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Femtosecond filamentation in chalcogenide glasses limited by two-photon absorption

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ABSTRACT

Filamentation of 800 nm femtosecond laser pulses in the conditions of strong two-photon absorption was first directly observed in $As_4Ge_{30}S_{66}$ chalcogenide glass, this effect being accompanied by increase in the pulse spectrum width from 8.5 to 11 nm and its modulation indicating the pulse temporal splitting. In contrast, there was no filamentation and pulse spectrum widening in stoichiometric As_2S_3 glass. The nonlinear figure of merit was shown to be as high as 0.5 and only ~0.1 in glassy $As_4Ge_{30}S_{66}$ and As_2S_3, respectively.

1. Introduction

Excellent nonlinear optical properties of chalcogenide glasses (ChG) combined with high photosensitivity make them an attractive media for 3D all-optical computing, data storage, IR telecommunication, biosensing and signal processing [1–3]. Recently, an essential progress in this field has been achieved, in fact, in the technology of ultrafast laser modification to achieve new functionality of ChG in photonic applications [4]. Tight-focused femtosecond (fs) laser pulses, particularly in filamented mode, were shown to be especially useful to fabricate ChG-based buried waveguides, optical couplers, gratings and memory systems [4,5]. Therefore, the study propagation features of fs laser pulses in ChG, including filamentation effect, is of high importance not only from fundamental point of view, but also in view of potential practical applications.

If the power of laser pulse propagating in a transparent material exceeds critical threshold

$$P_{cr} \equiv 3.72\lambda^2/8\pi n n_2 \quad (1)$$

where λ is laser pulse wavelength in free space, n and n_2 are refractive index of the material and its nonlinear refractive (Kerr) index at the same wavelength λ, respectively, then self-focusing overcomes diffraction divergence. This causes the beam collapse followed by filamentation [5], drastically changing thereby the laser pulse propagation mode.

Since nonlinear Kerr index n_2 in ChG is up to several hundred times higher than that in fused silica [1], the filamentation for 150 fs 800 nm laser pulses is expected to be observed there for pulse energies E_p exceeding several nJ. On the other hand, since the photon energy for most common wavelength λ = 800 nm of fs lasers ($h\nu$ = 1.55 eV) exceeds the half-band-gap energy E_g for the majority of ChGs (0.7–1.5 eV), the propagation of fs laser pulses in these materials are usually accompanied by strong two-photon absorption (TPA), which generally completely stops both self-focusing and filamentation phenomena.

Until now, only a few experimental studies of TPA-assisted filamentation have been performed in fused silica (E_g = 7.8 eV) and silicon (E_g = 1.13 eV) [6–9]. In Refs. [6–8], the filamentation and subsequent structural modification in fused silica was studied using 450 fs UV laser pulses ($h\nu$ = 5 eV). In Ref. [9], the spontaneous reshaping of fs Gaussian pulse into the Bessel one, featuring an absence of pulse temporal splitting, was recorded in silicon and fused silica in strong TPA regime. Recently [10], the elongated cross-section of the buried waveguide, written in Ga–La–S glass with 800 nm 150 fs laser pulses, was ascribed to the filamentation. Nevertheless, to the best of our knowledge, direct observation of filamentation phenomena in typical As/Ge-based ChG is still absent.

Here, we present experimental results on the first observed filamentation in the regime of strong TPA for two kinds of ChG, glassy g-As_2S_3 and g-$As_4Ge_{30}S_{66}$, having considerably different band-gaps and, consequently, nonlinear Figures of Merits (FOM):

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$$\mathrm{FOM} = n_2/\beta\lambda, \quad (2)$$

where β is TPA coefficient.

The former, the stoichiometric arsenic sulfide g-As_2S_3, belongs to typical network glass formers with fully saturated covalent bonding, known for a long time as model layer-structured vitreous semiconductor having a relatively wide optical band-gap (E_g = 2.4 eV) and refractive index $n \approx 2.35$ [11–14]. As it was predicted by Zachariasen in his pioneering work yet in 1932 [15], the main backbone-forming structural motives of this ChG are corner-sharing $AsS_{3/2}$ pyramids having only heteropolar As–S covalent bonds, assuring a quite dense space packing with minimal molar volume among other known S- and As-rich non-stochiometric ChG of this binary system. The average number of covalent chemical bonds per atom of formula unit reaches Z = 2.4, giving in terms of mean-field constraints theory [16–18] a so-called optimal glassy backbone (which is rigid but stress-free) with 3.0 Lagrangian constraints per atom corresponding to normal 3D-dimensionality of space. That is why g-As_2S_3 reflects some extremes in physical properties of multinary As-contained ChG [11,12,19]. One of essential disadvantages of this glass is low glass transition temperature T_g = 487 K [11–13], which significantly restricts its operating temperature range in potential device applications. Because of small E_g = 2.4 eV, we expect strong TPA in g-As_2S_3 for 800 nm fs laser irradiation due to low value of $E_g/h\nu$ = 1.55.

The latter, the g-$As_4Ge_{30}S_{66}$, is Ge-enriched composition within quasi-binary GeS_2–As_2S_3 cut-section, the germanium disulfide GeS_2 content achieving as high as 93.75 mol.%. Since stoichiomertric GeS_2 is too close to the border of glass-forming region [11,12], the role of As_2S_3 additives is to suppress strong crystallization processes ensuring a good melt-quenching ability. In contrast to g-As_2S_3, the structure of g-$As_4Ge_{30}S_{66}$ is more complicated, being presented by mixture of corner- and edge-sharing $GeS_{4/2}$ tetrahedrons interlinked with $AsS_{3/2}$ pyramids, but, probably, without direct Ge-As covalent bonds [11,12,19]. As a results, this glass having Z = 2.64 possesses a stressed backbone, which is simultaneously rigid with more than 3.0 Lagrangian constraints per atom. The g-$As_4Ge_{30}S_{66}$ is more thermally stable with characteristic T_g value close to 700 K and optical band-gap $E_g \approx 3.0$ eV [19–20]. So $E_g/h\nu$ ratio is 1.94 in this ChG indicating a regime of relatively weaker (in respect to g-As_2S_3) TPA for 800 nm fs laser pulses.

2. Glass samples preparation

The glassy samples of both above compositions As_2S_3 and $As_4Ge_{30}S_{66}$ were prepared by conventional melt quenching route [11,12]. The mixture of high-purity precursors was melted in evacuated quartz ampoules placed in a rocking furnace. The ingots were air-quenched to glassy state, which was controlled visually by a character conch-like fracture and data of X-ray diffraction, cut into disks of ~0.5 mm in thickness and polished to a high optical quality. To remove mechanical strains appeared after quick cooling, the freshly quenched samples were additionally annealed near glass transition temperature.

3. Glass samples characterization

Nonlinear optical properties of the studied ChG were tested with Z-scan technique [21].

The beam of fs regenerative amplifier (2.5 mJ, 150 fs, 800 nm, 1 kHz) of Gaussian transversal profile with 7 mm FWHM was attenuated with a set of calibrated neutral density filters (NDFs) and focused with a flat-convex lens of 15 cm focal length into a 0.5 mm thick polished ChG sample. This sample was moved along the beam through the focus position with PC-controlled translation stage, the digitized data of three photodetectors (the reference, open- and closed-aperture ones) being collected with the same PC.

The closed-aperture data were analyzed following a formula [21]:

$$\Delta T = 0.406\ kL_{\mathrm{eff}}n_2 I, \quad (3)$$

where ΔT is maximum–minimum difference of the normalized transmission curve, k is $2\pi/\lambda$, L_{eff} is $[1 - \exp(\alpha L)]/\alpha$, α is the linear absorption coefficient, L is the sample thickness, and I is the peak intensity of the laser beam inside the sample.

Neglecting α and comparing the obtained closed-aperture curve with that of fused silica (1 mm in thickness), we obtained $n_2 = 3.4 \times 10^{-14}$ cm^2/W for g-$As_4Ge_{30}S_{66}$ (the value of $n_2 = 3.2 \times 10^{-16}$ cm^2/W for fused silica [22] was taken as a calibration standard). In a similar way, by accounting the full depth of open-aperture transmission dip at Z = 0 with a formula [21]:

$$T_0 = 1 - \beta IL/(2\sqrt{2}), \quad (4)$$

we have found that TPA coefficient β in $As_4Ge_{30}S_{66}$ glass equals to 8×10^{-10} cm/W.

The refractive index n of g-$As_4Ge_{30}S_{66}$ determined from linear transmission measurements at 800 nm by assuming multiple reflections in plane-parallel sample was estimated to be near 2.2. It also follows from the above values of n_2 and β that FOM = 0.5 for g-$As_4Ge_{30}S_{66}$. Therefore, the critical power of laser pulse for self-focusing estimated with Eq. (1) was found to be 13 kW, that is equivalent to pulse energy of E_{cr} = 2.1 nJ at 150 fs duration.

The similar Z-scan measurements for g-As_2S_3 revealed a complete domination of TPA both in open- and closed-aperture measuring data. So we have been able to measure only TPA coefficient $\beta = 2.1 \times 10^{-9}$ cm/W, which occurs in close agreement with known value of $\beta = 2.0 \times 10^{-9}$ cm/W reported earlier for this glass at the same wavelength [23,24].

As to n_2 for g-As_2S_3, we have not found any data for 800 nm in the literature. The nearest experimental value is $n_2 = 4.5 \times 10^{-14}$ cm^2/W measured in g-As_2S_3 at 1.06 μm [25], which certainly differs from that at 800 nm because of strong dispersion of Kerr coefficient near band-gap. For this reason, starting from this value at 1.06 μm ($h\nu/E_g$ = 0.49) and using universal theoretical dependence of Kerr coefficient in glasses on $h\nu/E_g$ [26], we have found the expected value of $n_2 = 1.7 \times 10^{-14}$ cm^2/W at 800 nm ($h\nu/E_g$ = 0.65) in g-As_2S_3. Thus, the nonlinear FOM in this ChG is estimated to be only ~0.1 and critical power for self-focusing P_{cr} = 24 kW (or E_{cr} = 4.0 nJ) at 150 fs laser pulses).

4. Experimental

Previously we have studied the filamentation phenomena in wide-band-gap oxide glasses using fs time-resolved optical polarigraphy (FTOP) [27,28], which allows direct visualization of traveling laser pulse with 150 fs temporal resolution. Unfortunately, the FTOP turned out to be not applicable for ChG samples because of their residual optical anisotropy. Therefore, the self-focusing and filamentation have been studied here by recording time-integrated near-field profiles of the beam exiting the bulk sample. The scheme of experimental set-up is shown in Fig. 1.

Regenerative amplifier 1 delivers a train of 2.5 mJ, 150 fs, 800 nm, horizontally polarized pulses. A relatively low 50 s^{-1} pulse repetition rate was chosen to avoid heating and to slow down the photoinduced structural changes in the samples. A set of calibrated neutral density filters (NDFs) 2 provides the necessary pulse energy. Aperture 4 of 3.5 mm diameter cuts out the narrower beam before entering the lens 6 with 15 cm focal distance. The lens is fixed on a motorized Z-axis translation stage, which shifts it along the beam axis during the measurements. The distance between the