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Femtosecond-laser-induced submicron grating periodic structures on As_2S_3 and $As_{35}Se_{65}$ glasses

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ABSTRACT

Submicron periodic structures produced by femtosecond Ti:sapphire laser irradiation (1 kHz, 150 fs, 3–5 μm) on As_2S_3 and $As_{35}Se_{65}$ glasses at edges of damage holes are observed. Effects of changes in laser wavelength (λ), pulse number, power, and irradiation time on grating periodicity were studied. Regular groove-like ripples and distinct parallel submicron periodic gratings were seen in damaged areas. The damage grating period (Λ) is approximately $\lambda/2n$. The period increases with wavelength but is hardly influenced by irradiation time and pulse number. Samples with high refractive index and low bandgap are more prone to melting, and edge stripes are not obvious.

1. Introduction

Chalcogenide glass has high refractive index (n) and a wide transparent transmission window, making it a promising material for mid-infrared (MIR) applications [1]. Furthermore, chalcogenide glass shows structural flexibility and photo-induced phenomena that make it ideal for optical micromachining [2]. When the surface of chalcogenide glass is irradiated by a femtosecond laser, a periodic submicron stripe structure is formed when the energy density exceeds the ablation threshold [3]. A femtosecond-laser-induced periodic stripe structure (FLIPSS) having period far smaller than the laser wavelength (λ) has broad application prospects in fiber Bragg gratings [4], modification of materials [5], structural coloring [6], photonic crystals [7], and optical storage [8].

Femtosecond-laser-induced nanostructures have attracted extensive attention in recent years. In 1998, Mazur et al. first reported the rules for spike formation on a large-area Si surface upon irradiation with a femtosecond laser having pulse duration of 100 fs and pulse number of 500 in SF_6 and Cl_2 atmosphere [9]. In 2001, Efimov et al. reported the etching of optical waveguides on $As_{40}S_{60}$ chalcogenide glass by using a series of 850-nm femtosecond laser pulses. Raman spectra showed that As-S bonds were destroyed by the formation of As-As and S—S bonds [10]. In 2006, Bhardwaj et al. reported that self-organized nanostructures are only formed in a specific pulse energy range and the space of nanoplanes tend to $\lambda/2n$ [11]. In 2008, Guo et al. produced three nanostructures on the surface of ZnO crystals by femtosecond laser radiation. They believed that this structure was formed by *in-situ* second-harmonic generation (SHG) effects [12]. In 2010, Zhang et al. formed a 180-nm periodic nanograting on As_2S_3 chalcogenide glass surface by using 800-nm (1 kHz, 100 fs) multipulse laser irradiation and showed that the power intensity and pulse interval had no effect on the period [13]. In 2012, Messaddeq et al. irradiated a Ge-S chalcogenide glass by using a femtosecond laser (1 kHz, 34 fs, 806 nm) and described the self-organizing periodic stripe structure formed after irradiation. Scanning electron microscope (SEM) images showed that it produced periodic stripes close to the λ (~720 nm), and the direction was parallel to the electric field direction of light [14].

Femtosecond laser irradiation with linear polarization can be used to form stripes with two types of periods: low-spatial-frequency LIPSSs (LSFL: period is close to the incident wavelength) and high-spatial-frequency LIPSSs (HSFL: period is less than half of the incident wavelength) [12]. It is relatively easy to prepare a LSFL, but it is much more difficult to prepare a HSFL. LSFLs can be formed by the interference of light between the incident laser wave and the scattered wave caused by defects on the surface [15,16]. However, the formation of HSFLs remains unclear. The

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LIPSSs were potential integrated component in telecommunication fiber technology.

In this study, the submicron structure of As_2S_3 and $As_{35}Se_{65}$ glass surfaces was studied under different pulse number, time, wavelength, and power. The influence of the *n* of different samples on the period of the surface stripe was also investigated. The periodic structure of the sample surface was observed by scanning electron microscopy, and the mechanism by which the nanostructure was formed was explained.

2. Material and methods

The samples were prepared using the standard melt-quenching method, and they were double-polished to high optical quality (2 mm). Laser damage experiments were performed using a Ti:sapphire femtosecond laser (Mira 900D+, Coherent) and optical parametric amplifier (Legend Elite+ OpeaA Solo, Coherent) delivering 150-fs and 1-kHz pulses and a wavelength tunable laser. The power magnitude of the incident laser is controlled by a pair of polarizers, and a three-dimensional control platform was used to control the laser irradiation position. A shutter was used to control the laser irradiation time. After laser irradiation, the sample was immersed in an isopropanol solution and was ultrasonically cleaned for 10 min to remove particles generated by laser irradiation and sputtering. The morphology of the damage was observed by SEM (Nova NanoSEM 450, FEI) and AFM (Digital Instruments, D 3100, Veeco Metrology Group) after been gold-plated on the sample surface.

3. Results and discussion

Both the As_2S_3 and the $As_{35}Se_{65}$ glasses were irradiated by a femtosecond laser with repetition frequency of 1 kHz, pulse width

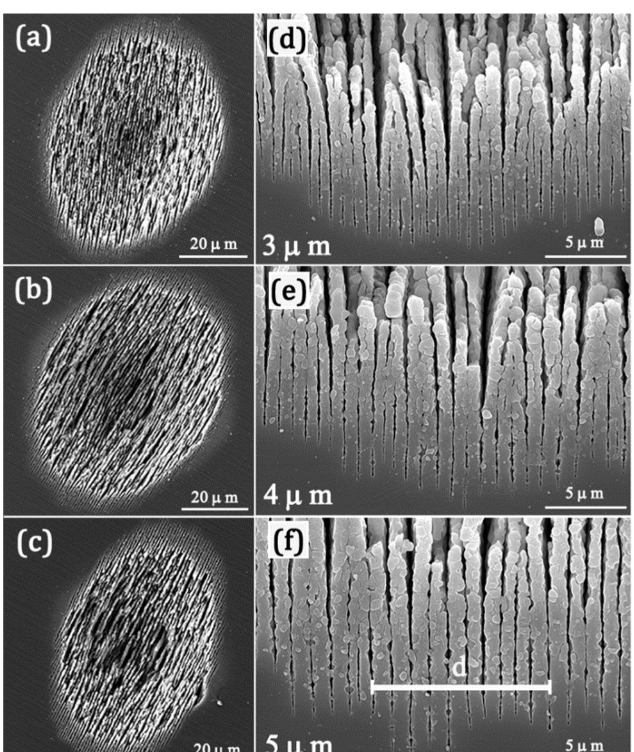

Fig. 1. SEM images of damage morphology caused by femtosecond laser irradiation: (a) 3 μm (4.14 kx), (b) 4 μm (3.96 kx), and (c) 5 μm (3.76 kx). (d)–(f) Partial enlargement of the damaged edges (10 kx).

Table 1
Refractive index and period of the corresponding stripe of the As-S glass under femtosecond laser irradiation with different powers and wavelengths and the theoretical values of the period.

λ (μm)	*n*	Λ (μm)			$\lambda/2n$ (μm)
		30 mW	25 mW	20 mW	
3	2.426	0.5992	0.6199		0.621
4	2.420	0.7693	0.8079	0.7759	0.826
5	2.416	0.9707	0.9252		1.04

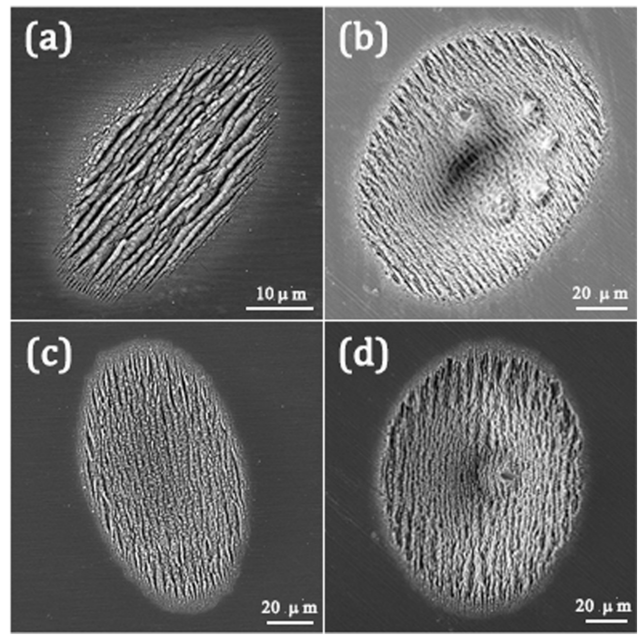

Fig. 2. Grating period of $As_{35}Se_{65}$ after femtosecond laser irradiation with wavelengths of (a) 3, (b) 4, (c) 5, and (d) 6 μm.

Table 2
Measured and calculated values of the marginal grating of As-Se glass under different wavelengths of femtosecond laser irradiation and the *n* at the corresponding wavelengths.

λ (μm)	*n*	Λ(μm)	$\lambda/2n$ (μm)
3	2.756	0.583	0.547
4	2.747	0.714	0.728
5	2.742	0.913	0.916
6	2.740	1.020	1.100

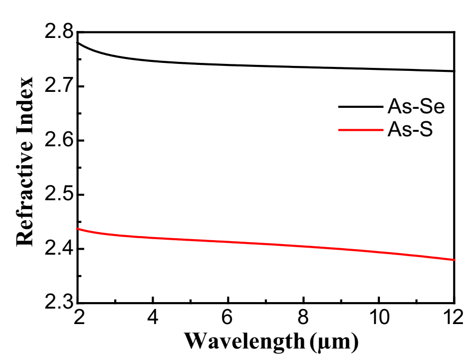

Fig. 3. Refractive index of As_2S_3 and $As_{35}Se_{65}$ glass.