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New quaternary Ge–Se–Sb–Ag optical materials: Blue shift in absorption edge and evaluation of optical parameters



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ABSTRACT

We report the effect of replacement of antimony by silver on the optical constants of new quaternary chalcogenide $Ge_{20}Se_{60}Sb_{20-x}Ag_x$ (x=0,5,10,15 and 20 at.%) thin films. Films of $Ge_{20}Se_{60}Sb_{20-x}Ag_x$ glasses were prepared by thermal evaporation of the bulk samples. The transmission spectra, $T(\lambda)$, of the films at normal incidence were obtained in the spectral region from 400 nm to 2500 nm. A straightforward analysis proposed by Swanepoel has been applied to derive the real and imaginary parts of the complex index of refraction. Increasing silver content is found to affect the refractive index and the extinction coefficient of the studied films. Optical absorption measurements show that the fundamental absorption edge is a function of composition. With increasing silver content the static refractive index decreases from 2.71 to 2.49 while the optical band gap increases from 1.37 eV to 1.55 eV.

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1. Introduction

In recent years, research has been more focused on chalcogenide glasses because these materials find applications in diverse sectors like fiber optics, xerography, semiconductor devices, transistors, detectors and novel memory devices [1]. Among various chalcogenide glasses, the ternary Ge-Se-Sb chalcogenide glasses are finding applications in far infrared region due to their high transparency in 2-14 µm region and high nonlinear properties [2,3]. The properties of glasses are worked out by adding fourth element (Ge-Se-Sb-Y, where Y = As, In, Te, etc.) to Se-rich Ge-Se-Sb chalcogenide glasses [4-7]. The addition of fourth element act as a glass modifier forming Y-Se bonds in the glass network. Thus, Ge-Se and Sb-Se bonds are reducing in the main network of Ge-Se-Sb glasses. This will create the configurational disorder in the network of glass and thereby will affect the structural, thermal, electrical and optical properties remarkably. This has been reported in literature [3]. Moreover, multi-component chalcogenide glasses having heavy elements are fascinating because of their plenteous structural features and typical physical properties that are relevant in thermoelectric devices [8].We

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have chosen silver as the fourth element. Since silver doped chalcogenide glasses are finding applications in various sectors like selective ion electrodes, phase change memories, holographic recording media and programmable metallization cell memories [9]. Ag can be doped using various methods like by either irradiation by UV light [10], thermal treatment [11] or melt quench technique [12].

In our present work, we report the synthesis and optical properties of new quaternary Ag doped Ge–Se–Sb chalcogenide glasses. We have employed the melt quench technique and the thin films of bulk alloys are synthesized from thermal evaporation technique. Transmission spectra of thin films are used to determine various optical parameters. The dispersion of refractive index has been analyzed using Wemple–DiDomenico single oscillator model [13,14]. We have employed two methods to evaluate high frequency dielectric constant [15,16]. Tauc plot has been used to estimate the optical band gap [17].

2. Experimental details

Different compositions of bulk $Ge_{20}Se_{60}Sb_{20-x}Ag_x$ (where x = 0, 5, 10, 15 and 20 at.%) chalcogenide glasses were prepared from high purity (99.999%) constituent elements by the melt-quenching technique. The elements were heated together in an evacuated (10^{-4} Pa) silica ampoule up to 1273 K at a heating rate of 1.7 K/min. Through the heating process, the ampoules were regularly shaken to ensure homogeneity of the melt. The melt was maintained at 1273 K for 20 h then at

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1073 K for 4 h before quenched into ice-cooled water to avoid crystallization. Thin films of $Ge_{20}Se_{60}Sb_{20-x}Ag_x$ were prepared by thermal evaporation of the bulk samples. The thermal evaporation process was performed inside a coating (Edward 306E) system, at a pressure of approximately 10^{-4} Pa. During the deposition process (at normal incidence), the substrates were suitably rotated in order to obtain films of uniform thickness.

The elemental compositions of the investigated specimens were checked using the energy dispersive X-ray (Link Analytical Edx) spectroscopy. The deviations in the elemental compositions of the evaporated thin films from their initial bulk specimens were found not to exceed 0.8 at.%. The amorphous state of the films was checked using X-ray (Philips type 1710 with Cu as a target and Ni as a filter, λ = 1.5418 Å) diffractometer. The absence of crystalline peaks confirms the amorphous state of the prepared samples. A double beam (Jasco V-630) spectrophotometer was used to measure the transmittance for the prepared films in the spectral range of wavelength from 400 nm to 2500 nm. Without a glass substrate in the reference beam, the measured transmittance spectra were used to calculate the optical constants of the films.

3. Results and discussion

To calculate the optical parameters, let us consider the film has a thickness t and a complex refractive index $(n^* = n - ik)$ where n is the refractive index of film and k is absorption index of film. The films are deposited on smooth glass substrates of refractive index (s) and the thickness of the substrate is very-very large in comparison to the film thickness. The large thickness of the substrate helps to destroy its own interference effects.

Fig. 1 shows the transmission spectra of Ge₂₀Se₆₀Sb_{20-x}Ag_x (x = 0, 5, 10, 15 and 20) thin films. T_s is the transmittance of the substrate alone. The smooth interference fringes as shown in figure confirm the homogeneity and uniformity of the deposited samples. The fringes in the high transmission region are due to the interference of light in the film sample. These fringes in the transmission spectrum can be used to determine the refractive index (n) and absorption index (k) by employing the Swanepoel method [18]. The refractive index shows a normal dispersion and has been found to decrease with an increasing content of silver (Fig. 2). The detailed procedure for the calculation of refractive index may be seen elsewhere [19]. We can clearly see a blue shift in the absorption edge with an increase in the silver content on replacing Sb in Ge₂₀Se₆₀Sb_{20_x}Ag_x glasses. It is emphasized here that following the fundamental Kramers–Kronig relation $(n(0) = 1 + (1/2\pi) \int_0^\infty \alpha d\lambda)$, the blue-shift in the transmission spectrum must inevitably give a decreased value of refractive index with an increase in silver content. Though these glasses have high percentage transmission i.e. >80% in the near infrared region (800-2500 nm) still they might have high nonlinearities in comparison to some other optical materials (discussed later).

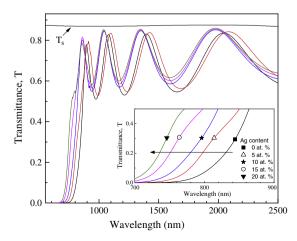


Fig. 1. Transmission spectra of $Ge_{20}Se_{60}Sb_{20-x}Ag_x$ (x = 0, 5, 10, 15 and 20 at.%) thin films. T_s is the transmission of the substrate alone.

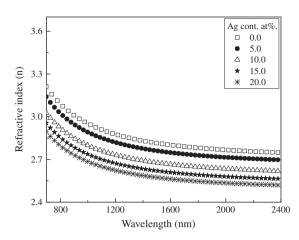


Fig. 2. Refractive index dispersion spectra for $Ge_{20}Se_{60}Sb_{20-x}Ag_x$ (x = 0, 5, 10, 15 and 20 at.%) thin films calculated according to Cauchy dispersion relationship [16].

If n_1 and n_2 are the refractive indices at two adjacent maxima or minima at two wavelengths λ_1 and λ_2 then the film thickness can be determined from the relation [18]; $t = \lambda_1 \lambda_2 / 2(\lambda_1 n_2 - \lambda_2 n_1)$. The calculated values of t have been used to find $t_{\rm correc}$ by making use of the basic interference equation $2nt_{\rm correc} = m\lambda$, where the order m is an integer for maxima and a half order for minima. The thicknesses of the films calculated from spectra are in the range 745–765 nm. These values are in good agreement with those observed from thickness monitor while depositing the thin films. The percentage variation in thickness is under 2%, so the effect of thickness can be neglected while studying the other optical parameters.

Furthermore, the dispersion of refractive index in the visible and near IR region has been analyzed using the single effective oscillator model proposed by Wemple–DiDomenico [14], which is mathematical expressed by; $n^2-1=E_dE_0/\{E_0^2-(h\nu)^2\}$, where E_0 is the energy of the effective single oscillator and also called as average energy gap, E_d is the dispersion energy which corresponds to the average strength of interband optical transitions and $h\nu$ is the photon energy. E_d is very nearly independent of E_0 . This is because E_d is proportional to the dielectric loss (ε_i) whereas E_0 does not depend on ε_i . Fig. 3 shows refractive index factor $(n^2-1)^{-1}$ vs. $(h\nu)^2$ plots. The plots are fitted for straight lines and the values of E_0 and E_d are obtained from the slope and intercept of fitted straight lines. The values of E_0 and E_d are given in Table 1. The values of E_0 go on increasing with an increase of silver

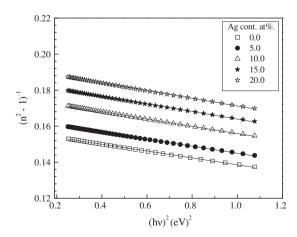


Fig. 3. Plots of refractive index factor $(n^2 - 1)^{-1}$ vs. $(hv)^2$ for $Ge_{20}Se_{60}Sb_{20-x}Ag_x$ (x = 0, 5, 10, 15 and 20 at.%) thin films.

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