.170	30.35	3.148	0.9944	56
710	3.195	94	0.759	0.496	2.680	3.231	30.51	3.191	0.9897	103
683	3.243	165	0.737	0.491	2.658	3.307	30.63	3.238	0.9825	176
658	3.292	289	0.710	0.484	2.631	3.412	30.68	3.291	0.9709	295
636	3.341	506	0.676	0.474	2.598	3.560	30.61	3.341	0.9498	515
616	3.390	889	0.631	0.458	2.559	3.791	30.14	3.392	0.9144	895
598	3.439	1563	0.570	0.432	2.511	4.175	28.83	3.423	0.8519	1603
581	3.488	2740	0.484	0.387	2.455	4.900	25.85	3.45	0.750	2877
566	3.537	4831	0.377	0.320	2.395	6.281	20.96	3.54	0.619	4796

in Δd. The maximum experimental accuracy of T'_M and T'_m is about 0.1% which means that Δd can be calculated to an accuracy of about 0.3%. In the example chosen α is never zero and it is strongly recommended that the transmission spectrum in the NIR region is obtained for calculating Δd. The purpose of equations (11) and (12) is only to calculate Δd and not n.

The values of T'_M and T'_m are now used in equations (18) and (19) to calculate n and x using a value of $\Delta d = 30$ nm. These values are shown as n_3 and x in table 1. The accuracy of the values decreases when the difference between T'_M and T'_m becomes small, but in this region the methods for calculating α in the region of strong absorption can be used.

The agreement between n_3 and the true values n_{tr} is very good and is better than 0.1%. These values can now be used to determine $\bar{d}$ from equation (7) and even further improved values for n as described in the previous work. If this is done a value for $\bar{d}$ is obtained to an accuracy better than 0.1% as

$$\bar{d} = 1000 \text{ nm}.$$

If the values of n are fitted to equation (20) the original equation used to generate the spectrum is exactly obtained.

The value of $\bar{d}$ can now be used to calculate the values of α from x using (17c). The values of the extinction coefficient k can be calculated using (2g) for the case where $k^2 \ll n^2$. The calculated values of α are shown in the last column of table 1. It is seldom possible to calculate α to an accuracy better than say about 5% from the transmission spectrum and values of α smaller than 100 cm^{-1} can seldom be obtained with reasonable accuracy. It is furthermore difficult to estimate the effect of errors in T'_M and T'_m on α since these errors are already included in the calculation of Δd. From table 1 it can be said that the accuracy of the calculated values of α is at least 5% and the values can be considered as very good.

The calculation of α in the region of strong absorption is identical to that discussed for a uniform film in the previous work and will not be repeated here.

2.5. *Relation with spectrum of a uniform film*

An alternative approach in treating the spectrum of figure 3 is to transform it to the spectrum of a uniform film with the same average thickness, as shown by the dotted-curve spectrum in figure 3. This can be done for the envelopes T'_M and T'_m of the non-uniform film and the envelopes T_{M0} and T_{m0} of the uniform film.

Substituting (2) in (17), (17) in (18) and (19), and using (3) and (16) the following compact relations between the envelopes around the two spectra are obtained:

$$T'_M = \frac{T_\alpha}{\theta} \tan^{-1}\left(\frac{T_{M0}}{T_\alpha} \tan\theta\right)$$

$$T'_m = \frac{T_\alpha}{\theta} \tan^{-1}\left(\frac{T_{m0}}{T_\alpha} \tan\theta\right)$$

or

$$T'_M = \frac{(T_{M0} T_{m0})^{1/2}}{\theta} \tan^{-1}\left[\left(\frac{T_{M0}}{T_{m0}}\right)^{1/2} \tan\theta\right] \qquad (23)$$

$$T'_m = \frac{(T_{M0} T_{m0})^{1/2}}{\theta} \tan^{-1}\left[\left(\frac{T_{m0}}{T_{M0}}\right)^{1/2} \tan\theta\right] \qquad (24)$$

where

$$\theta = \frac{2\pi n\, \Delta d}{\lambda} \qquad (25)$$

and

$$0 < \theta < \pi/2.$$

Equations (23) and (24) are again two independent transcendental equations in T_{M0}, T_{m0} and θ. In the transparent region $T_{M0} \equiv T_s$ where T_s is known from equations (14), equations (23) and (24) can thus be solved for T_{m0} and θ in the transparent region; n can be determined from equation (15) and Δd from equation (25), but a more accurate procedure exists to extrapolate the information obtained from the transparent region to the region of absorption, as follows.

It was shown in the previous work that equation (7) can be written as follows for the successive extremes, starting from the long-wavelength end:

$$\frac{l}{2} = \frac{2n\bar{d}}{\lambda} - m_1 \qquad l = 0, 1, 2, 3, \ldots \qquad (26)$$

m_1 is the order number of the first ($l=0$) extreme considered, integer for a maximum or half integer for a minimum. Substituting equation (25) in (26) yields

$$\frac{l}{2} = \left(\frac{\bar{d}}{\pi\, \Delta d}\right)\theta - m_1. \qquad (27a)$$

If $l/2$ is plotted as function of θ for the transparent region, the slope and m_1 of (27a) can be obtained as shown in figure 4. Equation (27a) can now be used to calculate θ for the extremes in the region of absorption and T_{M0} and T_{m0} can be calculated from equations (23) and (24), as shown in table 2.

Table 2 shows again the wavelengths at the extremes of the full-curve (or dotted) spectrum in figure 3. The true values of the maxima and minima of the spectrum of a uniform film, T_{Mtr} and T_{mtr}, as calculated from equation (3) using the film properties given in § 2.4 are given in table 2 for each wavelength. T_{m0} and θ are now calculated from equations (23) and (24) using the values of T'_M and T'_m of table 1 and putting $T_{M0} = T_s = 0.921$. The values of θ are shown as θ_1 in table 2 and θ_1 is plotted as a function of $l/2$ according to equation (27a) in figure 4. The best straight line through the few points of the transparent region passes unambigously through $l/2 = -7$ for $\theta = 0$. The straight line is now drawn *exactly* through the point $l/2 = -7$ and through the points of the transparent region. The deviation of the points for larger θ_1 from this straight indicates the onset of absorption and these points must be rejected. From figure 4

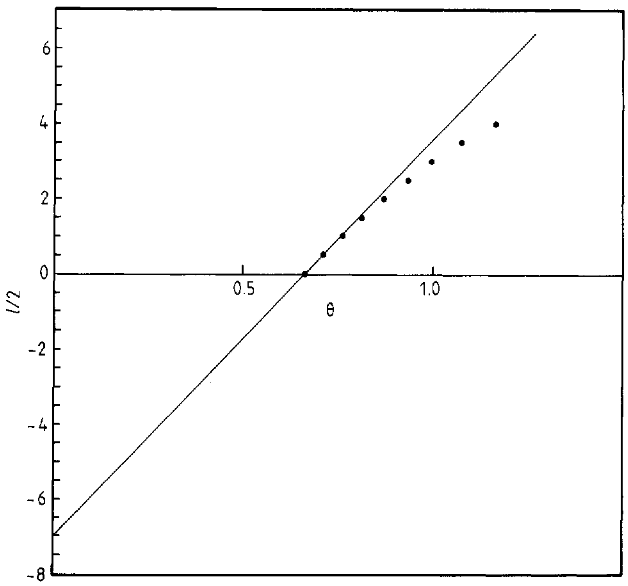

Figure 4. Plot of $l/2$ against θ_1 to determine θ in the region of absorption.

Table 2. Calculation of T_{M0} and T_{m0} from T'_M and T'_m.

λ	T_{Mtr}	T_{mtr}	θ_1	θ	T_{M0}	T_{m0}
859	0.919	0.480	0.665	0.660	0.919	0.480
814	0.918	0.470	0.711	0.707	0.919	0.469
775	0.916	0.459	0.763	0.754	0.917	0.459
740	0.913	0.448	0.817	0.801	0.913	0.488
710	0.908	0.437	0.872	0.848	0.907	0.438
683	0.899	0.426	0.932	0.895	0.897	0.426
658	0.881	0.413	1.002	0.942	0.880	0.413
636	0.853	0.398	1.077	0.990	0.852	0.398
616	0.804	0.379	1.165	1.037	0.804	0.379
598	0.728	0.354		1.084	0.727	0.354
581	0.610	0.318		1.131	0.609	0.318
566	0.461	0.268		1.178	0.460	0.268

equation (27*a*) can be represented by the equation

$$\frac{l}{2}=10.6\theta-7. \qquad (27b)$$

The value of θ at each extreme is calculated from (27*b*) is shown as θ in table 2. Using these values of θ, T_{M0} and T_{m0} are calculated from equations (23) and (24), using the values of T'_M and T'_m from table 1. These values are shown in the last two columns of table 2 and there is an excellent agreement between the calculated values of T_{M0} and T_{m0} and the true values.

The calculated values of T_{M0} and T_{m0} can now be used to calculate $\bar{d}$, $n(\lambda)$ and $\alpha(\lambda)$ using the formulae discussed in the previous work. This yields $\bar{d}=1000$ nm and substituting $\bar{d}$ in equation (27*a*) and using the value of the slope in equation (27*b*), gives a correct value for Δd of

$$\Delta d=30.0 \text{ nm}.$$

3. Effects of finite bandwidth

In the previous sections it was tacitly assumed that λ has a single sharp value. In practice a spectrophotometer has a finite bandwidth set by the slit of the instrument, which means that a band of wavelengths is incident on the sample such that

$$\lambda=\bar{\lambda}\pm\Delta\lambda. \qquad (28)$$

The case is now considered of a transparent film with uniform thickness d and constant n transmitting a band according to equation (28). Following the same procedure as in § 2.1 and using the approximation $(\bar{\lambda}^2-\Delta\lambda^2)=\bar{\lambda}^2$, the equations for the maximum and minimum envelopes in the transparent region become

$$T_{M\lambda}=\frac{\bar{\lambda}^2}{4\pi nd\,\Delta\lambda}\,\frac{a}{(1-b^2)^{1/2}}\times\tan^{-1}\left[\frac{1+b}{(1-b^2)^{1/2}}\tan\left(\frac{2\pi nd\,\Delta\lambda}{\bar{\lambda}^2}\right)\right] \qquad (29)$$

$$T_{m\lambda}=\frac{\bar{\lambda}^2}{4\pi nd\,\Delta\lambda}\,\frac{a}{(1-b^2)^{1/2}}\times\tan^{-1}\left[\frac{1-b}{(1-b^2)^{1/2}}\tan\left(\frac{2\pi nd\,\Delta\lambda}{\bar{\lambda}^2}\right)\right] \qquad (30)$$

where

$$0<\Delta\lambda<\bar{\lambda}^2/4nd. \qquad (31)$$

The constants in equations (29) and (30) are those given in equation (10). The effect of a finite bandwidth on a spectrum is thus similar to that of a variation in thickness and it also causes shrinking of the interference pattern. As far as the disappearance of the interference pattern is concerned, it follows from equations (13) and (31) that the two effects are equivalent according to $\Delta\lambda=\lambda\,\Delta d/d$. If a plot of a numerical integration of equation (9) is made for a film of $d=1000$ nm, $n=3.5$ and $\lambda=\bar{\lambda}\pm 22.4$ nm, a spectrum similar to the full-curve spectrum in figure 2 is obtained. The interference pattern also disappears at $\bar{\lambda}=560$ nm but for the rest of the spectrum the shrinkage is about 50% less than the case for $\Delta d=40$ nm. Plots of equations (29) and (30) also yield envelopes that fit exactly around the extremes of the spectrum.

When dispersion is present $n=n(\lambda)$ and the constant in equations (29) and (30) become functions of λ. This effect is rather severe and even with some approximations envelopes calculated from functions of the form of equations (29) and (30) do not agree well with the values obtained by numerical integration of equation (9). It is thus not worthwhile to pursue analytical expressions for the envelopes in the case where n is not constant or in the region of absorption.

The effect of slit width was also discussed in the previous work and formulae were given to correct for its effect. Although they are empirical formulae, they were found to work well in all parts of the spectrum for practical cases. The bandwidth is, however, under experimental control and its effect can be almost eliminated by using a small slit.

4. Variation in refractive index *n*

The case is now considered of a film with uniform thickness d, zero bandwidth for λ but non-uniformity in n according to the equation

$$n=\bar{n}\pm\Delta n. \qquad (32)$$

This case can be treated in the same way as the effect of Δd was treated in § 2.1. In the integration (9) the constants are however not constant but functions of n according to equation (2). Fortunately those functions are such that it is an excellent approximation to evaluate the constants at the value of $n=\bar{n}$. The equations for the maximum and minimum envelopes in the transparent region become

$$T_{Mn}=\frac{\lambda}{2\pi d\,\Delta n}\,\frac{a}{(1-b^2)^{1/2}}\times\tan^{-1}\left[\frac{1+b}{(1-b^2)^{1/2}}\tan\left(\frac{2\pi d\,\Delta n}{\lambda}\right)\right] \qquad (33)$$

$$T_{mn}=\frac{\lambda}{2\pi d\,\Delta n}\,\frac{a}{(1-b^2)^{1/2}}\times\tan^{-1}\left[\frac{1-b}{(1-b^2)^{1/2}}\tan\left(\frac{2\pi d\,\Delta n}{\lambda}\right)\right] \qquad (34)$$

where

$$0<\Delta n<\lambda/4d. \qquad (35)$$

The constants in equations (33) and (34) are the same as in equation (10) and the average value $\bar{n}$ is used to calculate them. Δn can now be determined from an experimental spectrum in the transparent region using equations (33) and (34) in the same way as Δd was obtained from equations (11) and (12). In the case where $\alpha>0$ equations (33) and (34) can be written in the same form as equation (18) and (19) and $\bar{n}$ and x can be determined from these equations.

There is a symmetry between the equations for Δd and Δn since the product nd occurs as optical path length in the phase angles of the formulae. The equivalence of their effects follows from equations (13) and (35):

$$\Delta n\cdot d\equiv n\cdot\Delta d. \qquad (36)$$

From equation (36) it follows that the spectrum in figure 2 for $\Delta d = 40$ nm should be similar to that for $\Delta n = 0.14$. Numerical integration also shows that these curves are identical and plots of equations (33) and (34) for $\Delta n = 0.14$ yield identical envelopes to those shown in figure 2 for $\Delta d = 40$ nm. Equations (23) and (24) are also valid for Δn using the equivalence (36) in equation (25).

Because of the equivalence of the two effects according to (36), there is unfortunately no way to say whether a spectrum as shown in figure 3 is caused by a variation in thickness or a variation in refractive index by considering only the transmission spectrum.

Equations (33) and (34) should be viewed with some caution. Equation (1) is strictly valid for one layer on a substrate and equation (33) and (34) are valid when the variation in n is lateral in the same way as d varies laterally in figure 1(a). In practical cases the variation in n can also be in the depth of the film resulting in a type of multilayer structure for which equation (1) is not strictly valid. In cases where Δn is not very large, equations (33) and (34) and their equivalent formulation in the absorption region should however be a good approximation. It can be used to model multiphase systems, for example the case where small crystallites of Si are embedded in an amorphous network. It can also be used as an approximation for multilayer systems that otherwise could not be solved analytically.

5. Conclusion

The effect of inhomogeneities in thin films is to cause a drastic shrinking of the interference fringes of the optical transmission spectrum. This may mistakenly lead to the conclusion that an absorption band tail exists in the long-wavelength region and serious errors occur if n, d and α are calculated from such a spectrum assuming the film to be uniform. Formulae were derived to calculate the magnitude of the inhomogeneities and to calculate the optical constants accurately for such spectra. These formulae were used on samples of α-Si:H (Swanepoel *et al* 1984) that displayed non-uniform thickness variations under certain conditions of preparation. The thickness variations were independently measured with a mechanical stylus instrument and the results agreed well with those obtained from the transmission spectra.

Acknowledgment

This work was supported by the Research Grants Division of the Council for Scientific and Industrial Research, South Africa.

References

Cisneros J I, Rego G B, Tomyiama M, Bilac S, Gonçalves J M, Rodriguez A E and Argüello Z P 1983 A method for the determination of the complex refractive index of non-metallic thin films using photometric measurements at normal incidence *Thin Solid Films* **100** 155–67

Clark A H 1980 *Polycrystalline and Amorphous Thin Film Devices* ed. L L Kazmerski (New York: Academic) chap. 4 pp 135–52

Freeman E C and Paul W 1979 Optical constants of RF sputtered hydrogenated amorphous Si *Phys. Rev.* B **20** 716–28

Heavens O S 1964 *Physics of Thin Films* ed. G Hass and R E Thun (New York: Academic) pp 193–304

Martin P J, Netterfield R P and Sainty W G 1983 Optical properties of thin amorphous silicon and amorphous hydrogenated silicon films produced by ion beam techniques *Thin Solid Films* **100** 141–7

Swanepoel R 1983 Determination of the thickness and optical constants of amorphous silicon *J. Phys. E: Sci. Instrum.* **16** 1214–22

Swanepoel R, Aharoni H and Swart P L 1984 Influence of argon partial pressure on the electrical and optical properties of sputtered α-Si:H *Thin Solid Films* (in press)

Szczyrbowski J and Czapla A 1977 Optical absorption in DC sputtered InAs films *Thin Solid Films* **46** 127–37