Journal of the Nigerian Association of Mathematical Physics
Volume 11 (November 2007), 423 - 428

Determination of the optical constants of a thin film supported on a thick transparent substrate

Samuel Ogo Azi[1] and Felix Okoro[2]
[1]Department of Physics, Ambrose Alli University
Ekpoma, Nigeria
[2]Department of Computer Science, Ambrose Alli University
Ekpoma, Nigeria
e-mail: soazi1@yahoo.co.uk

Abstract

The full expression for the minima of transmission and reflection spectra of a thin absorbing film are unwieldy transcendental functions of wavelength. Exact solutions of these equations for optical constants are, therefore, not single valued but can be uniquely determined at the extrema of the spectra instead. The solutions require elaborate computer iteration procedures. For ease of computation, we derived the equation for the transmission minima and reflection maxima With Mathematica™. film constants N and K of a $Ge_{28}As_{12}S_{60}$ thin film were then calculated by solving the non linear equations simultaneously. the solution converged for $1.5 < N < 24$ and $0.0 < K < 0.5$. Calculated values are at variance with published results by minkov [7] even with the same transmission and reflection data. The discrepancy arose from the r_m spectra. Interestingly, published values are in excellent agreement with straightforward calculation using another closed form equation by Swanepoel (1983) with only transmission data.

Keywords: Optical Constants, Thin Films, Semiconductors

1.0 Introduction

Optical constants of a semiconductor film point to its optoelectronic device application. Models for derivation of optical constants of evaporated polycrystalline thin films abound in scientific literature. These constants can be determined from reflectivity measurements using the KRAMERS-KRONIG dispersion relation. One advantage of this technique is that bulk specimens are suitable for reflectivity measurements. The method avoids the influence of the large number of imperfections, which can be present in thin films prepared by vacuum deposition techniques. However, results of the measurement are largely dependent on both the history and state of the surface. Heavens [1] gave a detailed review of the various methods used in the determination of optical constants. Two principal techniques are used for determination; intensity measurements using unpolarized light (usually at normal incidence) and measurements using polarized light. The first method is normally preferred because it is insensitive to polarization effects in the optical system and also, because irregularities of the surface do not produce large errors in the results. In principle, it is possible to obtain information on band structure from both reflectivity and transmission measurements.

Data obtained from optical measurements are studied mostly to determine film properties using some of the widely accepted models. Optical parameters of interest include absorption coefficient α, refractive index, n, and extinction coefficient, k. The detailed analysis of experimental data stems from the fact that these properties as reported are subtly dependent on experimental conditions. This makes difficult to compare published data in the literature [2]. On the other hand, accurate experimental data is a good test for existing models that based on idealized structure of a thin film as shown in Figure 1.

The figure depicts a thin film deposited on a thick transparent substrate. The refractive index of the surrounding air, n_o, is 1 while n_s is a constant depending on the substrate material. The film has a complex index of refraction $\hat{n}_f = n + ik$, where n is the refractive index and k the extinction coefficient.

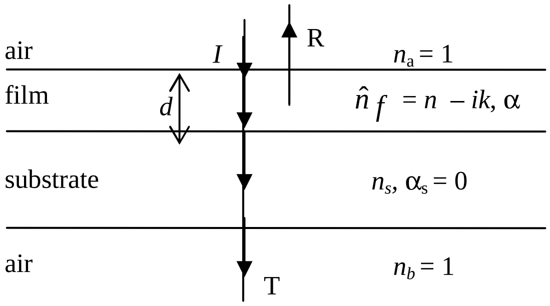

Figure 1. Thin film evaporated on a thick non-absorbing substrate

Optical constants of thin films are determined by solution of the simultaneous equation using either transmission or reflection spectra only and sometimes they are combined. Since the equations are transcendental functions of the wavelength, n(λ) and k(λ) are therefore not single valued. The solution require elaborate computer iteration procedures Lyashenko and Miloslavski [3]. Manifacier [4], Swanepoel [5] and Halindintwali [6] have uniquely determined n and k at the extrema by using only envelopes of transmission spectra. The straightforward expressions derived for calculations using this technique are sensitive to inevitable errors arising from tracing the envelopes. Minkov [7] have used transmission minima and reflection maxima instead to minimize the errors. However, unwieldy expressions involved in Minkov's method make it less elegant. Nevertheless, the cumbersome algebra can be handled with relative ease in Mathematica™.

A brief discussion of typical transmission spectra of thin films is given followed by theoretical models of the absorption spectra from which optical properties are derived. Calculated optical properties are then compared with published data on evaporated $Ge_{28}As_{12}S_{60}$ films [7].

2.0 Theory

A typical spectrum of a supported thin film on transparent substrate is shown in Figure 2. In the strong absorption region, α is sufficiently high to stop multiple reflection inside the film hence the spectra is damped and transmission decreases as with increasing film thickness. In the weakly absorbing region, transmission is weakly dependent on film thickness and is independent of thickness in the transparent region. Interference effects in the film give rise to the transmission T(λ), and reflection R(λ), spectra.

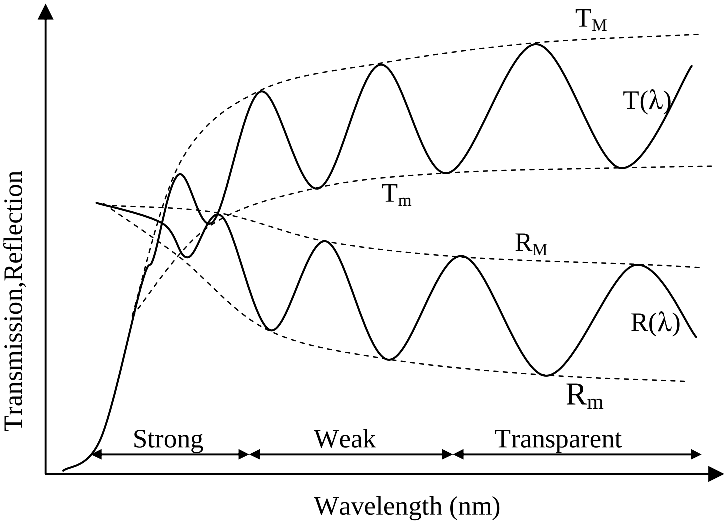

Figure 2: Typical transmission and reflection spectra of a CdS thin film evaporated on a glass substrate (full curves) and their envelopes (dashed curves).

Expressions for T(λ) and R(λ) of thin film semiconductors are derived by summing the complex amplitudes of the individual waves arising from multiple reflections at the air, film and substrate interfaces. These expressions are outlined following the method in [7]. The transmission and reflection equations, using signal flow graph are

$$T = \frac{\tau_{as}\tau_{ab}}{1-\rho_{sa}\rho_{ab}^{2}} \tag{2.1}$$

$$R = \rho_{as} + \frac{n_s^2\tau_{as}\rho_{sb}}{1-\rho_{sa}\rho_{ab}^{2}} \tag{2.2}$$

where τ_{ij} and ρ_{ij} are the amplitude coefficients of transmission and reflection between the respective media. As shown in Figure 1, the subscripts a and b refer to air; f is for the film and s for the substrate. The respective phase shifts as light crosses from air to film and film to substrate, are

$$\delta_{af} = ArcTan\left[\frac{2k}{n^2+k^2-1}\right]+\pi \tag{2.3a}$$

$$\delta_{fs} = ArcTan\left[\frac{2n_s k}{n^2+k^2-n_s^2}\right] \tag{2.3b}$$

The coefficients are further expressed as

$$\tau_{as} = \frac{\tau_{af}^2\tau_{fs}^2}{A^{-1}+\rho_{af}^2\rho_{fs}^2A+2\rho_{af}\rho_{fs}Cos[\delta_2]} \tag{2.4}$$

$$\rho_{as} = \frac{\rho_{af}^2A^{-1}+\rho_{fs}^2A+2\rho_{af}\rho_{fs}Cos[\delta_1]}{A^{-1}+\rho_{af}^2\rho_{fs}^2A+2\rho_{af}\rho_{fs}Cos[\delta_2]} \tag{2.5}$$

$$\rho_{sa} = \frac{\rho_{af}^2A+\rho_{fs}^2A^{-1}+2\rho_{af}\rho_{fs}Cos[\delta']}{A^{-1}+\rho_{af}^2\rho_{fs}^2A+2\rho_{af}\rho_{fs}Cos[\delta_2]} \tag{2.6}$$

where, $\delta_1 = -\delta_{af} + \delta_{fs} + 2\delta$; $\delta' = \delta_{af} - \delta_{fs} + 2\delta$; $\delta_2 = -\delta_{af} + \delta_{fs} + 2\delta$; $\delta = \frac{2\pi nd}{\lambda}$ and $A = e^{-\alpha d}$.

Similarly, the transmission coefficients are as follows;

$$\tau_{af} = \frac{2}{\sqrt{(1+n)^2 + k^2}} \tag{2.7a}$$

$$\tau_{fs} = \sqrt{\frac{n^2 + k^2}{(n+n_s)^2 + k^2}} \tag{2.7b}$$

$$\tau_{sb} = \frac{2n_s}{n_s + 1} \tag{2.7c}$$

while reflection coefficients are

$$\rho_{af} = \sqrt{\left(\frac{(1-n)^2 + k^2}{(1+n)^2 + k^2}\right)} \tag{2.8a}$$

$$\rho_{fs} = \sqrt{\left(\frac{(n-n_s)^2 + k^2}{(n+n_s)^2 + k^2}\right)} \tag{2.8b}$$

$$\rho_{ab} = \frac{n_s - 1}{n_s + 1} \tag{2.8c}$$

For the envelopes, T_m and R_M in the weak absorption and transparent regions, k<<n so that

$$\mathrm{Cos}[\delta_1] \approx \mathrm{Cos}[\delta'] \approx \cos[\pi + 2\delta] = 1. \tag{2.9}$$

Thus, equations (2.4), (2.5) and (2.6) subject to (2.9) give

$$\tau_{as} = \frac{\tau_{af}^2 \tau_{fs}^2}{A^{-1} + \rho_{af}^2 \rho_{fs}^2 A + 2\rho_{af}\rho_{fs} Cos[\delta_{21}]} \tag{2.10}$$

$$\rho_{as} = \frac{\rho_{af}^2 A^{-1} + \rho_{fs}^2 A + 2\rho_{af}\rho_{fs}}{A^{-1} + \rho_{af}^2 \rho_{fs}^2 A + 2\rho_{af}\rho_{fs} Cos[\delta_{21}]} \tag{2.11}$$

$$\rho_{sa} = \frac{\rho_{af}^2 A + \rho_{fs}^2 A^{-1} + 2\rho_{af}\rho_{fs}}{A^{-1} + \rho_{af}^2 \rho_{fs}^2 A + 2\rho_{af}\rho_{fs} Cos[\delta_{21}]} \tag{2.12}$$

where $\delta_{21} = \delta_{af} - \pi + \delta_{21}$. Equations (2.1) to (2.3) and (2.7) to (2.11) give rise to the full expressions for T_m and R_m as presented appendices 1 and 2 respectively. Non linear equations for T_m and R_M were solved simultaneously to determine n and k with Mathematica™ for apparent ease of calculation. The issue of singularity of the solution and convergence using Newton's iteration method has been fully discussed elsewhere [7]. The routine used in this work converged for initial choice of 1.5<n<24 and k=0.0. Swanepoel's method [5] was also used to compute optical constants using envelopes for T_M and T_m for comparison.

3.0 Results and discussions

The results are presented in Table 1. The column labeled (a) contains the calculated optical constants using T_M and T_m with MS Excel™ . Results with T_m and R_M for the constants in [7] is denoted as (b) and the calculated values in the present work as (c). Interestingly, the result in (a) is agrees well with those in (b). However, below 493nm in (a), n does not increase, as pointed out in [7], but appears to decrease to a value of 1.458 at 438nm. The value of 2.452 at 450nm likely to be an exception due to scanning error since a 0.1% decrease in T_M brings it lower than the corresponding value of 2.429 in column (b). Besides, n = 1.458 at 438nm in (a) is far less than 2.421 in (b) as reported. These facts raise a question about the remark that Swanepoel's method is less accurate because it gives higher values of n at lower wavelengths.

A more serious concern is that the values in columns (b) and (c) give different results regardless of the fact that both calculations follow a similar procedure. The results in column (b) were calculated via Newton's iteration while that in the present work for column (c) were calculated with Mathematica™. Considering the fact that the same transmission and reflection data were used for the calculations, the results from these two methods are not expected to differ. Furthermore, our recalculated values did not quite agree with simulated reflection spectra in [7]. As spot checks, we recomputed R_M at 539nm, 581nm and 683nm with the equation in appendix 2. This gave 0.2365, 0.1474 and 0.1537 instead of published values of 0.396, 0.542 and 0.564 respectively. However, the published results for T_m is consistent with the equation in appendix 1. Computed values for T_m were 0.1141, 0.3182 and 0.4260 which tallied with the published values of 0.111, 0.319 and 0.426 respectively. Meanwhile, efforts are being made to code the full equations for T_m and R_M in Fortran to repeat the calculations via Newton's iteration method.

Table1. Calculated values of the refractive index and extinction coefficient for $Ge_{28}As_{12}S_{60}$ film* using methods proposed by Swanepoel [5] and Minkov [7].

				(*a*)		(b)		(c)	
λ_{extr}	***T_M***	***T_m***	***R_M***	***n***	***k***	***n***	***k***	***n***	***k***
nm				T_M ,T_m	MS Excel™	T_m , R_M	published in [7]	T_m , R_M	present work
766	0.93	0.667	0.331	2.278	1.050E-04	2.278	9.930E-5	1.839	8.650E-03
729	0.93	0.663	0.335	2.290	9.963E-05	2.290	9.937E-5	1.855	8.220E-03
697	0.928	0.657	0.340	2.306	1.607E-04	2.307	1.434E-4	1.876	7.890E-03
668	0.926	0.651	0.344	2.322	2.164E-04	2.321	2.303E-4	1.894	7.640E-03
641	0.922	0.645	0.347	2.335	3.279E-04	2.334	3.555E-4	1.910	7.440E-03
617	0.916	0.638	0.351	2.348	4.898E-04	2.349	4.736E-4	1.927	7.270E-03
596	0.907	0.629	0.354	2.364	7.266E-04	2.364	7.128E-4	1.948	7.240E-03
575	0.892	0.617	0.356	2.380	1.113E-03	2.381	1.104E-3	1.968	7.350E-03
557	0.87	0.602	0.356	2.396	1.677E-03	2.395	1.685E-3	1.987	7.860E-03
540	0.836	0.582	0.354	2.409	2.555E-03	2.411	2.533E-3	2.006	8.220E-03
524	0.783	0.552	0.348	2.425	3.966E-03	2.427	3.942E-3	2.028	9.290E-03
509	0.704	0.508	0.336	2.442	6.222E-03	2.443	6.217E-3	2.051	1.115E-02
493	0.573	0.436	0.308	2.443	0.01060	2.444	0.01059	2.066	0.01487
477	0.372	0.318	0.265	2.322	0.02035	2.439	0.01986	2.103	0.02243
463	0.198	0.179	0.221	2.427	0.03372	2.436	0.03364	2.192	0.03562
450	0.061	0.059	0.188	2.452	0.05969	2.429	0.05986	2.326	0.06054
438	0.007	0.007	0.175	1.458	0.11281	2.421	0.10782	2.407	0.1079

* Reference [7]

(a) calculated: T_M and T_m – Eq. (5f, 11&12) [5] with MS Excel™
(b) reported: T_m and R_M - Eq. (1-8) [7]
(c) calculated: T_m and R_M - Eq. (1-3,7-11) with Mathematica™

4.0 Conclusion

An exact calculation of n(λ) and k(λ) for $Ge_{28}As_{12}S_{60}$ film thin film was carried out with Mathematica™ following Minkov [7]. The calculated optical constants with the same data set are at variance with published results in spite of the seemingly similar procedure. The discrepancy may have stemmed from the R_M spectra as calculations at three different wavelengths; 539nm, 581nm and 683nm, gave 0.2365, 0.1474 and 0.1537 instead of published values of 0.396, 0.542 and 0.564 respectively. However, results for T_m are quite consistent. As expected, straightforward calculations using only transmission maxima and minima by Swanepoel's method agree well with published values.

References

[1] Heavens, O. S., (1965), Optical Properties of Thin Solid Films.
[2] Petkov, K., (2002), "Compositional dependence of the photoinduced phenomena in thin chalcogenide films", J. Opt. and Adv. Matls, 4 (3), p. 621-629.
[3] Lyashenko, S. P., and Miloslavskii, V. K, (1964), Opt. Spectrocop. 16, pp. 80
[4] Manificier, J. C., Gasiot, J., and fillar, J. P., (1976), A simple method for the determination of the optical constants n,k and the thickness of a weakly absorbing thin film, ", J. Phys. E: Sci. Instrum., 9, pp1002-1004.
[5] Swanepoel, R., (1983),"Determination of the thickness and optical constants of amorphous silicon", J. Phys. E: Sci. Instrum., 16, pp1214-1250.
[6] Halindintwali, S. (2005), "A study of hydrogenated nanocrystalline silicon thin films deposited by hot-wire chemical vapour deposition (HWCVD)", Ph. D thesis, University of Western Cape, S. A.
[7] Minkov, D. A, (1989), "Method for determining the optical constants of a thin film on a transparent substrate, J. App. Phys. D, 22, pp199 – 205.