Computational method for determining the optical properties and its application for $CuIn(S,Se)_2$ thin films

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Abstract

We present in this work a computational method for determining the optical parameters of thin films quaternary $CuIn(S,Se)_2$ grown by Close-Spaced Vapor Transport and the variation of thickness (50 – 325 nm). The analysis of the basic properties of the films was carrie out by spectrophotometer, was measured between 800 – 2500 nm. The modified Abele-Theye method of numerical analysis was used to obtain the optical constants of the film.

Key words : Absorption, Close-Spaced Vapor Transport , quaternary $CuIn(S,Se)_2$, reflection, transmission

1. Introduction

Thin films of quaternary $CuIn(S,Se)_2$ compounds have desirable and exeptional properties for terresterial photovoltaic application [1-3]. Thus, it important that their optical constants be known. A need exists for a convenient method to determine the optical constants from relatively simple measurements made on easily used commercial apparatus. The accuracy of the determination should be better than the repeatability of the constants from one production run to another.

The reflectance $R_{(n,k)}$ and transmittance $T_{(n,k)}$ of an absorbing film of thickness d on a non-absorbing substrate at normal incidence are expressed by Murmann's exact equations [4]. Many author have applied difference methods for determining n and k [1]. The most recent methods consist of computerized algorithms intended to solve these complex equations. Several modifications have been made for spending up computation and enhancing accuracy. Bennett and Booty [5] used computer program which is basically a univariate search technique.

Their strategy is not good one of general optimization problems. Abele and Theye [6] used the Newton-Raphson method to obtain the solution of the two equations

$$T_{exp} - T_{(n,k)} = 0 \qquad R_{exp} - R_{(n,k)} = 0$$

where T_{exp} and R_{exp} are the experimentally determined values of T and R respectevely.

The main edvantage of their method is that n and k are varied simultaneously in the search for the solution and thus the convergence is improved. In the present work the Newton-Raphson algorithm was modified: hence a new computer program was applied to determine the optical constants of $CuIn(S,Se)_2$ thin films and the data were compared with those obtained by other authors.

2. Method of Computation

Fig. 1 shows the flowchart of the modified method. The program is intended to find the solution of the two simultaneous nonlinear equations

$$f_t(n,k) = T_{(n,k)} - T_{exp} = 0 \qquad (1)$$

$$f_r(n,k) = R_{(n,k)} - R_{exp} = 0 \qquad (2)$$

where $T_{(n,k)}$ and $R_{(n,k)}$ refer to Murmann's exact equations, i.e. the description of the iteration process is as follows.

a) The optical constants of the film material obtained from the literature are used as starting values, n_0 and k_0. The are entered together with the values of T_{exp}, R_{exp} and film thickness d.

b) Equations a or b is expanded in a Taylor series around the point (n_0, k_0). Ignoring higher-order partial derivatives, the resulting equations take the form

$$\left(\frac{\partial f_t}{\partial n}\right)_{no,ko} \Delta n + \left(\frac{\partial f_t}{\partial k}\right)_{no,ko} \Delta k = 0 \qquad (3)$$

$$\left(\frac{\partial f_r}{\partial n}\right)_{no,ko} \Delta n + \left(\frac{\partial f_r}{\partial k}\right)_{no,ko} \Delta k = 0 \qquad (4)$$

c) Devided the resulting linear equation c or d by the largest nonzero coefficient to get an expression for the variable, corresponding to the coefficient, in terms of the other variable.

d) Similarly, the second linear equation is processed through step b and c and the resulting expression for a variable has appeared.

e) When the processing of the two expressions is terminated, the two variables will have numerical values (i.e. a solution).

f) If the difference between the solution obtained in step e and the starting point used in step b is less than or equal to the tolerance desired, then the solution is output, otherwise the initial guess is updated by the solution and step b to step e are repeated.

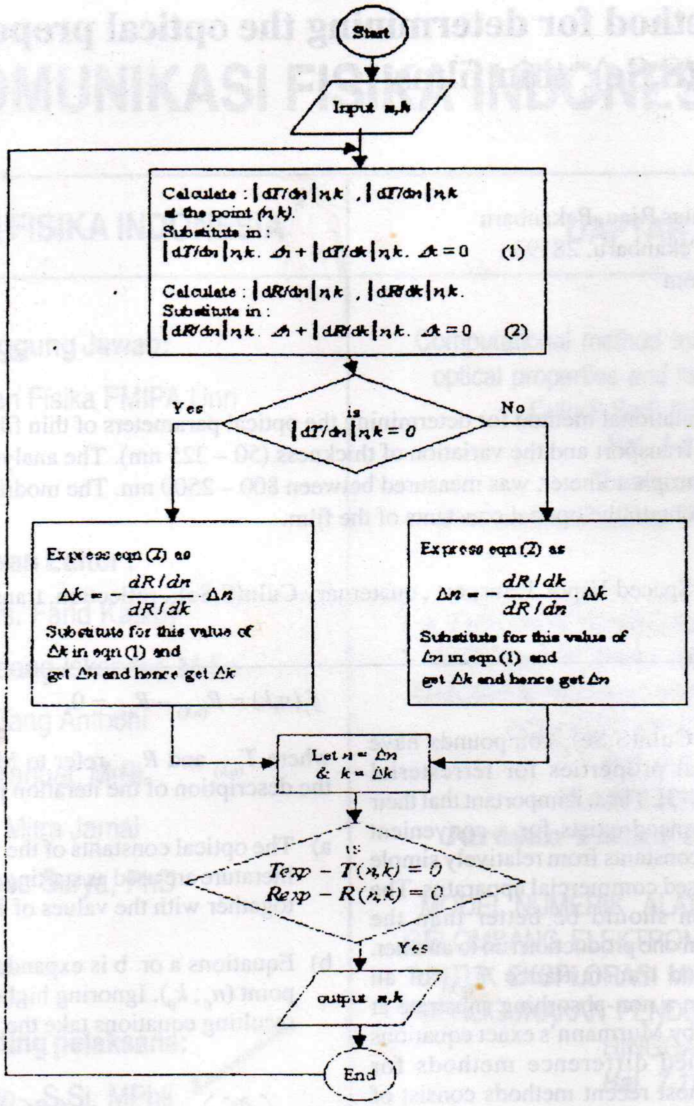

Figure 1 Flowchart of the computational method.

Results and Discussion

For comparison, data for five different wavelengths were proposed using two modes of computation: namely, the method of Abele and Theye and our modified method. Identical values for *n* and *k*, accurate to the third decimal place, were obtained. The only difference was the processing speed. The run time taken by each mode to the process the data was found to be 21 sec for Abele-Theye mode and 14 sec for our mode.

Thin films of $CuIn(S,Se)_2$ were formed by Close-Spaced Vapor Transport, five samples of difference thickness in the range 50 to 325 nm were prepared. The reflectance $R(\lambda)$ (Fig. 2) and transmittance $T(\lambda)$ (Fig. 3) at normal incidence in the wavelength range 800 to 2500 nm were carried out at room temperature using spectrophotometer (beckman UV 5270) provide with a reflection attachement, the obtained data together with the film thickness were fed to an IBM microcomputer.

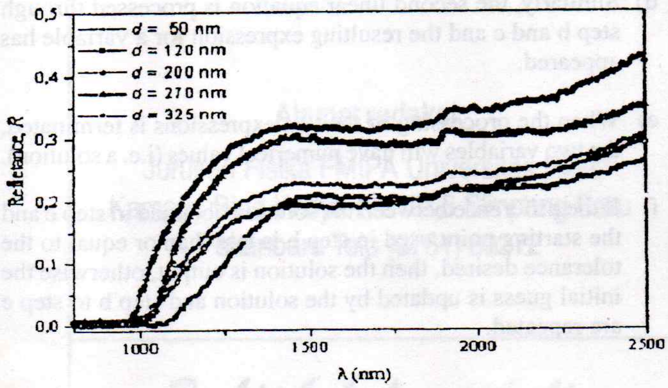

Figure 2 Spectral behaviour of the reflectance of $CuIn(S,Se)_2$ films.

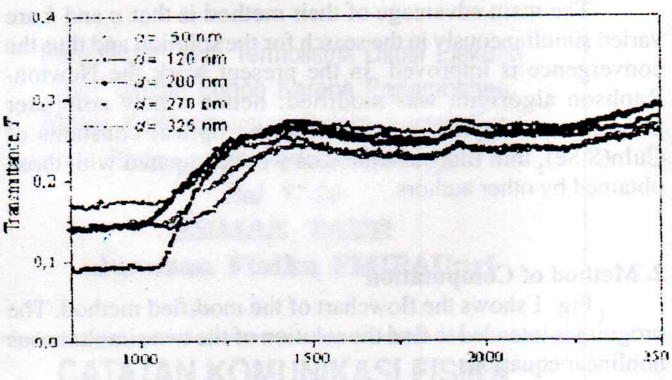

Figure 3 Spectral behaviour of the transmittance of $CuIn(S,Se)_2$ films.

The iterations proceeded using our modified Abele-Theye method, and the computed valuesof n and k are indenpendent of the film thickness. The variation lies within the associated experimental error. The mean dispersion curves of n and k are shown in Fig. 4.

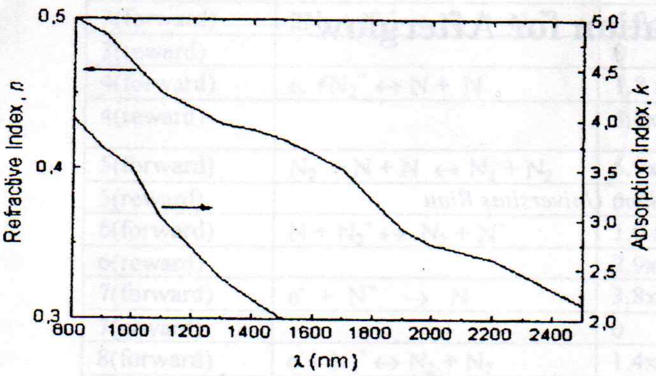

Figure 4 Main dispertion curves of the refractive index, n and the absorption index, k of $CuIn(S,Se)_2$ films.

The resʋlts are in agreement with those obtained for $CuIn(S,Se)_2$ crystal [7] and thin films was prepared by the sputtering technique [8], as indicated in Fig. 4. The spectral behaviour of the absorption coefficient, $a = 4\pi k/\lambda$ is shown in Fig. 5. The plot of a^2 against the photon energy *hí* at the absorption edge gives a straight line (Fig. 5), indicating direct allowed optical transition with eV, in complete agreement with the values eV obtained for $CuIn(S,Se)_2$ crystal for the composition x = 0.2 [9].

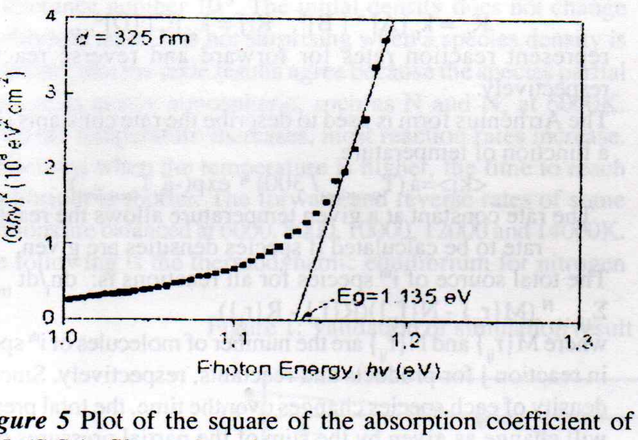

Figure 5 Plot of the square of the absorption coefficient of $CuIn(S,Se)_2$ film against photon energy.

Conclusion

Optical computational method on $CuIn(S,Se)_2$ thin films deposited by Close-Spaced Vapor Transport are reported for the first time. The optical constants were obtained by a numerical solution using an modified Abele-Theye method. The run time taken by each mode to the process the data was found to be 14 sec for our mode.

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References

1. T. L. Shay and J. M. Werwick, "Ternary Chalcopyrite Semiconductor: Growth, Electronics, Properties and Applications" (Pergamon Press, New York, 1976).
2. J. M. Meese, J. C. Manthuruthil and D. R. Locker, *Bull. Amer. Phys. Soc.* **20** (1975) 696.
3. H. Hahen and G. Strick, *Naturwissen* **54** (1967) 225.
4. O. S. Heavens. "Optical Properties of Thin Solid Films" (Dover Publications, New York 1965) p. 55.
5. J. M. Bennett and M. G. Botty, *Appl. Opt. Soc. Am.* **5** (1) (1966) 40.
6. F. Abele and M. L. Theye, *Surf. Sci.* **5** (1966) 325.
7. T. Ohashi, K. Inakoshi, Y. Hashimoto and K. Ito, *Sol. Energy Mat. and Sol. Cells* **50** (1998) 37-42.
8. Y. Yamamoto, T. Yamaguchi, Y. Demizu, T. Tanaka and A. Yoshida, *Thin Solid Films* **281-282** (1996) 372-374.
9. I.V. Bodnar, B.V. Korzun and A.I. Lukomskii, *Phys. Stat. Sol.* (b) **105**, K1 43 (1981).