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CO_2 laser induced two-photon absorption detected by near-infrared lasers in As_2Se_2S glasses

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Influence of CO_2 pulsed microsecond laser on two-photon absorption (TPA) at wavelengths of Nd:YAG (1060 nm and 1320 nm) and Er glass lasers (1540 nm) for As_2Se_2S glasses was studied. Dependence of the TPA versus the photoinducing power densities at wavelength 9.2 μm is studied. The highest values of the TPA β coefficient were achieved for perpendicular polarizations between the pumping and probing laser beams. The obtained values of TPA coefficients indicate a possibility of using the As_2Se_2S glasses as optically-operated IR laser limiters in a wide spectral range. The substantial role of anharmonic electron–phonon subsystem is demonstrated.

Keywords: nonlinear optics, infrared glasses.

1. Introduction

Recently, a growing interest in nonlinear optical properties based on complex glasses as materials for infrared (IR) quantum electronics [1–7] is observed. Their wider technological application is restrained by the following factors:

– low values of two-photon absorption nonlinear optical (TPA) coefficients;
– relatively high time response with respect to single crystals;
– high space non-homogeneity of the principal optical constants.

The main aim of this work is to search for a new type of glasses with improved technological parameters. From the previous considerations [1–6], one can expect that all of the optical constants mentioned might be varied in desirable directions by appropriate doping. Unfortunately, they are spectrally limited in the IR spectral range. More effective here may be chalcogenide crystals [7]. The main goal of this article consists in exploring the possibility of studying the photoinduced dependence of the TPA under influence of CO_2 photoinducing laser beam. Additionally, the pump--probe TPA will be explored.

2. Experimental part

2.1. Sample preparation

All the glasses were synthesized by a method similar to the one described in [1, 2]. The samples had sizes equal to $4\times3\times1$ mm^3. The surfaces were polished in order to obtain surface roughness better than 0.12 μm.

2.2. Nonlinear optical set-up

We have performed measurements of photoinduced TPA using as a source Nd:YAG laser at wavelengths 1060 nm and 1320 nm, with pulse duration about 12 ns, and frequency repetition about 12 Hz. CO_2 laser had wavelength $\lambda = 9.3$ μm, with pulse duration 3.5 μs, frequency repetition 11 Hz and maximal peak power about 12 MW. At the same time we have used the second and fourth harmonics of this laser at the wavelengths of 0.95 μm and 0.475 μm, respectively.

A principal experimental set-up for performing the photoinduced TPA measurements is shown in Fig. 1. The polarized light from the CO_2 laser creates relatively strong effective electric filed in the glass investigated. The system of polarizers and mirrors allows the incident angle to be tuned within 10–12° between the pumping CO_2-induced polarized light beam and the fundamental probing Nd:YAG and Er:glass lasers. The photoregistration system consisting of the fast response photomutiplier together with the electronic boxcar registration system makes it possible to monitor the kinetics of the output transparency versus intensity power of the probing laser beams.

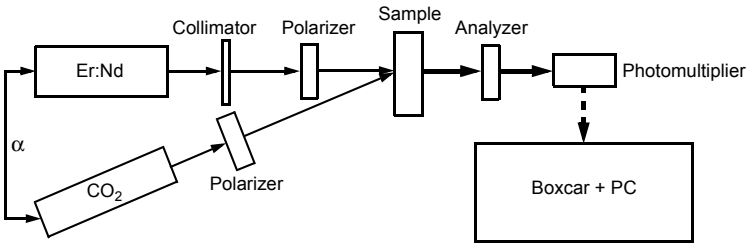

Fig. 1. Principal schema of IR-induced TPA set-up.

Because the wavelengths of the photoinducing lasers are near the phonon bands due to photoexcited phonons there occur enhanced dipole moments determining the corresponding third order susceptibilities determining the value of the TPA. Varying the polarization of the incident and output beams we were able to analyze different geometries of optical susceptibility corresponding to the TPA.

The TPA coefficient was evaluated with precision of up to 0.3 cm/GW from intensity-dependent transparency T following a method described in Ref. [1].

For more accurate control we have done all the measurements at varying angles between polarizations of the pump UV light and probe IR laser beams.

3. Results and discussion

The measured dependencies of the optical transparency versus the CO_2-induced power density are presented in Fig. 2 for three different fundamental lasers at 1540 nm, 1320 nm and 1060 nm. One can see that at the pump power densities varying within 0.38–0.59 GW/cm^2 substantial TPA coefficients in the corresponding pump power dependences were found. The higher angles correspond to the higher values of the TPA. The wavelength dependences are relatively strong. This one confirms that

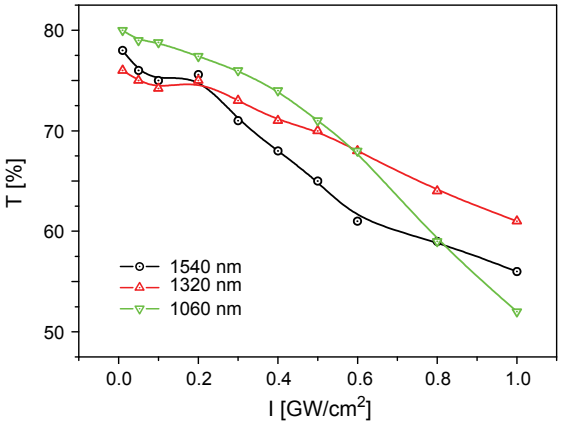

Fig. 2. Typical dependence of the intensity dependent transparency for different laser wavelengths.

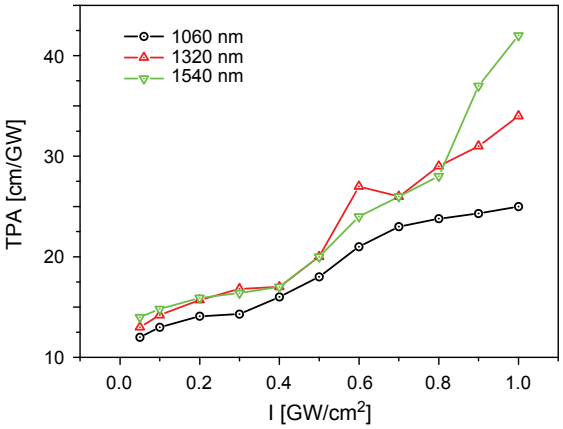

Fig. 3. Dependences of the photoinduced TPA versus pump power density at different fundamental wavelengths.

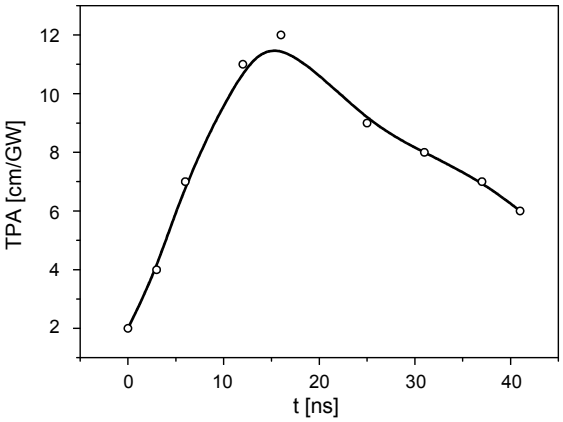

Fig. 4. Dependence of the TPA coefficients versus pump-probe delaying times.

the TPA dependences are spectrally dependent. Even from the qualitative consideration of the dependences one can see that there exists a drastic increase of the TPA with decreasing wavelength. A large increase of the TPA at lower wavelengths may be caused by the larger number of the phonons contributing to the TPA.

Another important factor determining the possible application of the glasses lies in the possibility of increasing the TPA up to 2 times in additional CO_2 laser treatment (see Fig. 3). It is crucial that this increase is more remarkable for lasers with higher wavelengths, which additionally confirms the principal role of the photoinduced phonons.

The optimal pump-probe delaying time (which corresponded to the maximal TPA value) was equal to about 18 ns (see Fig. 4). Generally, the observed pump-probe delaying TPA is substantially asymmetric. It is necessary to emphasize that maximal birefringence achieved (about 0.12) was observed for a CO_2-induced power density equal to about 0.6 GW/cm^2. Performing cyclic cooling–heating temperature measurements, no temperature hysteresis loop was found. All the temperature dependencies observed are generally similar to that ones for another chalcogenide glasses [1–11]. However, in this particular case, the optimal pump-probe delay time is at least 5 times higher. The maximal TPA signal is observed for collinear polarization of the CO_2 photoinducing pumping and fundamental beams. We have also discovered that minimal light scattering losses were observed for the spot diameter of the probe laser beam equal to about 75–120 μm. All the photoinducing changes of the optical constants were reversible with accuracy up to 0.041% (detected by phototransparency and light reflection).

All the effects were observed only during CO_2 laser pump treatment and the values presented correspond to maximum of the TPA output signal after statistical averaging through the specimen surface.

The TPA is described by the fourth-rank polar tensors and is connected with photoinduced dipole transition moments. The maximal output TPA signals were

observed for diagonal tensor component χ_{yyyy} where the *y*-direction corresponds to the polarization direction of the photoinducing beam. An increase of the TPA is observed near the long and short wavelength edges of the transparency windows. Such unusual spectral dependence of the TPA may be related to the exhaustion of the particular virtual states and is a consequence of the existed highly localized *f–d* rare-earth ion levels and nano-confined trapping states. Generally, the value of the TPA was better compared to those for the chalcogenide glasses [1–7].

From the experimental data (particularly, in the picosecond regime), we have revealed a crucial role of the electron–quasi-phonon subsystem in the phenomena observed. Hence, particular attention is paid to photoinduced changes of the electronic as well as quasi-phonon subsystems which are particularly important during the photo-induced changes.

In the case of the photoinduced TPA, the non-centrosymmetry does not play any role and the values of the dipole moments and their signs become crucial. The latter ones determine absolute values of imaginary part of the fourth rank optical tensors directly connected with the TPA. For the TPA there exists a maximum at pump-probe delay time at about 8 ps. This minimum is caused by occupation of particular praseodymium virtual nano-confined levels during the fourth-photon interactions. This feature is different from the three-photon interactions (the case of the photoinduced optical second harmonic generation) in pure amorphous-like phase. This fact reflects also a light macro-scattering that complicates an extraction of nonlinear optical susceptibilities.

During investigation of the photoinduced effects one cannot exclude completely the role played by irreversible changes [8]. The contribution of electron–phonon subsystems may be changed by variation of the anionic substitution [9, 10], similarly to the other nonlinear optical media [11–14].

4. Conclusions

We have found that the microsecond induced CO_2 laser pulses may play a crucial role for the TPA in the near-infrared spectral range for the As_2Se_2S glasses one can see that at the pump power densities varying within 0.38–0.59 GW/cm^2 substantial bending in the corresponding pump power dependences are observed. The higher angles correspond to the higher values of the TPA. The wavelength dependences are relatively strong. This one confirms that the TPA dependences are spectrally dependent. Even from the qualitative consideration of the dependences one can see that there exists a drastic increase of the TPA with decreasing wavelength. Another important factor determining the possible application of the glasses is the possibility of increasing the TPA up to 2 times at additional CO_2 laser treatment. The optimal pump-probe delaying time (which corresponded to the maximal TPA value) was equal to about 18 ns.

All the photoinduced changes are reversible with respect to IR illumination with precision up to 0.010%.

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