Optics Communications 403 (2017) 352–357

Contents lists available at ScienceDirect

Optics Communications

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

Measurement of ultrafast optical Kerr effect of Ge–Sb–Se chalcogenide slab waveguides by the beam self-trapping technique

Tintu Kuriakose [a,*], Emeline Baudet [b], Tomáš Halenkovič [c], Mahmoud M.R. Elsawy [d], Petr Němec [c], Virginie Nazabal [b,c], Gilles Renversez [d], Mathieu Chauvet [a]

[a] Department of Optics, FEMTO-ST Institute, UMR CNRS 6174, Université Bourgogne Franche-Comté, 15B avenue des Montboucons, 25030 Besançon, France
[b] Equipe Verres et Céramiques-Institut des sciences chimiques de Rennes (ISCR), UMR 6226 Université de Rennes 1-CNRS, Campus de Beaulieu, 35042 Rennes Cedex, France
[c] Department of Graphic Arts and Photophysics, Faculty of Chemical Technology, University of Pardubice, 53210 Pardubice, Czech Republic
[d] Aix Marseille Univ, CNRS, Centrale Marseille, Institut Fresnel, Marseille, France

ARTICLE INFO

Keywords:
Ultrafast nonlinear optics
Nonlinear optical materials
Slab waveguides
Kerr effect
Spatial solitons

ABSTRACT

We present a reliable and original experimental technique based on the analysis of beam self-trapping to measure ultrafast optical nonlinearities in planar waveguides. The technique is applied to the characterization of Ge–Sb–Se chalcogenide films that allow Kerr induced self-focusing and soliton formation. Linear and nonlinear optical constants of three different chalcogenide waveguides are studied at 1200 and 1550 nm in femtosecond regime. Waveguide propagation loss and two photon absorption coefficients are determined by transmission analysis. Beam broadening and narrowing results are compared with simulations of the nonlinear Schrödinger equation solved by BPM method to deduce the Kerr n_2 coefficients. Kerr optical nonlinearities obtained by our original technique compare favorably with the values obtained by Z-scan technique. Nonlinear refractive index as high as $(69 \pm 11) \times 10^{-18}$ m^2/W is measured in $Ge_{12.5}Sb_{25}Se_{62.5}$ at 1200 nm with low nonlinear absorption and low propagation losses which reveals the great characteristics of our waveguides for ultrafast all optical switching and integrated photonic devices.

1. Introduction

The characterization of nonlinear optical properties has an important role in modern photonic functionalities. Different techniques have been employed to determine the third order nonlinear optical constants of bulk/film samples including self-phase modulation (SPM) [1], Mach–Zehnder technique (MZT) [2], degenerate four wave mixing (DFWM) [3], two-photon absorption spectroscopy [4], optical Kerr gate [5], and Z-scan [6,7]. The latter technique is the most widely used one and is appropriate to characterize both bulk material and thin films. The Z-scan is suitable to determine both the real and imaginary part of the nonlinear refractive index. This method is based on the analysis of the diffraction modification due to nonlinear effect of a beam focused in the sample under study while the sample is moved longitudinally. For accurate measurements, it requires samples with very good homogeneity. In addition, this technique cannot be used in very thin layers since the induced beam change becomes indiscernible. Moreover, the thin film under test is often deposited on a substrate that can perturb or prevent the measurement. Unlike Z-scan, a single laser shot is enough in MZT technique to measure the nonlinearity. Nevertheless, the complex experimental setup based on the pump-probe experiment of the Mach–Zehnder interferometer can be cumbersome. Many techniques have also been developed to analyze nonlinearities in 2-D waveguides. For instance, the SPM technique, which is based on the analysis of the spectral broadening of a beam as a function of intensity, allows n_2 measurements of 2-D waveguides. Likewise, DFWM technique is also well suited to analyze third order susceptibility tensor of 2-D waveguides but it requires injection of synchronized pulses at different wavelengths. Principally, none of these techniques is well suited to study nonlinear properties in very thin layers that form planar waveguides. In this paper, we propose a technique that is convenient to determine the Kerr effect in such structures. The method is based on the direct analysis of the influence of the non-linear effect on the spatial light distribution of a beam propagating in the slab waveguide. The proposed method has several merits. The experimental setup is simple and one laser shot

---

[*] Corresponding author.
E-mail address: tintu.kuriakose@femto-st.fr (T. Kuriakose).

http://dx.doi.org/10.1016/j.optcom.2017.07.061
Received 6 June 2017; Received in revised form 20 July 2017; Accepted 21 July 2017
0030-4018/© 2017 Elsevier B.V. All rights reserved.

could be sufficient to deduce the Kerr coefficient. The sensitivity is very good especially when self-trapping leads to the formation of spatial soliton. The method can be used even in multimode waveguides and is relatively immune to perturbations due to the substrate. In addition, the proposed technique can be applied to characterize any materials that are deposited under thin films.

Moreover, identifying optical materials for ultrafast all-optical signal processing and more generally nonlinear photonic devices fabrication [8] have attracted researcher's attention over the past decades. Key materials such as silicon [9] or III–V compounds [10,11] have been investigated. Although excellent results have been obtained for given spectral range, the quest for better material with stronger Kerr coefficient, lower two-photon absorption (TPA), negligible free carrier absorption and low-cost processing techniques are still relevant. Chalcogenide glasses that have large Kerr nonlinearity, ultrafast response time, and optical transmittance in the infrared are among the materials that could fulfill part of these requirements. While few chalcogenide compositions and systems such as As_2S_3 [3], As_2Se_3 [12], Ge–As–S(Se) [13], and Ge–Sb–S [14,15] have been intensely explored for nonlinear optical properties purposes, new chalcogenide glasses are still synthesized, with the hope that a high-bit-rate optical processing system operating at low peak power can be reached. In the present work, the proposed characterization technique is applied to Ge–Sb–Se sputtered thin films. These sputtered films are of interest due to their low phonon energy, large glass forming region, excellent IR transmittance, and lower toxicity in comparison with arsenic based glasses. Moreover, the presence of antimony could reduce the detrimental photosensitivity of the material [16]. Recently, the linear and nonlinear optical properties of Ge–Sb–Se glasses have been studied at near and mid infrared wavelengths [17–21]. Krogstad et al. reported nonlinear properties of bulk and single mode strip waveguides made of $Ge_{28}Sb_{12}Se_{60}$ glass at a wavelength of 1030 nm [22]. Nevertheless, the Kerr nonlinear response of Ge–Sb–Se amorphous materials, when fabricated in thin film forms, needs further studies. The characterization technique we propose is based on the analysis of beam self-action and more specifically on beam self-trapping to measure optical nonlinearities in planar waveguides. Beam self-trapping occurs when diffraction is counteracted by nonlinear index change induced by the beam itself [23]. Such an effect can even lead to the formation of a spatial soliton when the trapped beam propagates without changing its shape. The technique is used to characterize ultrafast nonlinear properties of three different selenide waveguides at 1550 and 1200 nm.

2. Beam self-trapping technique

It consists in focusing a pulsed laser at the input face of a slab waveguide while the beam profile is monitored with a camera at the output face. If the launched beam is narrow, typically few 10's of micron wide, it clearly enlarges due to diffraction in the linear regime along few millimeters propagation distance. In the nonlinear regime, i.e. at higher power, diffraction is modified due to either self-focusing or self-defocusing. To properly model the propagation, the effects of both linear and nonlinear absorption must be considered. The nonlinear Schrödinger equation that includes absorption can be written as [10]

$$\frac{\partial E(x,y,z)}{\partial z} - \frac{i}{2k}\frac{\partial^2 E(x,y,z)}{\partial x^2} + \frac{\alpha}{2}E(x,y,z) - i\frac{2\pi}{\lambda}n_2 I E(x,y,z) = 0. \quad (1)$$

Here, $E(x,y,z)$ is the beam electric field distribution and is related to the beam intensity distribution by $I = 1/2c\varepsilon_0 n\left|E^2\right|$. x and y are the coordinates parallel and perpendicular to the chalcogenide layer, respectively, and z is the coordinate associated to the propagation direction. n_2 is the Kerr coefficient defined by $n = n_0 + n_2 I$ where n_0 is the effective linear refractive index of the guided mode, $k = 2\pi n_0/\lambda$ is the propagation constant in the medium at the wavelength λ. The second term of Eq. (1) corresponds to diffraction that only occurs along x since the beam is guided along y-axis. The third term accounts for absorption α expressed as $\alpha = \alpha_1 + \alpha_2 I$, α_1 and α_2 being the linear and TPA coefficients respectively. The contributions of three-photon and higher order absorption are neglected [24]. The last term of Eq. (1) accounts for the contribution of the Kerr nonlinearity. For a positive n_2 coefficient and within certain power constraints, self-focusing effect can compensate for diffraction and the fundamental solution of Eq. (1) leads to a spatial soliton [25]. In our work, the Kerr coefficient will be deduced by fitting the experimental results with simulations given by the nonlinear Schrödinger Eq. (1) solved with a beam propagation method (BPM) [26].

3. Optical characterization of slab waveguides

The proposed technique is applied to characterize slab waveguides that consist of Ge–Sb–Se films deposited on top of a 500 μm thick oxidized silicon substrate by radio frequency (RF) sputtering technique [27]. The deposition was carried out at a working pressure of 5.10^{-3} mbar. Three slab waveguides were fabricated by magnetron radio-frequency sputtering from three chalcogenide glass targets of the pseudo-binary system ($GeSe_2$–Sb_2Se_3): $Ge_{28.1}Sb_{6.3}Se_{65.6}$, $Ge_{19.4}Sb_{16.7}Se_{63.9}$ and $Ge_{12.5}Sb_{25}Se_{62.5}$, later called Se2, Se4, and Se6. The characteristics of these selenide waveguides are summarized in Table 1. The chalcogenide guiding layer thickness of 3.0–3.2 μm is determined by ellipsometry and scanning electron microscope techniques. The structure and physicochemical properties of the RF sputtered selenide films were analyzed using micro-Raman spectroscopy, Energy Dispersive X-ray Spectroscopy and X-ray Photoelectron Spectroscopy [28,29] and they are found to present some similarity to the bulk glass target prepared by the conventional melting and quenching technique depending on deposition conditions [21]. The material band gap energy from 1.70 to 2.11 eV was deduced by the variable angle spectroscopic ellipsometry method. The high refractive index of chalcogenide film and the presence of lower refractive index SiO_2 layer form step index waveguides with high index contrast. Prism coupling technique is used to analyze the guiding properties. The slab waveguide is multimode at 1550 nm. The measured refractive index at 1550 nm is 2.93, 2.68, and 2.47 for respectively Se6, Se4, and Se2. By cleaving the crystalline silicon substrate, samples with high quality end faces are produced. Efficient light end-fire coupling into the waveguide is thus possible.

As shown in Fig. 1, the optical nonlinear measurements are performed with 200 fs laser pulses from a tunable optical parametric oscillator (OPO) with an 80 MHz repetition rate. The OPO is tuned to operate either at 1550 nm or 1200 nm. The laser beam is reshaped to an elliptical spot by a cylindrical lens and focused by a X40 microscope objective. The spot size at the entrance of the waveguide at a wavelength of 1.55 μm is 4×33 μm (FWHM) in the guided (y) and transverse dimension (x), respectively. It is slightly smaller at 1200 nm in accordance with the wavelength dependence. The impact of the cumulative thermal effect is excluded with an optical chopper as it will be shown later. The sample is mounted on an XYZ translation stage to get the maximum light coupling. The spot size allows end fire coupling to the fundamental mode of the waveguide. A coupling efficiency of ~21% is measured in our waveguides. The combination of a half-wave plate and a polarizer is used to vary the coupled power. Beam distribution at the output face of the chalcogenide film is monitored with a Vidicon camera using a X10 microscope objective while two calibrated power meters measure the input and output powers.

In order to accurately fit the experimental data with Eq. (1), we first determine the linear absorption and TPA of the waveguides. Cutback method is first used to measure the linear loss. The measurements are performed by cutting the waveguide into two different lengths, starting from the long propagation length $z_2 = 1$ cm to a small length $z_1 = 0.5$ cm. We make sure that the same power is coupled in both waveguides by optimizing the coupling efficiency. The following equation is then used to calculate the linear losses,

$$\alpha_1 = \frac{\ln\left(\frac{p_1}{p_2}\right)}{z_2 - z_1}, \text{ for } z_2 > z_1, \quad (2)$$