Physica B 454 (2014) 189–196

Contents lists available at ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Determination and analysis of optical constants for $Ge_{15}Se_{60}Bi_{25}$ thin films

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ARTICLE INFO

Article history:
Received 21 May 2014
Received in revised form
16 July 2014
Accepted 23 July 2014
Available online 4 August 2014

Keywords:
Optical constants
Approximate methods
Accurate method
Absorption edge
Dielectric constants
Relative errors

ABSTRACT

Thin films of $Ge_{15}Se_{60}Bi_{25}$ were deposited, at room temperature, on glass substrates by thermal evaporation technique. The optical reflectance and transmittance of amorphous $Ge_{15}Se_{60}Bi_{25}$ films were measured at normal incident in the wavelength range (500–2500 nm). The optical constants, the refractive index n and the absorption index k, were determined and analyzed according to different approximate methods using the transmittance measurements only and accurate method using the transmittance and reflectance measurements. Analysis of the absorption index k data reveal the values of the optical band gap E_g^{opt}, the width of tails E_e and the type of transitions. Some optical parameters such as, high frequency dielectric constant ε_∞, dispersion parameters (oscillation energy E_s and the dispersion energy E_d), real and imaginary parts of complex dielectric constant (ε_1 and ε_2) and dielectric parameters (dissipation factor tan δ, dielectric relaxation time τ, the volume and surface energy loss functions) were estimated by analyzing the refractive index n data.The relative errors for all optical parameters depending on different approximate methods were identified and discussed.

1. Introduction

Chalcogenide glasses of Ge–Se alloy are interesting materials for infrared optics. They have a wide range of transparency extending from 0.6 to 3 μm. Tohge and co-workers [1,2] were the first to point out the role of Bismuth in the appearance of n-type conduction in chalcogenide glasses for a Ge–Se–Bi system. In the Ge–Se–Bi system, Ge is known to contribute to long term room temperature stability, Se is considered as an interesting material due to its wide commercial importance and the addition of Bi to obtain n-type conduction [3]. Nagels et al. [4] studied the p-type to n-type transition in a $(Ge_3Se_5)_{100-x}Bi_x$ system. Rajagopalan et al. [5] studied the thermal and optical properties of $Ge_5Se_{77}Bi_{18}$. Kumar et al. [6] reported the decrease of the values of the optical gap with increasing Bi content in Ge–Se–Bi system. The optical properties of $Ge_{20}Se_{80-x}Bi_x$ have attracted the attention of various investigators [7–10].

Several approximate methods were used to calculate the optical constants (refractive index n and absorption index k) for semiconducting absorbing thin films deposited in transparent substrates using the data of transmittance spectrum only. Manifacier et al. [11] proposed a traditional method for extracting the optical constants and film thickness from the fringe pattern of transmittance spectrum of films deposited in the non-absorbing substrates. The particular interest of this method, besides its easiness, is that it makes a directly programmable calculation possible. Swanpoel [12,13] proposed a method based on the use the maxima and minima of the interference fringes in the transmittance spectrum to calculate the values of the refractive index n and film thickness in the weakly absorbing and transparent regions. The values of absorption index k have been determined in weak and strong absorption regions of the transmittance spectrum. Demiryont et al. [14] suggested a method to determine the optical constant and film thickness, which were deduced from transmission measurements only. An accurate method, used to determine the optical constants, was proposed by the Murmann exact equations [15]. This method is complicated in calculations of optical constants and require the measurements of reflectance as well as transmittance data.

The present work aims to determine and analyze the optical constants depending on three approximate methods as well as accurate method for amorphous $Ge_{15}Se_{60}Bi_{25}$ films with different thicknesses. The relative error of the obtained data for each methods were calculated and discussed to evaluate the accuracy of different approximate methods for studied films.

2. Experiential technique

2.1. Material preparation

Bulk $Ge_{15}Se_{60}Bi_{25}$ material was prepared from highly pure Ge, Se and Bi elements of purity 99.999%. These components were

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http://dx.doi.org/10.1016/j.physb.2014.07.068
0921-4526/

weighted in accordance with their atomic percentage. The weighting materials were placed in a clean sealed evacuated (10^{-5} Torr) silica tube, then the tube was heated to 1273 K (> m.p. of Ge) and kept constant for 24 h in an oscillatory furnace [16]. Long time of synthesis and oscillation of the tube are necessary for the homogeneity of the synthesized samples. The tube is then quenched in icy water to obtain the samples in the glassy form. Thin films of the investigated sample were obtained from bulk samples by thermal evaporation technique under vacuum and subsequent deposition on highly cleaned glass substrates. The substrate temperature was held at that of the room temperature during deposition process. The thickness of film was measured after deposition by Tolansky's interferometric method [17].

2.2. Structural identification

The chemical composition of the investigated samples was checked by energy dispersive X-ray analysis (EDX) using a JEOL 5400 scanning electron microscope. The EDX diagrams of our composition in thin film forms of different thicknesses possess a nearly stoichiometric composition $Ge_{15}Se_{60}Bi_{25}$ and there is no impurity elements in the studied composition [16]. The structure of the investigated films was examined by X-ray diffraction analysis (Shimadzu XD-D2) with Cu target and Ni filter. The loss of any sharp peak in XRD pattern reveals the amorphous nature of the studied films [18].

2.3. Optical measurements

The optical transmittance T and reflectance R of $Ge_{15}Se_{60}Bi_{25}$ films, deposited in glass substrate, were measured at room temperature (303 K) with unpolarized light at normal incidence in the wave length range (500–2500 nm) using a dual beam spectrophotometer (UV-3101 PC Shimadzu). From the experimentally data of transmittance T_{exp}, reflectance R_{exp} and the film thickness, the optical constants (refractive index n and absorption index k) were calculated using several approximate methods as well as accurate method.

2.4. Methods of calculation

2.4.1. The first approximate method

The optical constants (refractive index n and absorption index k) were determined using Swanepoel's method [12,13]. If the thickness d is uniform, interference effects give rise to the spectrum showing a maxima and minima of the transmission curve. These interference fringes can be used to calculate the optical constants of $Ge_{15}Se_{60}Bi_{25}$ films according to the following basic Eqs. [12,19–21].

2.4.1.1. In the transparent region the refractive index n is given by.

$$n = [M + (M^2 - n_s^2)^{1/2})^{1/2} \tag{1}$$

where

$$M = \frac{2n_s}{T_{min}} - \frac{n_s^2+1}{2} \tag{2}$$

2.4.1.2. In the weak and medium absorption regions, the refractive index n is given by [19,20].

$$n = [N + (N^2 - n_s^2)^{1/2})^{1/2} \tag{3}$$

where

$$N = 2n_s \frac{T_{max} - T_{min}}{T_{min}T_{max}} - \frac{n_s^2+1}{2} \tag{4}$$

To calculate the absorption index k, the absorbance x must be given in terms of the interference extremes using the following relations:

$$x = \frac{E_M - [E_M^2 - (n^2-1)^3(n^2-n_s^4)]^{1/2}}{(n-1)^3(n-n_s^2)} \tag{5}$$

where

$$E_M = \frac{8n^2 n_s}{T_{max}} + (n^2-1)(n^2-n_s^2) \tag{6}$$

and

$$x = \exp\left(\frac{-4\pi kd}{\lambda}\right) \tag{7}$$

2.4.1.3. For the strong absorption region, where the interference maxima and minima converge to a single curve T_O, the absorbance x is given by.

$$x = \frac{(n+1)^3(n+n_s^2)}{16\, n^2 n_s} T_O \tag{8}$$

2.4.2. The second approximate method

This method was proposed by Manifacier et al. [11], according to the following equations to determine the optical constants [11,21]:

2.4.2.1. For the refractive index n.

$$n = [N + (N^2 - n_o^2 n_s^2)^{1/2})^{1/2} \tag{9}$$

where

$$N = \frac{n_o^2 + n_s^2}{2} + 2n_o n_s \frac{T_{max} - T_{min}}{T_{min}T_{max}} \tag{10}$$

2.4.2.2. For the absorption index k.

$$x = \frac{c_1[1 - (T_{max}/T_{min})^{1/2}]}{c_2[1 + (T_{max}/T_{min})^{1/2}]} \tag{11}$$

where

$$c_1 = (n+n_0)(n_s+n) \ , \ c_2 = (n-n_0)(n_s-n) \ \text{and} \ x = \exp\left(\frac{-4\pi kd}{\lambda}\right) \tag{12}$$

2.4.3. The third approximate method

This method was proposed by Demiryont et al. [14]. The basic equations for determination of the optical constant according to this method are written as follows [14,21]:

2.4.3.1. For refractive index n.

$$n = \left(\frac{1}{2}\right)[8n_s C + \{n_s+1\}^2)^{1/2} + (8n_s C + \{n_s-1\}^2)^{1/2} \tag{13}$$

where

$$C = \frac{T_{max} - T_{min}}{2T_{max}T_{min}} \tag{14}$$

2.4.3.2. For absorption index k.

$$k = \left(\frac{\lambda}{4\pi d}\right) L \tag{15}$$

where

$$L = \ \ln \frac{U + [U^2 - C^2 + \sigma]^{1/2}}{2a} \tag{16}$$