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Optical Absorption in Polyerystalline CdTe Thin Films

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Abstract

The spectral dependence of the absorption coefficients in vacuum deposited polycrystalline CdTe thin films on transparent glass substrate using envelope method is presented over incident photon energies ranging between 0.5-1.6 eV. The method required only a measurement of optical transmittance at normal incidence and is based on the evaluation of the spectral position and the contrast of the interference fringes over a wide range of the incident photon energies. Light absorption at lower photon energies (below bandgap) may be ascribed to the structural imperfections. In this spectral region hypothetical transmittance spectrum, in the case of non-zero absorption displayed a fairly good agreement with the experimental spectrum. As is typical in compound semiconductors, a disorder related exponential absorption (Urbach) tail was observed below the fundamental (mobility) gap. Optical bandgap of as-deposited layers was determined using Tauc equations as ~1.53 eV.

Keywords: Absorption coefficient, CdTe thin films, Optical bandgap.

Özet

Saydam cam lam üzerine vakumda buharlaştırma tekniğiyle büyütülen çoklu kristal CdTe ince filmlerin soğurma katsayılarının 0.5-1.6eV foton enerji aralığındaki spektral dağılımı verilmektedir. Kullanılan metot sadece normal açı altında optiksel transmisyon ölçümünü gerekli kılar ve girişim saçaklarının spektral genişliklerinin ve spektral konumlarının incelenmesine dayanır. Foton enerjisinin düşük olduğu bölgede ışığın soğurumu yapısal kusurlardan dolayı olabilir. Optik band aralığının dışındaki bölgede teorik olarak hesaplanan transmisyon deneysel olarak elde edilen spektruma oldukça yakın olduğu saptandı. Bileşik yarı-iletkenlere özgü yasak enerji aralığının kenarında yapı düzensizliğiyle ilişkili üstel (Urbach) kuyruğu gözlendi. İşlenmemiş filmlerin optiksel band aralığı Tauc denklemleri kullanılarak ~1.53 eV olarak hesaplandı.

Anahtar kelimeler: Soğurum katsayısı, CdTe ince filmi, Yasak enerji aralığı.

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1. Introduction

Accurate measurements of optical constants and the thickness of semiconducting and dielectric thin layers are great importance, particulary for optical coatings and photo-optical devices. The performance of optoelectronic devices strongly depends on the wavelength dependence of the optical constants (i.e., refractive index n_f and absorption coefficient, α_f) of the absorber layer. The stoichiometry, doping, uniformity etc. parameters of importance for device performance, also affect the factors such as the spectral location and Urbach absorption tail, the contrast of the fringes, both of which can be deduced from the optical transmission and/or reflection spectrum of which is potentially useful as a non-destructive tool in layer characterisation.

The evaluation of the optical constants and the thickness of the absorbing layer in the visible and infrared wavelength regions from the various types of the optical measurements is a field of widespread interest, and therefore it has been the subject of numerous studies. Many methods for determining optical constants and the layer thickness of weakly absorbing coating material deposited onto substrate with finite thickness have been published (Laaziz et al, 2000, 149-155; Kushev et al, 1986, 385-393; Vedam, 1998, 1-9). Mostly of these involve measurements of transmission and reflection at normal or oblique incidence with or without perpendicular and parallel polarization components. Vriens and Rippens used $(1 \mp R)/T$ or $(1 \mp R')/T$ functions to deduce n_f and α_f of a thin solid film on a slightly absorbing substrate with use of an iterative method (Vriens et al, 1983, 4105-4110). In their analysis transmittance, T air-incident reflectance, R and substrate-incidence reflectance, R' are utilized. Minkov used the reflectance spectrum for different angles of incidence of nonpolarised or s (or p) polarised light to compute the optical constants and the thickness (Minkov, 1991, 306-310). Moreover optical constants and the layer thickness of a layer coated on a semi-infinite transparent layer were estimated using transmission spectrum alone by Manifacier et al (so-called envelope function method of which takes into account of interference fringe pattern) (Manifacier et al, 1976, 1002-1004). Swanepoel improved this method further in the case of finite substrate thickness (Swanepoel, 1983, 1214-1222). From a practical point of view, transmittance data are usually preferred to the reflectance data because the measurements are more accurate and straightforward. In addition, they are less sensitive to surface inhomogeneities.

CdTe has been shown to be the most promising semiconducting material for a large number of applications in the field of photovoltaic devices and X- and γ-ray detectors (Paulson et al, 2000, 299-306; Burgelman et al, 2005, 392-398; Limousin, 2003, 24-37). It has high absorption coefficient, direct bandgap which is close to the optimum value for low cost efficient solar cells and its ability to be doped p-type and the ease with which it may be produced in thin film form. Device grade CdTe thin films can be formed in both homojunction and heterojunction configuration. Different techniques of deposition have been reported in the literature, among which sputtering (Compaan et al, 2004, 815-822), closed vapour deposition (Britt et al, 1993, 2851-2852) and electrodeposition (Turner et al, 1994, 263-270) are worth mentioning. All these techniques have their own merits in producing high quality CdTe thin films.

This paper is organized as follows: In section 2 we provide a short overview on the envelope function model to extract the optical constants using transmission spectra alone. The details of preparation methods and the measurements are briefly given in section 3. Finally in section 4 we present the findings of the optical analysis and discuss them, particularly in the case of weak absorption. In this paper, we are motivated to measure and to analyze the transmission spectra to extract the optical constants and the film thickness accurately of absorbing singly refractive thin films of CdTe on a transparent substrate. To that aim, unpolarised normal incidence transmittance spectra yielding multiple reflections at interfaces which are coherent in the thin film and incoherent in the thick substrate are utilized. Special attention is focused on the estimation of absorption coefficients as a function of wavelength for the fundamental and technological point of view.

2. Theoretical aspects

A monochromatic beam is incident normally on a weakly absorbing thin film with a complex refractive index deposited on a non-absorbing substrate. If we assume smooth and parallel interfaces and multiple reflections to be coherent in the film and incoherent in the substrate, the optical constants of thin layer can be easily estimated using a simpler and straightforward process based on well-known turning point (or envelope) method, using transmittance spectra alone. This is suitable for the materials with a refractive index which differs considerably from that of the substrate. The basis of the analysis is based on the assumptions that;

(1) radiation of wavelength, λ which is normally incident on the sample has components in the wavelength range, $\Delta\lambda$ (i.e., finite bandwidth).

(2) interference of internally reflected radiation occurs in a semiconductor thin film [$\Delta\lambda << \lambda^2/(2n_f d_f)$] and is negligible in the substrate [$\Delta\lambda >> \lambda^2/(2n_s d_s)$] where d_s and n_s represent the thickness and refractive index of the substrate.

(3) the change in the real part of the refractive index, Δn_f of the film is negligible over the illuminated sample area:

$$\frac{\sigma_f}{d_f} + \frac{\Delta\lambda}{\lambda} >> \frac{\Delta n_f}{n_f}$$

where σ_f is the standard deviation of the film thickness over the illuminated sample area. In the weak absorption region, ($\alpha_f d_f << 1$), transmission decreases with decreasing wavelength mainly due to the effect of σ_f and may be briefly, determined from the continuous envelopes of the interference fringes, $T_M(\lambda)$ and $T_M(\lambda)$, as;

$$T_M(\lambda) = \frac{Ax}{B - Cx + Dx^2} \tag{1}$$

and

$$T_m(\lambda) = \frac{Ax}{B + Cx + Dx^2} \tag{2}$$

where, $A = 16n_f^2 n_s$, $B = (n_f + 1)^3 (n_f + n_s^2)$, $C = 2(n_f^2 - 1)(n_f^2 - n_s^2)$, $x = exp(-\alpha_f d_f)$ and $D = (n_f - 1)^3 (n_f - n_s^2)$ Solving equation (1) gives;

$$x = \frac{E_M - \left[E_M^2 - (n_f^2 - 1)^3 (n_f^2 - n_s^4)\right]^{1/2}}{(n_f - 1)^3 (n_f - n_s^2)} \tag{3}$$

where

$$E_M = \frac{8n_f^2 n_s}{T_M} + (n_f^2 - 1)(n_f^2 - n_s^2) \tag{3a}$$

Alternatively, adding the reciprocals of equations (1) and (2) yields;

$$\frac{2T_M T_m}{T_M + T_m} = \frac{Ax}{B + Dx^2} \tag{4}$$

and solving for x gives

$$x = \frac{F - \left[F^2 - (n_f^2 - 1)^3 (n_f^2 - n_s^4)\right]^{1/2}}{(n_f - 1)^3 (n_f - n_f^2)} \tag{5}$$

where

$$F = \frac{8n_f^2 n_s}{T_i} \tag{6a}$$

and

$$T_i = \frac{2T_M T_m}{T_M + T_m} \tag{6b}$$

Note that equation (5) encounters the influences of both extremes, $T_M(\lambda)$ and $T_M(\lambda)$ on the optical absorption.

3. Experimental

The CdTe thin films were deposited onto cleaned glass substrates by vacuum deposition. Immediately prior to loading into the deposition chamber, the substrates were first pre-cleaned by washing in a solution of detergent in deionised water followed by widely known successive refluxing, ultrasonic bath and rinsing routines in trichloroethylene, isopropyl alcohol and deionised water (Bayhan, 1994, 42). The CdTe starting material used was synthesised in house by direct combination of high purity double zone refined Cd and Te (99.9999%) supplied by MCP Electronic Materials. The layers were typically grown with substrate and source temperatures of 220°C and of 750°C respectively and were in general, 1.5–2.5 μm thick.

The optical transmission and absolute reflection spectra were obtained using a Perkin Elmer Lambda 19 UV/VIS/NIR spectrophotometer. Transmission spectrum was taken at normal incidence with reference to air. The area where the light upon was minimized using a suitable mask to eliminate the effects of layer inhomogeneities. The absolute reflectance spectrum was measured with an incidence angle of 8° and was normalized with respect to a perfectly polished mirror supplied by Melles-Geriot.

4. Results and discussion

4. 1. Determination of α_f in the weak absorption region

A typical normal incidence transmittance spectrum for an as-deposited CdTe thin layer measured over the spectral range 0.5-1.6 eV is shown in figure 1 (solid line). In the weak absorption region, sharp interference fringes were apparent and indicated that the interfaces, air/layer and layer/glass were flat and parallel. Strong absorption was observed at photon energies higher than 1.45 eV where interference effects suppressed almost completely due to a well defined band edge. The broken spectra are obtained theoretically in the case for a non-absorbing thin layer (i.e., $\alpha_f = 0$) with different thicknesses. It can be seen that the position of the fringe extreme of the transmittance spectra of a hypothetical non-absorbing layer of about 2.1737μm in thickness demonstrated a close agreement over the measured spectral range with the experimental spectrum. The transmittance, *T* was calculated theoretically from equation (11) using least-squared best fitting of Sellmeier function to the values of n_f. The procedure for the extraction of n_f data from the extremes of the interference fringes is in detail reported elsewhere (Swanepoel, op.cit., Ref. 7, pp. 1214-1222).

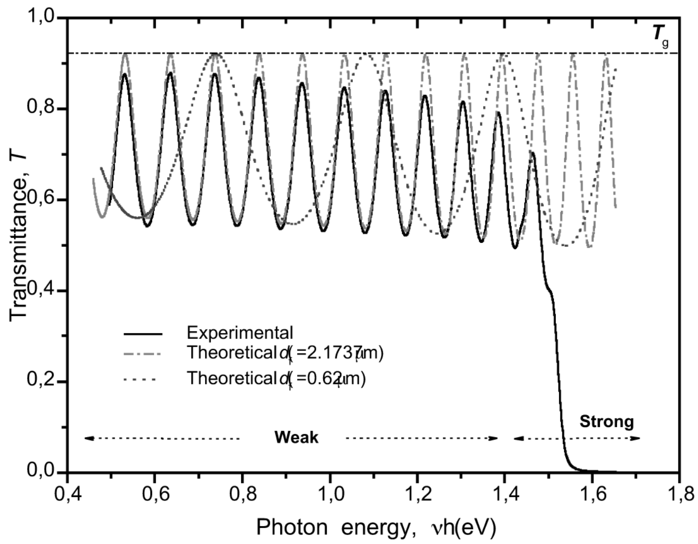

Figure 1. Transmittance spectrum obtained from a typical as-deposited CdTe layer on glass substrate (solid line). Dotted lines refer hypothetical spectrum for 2.1737μm thick and 0.62μm thick CdTe layer in the case of zero absorption (i.e., $\alpha_f = 0$). T_g denotes the transmittance of glass substrate.

It can be seen from figure 1 that there was some enhanced light absorption at photon energies below bandgap giving rise to a reduction in the transmittance. This may have been due mainly to diffuse light scattering at textured film surface and grain boundaries or other structural defects (impurities and compositional fluctuations). CdTe layers exhibited columnar growth in feature with high preferential orientations and with noticeable small values of grain size compared to the dimension of the wavelength upon the layer. Therefore scattering at grain boundaries was probably of minor importance and can be neglected. Nevertheless, residual absorption at lower photon energies was more probably due to other defects and impurities arising from the vacuum environment and glass substrate. Surface roughness may also have pronounced effects on the propagation of the incident light upon the surface mainly due to diffused layer and light scattering (Poruba et al, 2000, 148-160). The implication of the roughness is to reduce the optical interference effects due mainly to extended propagation paths at film surface and interfaces. This causes in particular, a reduction at reflectance leading to an enhancement at the optical absorption. As-deposited CdTe layers have a typical surface roughness of about 20 nm as estimated from the absolute reflectance spectra indicating that layers were relatively smooth with a specular surface as in the natural growth condition.

In practice a spectrophotometer always has a finite spectral width (band width). The influence of finite band width on the transmission spectrum is to shrink the interference fringes. The effect can be minimised by experimentally reducing the slit width but the noise problems reducing the accuracy of the transmittance may arise. In our case it appears to be rather less important since $S << n_f d_f$ and therefore can be neglected. According to the discussion above, a correct procedure to make spectrometric measurements should begin by lowering the size of the spot incident onto film surface which reduces efficiently the inhomogeneity effect, as in this case. Treatment of layer inhomogeneities is an own area of research and goes beyond the scope of this work.

In the weak absorption region, the absorption coefficients were calculated separately from equations (2) and (3) for a typical as-deposited CdTe layer. The plot of averaged values of $\overline{\alpha}_f$ as a function of wavelength displayed a progressive increase as the wavelength decreased (see figure 2). A least-squares best fit of the Sellmeier expression to the data for a typical layer yielded $\log\alpha_f = [4.14 + 1.24\,\lambda^2/(\lambda^2 - 0.39)]^{1/2}$ (for λ in μm).

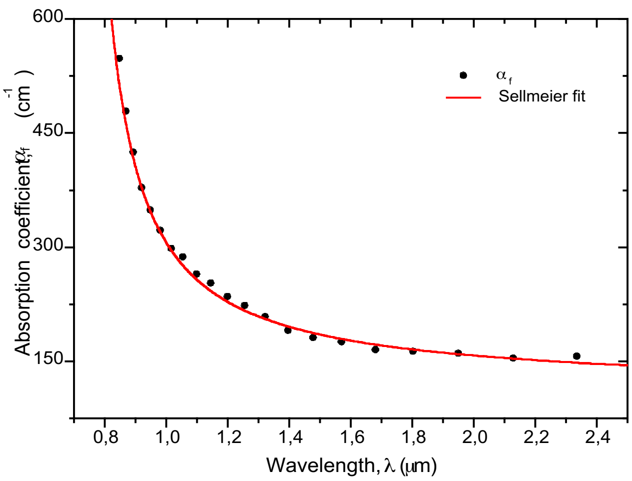

Figure 2. Absorption coefficient versus wavelength characteristics of as-deposited CdTe layer in the weak absorption region. The dashed line displays a least-squares best fit of the Sellmeier expression to the data.

Figure 3 shows calculated transmittance spectra in the case of non-zero absorption using Eq. (11). This was done by fitting Sellmeier function of the form;

$[a + b\lambda^2/(\lambda^2 - c^2)]$ to values of $\log\alpha_f$ in the weak absorption region and then using the function to provide the values of absorbance, x. A fairly good agreement between experimental and calculated spectra is obtained as to the position of the fringe extremes over the measured spectral range.

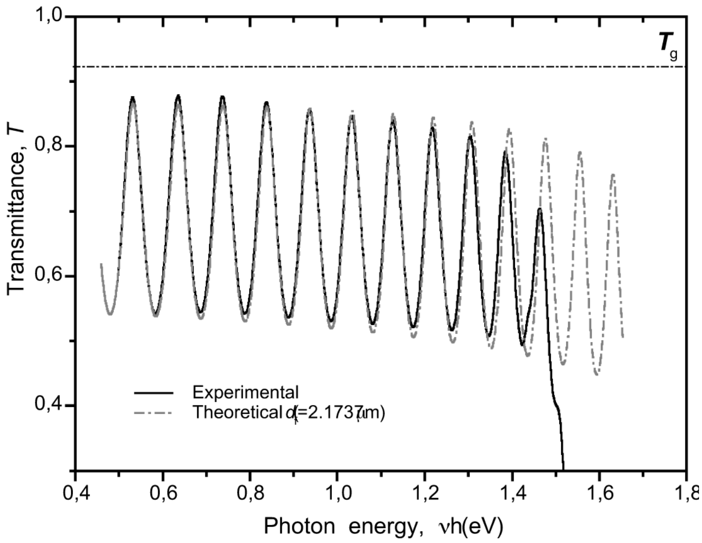

Figure 3. Experimental (solid line) and hypothetical (dotted line) transmittance spectra for typical 2.1673μm thick film of as-deposited CdTe on glass, in the case absorption (i.e., $\alpha_f \neq 0$).

If n_i and n_{i+1} are the refractive indices at two adjacent maxima (or minima) at λ_i and λ_{i+1}, it follows from the basic interference equation, $2n_f d_f = m\lambda$ that film thickness is given by;

$$d_f = \frac{\lambda_i \lambda_{i+1}}{2(\lambda_i n_{i+1} + \lambda_{i+1} n_i)} \qquad (7)$$

A mean value of d_f using above equation was obtained as 2.1673μmwith a negligible dispersion. This was in good agreement with the value $(2.1737\mu\text{m})$ which was initially used in the equation (11) to obtain absorption free transmittance. Table I tabulates the values at the fringe extremes of the spectrum given in figure 1 (solid line) of λ, n_f, T_M, T_m and $\overline{\alpha}_f$.

4. 2. Determination of α_f in the strong absorption region

Abele's exact expressions for the reflectance and the transmittance for a weakly absorbing thin layer on a transparent substrate with finite thickness reduce to (Hishikawa et al, 1991, 1008-1014)

$$\frac{1-R}{T}=\exp(\alpha_f d_f) \tag{8}$$

over the spectral range where interference effects has been suppressed to a great extent. The increase in α_f with photon energy $h\nu$ near band edge depends on the type of transition taking place and is given by the following proportionality (Pankove, 1971, 36);

$$\alpha_f \propto \frac{(h\nu - E_g)^{\xi}}{h\nu} \tag{9}$$

where ξ= 1/2, 3/2, for allowed direct and forbidden direct transitions respectively (since CdTe is a direct gap semiconductor, indirect transitions are not important) and E_g is the optical bandgap. At the absorption edge, the values of absorption coefficient have been found to be in the order of $10^4\ \mathrm{cm}^{-1}$. Optical bandgap of the layers may be easily determined using well known Tauc equations by plotting $(\alpha_f h\nu)^2$ versus $h\nu$ in the strong absorption region ($h\nu$ >1.5 eV) and extrapolating the linear portion of the plot to $(\alpha_f h\nu)^2=0$.

λ (μm)	T_M	T_m	n_f	$\overline{\alpha_f}$ (cm^{-1})
2.3307	0.8825 *m=5*	0.5421	2.6886	156.69
2.1293	0.8813	0.5406 *m=5.5*	2.7019	154.45
1.9526	0.8804 *m=6*	0.5400	2.7029	160.84
1.8075	0.8784	0.5392 *m=6.5*	2.7105	163.32
1.6822	0.8775 *m=7*	0.5383	2.7167	165.40
1.5722	0.8746	0.5377 *m=7.5*	2.7204	175.88
1.4777	0.8717 *m=8*	0.5363	2.7273	181.42
1.3942	0.8667	0.5348 *m=8.5*	2.7340	190.87
1.3216	0.8615 *m=9*	0.5333	2.7441	208.83
1.2557	0.8571	0.5304 *m=9.5*	2.7521	223.51
1.1998	0.8513 *m=10*	0.5275	2.7680	235.44
1.1458	0.8469	0.5246 *m=10.5*	2.7756	252.92
1.1018	0.8410 *m=11*	0.5231	2.7961	264.86
1.0556	0.8358	0.5188 *m=11.5*	2.8007	287.47
1.0182	0.8294 *m=12*	0.5158	2.8189	298.36

0.98	0.8235	0.5129 $^{m=12.5}$	2.8262	322.39
0.9479	0.8163 $^{m=13}$	0.5100	2.8429	348.94
0.9171	0.8046	0.5042 $^{m=13.5}$	2.8564	378.33
0.8929	0.7900 $^{m=14}$	0.4983	2.8840	425.03
0.8718	0.7716	0.4887 $^{m=14.5}$	2.9164	478.97

Table I: Values of λ, n_f, T_M, T_m and $\overline{\alpha}_f$ at the fringe extremes for the experimental spectrum given in figure 1 (solid line). m refers to the fringe order number.

Figure 4 shows such a plot and a value of 1.53 eV corresponding to allowed direct transitions occurring in the band was estimated. However this was found to be somewhat ~20 meV higher compared to that of bulk crystalline CdTe determined by Olego et al (1.51 eV) (Olego et al, 1985, 1172-1174), Enloe et al (1.51 eV) (Enloe et al, 1987, 2005-2010), and Pikhtin and Yas'kov (1.50 eV) (Pikhtin et al, 1988, 613-626). This can be ascribed to the band-tail effects and possibly to the deviations in the stoichiometry resulting relatively poor layer crystallinity.

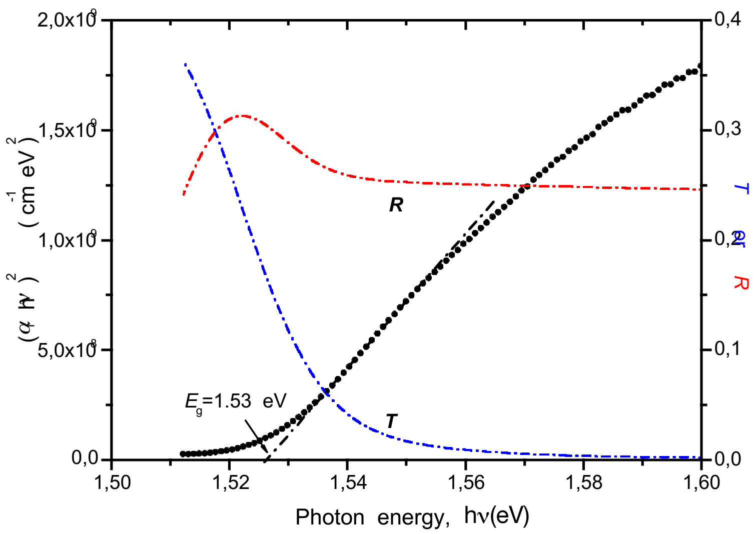

Figure 4. $(\alpha_f hv)^2$ versus hv characteristics of as-deposited CdTe layer in the strong absorption region.

5. Conclusion

The envelope method to extract the optical constants using normal incidence transmittance spectrum alone has been successfully applied to the vacuum evaporated CdTe thin layers on transparent substrate with finite thickness. In the weak absorption region, sharp interference fringes due to parallel surface and interfaces were apparent. Spectral region below the bandgap there was some excess absorption of which could be ascribed to the variations in stoichiometry and impurities originating possible from the vacuum environment and glass substrate. Hypothetical transmittance spectrum in the case of non-zero absorption yielded a fairly good agreement with the experimental data. In association with α_f estimation, a least-squares best fit of Sellmeier function is utilized. That the value is lower by ~20 meV from the bandgap of the corresponding bulk single crystal CdTe can be attributed to band-tail effects and the deviations in the stoichiometry.

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Appendix

Analytical expressions of which envelope function method utilizes for weakly absorbing thin layers on transparent substrates with finite thickness are briefly presented. The advantages of the envelope function method over other reported procedures can be summarized as follows:

(1) A unique and simple analytical expression of the transmittance is used over the entire spectrum.

(2) A non-linear least-squares smoothing of the experimental spectrum is performed to reduce the noise in the experimental apparatus: this enhances the precision of the determination of the turning point wavelength positions.

(3) The fringe orders are determined in a straightforward manner without the need for any iterative computations.

Figure 5 represents a semi-infinite thin layer with a complex refractive index, $n_c = n_f + ik_f$ bounded by two transparent media with refractive indices, n_o and n_1. Normal incidence transmittance of the layer, T is given in the case of weak absorption (i.e., $k_f^2 << (n_f - n_o)^2$) *and* $k_f^2 << (n_f - n_1)^2$) (Manifacier, op.cit., Ref. 6, pp. 1002-1004);

$$T = \frac{16 n_o n_1 n_f^2 x}{A^2 + 2ABx\cos\varphi + B^2 x^2} \quad (10)$$

where $A = (n_f + n_o)(n_1 + n_f)$, $B = (n_f - n_o)(n_1 - n_f)$, $x = \exp(-\alpha_f d_f)$ and $\varphi = 4\pi n_f d_f / \lambda$.

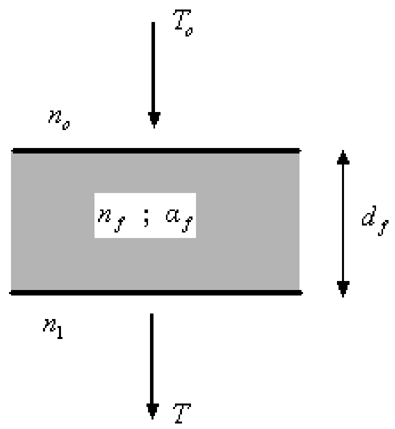

Figure 5. Transmission of the light by a single absorbing thin film.

One of the frequently employed assumptions in the estimation of n_f and α_f values is the assumption of semi-infinite substrate. However, this is valid only for absorbing substrates which are thick enough so that there is no reflection from the back side of the substrate and is usually not the case. It is therefore necessary to correct the formulae for T in the case of finite substrate. The commonly encountered case is that of optically thick thickness to wavelength ratio is so large that there is insufficient resolution to sense the phase coherence of the transmitted beams through the substrate. According to Swanepoel, the situation for a thin film on a transparent substrate can be represented in figure 6. The film has a thickness, d_f, index of refraction, n_f and has absorption coefficient, α_f. The transparent substrate has a thickness several orders of magnitude larger than d_s and has absorption coefficient $\alpha_s \cong 0$ over the spectral range examined. The index of the substrate and surrounding air are 1 and 1.51 respectively. Rigorous analysis has to take into account all the multiple reflections at the interfaces (i.e. air/film, film/substrate, and substrate/air) when calculating T .

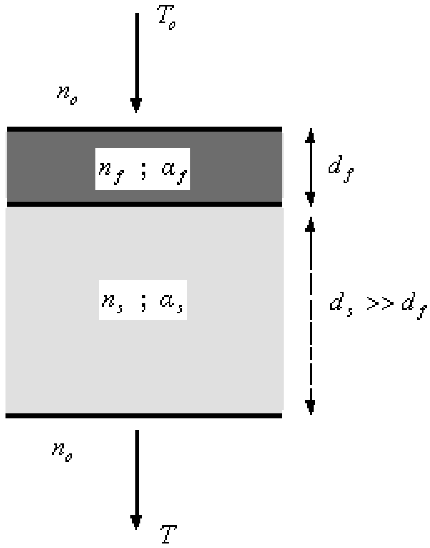

Figure 6 . System of a weakly absorbing thin film on a transparent substrate immersed in air.

Normal incidence transmittance, T for a system of a weakly absorbing thin film ($k_f^2 << n_f^2$) on a finite transparent substrate is given by;

$$T = \frac{Ax}{B - Cx\cos\varphi + Dx^2} \tag{11}$$

where

$$A = 16 n_f^2 n_s$$

$$B = (n_f + 1)^3 (n_f + n_s^2)$$

$$C = 2(n_f^2 - 1)(n_f^2 - n_s^2)$$

$$D = (n_f - 1)^3 (n_f - n_s^2)$$

$$\varphi = 4\pi n_f d_f / \lambda$$

$$x = \exp(-\alpha_f d_f)$$

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