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A new method for calculating the refractive index of semiconductor thin films retrieved from their transmission spectra

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ABSTRACT

A **simple new** method to calculate the refractive index dispersion **n(λ) from the** transmission spectrum of a semiconductor thin film is presented. The proposed method is based on analyzing the fringes of equal chromatic order (FECO) of thin film by solving the interference equation of light waves interfering coherently at two successive minimum and the one maximum between them. **A direct** relations for FECO order **p**, refractive index dispersion **n(λ)** at each FECO peaks and refractive index variation **δn(λ)** from one FECO peak to the next one are explicitly obtained. The method of calculation is successively applied to **a** fixed thickness ZnO, ZnS, ZnSe, ZnTe and SiO_2 as well as wedge-shape $As_{30}Se_{70}$ semiconducting thin films. The obtained refractive index dispersion data for all aforementioned semiconducting thin films agree very well with the previously reported data. In wide wavelength range (500–2500 nm), the **obtained** refractive index **dispersions n(λ) data** are fitted to Cauchy dispersion function with relative errors in calculating refractive index of order 5.4×10^{-3} at $\lambda = 800$ nm and 5.6×10^{-3} at $\lambda = 2500$ nm, respectively. Our calculation procedure yields refractive index **data** agree very well with the refractive index **data** calculated from the well known Swanepoel method. Finally, a procedure to calculate the film geometrical thickness **t** is also presented.

1. Introduction

The optical properties of thin film are very important topics in basic and applied research. For example in basic research, obtaining the material information like band structure, optical band gap, absorption coefficient, dispersion of refractive index. On the other hand, in applied research like thin film transistors, solar cells, active matrix display, image sensors and optical multilayer interference filter [1–3]. Obtaining the optical parameters of thin film stack (semiconductor layer deposited on a transparent substrate) is very active field of research and attracting wide range of experimental techniques and new methods of calculations [4]. In thin film terminology, thin film stack consists of thin absorbing layer ($t_{film} \approx 600$–1200 nm) of semiconducting material deposited on thick substrate ($t_{Subs.} \approx 1$–2 mm). Therefore, thin film stack is actually consists of three successive interfaces, which are: air-film, film-substrate and substrate-air. Due to multiple reflections and transmissions of the incident light waves at the film stack interfaces FECO fringes of thin film are produced [5]. These FECO's are observed as periodic variation of intensity in either transmission or reflection spectra. Such FECO's have been extensively analyzed to obtain a rich information about the optical properties (band gap, refractive index dispersion and spectral behavior of absorption coefficient) of thin film under investigation [6–8]. In the work published by J. C. Manifacier et al. and R. Swanepoel, they suggested so called envelop method. In this method, they proposed a construction of two continuous functions bounding the transmission or reflection spectra known as the envelop functions. The refractive index of the film is calculated by accurately identifying the contacting points between the upper and lower envelopes with maximum and minimum intensities of the FECO fringes. Applying envelop method requires difficult preparation and measurements conditions, e.g. the film under investigation must has homogenous structure and uniform thickness with very low surface roughness [9]. Using envelop method approach; the optical constants **n** and **k** and film thickness **t** have been determined at the corresponding

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maximum and minimum of FECO fringes [10–16]. However, the envelope method working well and gives good result only in the transparency region (region of weak and medium absorption ($\mathbf{k}^2 << \mathbf{n^2}$) [9,11,17]. However, the envelope method gives incorrect result for the refractive index dispersion in the region of strong absorption. Therefore, a new method for calculating the refractive index dispersion of thin film in the weak, medium as well as strong absorption region is of great demand.

Transmission spectrum of thin film stack is a relation between light intensity transmitted through the film as a function of wavelength. Most of the research groups using envelope method concentrating on developing numerical algorithms to increase the accuracy of identifying the contacting points. Consequently, **calculation of** the refractive index dispersion of thin film will be accurately identified. In contrast, the wavelength (wavenumbers) at which the FECO's maxima and minima observed are measured with high accuracy. In addition, the spectral positions of the FECO's maxima and minima are independent of any inhomogeneity or any other experimental factors could affect the intensities of the FECO's peaks. Therefore, it would be very helpful to benefit from the existing accuracy in measuring the wavelengths (wavenumbers) of the FECO's maxima and minima to extract the refractive index dispersion $\mathbf{n}(\lambda)$ and film thickness $\mathbf{t}$ up to the band edge of the investigated thin film.

In the present manuscript, a simple calculation scheme to extract the refractive index dispersion $\mathbf{n}(\lambda)$ and thickness $\mathbf{t}$ from measuring the transmission spectra at normal incidence is proposed. This choice of the angle of incidence is justified because no polarization effects will take place through the measurements. Our calculation procedure is based on solving interference equation of coherently interfering waves for two successive minimum peaks and the one maximum peak between them or visa versa. The outcome of the proposed method gives the interference orders $\mathbf{p} = \mathbf{p}(\lambda)$ of the FECO's peaks, the refractive index $\mathbf{n} = \mathbf{n}(\lambda)$ at each FECO peaks as well as the refractive index variation from one FECO peak to the next one peak $\delta\mathbf{n} = \delta\mathbf{n}(\lambda)$. In addition, our calculation method produces the optical thickness ($\mathbf{nt}$) from which the geometrical thickness of the investigated films $\mathbf{t}$ can be obtained. The proposed calculation method is applied to two groups of samples. The first group consists of four polycrystalline ZnO, ZnS, ZnSe and ZnTe semiconductor thin films of fixed thickness ($\approx$600 nm). The second group consists of two amorphous samples which are SiO_2 of fixed thickness ($\approx$592 nm) and $As_{30}Se_{70}$ of wedge like shape of average thickness ($\approx$1168 nm).

The present article will be organized as follows:

- In Sec. 2, the experimental preparation conditions and the measurements are given in detail.
- In Sec. 3, the crystal structure parameters of the investigated films are extracted from the x-ray diffraction (XRD) technique.
- In Sec. 4, the formation of the FECO's of amorphous and polycrystalline semiconductor thin films are outline.
- In Sec. 5, the calculations procedure used to calculate the order of interference, spectral variation of refractive index and refractive index change from one FECO to the next are given in detail. In addition, the refractive index dispersions for all investigated films are fitted to Cauchy dispersion function.
- In Sec. 6, the films geometrical thickness is obtained experimentally.

2. Sample preparation

Since our aim in the present manuscript is to introduce a simple method for calculating spectral variation of the refractive index of semiconductor thin film with reasonable accuracy. Therefore, our experimental part will be orientated towards producing high quality semiconducting thin film samples. The starting materials used to fabricate different types of semiconductor thin films (SiO_2, ZnO, ZnS, ZnSe, ZnTe, As and Se) were purchased from Aldrich & sigma chemical company of purity 99.999%. The starting powders were compressed in the form of circular pellets from which thin films will be deposited. On the other hand, the $As_{30}Se_{70}$, bulk material was prepared using known melt-quench technique [18]. After that, the SiO_2, ZnO, ZnS, ZnSe, $As_{30}Se_{70}$ and ZnTe bulk pellets were loaded one after the next into graphite boats positioned in the path of an accelerated electron beam (e-beam). On a very clean corning glass substrate of No. 1022, the different semiconductor films of nearly fixed thickness were deposited by an e-beam deposition technique using coating unit of type Edward 306Auto. Before the deposition process, the substrates were carefully cleaned using ultrasonic hot bath, distilled water and technical graded pure alcohol. During deposition, the vacuum system was pumped to a pressure as law as 5×10^{-7} mbar. The homogeneity and uniformity of the deposited films were fully controlled by rotating substrate holder at low speed (5 rpm) with the source to the substrate distance was set at 20 cm. During deposition process, the substrate temperature was kept at 300 k. In addition, the conditions of evaporation were fully monitored by controlling the rate of evaporation which was set at 2 nm/s. During evaporation process, the film thickness and rate of evaporation were fully monitored using thickness monitor (model: FTM6 quartz crystal) attached to the vacuum system. To produce wedge like film from $As_{30}Se_{70}$ the rotation of the substrate was stopped during the deposition. Independently, the thickness and surface quality of the produced thin films are finally studied using spectroscopic ellipsometr of model M2000V (produced by J. A. Woollam), which give thickness value for the deposited films of order 600 nm, 592 nm and 1168 nm with accuracy of about ±4.0 nm. The experimental detailed used here can be found in our previously reported results [19–21].

The crystal quality, type of structure and phase purity of the deposited films were examined by means of conventional XRD (of type Shimadzu Diffractometer XRD 6000, Japan) with Cu-$K_{\alpha 1}$ radiation ($\lambda = 1.54056$ Å). The X-ray measurements were collected in step-scan modes, in a 2θ mode between 20° and 60° (step-size of 0.02° and step time of 0.6 s). Pure Silicon~ Si 99.9999% was used as an internal standard.

The optical measurements (transmittance and reflectance) were performed in a wide wavelength range (300–2500 nm) using JASCO V670 double beam spectrophotometer. For the deposited films, all the transmitted spectra were recorded at normal incidence.

3. Structure properties of the deposited films

In Fig.1a, the XRD patterns of the as deposited polycrystalline ZnO, ZnS, ZnSe and ZnTe thin films are plotted. For the ZnO thin film, the diffraction pattern shows a strong peak originate from (002) plane ($2\theta = 34.4°$) with many peaks come from (100) ($2\theta = 31.8°$), (101) ($2\theta = 36.28°$), (102) ($2\theta = 47.6°$) and (110) ($2\theta = 56.64°$) planes. The observed peaks of the ZnO confirm the hexagonal type structure. Furthermore, for ZnO thin film the data is in very good agreement with the previously reported results based on the standard JCPDS data card No.80-0075 [22,23]. For ZnS thin film, the diffraction pattern depicts a strong peak originate from (111) plane ($2\theta = 28.6°$) with two extra peaks related to (220) ($2\theta = 47.56°$) and (311) ($2\theta = 56.36°$) planes. The observed reflection lines of the as-deposited ZnS thin film verify the cubic type structure. The result of ZnS thin film is also in very good agreement with the previously reported JCPDS data card No.80-0020 [24,25].