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Study of third order nonlinearity of chalcogenide thin films using third harmonic generation measurements

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ABSTRACT

Third order nonlinear susceptibility of $(GeSe_{3.5})_{100\text{-}x}Bi_x$ (x = 0, 10, 14) and $Zn_xS_ySe_{100\text{-}x\text{-}y}$ (x = 2, y = 28; x = 4, y = 20; x = 6, y = 12; x = 8, y = 4) amorphous chalcogenide thin films prepared using thermal evaporation technique is estimated. The dielectric constant at incident and third harmonic wavelength is calculated using "PARAV" computer program. 1064 nm wavelength of Nd: YAG laser is incident on thin film and third harmonic signal at 355 nm wavelength alongwith fundamental light is obtained in reflection that is separated from 1064 nm using suitable optical filter. Reflected third harmonic signal is measured to trace the influence of Bi and Zn on third order nonlinear susceptibility and is found to increase with increase in Bi and Zn content in $(GeSe_{3.5})_{100\text{-}x}Bi_x$, and $Zn_xS_ySe_{100\text{-}x\text{-}y}$ chalcogenide thin films respectively. The excellent optical nonlinear property shows the use of chalcogenide thin films in photonics for wavelength conversion and optical data processing.

1. Introduction

For many optical device applications like optical switching and optical signal processing, highly efficient nonlinear optical materials are required. Amorphous chalcogenide glasses possess unique property of infrared transmission and high third order optical nonlinearity that is 27,000 times than that of silica glasses [1,2]. Chalcogenide glasses are one of the most important materials in all-optical signal processing also due to very low two-photon absorption and no free carrier absorption at telecommunication walelengths [3,4]. Nonlinear optical properties of $Sn_1As_{20}S_{79}$ strip waveguide has been studied by L. E. Zou et al. [5] for its use as integrated optical component in supercontinuum generation, parametric conversion etc. Nonlinear optical effects in strip waveguide are also enhanced by induction of band gap light due to increase in full width at half maximum spectral width [5]. Furthermore different chalcogenide microstructured optical fibers have been designed with promising applications in mid-IR guiding, infrared light transport and delivery, generation of new infrared sources, and infrared spectroscopy [6].

The study of optical nonlinearity in Bi, Zn added chalcogenide glasses is interesting due to their high glass transition temperature, high crystallization temperature, thermal stability, large IR transmission range, small ageing effects, high linear and nonlinear refractive indices [7–10]. The motivation for this study is to deeply investigate the nonlinear properties of thin films by third harmonic generation method [11]. Armstrong et al. [12] first reported the theoretical analysis of third harmonic generation in 1962 and Maker et al. [13] performed THG experiments in isotropic crystals.

The objective of the present work is to study the optical nonlinear properties of $(GeSe_{3.5})_{100\text{-}x}Bi_x$ (x = 0, 10, 14) and $Zn_xS_ySe_{100\text{-}x\text{-}y}$ (x = 2, y = 28; x = 4, y = 20; x = 6, y = 12; x = 8, y = 4) chalcogenide thin films. Transmission spectra is used as input file in "PARAV" computer program for estimation of optical parameters like refractive index, optical band gap and linear absorption coefficient of $(GeSe_{3.5})_{100\text{-}x}Bi_x$ chalcogenide films. Reflected third harmonic signal in harmonic generation is analyzed for measurement of third order nonlinear optical susceptibility ($\chi^{(3)}$).

2. Experimental

Chalcogenide glasses are prepared by melt quench method that involves melting of high purity elemental constituents in sealed ampoules and then quenching of melt in ice cold water [14]. Thin films of bulk chalcogenide glass are deposited on ultrasonically cleaned glass substrate by thermal evaporation technique.

1064 nm wavelength is incident on chalcogenide thin film using Nd:

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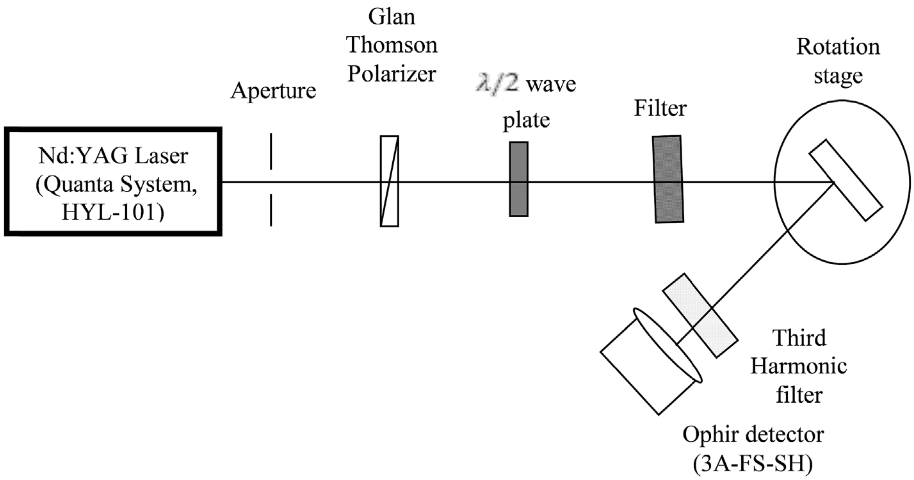

Fig. 1. Measurement of reflected third harmonic signal power.

Table 1
The optical parameters of amorphous $(GeSe_{3.5})_{100-x}$ Bi_x chalcogenide thin films.

Parameters	$(GeSe_{3.5})_{100}Bi_0$	$(GeSe_{3.5})_{90}Bi_{10}$	$(GeSe_{3.5})_{86}Bi_{14}$
$E_g(eV)$	1.62	1.86	1.75
n_{1064}	2.53	2.49	2.71
n_{355}	3.75	4.73	5.19
$\alpha_{1064}(10^5cm^{-1})$	0.01	0.02	0.04
$\alpha_{355}(10^5cm^{-1})$	1.46	1.01	1.59
$\chi^{(3)}$ $(10^{-6}esu)$	2.73 ± 0.13	4.75 ± 0.29	14.06 ± 0.22

Table 2
Optical parameters of amorphous Zn_x-S_y-$Se_{100-x-y}$ chalcogenide thin films.

Parameters	$Zn_2S_{28}Se_{70}$	$Zn_4S_{20}Se_{76}$	$Zn_6S_{12}Se_{82}$	$Zn_8S_4Se_{88}$
$E_g(eV)$	1.99	1.96	1.94	1.85
n_{1064}	2.46	2.64	2.59	2.56
n_{355}	3.08	2.94	3.21	3.42
$\alpha_{1064}(10^5cm^{-1})$	0.02	0.04	0.03	0.05
$\alpha_{355}(10^5cm^{-1})$	1.04	1.12	0.99	0.73
$\chi^{(3)}$ $(10^{-7}esu)$	1.91 ± 0.06	2.53 ± 0.13	3.79 ± 0.06	6.18 ± 0.04

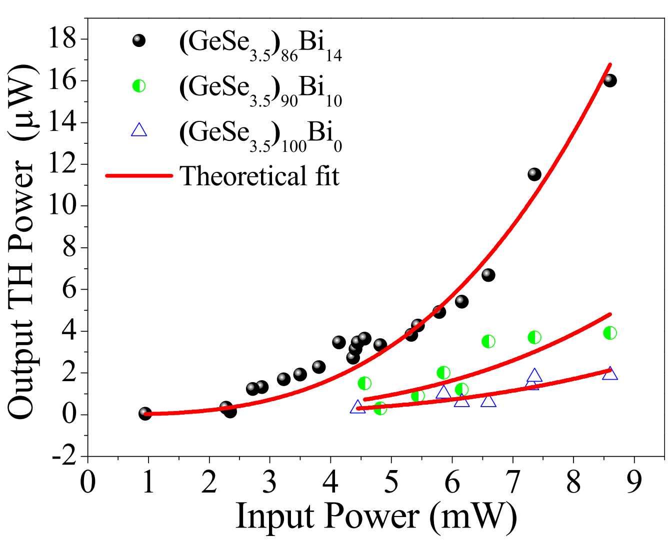

Fig. 2. Third harmonic beam power as a function of input beam power in $(GeSe_{3.5})_{100-x}$ Bi_x chalcogenide thin films.

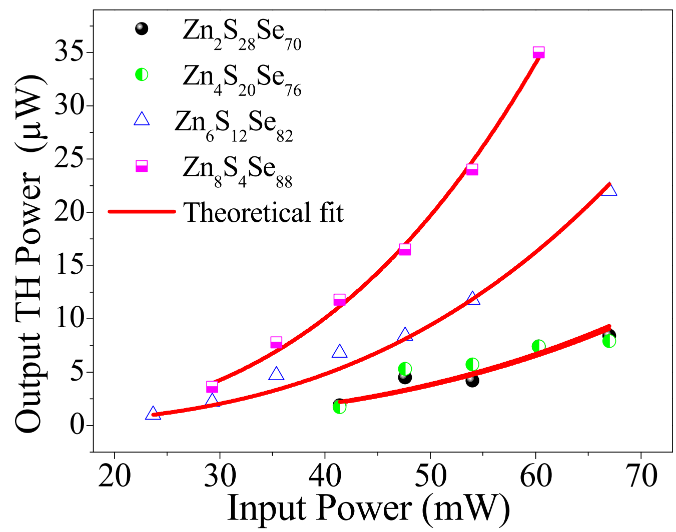

Fig. 3. Third harmonic beam power as a function of input beam power in Zn_x-S_y-$Se_{100-x-y}$ chalcogenide thin films.

YAG laser (Quanta System, HYL-101) of 5ns pulse duration and 10 Hz repetition rate as shown in Fig. 1. S-polarization of incident laser beam is obtained using Glan Thomson polarizer followed by a half wave plate. Thin film is placed on a rotation stage with resolution of 0.04^0 for particular angle of incidence (θ_i). Third harmonic light at 355 nm alongwith incident light is obtained in reflection that is used for estimation of third order nonlinear susceptibility. Suitable optical filter separates third harmonic signal from incident light and Ophir detector (3A-FS-SH) is employed for measuring power of separated third harmonic light.

3. Results and discussion-

To calculate optical constants like refractive index and linear absorption coefficient of $(GeSe_{3.5})_{100-x}Bi_x$ (x = 0, 10, 14) chalcogenide films, transmission spectra is used as input file in "PARAV" program [15]. The estimated values of linear refractive indices (n_{1064}, n_{355}) and absorption coefficients (α_{1064}, α_{355}) of chalcogenide thin films at two different wavelengths (1064 nm and 266 nm) are tabulated in Tables 1 and 2. As atomic size of Bi is larger than Ge and Se, polarizability increases with increasing Bi concentration and according to Lorentz–Lorenz relation, the refractive index will become larger with the increase of polarizability [16]. Same is observed in Zn_x-S_y-$Se_{100-x-y}$ thin