Lithography-free surface enhanced nonlinear absorption of chalcogenide film

Jianxing Zhao[1] and Jianhong Zhou[1, 2, *]

[1]School of Photoelectric Engineering, Changchun University of science and technology, Changchun, 130022, China

[2]Key Laboratory of Optoelectric Measurement and Optical Information Transmission Technology of Ministry of Education, Changchun University of science and technology, Changchun 130022, China

[*]Corresponding author: zjh@cust.edu.cn.

Keywords: Nonlinear optics, reverse saturated absorption, chalcogenide.

Abstract: We experimentally study the nonlinear absorption enhancement of chalcogenide $Ge_{28}Sb_{12}Se_{60}$ (GSS) films by using lithography-free metasurface structure. The antennas that support localized surface plasmon are randomly distributed on the substrate, which is generated by annealing the silver thin film on a quartz substrate. Z-scan technique is carried to measure the nonlinear absorption of the samples. The measured result show that the nonlinear absorption of GSS film can be enhanced by 2 folds at 532 nm laser excitation. This method of enhancing nonlinearity of materials have avoided the complex lithography technique and reduced the cost in fabrication, providing a new way in achieving the high throughput fabrication for nonlinear optical devices.

1. Introduction

Chalcogenide, refer to the amorphous materials containing S, Se or Te element, is excellent candidate for ultrafast optical devices due to its high infrared transmittance, high refractive index and high nonlinearity [1-3]. The nonlinearity of chalcogenide that is significant to the modulation of light and light matter interaction has been investigated widely in recent years [4, 5]. Generally, the nonlinearity can be enhanced by the localized surface plasmon (LSP) originated from the interaction between light and sub-wavelength metal or semiconductor structures [6, 7]. However, the study about LSP enhanced chalcogenide nonlinearity has seldom been reported.

In this paper, we experimentally investigate the nonlinear absorption enhancement of annealed Ag film to $Ge_{28}Sb_{12}Se_{60}$ (GSS) film. By annealing the 20 nm film, the randomly distributed island-shape structure that support LSP is generated, which can be utilized to enhance the nonlinear response of GSS film. Z-scan system is presented to measure the nonlinear response of the proposed samples. Experimental results show that the annealed Ag structure can enhance the nonlinear absorption coefficient by 2 folds. We also investigate the spectral responses of the fabricated samples to further validate the design. This work is beneficial for developing lithography-free nonlinear optical devices.

2. Experiment

2.1 Sample preparation

The quartz substrate is cleaned using deionized water, alkali liquor and acid liquor successively. In sample 1, GSS film with a thickness of 20 nm is deposited directly on the quartz substrate. In sample 2, the Ag film with a thickness of 20 nm is firstly deposited on the quartz substrate, then the GSS film with a thickness of 20 nm is deposited onto the Ag film. In sample 3, the Ag film with a thickness of 20 nm is firstly deposited on the quartz substrate. Then the Ag-coated sample is placed in muffle furnace, heated rapidly to 270 ℃ and kept for 1 min then cooled naturally to room temperature, forming the randomly distributed island-shape structure [8]. Finally the GSS film with a thickness of 20 nm is deposited onto the annealed sample. The fabrication process is shown in Fig. 1.

  DOI: 10.25236/isaiee.2020.029

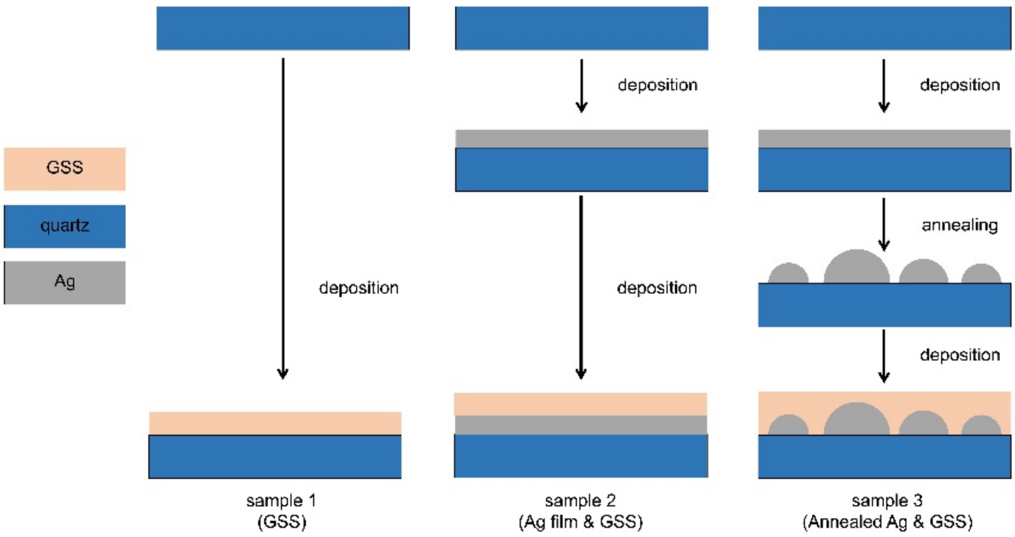

Fig 1. The fabrication process of the proposed samples.

In the fabrication process, the alkali liquor is composed of $NH_3{\bullet}H_2O$, H_2O_2 and H_2O with a volume ratio of 1:2:5. The acid liquor is composed of HCL, H_2O_2 and H_2O with a volume ratio of 1:2:8. The deposition process of GSS and Ag film are carried out by using thermal evaporation system (Angstrom Engineering Inc. Canada) with the depositing rates of $0.015\,nm/s$ and $0.01\,nm/s$, respectively. The materials are chosen as purchased Ag particles (99.999%) and $Ge_{28}Sb_{12}Se_{60}$. In the Z-scan measurement, the nonlinear response of samples is studied using Nd:YAG laser with a wavelength of 532 nm and pulse width of 7.8 ns. The Z-scan system is shown in Fig. 2, in which a beam expander is added due to the poor light spot.

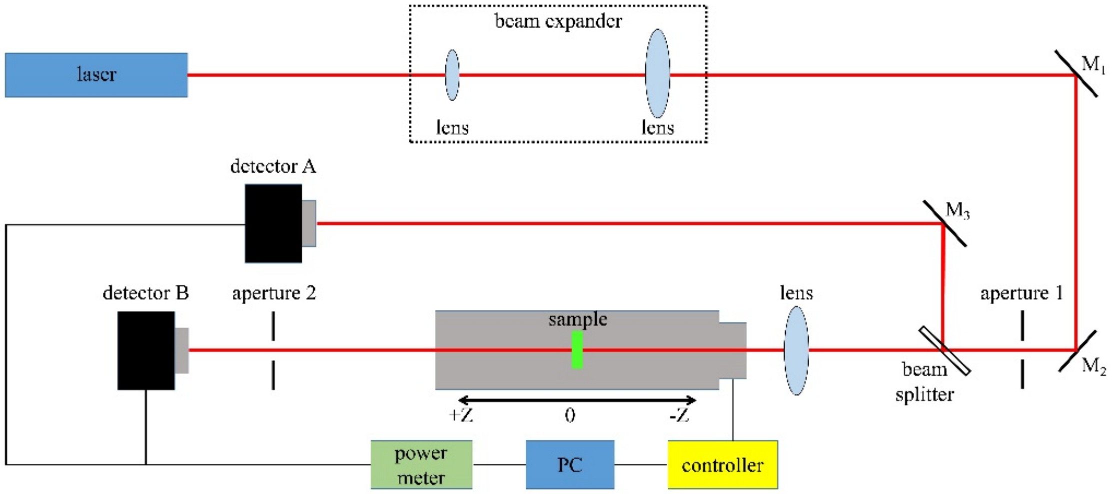

Fig 2. Z-scan system for optical nonlinearity measurement.

2.2 Results and discusstion

The far field transmittance in open and closed aperture can be expressed as Eqs. (1) and (2)

$$T(z)=\sum_{m=0}^{\infty}\frac{\left(-\frac{q_{(z,0)}}{(1+\gamma^2)}\right)^m}{(m+1)^{\frac{3}{2}}}, \tag{1}$$

And

$$T(z)=1-\frac{4\Delta\phi\gamma}{(1+\gamma^2)(9+\gamma^2)}, \tag{2}$$

where m is an integer, $q(z,0)=\beta I_0(t)L_{eff}$, β is the nonlinear absorption coefficient, $L_{eff}=\frac{1-e^{-\alpha l}}{\alpha}$ is the efficient thickness, l is the sample length, $\gamma=\frac{z}{z_0}$ with z and z_0 being the longitudinal displacement of the sample from focus and the Rayleigh length, I_0 is the laser power at focal point, $\Delta\phi=2\pi n_2 I_0(t)L_{eff}/\lambda$ is the phase shift at focal point.

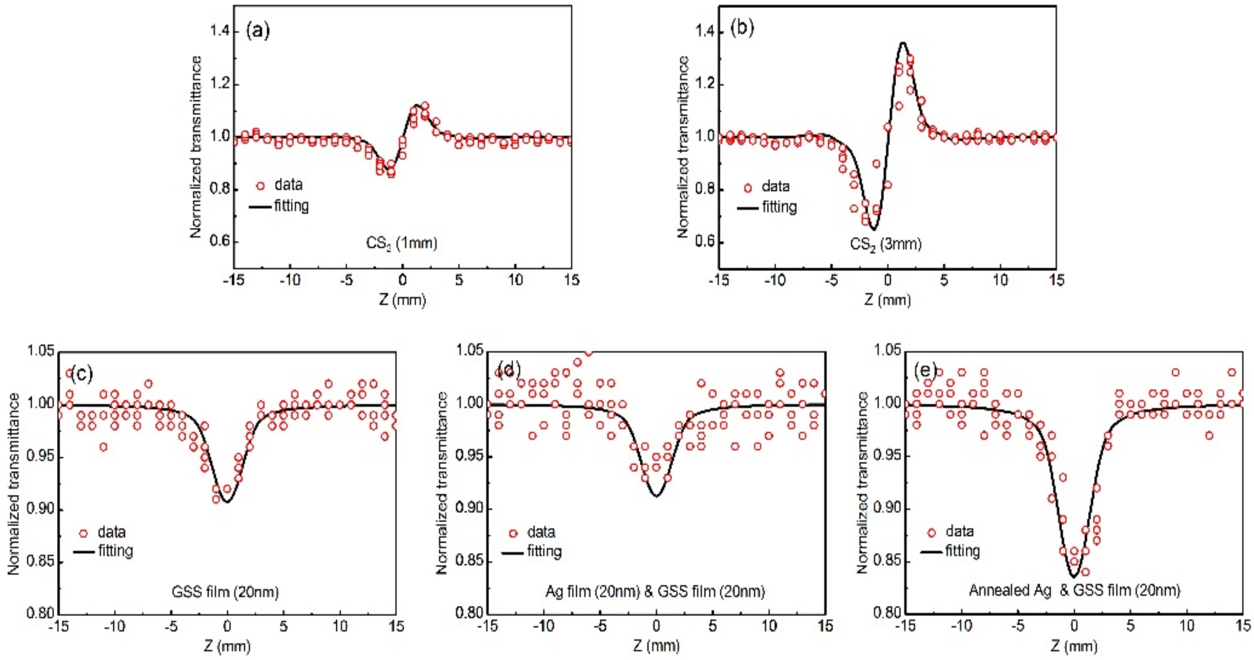

Fig 3. Z-scan measurement results of CS_2 reference and fabricated samples. Closed aperture results of (a) 1 mm and (b) 3 mm CS_2. Open aperture results of (c) sample 1, (d) sample 2 and (e) sample 3

Figures 3 (a) and (b) show the closed aperture Z-scan results for 1 mm CS_2 and 3 mm CS_2 samples, respectively. The red circles are measured results and the black curves are fitted results, which agree well with the former reports [9]. Utilizing the calibrated Z-scan system, we measure the nonlinear absorption response of fabricated samples when the aperture 2 is open. For sample 1, 2 and 3, the measured results are shown by the red circles in Fig. 3 (c), 3(d) and 3(e), where the reverse saturation absorption responses are acquired. After theoretical fitting, shown by the black curves, the nonlinear absorption coefficients are calculated to be $\beta_1=3.1\times10^{-4}\ m/W$, $\beta_2=2.9\times10^{-4}\ m/W$ and $\beta_3=6.3\times10^{-4}\ m/W$, respectively. By comparing sample 1 with 2, we find that the 20 nm Ag film that is not annealed has no effect on the nonlinear absorption response of the GSS film. And by comparing sample 1 with 3, we find that the annealed 20 nm Ag film enhanced the nonlinear absorption coefficient by nearly 2 folds (the annealed 20 nm Ag film presents no nonlinear absorption response which is not shown here). Because after annealed, the 20 nm Ag film is converted to randomly distributed island-shape antennas [8], which support the LSP resonance that enhances the nonlinear absorption effect [6, 7]. The transmission spectra of the Ag film and the annealed Ag film are shown by the black dashed curve and the red solid curve, respectively in Fig. 4(a). We can see that the transmission dip around 430 nm is acquired after annealing, indicating a LSP resonance. In Fig. 4(b) we also find that the spectrum of GSS with annealed Ag is not simple overlap of spectra of GSS and annealed Ag. The resonance dip of annealed Ag (red dotted curve) undergoes a red shift after coated with the GSS film, which validates better the LSP property of shifting with the surrounding refractive index [10].

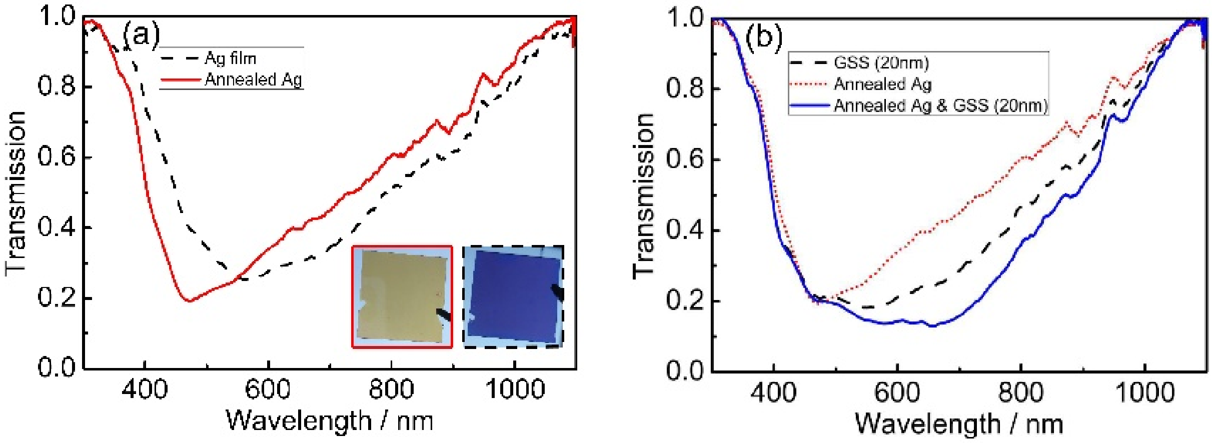

Fig 4. (a) Transmission spectra of the Ag film and the annealed Ag film. Inset show the corresponding sample pictures with a background of sky. (b) Transmission spectra of GSS only, annealed Ag and GSS with annealed Ag.

3. Conclusions

In this paper, the effect of the annealed Ag film to the nonlinear absorption of GSS film is studied. Experimental results show that a 2-fold enhancement of nonlinear absorption coefficient is attained at 532 nm wavelength. The enhancement origins from the LSP on the randomly distributed structure that is acquired by annealing the 20 nm Ag film. It should noticed that the proposed structure is easy in fabrication, because we do not need the complex lithography technology in the fabrication process, which presents great potential in volume production of nonlinear optical devices.

Acknowledgements

The authors gratefully acknowledge the financial support from Scientific and Technological Developing Scheme of Jilin Province (20180101281JC); “135” Research Project of Education Bureau of Jilin Province (JJKH20190579KJ); “111” Project of China (D17017).

References

[1] Wuttig M. and Yamada N., “Phase-change materials for rewriteable data storage,” Nat. Mater. 6, 821-832 (2007).

[2] Shportki K., Kremers S., Wods M., Lencer D., Robertson J., and Wuttig M., “Resonant bonding in crystalline phase-change materials,” Nat. Mater. 7, 653-658 (2008).

[3] Ding F., Yang Y., and Bozhevolnyi S. I., “Dynamic Metasurfaces Using Phase-Change Chalcogenides,” Adv. Opt. Mater. 7(14), 1801709 (2019).

[4] Pradhan P., Khan P., Aswin J. R., et al. “Quantification of nonlinear absorption in ternary As-Sb-Se chalcogenide glasses,” Journal of Applied Physics 125, 015105 (2019).

[5] Viswanathan A., Thomas S., “Tunable linear and nonlinear optical properties of GeSeSb chalcogenide glass with solute concentration and with silver doping,” Journal of Alloys and

[6] Compounds 798, 424-430 (2019).

[7] Miao R., Shu Z., Hu Y., et al., “Ultrafast nonlinear absorption enhancement of monolayer MoS2 with plasmonicAu nanoantennas,” Optics Letters 44(13), 3198-3201 (2019).

[8] Alam M. Z., Schulz S. A., Upham J., et al., “Large optical nonlinearity of nanoantennas coupled to an epsilon-near-zero material,” Nature Photonics 12, 79-83 (2018)

[9] Kong X., Fu Y., Zhang W., et al., "Analysis of random antireflective structures fabricated by silver dewetting to enhance transmission," Journal of Nanophotonics 11(3), 036019 (2017).

[10] Sheik-Bahae M., Said A. A., Wei T., "Sensitive Measurement of Optical Nonlinearities Using a Single Beam," IEEE Journal of Quantum Electronics 26(4), 760-769 (1990).

[11] Liu N., Weiss T., Mesch M., et al., "Planar Metamaterial Analogue of Electromagnetically Induced Transparency for Plasmonic Sensing," Nano Letters 10, 1103-1107 (2010).