CHAPTER THREE

CALCULATION OF THICKNESS AND OPTICAL PARAMETERS OF THIN FILM BY USING OPTICAL TRANSMISSION DATA

3.1 Introduction

Optical characterization of semiconductor and dielectric layers requires the determination of thickness and the refractive index of the layer within a certain range of the electromagnetic spectrum. Several methods have been proposed for the determination of the thick m . ness and the complex refractive index $n^* = n + i\,k$ of a thin film deposited onto a transparent substrate using experimental values of the reflectance R_f and/or transmittance T_f at normal incidence [81-88]. However, using this type of calculation frequently leads to multiple solutions, i.e. several pairs of values of (n, k). It is necessary to apply an efficient criterion in order to decide which solution is the correct one. For instance the solution which gives a nearest value to an approximate value of n such as the bulk value will be chosen. Manifacier *et al.* [82] have proposed a method of calculation, which is known as the envelope method, for deducing optical constants and thickness from the fringe pattern of the transmission spectrum of a thin transparent film surrounded by non-absorbing media. Davis *et al.* [87] have used the values of the interference maxima, at long wavelengths (where the film is essentially transparent i.e. $R_f + T_f = 1$), to calculate the optical thickness ($t_o = nt$), where t is the geometrical film thickness. Then they have used the method proposed by Cisneros *et al.* [85] to calculate the refractive index n using the magnitude of interference fringes in the corresponding reflection spectrum. For the application of this method, both the film and the substrate are assumed to be transparent, i.e. non-absorbing.

In this chapter a new method is developed to estimate the thickness and optical constants of transparent thin film deposited onto transparent substrate by using the measured transmission spectrum at normal incidence. The condition for the application of the proposed method is the existence of a region in the transmission spectrum in which the assumption $n >> k$ is valid, and n is almost independent of the incident photon wavelength. For semiconductor as well as dielectric films there is always a region in which these conditions are satisfied. While a large number of interference maxima and minima in the transmission spectrum are required for accurate determination of optical constants in the previous methods, two interference edges are sufficient to calculate t by the proposed method. Besides this advantage, the method can be used to calculate n and k from the transmission spectrum without any interference effect. The results of the proposed method will be compared to those obtained by using the envelope method. The analysis shows that the measurement of T_f over the visible or infrared region is adequate for the determination of the thickness and the optical constants of thin film by the proposed method.

3.2 Theory

Figure 3.1 represents a plane-parallel film, with a complex refractive index $n^* = n + ik$ (where n is the real part of the refractive index and k is the extinction coefficient), sandwiched between two transparent semi-finite media with refractive indices n_1 and n_3. For normal incidence the transmittance T_f and the reflectance R_f of the homogenous film are given by [89].

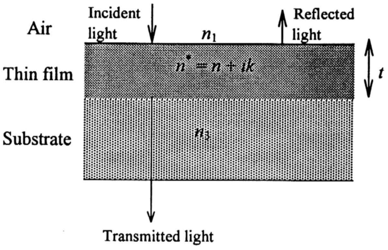


Figure 3.1: Reflection and transmission of light by a single film.

$$T_f = \frac{n_3}{n_1}\left|\frac{t_{12}t_{23}\exp(i\beta)}{1+r_{12}r_{23}\exp(2i\beta)}\right|^2 \quad \text{.....(3.1)}$$

and

$$R_f = \left|\frac{r_{12}+r_{23}\exp(2i\beta)}{1+r_{12}r_{23}\exp(2i\beta)}\right|^2 \quad \text{.....(3.2)}$$

The indices 1, 2 and 3 correspond to air, film and substrate respectively; r_{ij} and t_{ij} are the Fresnel coefficients, which for normal incidence are

$$t_{ij} = \frac{2N_i}{N_i + N_j} \quad \text{.....(3.3)}$$

$$r_{ij} = \frac{N_i - N_j}{N_i + N_j} \quad \text{.....(3.4)}$$

If N_2 is written as $N_2 = n + ik$, with n and k are real functions of λ, the complex angle β is given by

$$\beta = \frac{2\pi t N}{\lambda} = \frac{2\pi t n}{\lambda} + i\frac{2\pi t k}{\lambda} \qquad (3.5)$$

where t is the film thickness and λ is the vacuum wavelength of the incident radiation.

Since the surrounding media are assumed to be transparent then N_1 and N_3 are real quantities denoted as n_1 and n_3, respectively, where n_3 is assumed to be a known function of the wavelength. By replacing Fresnel coefficients (equations 3.3-3.5) in equations 3.1 and 3.2 and considering a very weak absorption i.e. $k^2 << (n - n_1)^2$ and $k^2 << (n - n_3)^2$, equations 3.1 and 3.2 can be approximated to

$$T_f = \frac{16 n_1 n_3 n^2 A}{C_1^2 + C_2^2 A^2 + 2C_1 C_2 A \cos(4\pi n t/\lambda)} \qquad (3.6)$$

$$R_f = \frac{B_1^2 + B_2^2 A^2 + 2C_1 C_2 A \cos(4\pi n t/\lambda)}{C_1^2 + C_2^2 A^2 + 2C_1 C_2 A \cos(4\pi n t/\lambda)} \qquad (3.7)$$

where $C_1 = (n + n_1)(n + n_3)$, $C_2 = (n - n_1)(n_3 - n)$, $B_1 = (n - n_1)(n + n_3)$, $B_2 = (n + n_1)(n_3 - n)$ and $A = exp(-4\pi kt/\lambda) = exp(-\alpha t)$, where α is the absorption coefficient of the film. Equations 3.6 and 3.7 will be used now to calculate the thickness of $ZnS_{0.9}Se_{0.1}$ film as well as n and k from the transmission spectrum obtained at normal incidence.

3.3 The proposed method of calculation

The estimation of thickness and refractive index of thin film from the transmission spectrum is presented below.

3.3.1 Calculation of thickness

This calculation is based on the existence of two interference fringes, which frequently appear in the transparent region of the transmission spectrum. Generally the transmission T_f is a function of n, k, and λ i.e. $T_f \equiv T_f(n, k, \lambda)$, thus the derivative of T_f takes the form

$$\frac{dT_f}{d\lambda} = \frac{\partial T_f}{\partial \lambda} + \frac{\partial T_f}{\partial n}\frac{dn}{d\lambda} + \frac{\partial T_f}{\partial k}\frac{dk}{d\lambda} \qquad (3.8)$$

In the region of the spectrum below the absorption edge $k \cong 0$, so that the term $dk/d\lambda$ in equation 3.8 can be ignored. Furthermore, in this region, n is a slowly varying function of λ, hence the variation of n with respect to λ can be neglected in equation 3.8. Thus the slope of an interference edge is due mostly to the term $\partial T_f/\partial\lambda$ i.e. $dT_f/d\lambda \cong \partial T_f/\partial\lambda$. From the expression of T_f (equation 3.6) and its derivative with respect to λ one can deduce two equations for thickness in terms of T_f, n, λ and $dT_f/d\lambda$ as follow

$$t = \frac{\lambda}{4\pi n}\cos^{-1}\left[\frac{16n_1n_3n^2 - T_f\left(C_1^{\,2} + C_2^{\,2}\right)}{2T_fC_1C_2}\right] - \frac{M\lambda}{2n} \qquad (3.9)$$

$$t = \frac{-2\lambda^2 n_1 n_3 n\, dT_f/d\lambda}{T_f^{\,2}\pi C_1C_2}\left[1 - \left\{\frac{16n_1n_3n^2 - T_f\left(C_1^{\,2} + C_2^{\,2}\right)}{2T_fC_1C_2}\right\}^2\right]^{-1/2} \qquad (3.10)$$

where M is an integer number. Equations 3.9 and 3.10 can be solved to determine n and t for any point at any interference edge. The slope of an interference edge, $dT_f/d\lambda$, can be found for a particular wavelength with known corresponding value of T_f of the edge. By solving Equations 3.9 and 3.10 at that particular wavelength a pair of solutions of n and t for each value of M can be obtained. These pairs of solutions can be matched with other pairs of solutions calculated for another wavelength of another interference edge but with the same magnitude of T_f as that for the previous edge. The correct pair of solution is that one with closest values of n and t for the two wavelengths, such that the difference between the values of M for two wavelengths having same values of T_f and lying onto two consecutive edges is unity and M is larger for the longer wavelength.

The transmission spectrum shown in Figure 3.2 is used as an example to calculate t and n for $ZnS_{0.9}Se_{0.1}$ thin film. Two points are chosen from two consecutive edges with $T_f = 0.825$ corresponding to wavelengths of $\lambda_1=1530$ and $\lambda_2 = 1318$ nm. The values of T_f, λ and d T_f /dλ for the two points are used to solve equations 3.9 and 3.10 using FORTRAN program. For each value of M pairs of values of n and t are obtained. Table 3.1 shows the pairs of solutions and the values of M for the points. The chosen solution gives closest values of n and t for the two points and the difference between the corresponding values of M is unity. The correct solutions are $n = 2.679$ and $t = 1760$ nm for $\lambda_1 = 1530$ nm, and $n = 2.671$ and $t = 1770$ nm for $\lambda_2 = 1318$ nm, giving an average thickness $t = 1765$ nm.

3.3.2 Calculation of *n* and *k*

Equation 3.6 can be rewritten as

$$\left(C_2^2 T_f\right)A^2 + \left(2C_1C_2T_f \cos\left[\frac{4\pi nt}{\lambda}\right] - 16n_1n_3n^2\right)A + C_1^2\, T_f = 0 \qquad (3.11)$$

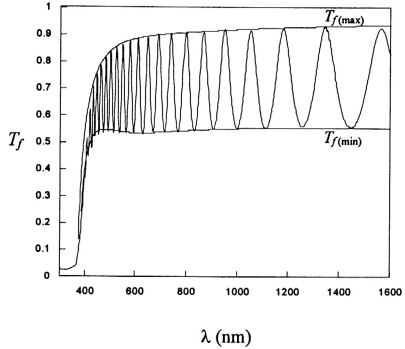


Figure 3.2: Transmittance spectrum for $ZnS_{0.9}Se_{0.1}$ film prepared by electron beam evaporation onto glass substrate at 60 °C, $T_{f(max)}$ and $T_{f(min)}$ are the fitting curves for extreme values.

Table 3.1: Values of *M*, *t* and *n* for $\lambda_1 = 1530$ nm and $\lambda_2 = 1318$ nm obtained from the proposed method.

$\lambda_1 = 1530$ nm			$\lambda_2 = 1318$ nm		
M	*t* (nm)	*n* (nm)	*M*	*t* (nm)	*n*
-8	2623	2.393	-9	2530	2.397
-7	2189	2.511	-8	2160	2.499
-6	1760	2.679	-7	1770	2.671
-5	1341	2.933	-6	1425	2.843

The roots of equation 3.11 are

$$A=\frac{-\left(2C_1C_2T_f\cos\left[\frac{4\pi nt}{\lambda}\right]-16n_1n_2n^2\right)\pm\left(\left\{2C_1C_2T_f\cos\left[\frac{4\pi nt}{\lambda}\right]-16n_1n_2n^2\right\}^2-\left(2C_1C_2T_f\right)^2\right)^{1/2}}{2C_2^{\,2}T_f}\quad\text{......(3.12)}$$

The value of t, which was calculated previously, can be used in equation 3.12 to calculate n and k for each value of λ in a two-parameter search. A FORTRAN program was used to do this search with the following steps:

(i) For a particular value of λ and its corresponding measured value of T_f, a range of n values that is in the domain of the value for the bulk material. For $ZnS_{0.9}Se_{0.1}$ film, a range of 1.9 up to 3.4 in a step of 0.0005 was chosen for n values. Each value of n as well as the values of λ and T_f were inserted in equation 3.12 to calculate the two values of A. Since

$$A=\exp\left[\frac{-4\pi kt}{\lambda}\right]\quad\text{...... (3.13)}$$

then

$$k=-\frac{\lambda}{4\pi t}\ln A\quad\text{......(3.14)}$$

So, the criteria for correct value of A is that $k \geq 0$ i.e. $0 < A \leq$. The calculation continued for the whole range of n. Many pairs of solutions of n and A were obtained for each value of λ.

(ii) These pairs of solutions of n and A were used in equations 3.6 and 3.7 to calculate $(T_f)_{cal}$ and $(R_f)_{cal}$. The correct pair, which was adopted, is the one that gave $(T_f)_{cal} \cong (T_f)_{exp}$ and $(T_f)_{cal} + (R_f)_{cal} \cong 1$.

(iii) After the value of n and A; and hence k were found for a particular λ, the calculation proceeded to the next value of λ till the whole range of λ was covered. Then the determined values of n were fitted to the Cauchy relation

$$n^2 = d_0 + d_1/\lambda^2 + d_2/\lambda^4 \qquad (3.15)$$

where d_0, d_1 and $d\ 3$ are constants

3.4 The envelope method

This method has been proposed by Manifacier *et al.* [82]. This method is based on the continuous function of the maxima $T_{f(max)}$ and minima $T_{f(min)}$ of the transmission spectrum as a function of λ through n (λ) and A (λ). The refractive index n was calculated using

$$n = \left(G + \left[G^2 - n_1^{\ 2} n_3^{\ 2} \right]^{\frac{1}{2}} \right)^{\frac{1}{2}} \qquad (3.16)$$

where $G = \dfrac{n_1^{\ 2} + n_3^{\ 2}}{2} + 2n_1 n_3 \dfrac{T_{f(max)} - T_{f(min)}}{T_{f(max)} T_{f(min)}}$

and then A was calculated from

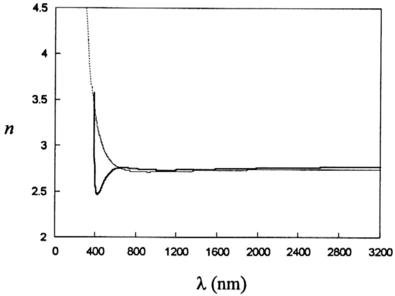
$$A = \frac{C_1\left(1 - \left[T_{f(max)}/T_{f(min)}\right]^{\frac{1}{2}}\right)}{C_2\left(1 + \left[T_{f(max)}/T_{f(min)}\right]^{\frac{1}{2}}\right)} \qquad (3.17)$$

Furthermore, the thickness t was calculated from two maxima or minima through

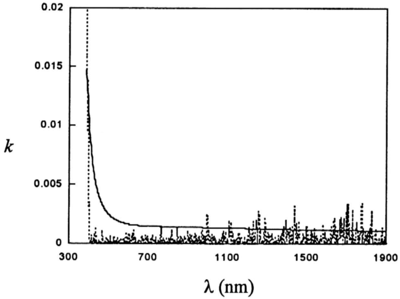
$$t = \frac{M_1 \lambda_1 \lambda_2}{2[n(\lambda_1)\lambda_2 - n(\lambda_2)\lambda_1]} \qquad (3.18)$$

where $M_1 = 1$ for two consecutive maxima or minima; λ_1, $n(\lambda_1)$, and λ_2, $n(\lambda_2)$ are the corresponding wavelengths and indices of refraction. The extinction coefficient k was calculated using the value of t and A in equation 3.14. This method was applied to the spectrum in Figure 3.2. The envelopes of the maxima $T_{f(max)}$ (λ) and the minim $T_{f(min)}$ (λ) are shown in Figure 3.2. Film thickness was calculated using equation 3.18 for two consecutive maxima and was found to be 1732 nm, which is very close to the value determined using the proposed method.

The dispersions of n and k calculated from the proposed and the envelope methods are shown in Figures 3.3 (a) and (b), respectively. Figure 3.3 (a) shows that the two methods give almost equal values of n in the range of λ > 800 nm. In the vicinity of the absorption edge, the results of the proposed method show a smooth increase in the refractive index by decreasing the wavelength. But in the envelope method calculation, the refractive index decreases slightly in the vicinity of the absorption edge and then increases drastically with decreasing λ. The accuracy of the envelope method depends on the accuracy of the fitting of the extreme values of T_f to calculate $T_{f(max)}$ (λ) and $T_{f(min)}$ (λ) functions. The valley in n values appears near 500 nm in Figure 3.3 (a) is a result of the small peak of $T_{f(min)}$ (λ) function (see Figure 3.2). However, when the minimum values of T_f near 500 nm excluded from the fitting to determine $T_{f(min)}$ (λ), the valley in n values disappeared. In Figure 3.3 (b), the slight difference in k values, which obtained from the proposed and envelope methods in the range of λ > 600 NM, is not significant while the significant difference appears in the vicinity of the fundamental optical absorption edge. It is also observed in Figure 3.3



(a)



(b)

Figure 3.3: Dispersions of n **(a)** and k **(b)** for $ZnS_{0.9}Se_{0.1}$ film as calculated using the proposed method ------- and the envelope method ____.

(b), that the proposed method gives $k \approx 0$ followed by a sharp increase in k values in the onset of the absorption edge. While the envelope method gives higher values of k. This indicates that the envelope method does not give accurate values of k near the absorption edge and this is because of the difficulty of fitting $T_{f(max)}$ and $T_{f(min)}$ in that region. The results of n, k and t estimated from the proposed and envelope methods were used in expression 3.6 to calculate T_f. Figure 3.4 shows the values of T_f, as calculated by using the proposed and the envelope methods, and R_f, as calculated from expression 3.7, along with the measured values of T_f. As can be seen from Figure 3.4, the proposed method gives exactly the same values of T_f as compared to the measured ones while there is a significant shift of the T_f values calculated by the envelope method compared to that of the measured values. This is because the proposed calculation is based on the use of the measured T_f values to solve the theoretical expression for T_f (equation 3.6), while the envelope method is extracted from continuous functions of $T_{f\,(max)}$ and $T_{f\,(min)}$ as functions of λ to solve expression 3.6. Understandably the accuracy of the calculation by the envelope method dependant on the accuracy of the fitting for $T_{f(max)}$ and $T_{f(min)}$.

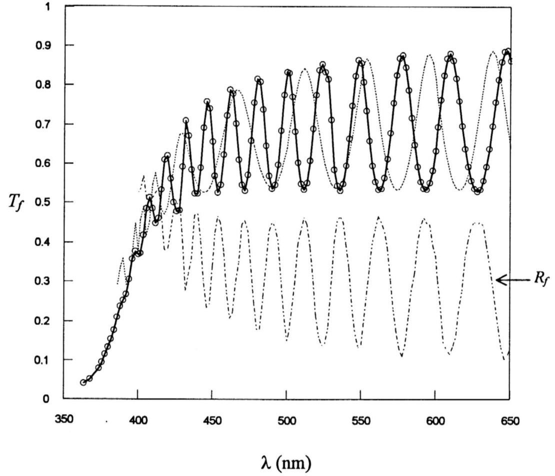


Figure 3.4: Transmittance and reflectance spectra for $ZnS_{0.9}Se_{0.1}$ thin film. T_f(measured) ○ , T_f(calculated according to expression 3.6 using the results of) the proposed method —— , the envelope method ········ and R_f ––– (calculated from expression 3.7 using the results of the proposed method).