Determination of thickness, refractive index, and thickness irregularity for semiconductor thin films from transmission spectra

Akram K. S. Aqili and Asghari Maqsood

A simplified theoretical model has been proposed to predict optical parameters such as thickness, thickness irregularity, refractive index, and extinction coefficient from transmission spectra. The proposed formula has been solved for thickness and thickness irregularity in the transparent region, and then the refractive index is calculated for the entire spectral region by use of the interference fringes order. The extinction coefficient is then calculated with the exact formula in the transparent region, and an appropriate model for the refractive index is used to solve for the extinction coefficient in the absorption region (where the interference fringes disappear). The proposed model is tested with the theoretical predicted data as well as experimental data. The calculation shows that the approximations used for solving a multiparameter nonlinear equation result in no significant errors. © 2002 Optical Society of America

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1. Introduction

Many methods have been proposed in the past 25 years to determine the optical parameters of thin films from transmission data, 1-12 rather than from transmission and reflection data. This is due to the simplicity of calibration of the spectrophotometer, which yields accurate experimental results. One of the simplest methods, introduced by Swanepoel,1 used the transmission envelope to solve the equation, analytically, for the thickness and the refractive index. However, including the thickness variation in his formula² required a numerical solution for every data point. Another interesting method by Cisneros^{3,4} for a uniform film includes the effect of substrate and solves an equation numerically for the optical parameters. In this paper we present a simple approach for determining thickness, thickness irregularity, refractive index, and extinction coefficient for semiconductor thin films.

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2. Theoretical Background

The expression for transmittance, including the reflection from the second interface of the substrate and the effect of a finite substrate, which is valid for transparent as well as weakly absorbing substrates,³ is

$$T = \frac{(1 - \rho)T_{123}U}{1 - \rho R_{321}U^2},\tag{1}$$

$$R_{321} = r_{321} r_{321}^*, (2)$$

$$T_{123} = (n_3/n_1)t_{123}t_{123}^*, (3)$$

where r_{321} and t_{123} are the amplitude of the electric field of the wave reflected and transmitted in 321 and 123 directions, respectively (illustrated in Fig. 1). These parameters are given by

$$t_{123} = \frac{t_{12}t_{23} \exp(i\psi/2)}{1 + r_{12}r_{23} \exp(i\psi)},$$
 (4)

$$r_{321} = \frac{r_{32} + r_{21} \exp(i\psi)}{1 + r_{32}r_{21} \exp(i\psi)}.$$
 (5)

A. K. S. Aqili and A. Maqsood (tpl.qau@usa.net) are with the Thermal Physics Laboratory, Department of Physics, Quaid-i-Azam University, Islamabad, Pakistan.

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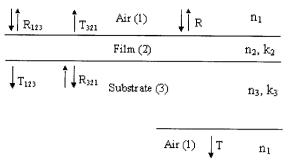


Fig. 1. Optical parameters and directions of the transmittance and reflectance, adopted in Eqs. (1)–(6).

Here r_{ij} and t_{ij} are the Fresnel coefficients of the reflected and the transmitted wave, respectively, in different regions^{3,13}

$$r_{ij} = \frac{N_i - N_j}{N_i + N_j}, \qquad t_{ij} = \frac{2N_i}{N_i + N_j}.$$
 (6)

The complex refractive index is

$$N_i = n_i + ik_i,$$

where n_i is the real part and k_i is the imaginary part (extinction coefficient) of the complex refractive index of air (n_1, k_1) , film (n_2, k_2) , and substrate (n_3, k_3) . ψ is the phase difference of the wave between two interfaces,

$$\psi = 4\pi N_2 d/\lambda = 4\pi n_2 d/\lambda + 4\pi k_2 di/\lambda = \phi + i\alpha d,$$

where d is the film thickness, λ is the wavelength, α is the absorption coefficient, and ϕ is the phase angle.

The modified Fresnel coefficients of reflected and transmitted waves at a rough film surface, ¹⁴ (1–2)

where the parameters A_1 , B_1 , B_2 , C_1 , C_2 , C_3 , D_1 , D_2 , D_3 , U, and ρ are given in Appendix A.

3. Solving for d, σ , n, and α

In the transparent region of the transmission spectra, maximum and minimum transmission occur at $^{1-5}$ $\phi = m\pi$, where $m = 1, 2, 3, \ldots$,

$$4\pi nd/\lambda_m = m\pi. \tag{11}$$

For $n_2 > n_3$, m is even at maximum transmission and odd at minima. Equation (11) could be rewritten as

$$n_2(m, \lambda) = m\lambda/4d. \tag{12}$$

We can calculate the interference fringes order (m) by assuming that the refractive index varies slowly with the wavelength^{3,4} in this region so that

$$(m-1)\lambda_{m-1} \approx m\lambda_m \approx (m+1)\lambda_{m+1} \to m$$

$$\cong \frac{\lambda_{m-1}}{\lambda_{m-1} - \lambda_m} \cong \frac{\lambda_{m+1}}{\lambda_m - \lambda_{m+1}}.$$
 (13)

Using the conditions that m is even for maxima and odd for minima makes it easy to find the value of m. In some cases, in which it is not easy to decide the value of m, for example, in thicker films, one could use a model for variation of the refractive index with the wavelength^{1,2,6} as

$$n = n_0 + g/\lambda^2, \tag{14}$$

where n_0 and g are constants, and Eq. (12) may be written as

$$4dn_m = m\lambda_m$$
, $4dn_{m+1} = (m+1)\lambda_{m+1}$, $4dn_{m-1} = (m-1)\lambda_{m-1}$. (15)

Substituting Eq. (14) into Eq. (15) and solving for m gives

$$m \approx \frac{\lambda_{m-1}^3 \lambda_{m+1}^2 + \lambda_{m-1}^2 \lambda_{m+1}^3 - \lambda_{m-1}^3 \lambda_m^2 - \lambda_{m+1}^3 \lambda_m^2}{(\lambda_{m+1} - \lambda_m)(\lambda_{m-1}^3 \lambda_{m+1} - \lambda_{m-1}^2 \lambda_{m+1}^2 + \lambda_{m-1}^3 \lambda_m - \lambda_{m-1}^2 \lambda_{m+1}^2 \lambda_m - \lambda_{m-1}^2 \lambda_m^2 + \lambda_{m+1}^2 \lambda_m^2)}.$$
 (16)

interface, where the rms height of surface irregularity $\sigma \ll \lambda$, are

$$r'_{12} = r_{12} \exp[-2(2\pi\sigma/\lambda)^2 n_1^2] = \eta r_{12},$$
 (7)

$$r'_{21} = r_{21} \exp[-2(2\pi\sigma/\lambda)^2 n_2^2] = \beta r_{21},$$
 (8)

$$t'_{12} = t_{12} \exp[-1/2(2\pi\sigma/\lambda)^2(n_1 - n_2)^2] = \gamma t_{12}.$$
 (9)

Substitution of Eqs. (2)–(9) into Eq. (1) and proceeding with careful and lengthy calculation will result in an expression for transmittance in the following simplified form,

$$T = \frac{A_1 \exp(\alpha d)}{B_1 \exp(2\alpha d) + C_1 \exp(\alpha d) + D_1} \times \frac{B_2 \exp(2\alpha d) + C_2 \exp(\alpha d) + D_2}{B_2 \exp(2\alpha d) + C_3 \exp(\alpha d) + D_3}, \quad (10)$$

Then m could be listed for all the spectra where the interference fringes appear. Knowing the value of m, we replace the refractive index of the film (n_2) at the extremes in Eq. (10) with

$$n_2(m, \lambda) = m\lambda_m/4d. \tag{17}$$

Substituting Eq. (17) into Eq. (10) and setting $k_2=0$ gives two equations for T_M [for m even $\to \cos(\phi)=1$] and T_m [for m odd $\to \cos(\phi)=-1$]. We solve these two equations by minimizing Δ , where

$$\Delta = (T_m - T_{me})^2 + (T_M - T_{Me})^2. \tag{18}$$

 T_{Me} and T_{me} are the experimental transmittance data at maxima and minima, respectively. Solving Eq. (18) for one consecutive maxima and minimagives the value of d and σ . Then the refractive index can be calculated from Eq. (17) for all maxima and

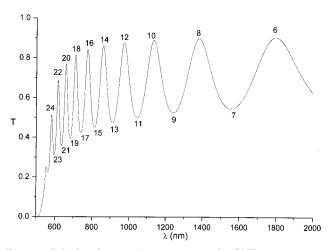


Fig. 2. Calculated transmittance curve of α -Si:H versus wavelength (λ), along with interference fringes order (m).

minima, since this equation is valid for absorption films for which the value of m could neatly be defined.³ Fitting the values of the refractive index to some known model gives the values of the refractive index for the entire spectra.

Knowing the values of d, σ , and n_2 , we can solve Eq. (10) for k_2 by minimizing Δ_1 ,

$$\Delta_1 = [T(k) - T_e]^2. \tag{19}$$

Here T(k) is the transmittance formula (10), and T_e is the experimental transmittance value.

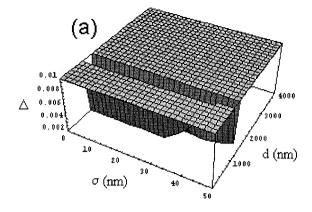
4. Simulation for α -Si:H

For checking the accuracy of the presented method, typical values 2,6 for $n(\lambda),\,\alpha(\lambda),\,d,$ and $\sigma,$ were used to reproduce the transmission curve with Eq. (10). The values were $n_2=2.6+3\times 10^5/\lambda^2,\, {\rm Log}(\alpha)=-8+1.5\times 10^6/\lambda^2,\,d=1000$ nm, $\sigma=15$ nm, $n_3=1.53,\,{\rm and}\,n_1=1.$ Interference fringes (m) calculated with relation (13) and listed for all extremes are shown in Fig. 2. Solving Eq. (18) in the transparent region as shown in Fig. 3 gave d=999.63 nm and $\sigma=15.014$ nm. The value of d was used to determine the refractive index by Eq. (17). A plot of $n_2(\lambda)$ versus $1/\lambda^2$ is shown in Fig. 4. A linear fitting gives $n_2=2.60138+3.00859\times 10^5/\lambda^2.$

From Table 1, which shows the calculated values of the thickness (d) and thickness irregularity (σ) for different values of the interference order (m) in the transparent and in the low-absorption regions, one can note that the calculated values of d are within good accuracy, better than 0.1%, whereas the accuracy of σ is better in the transparent region.

The calculated values of d, σ , and n_2 are used to determine the absorption coefficient (α) from Eq. (19) in the transparent and in the low-absorption regions, whereas in the high-absorption region, where the interference fringes disappear, the fitting parameters of the refractive index have been used.

Table 2, which gives the comparison of the calculated and the exact values of the absorption coeffi-



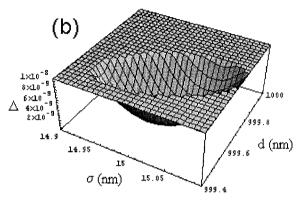


Fig. 3. Solution of Eq. (18) for d and σ , (a) large range and (b) small range.

cient for different wavelengths, shows that the calculated values of α have good accuracy in high- and low-absorption regions, whereas in the highly transparent region the accuracy was less because of a small value of α ($\sim\!10^{-6}\text{--}10^{-8}~\text{nm}^{-1}$). However, fitting the calculated values (Fig. 5) gave good accuracy, since the values of α in the transparent region have less weight than in the other regions.

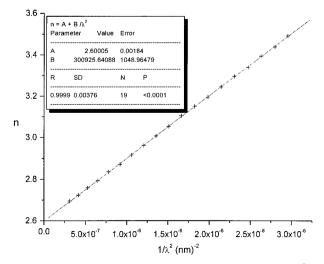


Fig. 4. Linear fitting of the refractive index (n) versus $1/\lambda^2$ for $\alpha\textsc{-Si:H}$.

Table 1. Calculated Values of Thickness (d) and Thickness Irregularity (σ) Obtained by Solution of Eq. (18) for Different Values of Interference Fringes Orders, for α -Si:H

Interference Orders (m)	λ_{M} (nm)	T_{M}	λ_m (nm)	T_m	d (nm)	σ (nm)
6,7	1796	0.90690	1556	0.54687	999.63	15.014
8,9	1379	0.89955	1241	0.52515	999.91	15.029
10,11	1134	0.88973	1044	0.50099	999.631	15.048
12,13	973	0.87727	911	0.47532	999.764	15.098
14,15	859	0.86191	814	0.44872	999.90	15.190
16,17	775	0.84243	741	0.42120	1000.58	15.432
18,19	710	0.81564	683	0.39211	1000.60	15.996
20,21	659	0.77156	636	0.35808	1000.7	17.3682
22,23	617	0.68630	597	0.31002	1000.83	20.571

Table 2. Comparison between Calculated and Exact Values of the Absorption Coefficient in Different Regions, for α-Si:H

λ (nm)	$_{(nm)^{-1}}^{\alpha_{\mathrm{exact}}}$	$\begin{array}{c} \alpha_{\rm calculated} \\ {\rm (nm)}^{-1} \end{array}$	$\lambda \; (nm)$	$\begin{array}{c} \alpha_{\rm exact} \\ \left(nm\right)^{-1} \end{array}$	$rac{lpha_{ m calculated}}{{ m (nm)}^{-1}}$
500	0.01000	0.00999	582	$2.682 imes 10^{-4}$	$2.695 imes 10^{-4}$
505	0.00762	0.00752	595	$1.783 imes 10^{-4}$	$1.878 imes 10^{-4}$
510	0.00585	0.00584	597	$1.670 imes10^{-4}$	$1.592 imes 10^{-4}$
515	0.00452	0.00452	600	$1.468 imes10^{-4}$	$1.319 imes 10^{-4}$
520	0.00353	0.00352	617	$8.714 imes10^{-5}$	$8.841 imes 10^{-5}$
525	0.00277	0.00276	636	$5.109 imes10^{-5}$	$5.138 imes 10^{-5}$
530	0.00219	0.00220	659	$2.844 imes10^{-5}$	$2.901 imes10^{-5}$
535	0.00174	0.00174	683	$1.643 imes10^{-5}$	$1.473 imes 10^{-5}$
540	0.00139	0.00139	710	$9.454 imes 10^{-6}$	$9.262 imes 10^{-6}$
545	0.00112	0.00110	741	$5.390 imes10^{-6}$	$3.297 imes 10^{-6}$
550	$9.088 imes10^{-4}$	$8.996 imes 10^{-4}$	776	$3.090 imes 10^{-6}$	$2.902 imes 10^{-6}$
555	$7.400 imes 10^{-4}$	$7.558 imes10^{-4}$	814	$1.835 imes 10^{-6}$	$1.105 imes 10^{-6}$
560	$6.069 imes 10^{-4}$	$6.301 imes10^{-4}$	859	$1.078 imes10^{-6}$	$9.538 imes10^{-7}$
565	$4.999 imes 10^{-4}$	$5.024 imes10^{-4}$	911	$6.410 imes10^{-7}$	$2.759 imes 10^{-7}$
570	$4.138 imes 10^{-4}$	$3.875 imes 10^{-4}$	972	$3.890 imes10^{-7}$	$3.659 imes 10^{-7}$
			1044	$3.516 imes10^{-6}$	$1.878 imes10^{-7}$
			1134	$1.467 imes10^{-7}$	$9.973 imes10^{-8}$

5. ZnTe Thin Film

The transmission spectra ¹⁵ of ZnTe thin film, prepared by two-sourced thermal evaporation on Corning 7059 glass substrate, recorded with a Perkin-Elmer, Lambda 19, UV-VIS-NIR (UV-visible—near-IR) spectrophotometer with UV-WinLab software, for range 400-2000 nm is shown in Fig. 6. The interference order (m) calculated from relation (13) is also listed for all extremes in Fig. 6.

The solution of Eq. (18) gives d = 745.8 nm and $\sigma = 6.17$ nm, where the refractive index of the substrate (n_3) was 1.53. The calculated refractive index of the ZnTe film, using Eq. (17), is shown in Fig. 7.

Many models^{3,4,8,10,12,16-21} could be used for fitting of such film, but the single-oscillator model^{3,4,12,16-19} is found to have the best fitting for the refractive-index values.

$$n^{2} = 1 + \frac{E_{m}E_{d}}{E_{m}^{2} - (h\nu)^{2}} = 1 + \frac{(n_{0}^{2} - 1)E_{m}^{2}}{E_{m}^{2} - (h\nu)^{2}}, \quad (20)$$

where E_m , E_d are the oscillator and dispersive energies, h is the Planck constant, ν is the photon fre-

quency, and n_0 is the refractive index of an empty lattice at infinite wave length. The calculated values of the refractive index along with fitting to Eq. (20) is shown in Fig. 7.

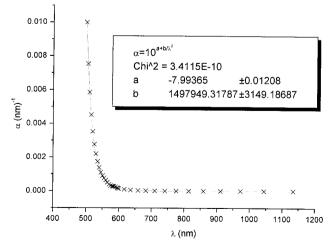


Fig. 5. Fitting of calculated absorption coefficient (α) values for α -Si:H.

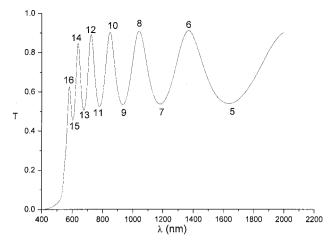


Fig. 6. Experimental transmission spectra of ZnTe film with interference fringes order (m).

The values of $\alpha,$ calculated by Eq. (19), are fitted to the Urbach relation, 16,22

$$\alpha = \alpha_0 \exp(h\nu/E_e) = \exp(a + b/\lambda), \qquad (21)$$

where α_0 , E_e , α , and b are constants related to the characteristic slope of α . It is obvious, from Fig. 8 that the Urbach relation has good fitting for $\alpha < 0.003$ nm⁻¹ (30,000 cm⁻¹).

Near the absorption edge the optical energy gap (E_g) for allowed direct transition could be calculated, with the well-known dependence $\alpha^2 \sim (h\nu - E_g)$. The energy gap is obtained by means of extrapolating the square of the absorption coefficient (α^2) versus incident photon energy $(h\nu)$. Figure 9 shows the energy gap with the values of α calculated by Eq. (19), $E_g = 2.47$ eV, and α is calculated with the following approximation α^{23-25} near the absorption edge,

$$T \sim \exp(-\alpha d)$$
. (22)

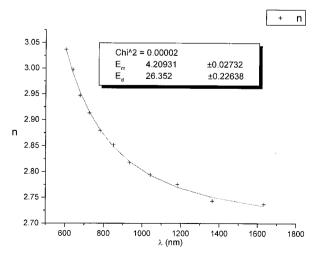


Fig. 7. Calculated values of the refractive index of ZnTe film, with fitting to Eq. (20).

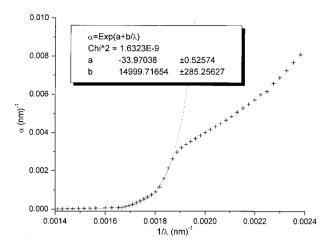


Fig. 8. Calculated values of the absorption coefficient, with fitting to Urbach relation.

 E_g obtained was 2.234 eV. Figure 10 shows the transmittance curve reproduced by Eq. (10) by use of calculated values of d and σ and the fitting parameters of n_2 and α , in the Urbach region, along with experimental transmission data, which have a good matching.

6. Discussion and Comments

The following points summarize our findings:

- 1. The formulas include most of the film and substrate parameters, which affect the transmission spectra, and are given in a simplified form.
- 2. The values of the interference fringes order (m), for films of thickness below ~ 2000 nm, could simply be determined with relation (13) by approximation of the resulting value to the closest even integer for maxima and odd integer for minima. For thicker films where the interference fringes are so close, relation (16) could be used.
- 3. The thickness, the thickness irregularity, and the refractive index of the film (with good accuracy)

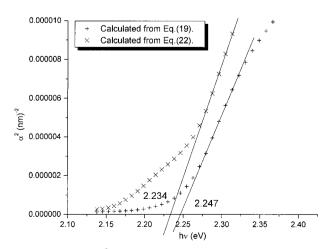


Fig. 9. Plot of α^2 versus photon energy $(h\nu)$ for ZnTe film.

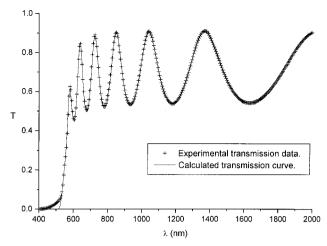


Fig. 10. Calculated transmittance curve, with experimental transmission data, versus wavelength for ZnTe film.

can be calculated by simple solution of one numerical equation.

- 4. Calculated values of the absorption coefficient or the extinction coefficient of the film, in the absorption region, where the interference fringes no longer exist, depend on the selection of model for the refractive index. With the transparent region it is calculated by use of the values of the refractive index calculated in this region.
- 5. Simulation of a tested theoretical model proved that there is no significant error due to approximation used, consider k = 0 in transparent region and m is exactly integer, even at wavelengths below 1000 nm (but not at lower wavelengths where the absorption is too high).
- 6. Comparing the rms of thickness irregularity (σ) with the value of thickness variation (Δd) calculated with the Swanepoel formula, with similar a approach, indicates that $\Delta d \sim 2\sigma$, which is due to the definition of the parameter in both formulas, whereas the values of the thickness and the refractive index were approximately same.

Appendix A

The values of the parameters in Eq. (10) are as follows:

$$\begin{split} A_1 &= \gamma^2 [16n_1n_3(1-\rho)(n_2^2+k_2^2)U], \\ B_1 &= st - \rho s \nu U^2, \qquad B_2 = st, \\ C_1 &= \beta \{ [2(4n_3k_2^2-ZY)\cos\phi+4k_2(n_3Y+Z)\sin\phi] \\ &- \rho U^2 [4k_2(Z-n_3Y)\sin\phi-2(ZY+4n_3k_2^2)\cos\phi] \}, \\ C_2 &= \beta [2(4n_3k_2^2-ZY)\cos\phi+4k_2(n_3Y+Z)\sin\phi], \\ C_3 &= \eta [2(4n_3k_2^2-ZY)\cos\phi+4k_2(n_3Y+Z)\sin\phi], \\ D_1 &= \beta^2 [u\nu-\rho t u U^2], \qquad D_2 &= \beta^2 [u\nu], \\ D_3 &= \eta^2 [u\nu], \qquad u = (n_1-n_2)^2+k_2^2, \\ \nu &= (n_2-n_3)^2+k_2^2, \qquad s = (n_1+n_2)^2+k_2^2, \end{split}$$

$$\begin{split} t &= (n_2 + n_3)^2 + k_2^2, \\ Y &= n_2^2 - n_1^2 + k_2^2, \qquad Z = n_2^2 - n_3^2 + k_2^2, \\ \rho &= [(n_1 - n_3)^2 + k_3^2]/[(n_1 + n_3)^2 + k_3^2], \\ n_3 &= n_1[1/T_s + (1/T_s^2 - 1)^{1/2}], \\ U^{-1} &= \frac{(1 - \rho)^2}{2T_s} + \left[\frac{(1 - \rho)^4}{4T_s^2} + \rho^2\right]^{1/2}, \end{split}$$

where T_s is the transmittance of the substrate and, for transparent substrate U=1, and $k_3=0$.

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