Near-infrared nonlinear properties of antimony and lead oxyhalide glasses

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Abstract

Nonlinear optical properties of antimony and lead oxyhalide glasses were studied using picosecond Z-scan and femtosecond optical Kerr gate techniques at 1064nm and 800nm, respectively. The full width at half maximum of the third-order correlation signal was ~100fs which implies a fast temporal response of the samples. Nonlinear absorption was negligible in the range of intensities used. The results show a large nonlinear refractive index (n_2).

Introduction

Highly nonlinear optical materials are of large interest for a number of current photonic applications including optical limiting, all-optical switching and other uses. Amongst the materials available to date antimony based glasses are emerging as promising systems for such applications [1-4]. These glasses present high refractive index, large infrared transmission window and large thermal stability. Antimony oxide, Sb_2O_3, is the main glass component and is considered as the glass former.

Recently the nonlinear optical absorption of antimony and lead oxyhalide glasses (ALOG) were studied using laser pulses of 80 ps at 532 nm [4]. Large nonlinear absorption coefficients up to 20 cm / GW were measured which illustrate the possibility of using these glasses for optical limiting. Also, photo induced modifications of these glasses were observed which suggests that inscription of Bragg gratings can be made using green lasers of moderate powers.

In the present work, using femtosecond heterodyne optical Kerr gate and picosecond standard Z-scan techniques, we report measurements of the infrared third-order nonlinear optical response of new glasses compositions.

Experimental Setup

In this presentation, we report on nonlinear optical experiments performed with glass samples with the following composition:

Sample	Composition (% mol)
A	70% Sb2O3 – 30% SbPO4
B	60% Sb2O3 – 40% SbPO4
C	50% Sb2O3 – 50% SbPO4
D	40% Sb2O3 – 60% SbPO4
E	50% Sb2O3 – 40% SbPO4 – 10% PbO
F	40% Sb2O3 – 59% SbPO4 – 1% Ga2O3
G	30% Sb2O3 – 50% SbPO4 – 20% PbO
H	70% Sb2O3 – 30% Sb(PO3)3
I	70% Sb2O3 – 20% Sb(PO3)3 – 10% PbO

The purity for starting materials was at least 99% according to supplier's specifications. The glass synthesis was carried out by melting Sb_2O_3 (purity larger than 99%) and the others compounds in glassy carbon crucibles via an electrical furnace for 10 min at 700-1000°C in room atmosphere. Then, when a clear liquid is obtained, it is cast into a brass mold at 20°C and glass samples obtained after cooling. Finally, samples are annealed for few hours at a temperature lower than the glass transition temperature. The samples appear yellow in transmission and scattering defects limit the optical quality of large samples. For more details on the sample's preparation see ref. [2, 3].

Fig.1 shows the absorption spectra of the samples at room temperature which show a high transparency window for wavelengths larger than ≈ 500 nm.

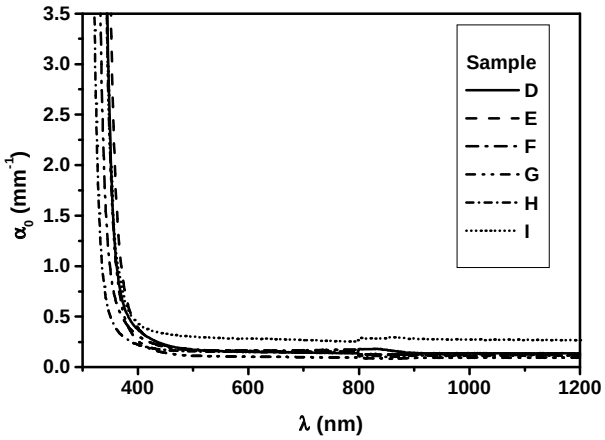

Figure 1: Absorption spectra of the antimony and lead oxyhalide glasses at different compositions (samples D, E, F, G, H, I).

Nonlinear refraction and nonlinear absorption were investigated using the standard Z-scan technique [7]. To determine n_2 and α_2 the sample is moved along the beam propagation direction (direction Z) using a computer controlled translation stage. Negative values of Z correspond to locations of the sample between the focusing lens and its focal plane. Measuring the variation of the transmitted beam intensity through an aperture placed in front of a detector in the far-field region (*closed aperture Z-scan*), it is possible to determine n_2. When the beam passing through the sample is unblocked (no aperture), and all the light is detected as a function of Z (*open-aperture Z-scan*), it is possible to measure α_2. The light source used for the Z-scan measurements was a Q-switched and mode-locked Nd:YAG laser delivering bursts of pulses of 100 ps duration at 1064 nm. Single pulses at low repetition rate (100 Hz) were selected using a pulse-picker. A lens with focal distance of 10 cm was used to focus the light beam in the sample. Slow detectors connected to a boxcar integrator were used for signal analysis.

The temporal response of the ALOG was evaluated using a typical Kerr shutter setup [5]; however in the present case we exploit an optical heterodyne detection. For these experiments a Ti:Sapphire laser system operating at 800 nm and delivering 100 fs pulses at a repetition rate of 82 MHz was employed. In this case, the light beam was split into two beams with 1:10 intensities ratio. The polarization of the stronger beam (pump) was set at 45° with respect to the polarization of the weaker beam (probe). Both beams were focused by a 10 cm focal length lens. The temporal overlap between the pulses is adjusted by a computer controlled delay line. The sample was placed at a position where the two beams cross and when they overlap temporally the probe beam polarization rotates due to the birefringence induced in the sample by the pump beam. In this situation a fraction of the transmitted probe beam passes through a polarizer crossed relative to the input probe beam polarization. A photo detector and a lock-in amplifier were used for signal processing.

For both experiments, the samples were submitted to laser intensity around 1.5 GW/cm^2.

Results and Discussions

Fig. 2 shows the behavior of the Kerr shutter signal as a function of τ for the sample D. For the assumed Gaussian pulse shape, the symmetric correlation signal implies that the samples have a temporal response faster than 100 fs. This means that the origin of the nonlinearity may be attributed to electronic processes either alone or in combination with other processes whose characteristic times are less than 100 fs. This signal was obtained by scanning a computer controllable delay line and monitoring the switched out probe beam intensity as a function of the delay time,τ between pump and probe pulses arriving on the sample position.

Fig. 3 (a) illustrates the temporal behavior of the Kerr signal as a function of the laser intensity while Fig. 3 (b) shows the linear behavior of the signal amplitude as expected for a heterodyne Kerr shutter signal. Here we used the sample I.

The magnitude of n_2 is obtained by comparison with the nonlinear refractive index of fused quartz of 2.2 x 10^{-16} cm^2/W [6] which was used as a calibration standard. The values of n_2 for the studied samples are given in table 2.

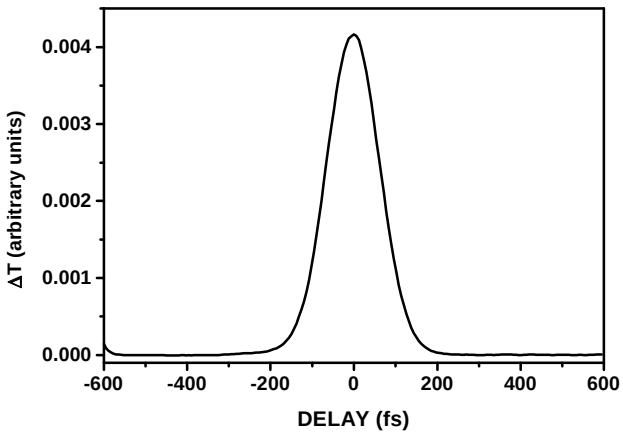

Figure 2: Behavior of the Kerr shutter signal for the sample D.

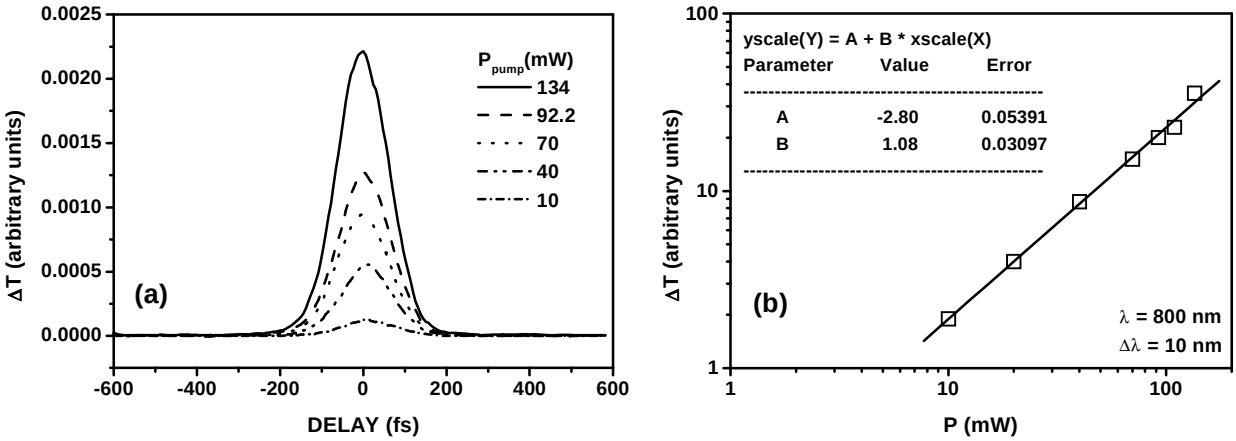

Figure 3:.Laser intensity dependence for sample I: (a) temporal behavior of the Kerr shutter signal; (b) linear behavior of the Kerr signal amplitude.

Sample	$\|n_2\|$ x 10^{-14} (cm^2/W)	Composition (% mol)
A	3.20	70% Sb2O3 – 30% SbPO4
B	1.32	60% Sb2O3 – 40% SbPO4
C	1.19	50% Sb2O3 – 50% SbPO4
D	2.02	40% Sb2O3 – 60% SbPO4
E	1.22	50% Sb2O3 – 40% SbPO4 – 10% PbO
F	0.64	40% Sb2O3 – 59% SbPO4 – 1% Ga2O3
G	1.18	30% Sb2O3 – 50% SbPO4 – 20% PbO
H	1.57	70% Sb2O3 – 30% Sb(PO3)3
I	2.16	70% Sb2O3 – 20% Sb(PO3)3 – 10% PbO

Table 2: n_2 values

Conclusions

In summary, we described the characterization of the response time and the value of the effective nonlinear refractive index of antimony polyphosphate glasses as well as demonstrate its application as an ultra fast Kerr gate. The response time in the experiments was limited by the laser pulse duration and indicates a fast response for the nonlinearity of the samples. On the other hand, the value of the nonlinearity is high in comparison to other optical glasses commonly studied [8].

Acknowledgements

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