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Optical limiting behavior of bismuth oxide-based glass in the visible range

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The authors report experimental results on the optical limiting behavior of a bismuth oxide-based glass by exciting the samples with nanosecond laser pulses at 532 and 598 nm. The results show that two-photon and free-carrier absorption processes contribute for the nonlinear absorption. Values for β, the two-photon absorption coefficient, and σ_e, the absorption cross section due to free carriers, were determined. The values of β and σ_e are dependent on the amount of bismuth oxide in the glass composition. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2393161]

Presently, there is an increasing effort in integrating all-optical devices in more complex structures that allow controlling light signals through amplification, modulation, and switching. Besides these characteristics, limiting the amplitude of a light signal is extremely relevant because optical elements have to be protected against overillumination.[1] Approaches to achieve this are based on the use of active and passive devices. Active devices rely on feedback loops, which control an aperture allowing that a greater or smaller amount of light pass through it, regulating the incoming light flux. Due to the electronic circuits required to drive such systems, they are in general too slow to follow the steep intensity variation of a nanosecond or a picosecond light pulse. On the other hand, passive devices that exploit the nonlinear (NL) optical susceptibility of a given substance to the incoming light field are more appropriate systems because the NL response may be ultrafast.

There are different mechanisms that allow operating passive optical limiters such as NL refraction,[2] induced scattering,[3] free-carrier absorption (FCA),[4] reverse saturable absorption,[5] and two-photon absorption (TPA).[6] There is also the possibility of improving the performance of an optical limiter by combining some of the above mentioned processes in a single device,[7,8] as well as by exploiting material's nonlinearity in a multipass configuration.[9] In any case, the NL properties of the material used determine the performance of the device.

In this work, we report on the optical limiting behavior of oxide-based glasses. The samples were fabricated following the procedure described in Ref. 10. The glasses were synthesized by melting and quenching the powdered starting materials: bismuth oxide (Bi_2O_3), zinc oxide (ZnO), and boron oxide (B_2O_3). The concentration of Bi_2O_3 was varied in the ternary system (Bi_2O_3–ZnO–B_2O_3) and the glass compositions studied in this work were (in mol %) as follows: 25.0 Bi_2O_3-37.5 ZnO-37.5 B_2O_3 (sample BZH2) and 12.5 Bi_2O_3-43.75 ZnO–43.75 B_2O_3 (sample BZH7). The samples with a thickness $L=1.0$ mm were excited either by the second harmonic (SH) of a Q-switched Nd:YAG (yttrium aluminum garnet) laser (7 ns, 5 Hz) or at 598 nm, obtained from a homemade dye laser pumped by the SH of the same Nd:YAG laser. To control the incident intensity on the sample, the laser beam was made to pass through a half-wave plate combined with a polarizer. Then, a 5 cm focal length lens focused the beam on the sample, which was positioned at the place corresponding to the smaller beam-waist size. Two large area photodiodes were used to measure independently the incident intensity and the transmitted intensity by the sample. Care was taken to collect all transmitted light to avoid errors in measuring the TPA coefficient β and the FCA cross section σ_e.

Figure 1 shows samples' linear absorption spectra obtained with a double beam spectrophotometer. Taking into account samples' reflectivities and their thicknesses, it was possible to determine a negligible linear absorption coefficient α_0. Table I shows the values of the reflectance R and samples' energy gap values E_g.

The experimental results for the NL transmission are shown in Fig. 2(a) and 2(b), for excitation wavelengths of 532 and 598 nm, respectively. The magnitude of the TPA coefficient and the FCA cross section was determined for each sample and for each excitation wavelength by analyzing the light attenuation through the samples, that is described by

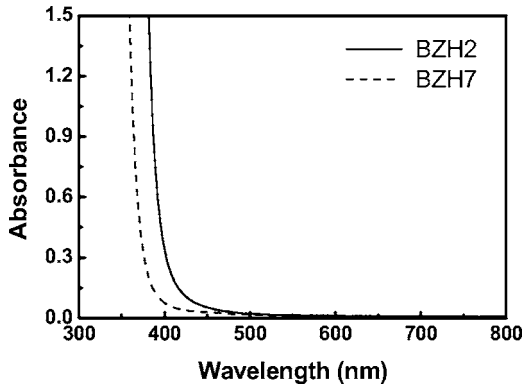

FIG. 1. Absorption spectra at room temperature. Samples' thicknesses are 1.0 mm.

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TABLE I. Samples' characteristics. The reflectivities R were calculated from the measured values of the linear refractive index n_0 considering normal incidence. The TPA coefficients β and the FCA cross section σ_e were obtained by fitting Eq. (3) to the experimental data. The values of β at 532 nm were measured in the picosecond regime (see Ref. 11).

		R		β (cm/GW)		σ_e ($\times 10^{-19}$ cm^2)	
Sample	E_g (eV)	532 nm	598 nm	532 nm	598 nm	532 nm	598 nm
BZH2	3.31	0.113	0.110	5.50	1.20	6.7	1.0
BZH7	3.50	0.089	0.087	1.50	0.36	3.1	0.5

$$\frac{dI(z)}{dz} = -\beta I^2(z) - \sigma_e N I(z), \tag{1}$$

where $I(z)$ is the light intensity along the propagation direction z, considering $\alpha_0=0$. The last term represents the FCA, with N being the photogenerated free-carrier density governed by the equation $dN/dt=[\beta I(z)/2\hbar\omega]I(z)$. For a Gaussian laser pulse represented by $I(z,t)=I(z)\exp(-t^2/\tau^2)$, where τ is the laser pulse duration, one obtains $N=\sqrt{\pi}/2(\beta\tau/2\hbar\omega)I^2(z)$. That leads to

$$\frac{dI(z)}{dz} = -[\beta I(z) + \gamma I^2(z)]I(z), \tag{2}$$

where $\gamma=\sqrt{\pi/2}(\beta\tau\sigma_e/2\hbar\omega)$. Thus an effective intensity dependent NL absorption coefficient can be defined as $\alpha_2^{\text{eff}}(I)=\beta+\gamma I$.

Integrating Eq. (2) and taking into account samples' reflectivity R one obtains the transmitted intensity through the sample given by

$$I(z=L) = \frac{(1-R)^2 I_0}{1+(1-R)I_0\{\beta L + (\gamma/\beta)\ln[[\beta+\gamma I(L)]/[\beta+\gamma(1-R)I_0][(1-R)I_0]I(L)]\}}, \tag{3}$$

where $I_0=I(z=0)$.

Equation (3) has two parameters, β and γ, that could be found using a NL curve fitting procedure to obtain the best agreement with the experimental data. However, the values for β at 532 nm, shown in Table I, were previously measured using the Z-scan technique in the picosecond regime.[11] Assuming that the picosecond measurements are not much affected by the FCA, those values were considered in the analysis of the present experiment to obtain the value for σ_e at 532 nm. Then, the value of σ_e obtained for 532 nm was used as the starting value in the fitting procedure to determine the NL parameters at 598 nm. The solid lines in Figs. 2(a) and 2(b) represent the solution of Eq. (3) with the best-fit values obtained using the MATCHCAD® software. The NL parameters obtained are summarized in Table I.

The experimental data presented in Figs. 2(a) and 2(b) and the results shown in Table I indicate that the TPA coefficient grows considerably when doubling the Bi_2O_3 molar concentration. Notice also that the effective NL absorption coefficient is larger for two-photon excitation deeper in the conduction band, where one has a larger free-carrier density of states.

Concerning materials' potential as an optical limiter, the sample BZH2 is somewhat promising, as illustrated in Fig. 3. An appropriate quantity to evaluate the performance of the sample is the dynamic range (DR), defined as the ratio between the maximum intensity used in the experiment and the threshold intensity.[12] For the sample BZH2 at 532 nm (at 598 nm) the DR was estimated to be $\approx 5(\approx 2)$, which is the same order of magnitude as obtained in experiments with

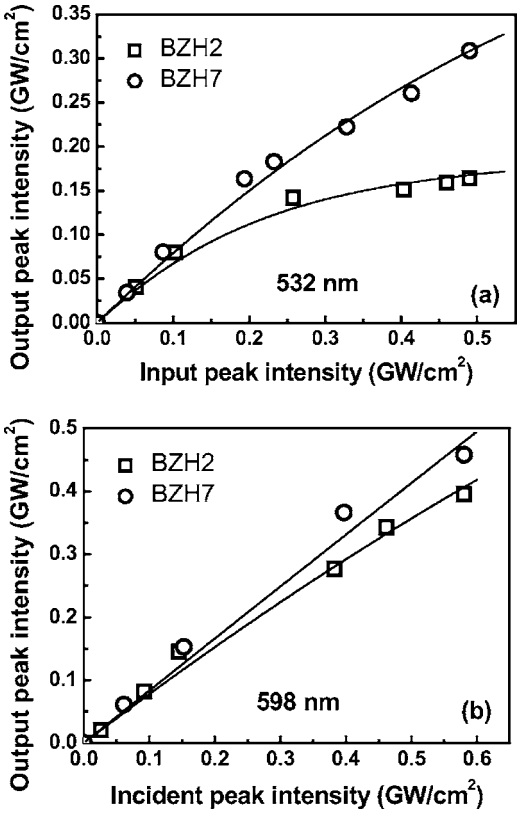

FIG. 2. Transmitted intensity as a function of the laser intensity. Wavelength of excitation: 532 nm (a) and 598 nm (b).

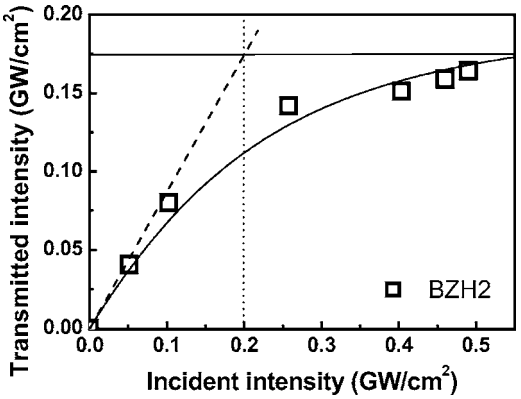

FIG. 3. Optical limiting behavior of the sample BZH2 for excitation at 532 nm.

organic materials. Nevertheless, the BZH glass does not suffer from drawbacks associated with organic systems such as large linear absorption, low damage threshold, and poor chemical stability. Notice that for the sample BZH2, the TPA coefficient has the same order of magnitude of those measured in tungstate fluorophosphates[13] and chalcogenide glasses.[14] However, the performance of BZH glass is better as an optical limiter due to the FCA contribution.

In summary, we have measured the NL absorption coefficients of bismuth-based glasses at 532 and 598 nm. It was observed that increasing the amount of bismuth oxide in the glass composition by 100% produces an enhancement of the TPA coefficient of $\approx 250\%$ and an increase of $\approx 100\%$ in the free-carrier absorption cross section. The sample with 25% of Bi_2O_3 shows a large dynamic range for single pass optical limiter in the nanosecond regime.

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