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Nonlinear optical studies in semiconductor-doped glasses under femtosecond pulse excitation

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Abstract. Nonlinear optical studies in semiconductor-doped glasses (SDGs) are performed under femtosecond laser pulse excitation. Z-scan experiments with 800 nm wavelength pulses are used to excite SDG samples in the resonance and non-resonance regimes. Schott colour glass filter OG 515 shows stronger two-photon absorption than GG 420 and both the samples exhibit positive nonlinearity. However, in resonantly excited RG 850 the intensity-dependent Z-scan shows transition from saturable to reverse saturable absorption behaviour with the increase in intensity.

Keywords. Nonlinear optics; semiconductor-doped glasses; Z-scan.

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1. Introduction

Understanding nonlinear optical properties of semiconductor-doped glasses (SDGs) are interesting because of the possibility of using them for all-optical data processing applications [1,2]. In SDGs, semiconductor nanoparticles are embedded in glass matrix. Commercially available Schott sharp cut-off colour glass filters contain CdS_xSe_{1-x} and $CdSe_xTe_{1-x}$ nanoparticles. Nonlinear optical properties, specifically nonlinear refractive index (n_2) and two-photon absorption (TPA) coefficient corresponding to the real and the imaginary parts of the third-order susceptibility $\chi^{(3)}$, respectively were studied in various colour glass filters [3–6]. Most of the studies in SDGs are carried out in the transparency region, $2\hbar\omega > E_{\rm g} > \hbar\omega$, where $E_{\rm g}$ is the band gap of the semiconductor and $\hbar\omega$ is the excitation photon energy. Very limited studies were conducted in resonance excitation region in SDGs with high-intensity femtosecond laser pulses.

Z-scan is now recognized as a standard technique [7] to study nonlinear optical properties and is widely used because of its simplicity and high sensitivity as well as the capability to simultaneously measure the magnitude and sign of the nonlinear refraction and absorption. The Z-scan technique is widely used with both CW and pulsed lasers ranging from nanosecond to femtosecond to measure nonlinear optical

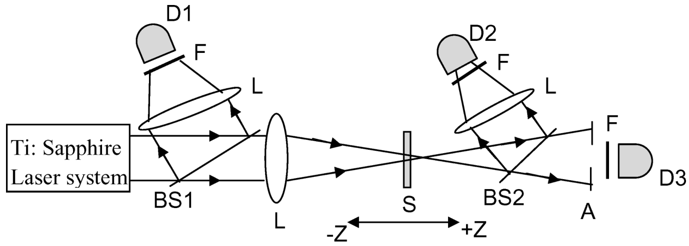

Figure 1. Experimental Z-scan set-up. L – lens, F – neutral density filters, BS – beam splitter, A – aperture, D – photodiode, S – sample and Z – position.

properties of various classes of materials including bulk semiconductors, dielectrics and semiconductor-doped glasses, organic molecules, liquid crystals and solvents.

Here, we present nonlinear optical studies in different Schott colour glass filters under femtosecond laser pulse excitation. The SDG samples are chosen with band-gap energies which are in resonance and transparency regions with respect to the excitation photon energy. Z-scan profiles are very different in resonance and transparency regions. Furthermore, the Z-scan experiments performed at different intensities show interesting results in RG 850. At low intensity (energy) the sample exhibits saturable absorption only in the open aperture Z-scan, but for open aperture performed at high intensity (energy), the sample exhibits transition from saturable absorption to reverse saturable absorption at $Z = 0$.

2. Experimental details

Z-scan set-up shown in figure 1 was used to simultaneously measure open- and closed-aperture Z-scans. Incident laser pulses from amplified femtosecond laser system that delivered 100 fs pulses centred around 800 nm operating at 1 kHz repetition rate were used. Experimentally it has been shown that at 1 kHz repetition rate, the thermal nonlinearity does not contribute to the nonlinearity. Incident beam was focussed by lens L onto the sample S. Sample kept on a translating stage was moved from $-Z$ to $+Z$ direction across the focal region. Transmitted beam through the sample was detected at two places, one through an aperture A placed in the far field by a detector D3 to collect closed-aperture Z-scan data and other, using the beam splitter BS2. The entire reflected light from BS2 was collected on the photodetector D2 to simultaneously record open-aperture Z-scan data. A small portion of the incident beam reflected from the beam splitter BS1 as shown in figure 1 was collected by the photodetector D1. The photodetectors D1, D2 and D3 are large-area photodiodes configured to monitor the incident pulse energy of single femtosecond pulse.

3. Results and discussion

Figure 2 shows closed- and open-aperture Z-scan data of the Schott colour glass filter GG 420. Squares and circles show experimental closed- and open-aperture

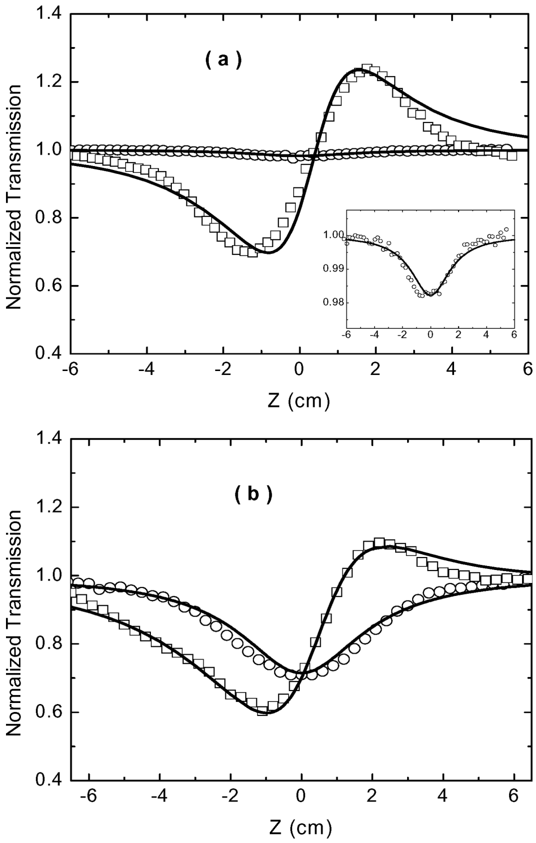

Figure 2. Closed- and open-aperture Z-scan profiles for (**a**) GG 420 and (**b**) OG 515 at peak intensities of 175 GW/cm^2 and 400 GW/cm^2, respectively. Squares and circles are experimental closed- and open-aperture Z-scan data points, respectively and solid lines are numerically fitted curves.

Z-scan data, respectively. Closed-aperture scan shows valley followed by a peak indicative of positive nonlinearity. GG 420 is transparent in the excitation wavelength region; hence the excitation is through simultaneous absorption of two photons. A dip in the open aperture Z-scan curve is clearly shown in the inset of figure 2a indicating the presence of two-photon absorption in the sample. Z-scan experiment with CS_2 was performed to standardize the set-up and to calibrate the laser intensity. Theoretical fitting to the experimental data of GG 420 was performed and is shown as a solid line in figure 2a. Nonlinear intensity-dependent refractive index (n_2) estimated from numerical fitting of the data was found to be 3.4 $\times$ 10^{-16} cm^2/W and two-photon absorption coefficient was 7×10^{-4} cm/GW.

Figure 2b shows open- and closed-aperture Z-scan data of the Schott colour glass filter OG 515. Squares and circles show experimental closed- and open-aperture Z-scan data, respectively. As absorption edge is red-shifted in OG 515 compared to GG 420 filter, this results in stronger two-photon absorption that is clearly evident

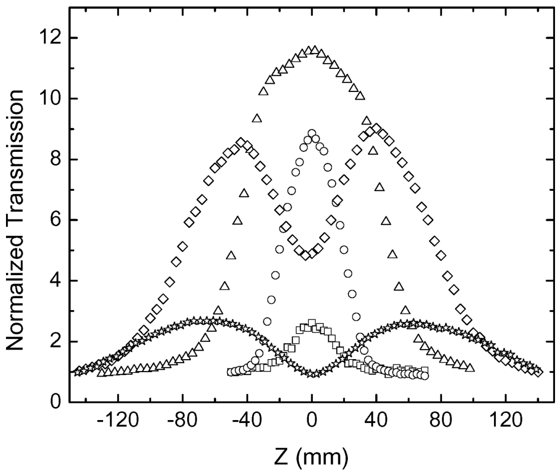

Figure 3. Open-aperture Z-scan profiles for RG 850 performed at intensities of 14 (squares), 50 (circles), 290 (triangles), 650 (diamonds) and 2.5×10^3 (stars) GW/cm^2.

from open-aperture Z-scan profile shown in figure 2b. Because of the large nonlinear absorption, the closed-aperture Z-scan curve becomes asymmetric. In this case, the valley gets enhanced and the peak gets suppressed. On numerical fitting to the experimental data, n_2 and TPA values were estimated to be 1.8×10^{-16} cm^2/W and 8×10^{-3}cm/GW, respectively. It should be noted that the TPA coefficient for OG 515 is much larger compared to GG 420.

Schott colour glass filter RG 850 absorbs 800 nm laser excitation beam resonantly. In this case, Z-scan experiment was performed at various excitation energies as shown in figure 3. At low energy, open-aperture Z-scan shows saturable absorption. At low intensities far from focal point, transmission through the sample is low due to strong linear absorption. As the intensity increases near the focal point, population of electrons saturates the conduction band, thus blocking further excitation from valance band, hence, transmission through the sample increases. This is one of the phenomena responsible for passive mode locking in lasers. As the excitation pulse energy increases saturable absorption in the sample becomes stronger. Further, the Z-scan experiment performed at a peak intensity of 650 GW/cm^2 or higher shows reverse saturable absorption near the focal point region and saturable absorption away from focal region. At this high intensity generated free carrier density increases significantly and starts absorbing excitation pulse itself, resulting in reduction of transmission through the sample. Dip in the transmission at $Z = 0$, may also be due to TPA process. It is evident from the figure that the Z-scan experiment performed at low-energy ΔZ at FWHM is ∼30 mm. In Z-scan experiments performed at high energy, the excitation pulse has sufficient intensity to induce saturable absorption in the sample away from the focus resulting in an increase in ΔZ value. Although the phenomenon of transition from saturable absorption to reverse saturable absorption with increase in intensity has been observed in organic molecules [8] and nanoparticles [9], it has not been reported, to our knowledge, in SDGs.

4. Conclusion

We have performed Z-scan experiments with femtosecond laser pulses in semiconductor-doped glasses having band gap in the resonance and non-resonance excitation photon regimes. RG850 shows the interesting behaviour of changing saturable absorption to reverse saturable absorption with increase in excitation pump energy.

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