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► To cite this version:

Cid de Araújo, Edilson Falcao-Filho, A. Humeau, Dominique Guichaoua, Georges Boudebs, et al.. Picosecond third-order nonlinearity of lead-oxide glasses in the infrared. *Applied Physics Letters*, 2005, 87 (22), pp.221904. <10.1063/1.2137457>. <hal-01389338>

HAL Id: hal-01389338

<https://hal.science/hal-01389338v1>

Submitted on 8 Oct 2021

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Picosecond third-order nonlinearity of lead-oxide glasses in the infrared

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(Received 12 July 2005; accepted 4 October 2005; published online 22 November 2005)

Heavy metal-oxide glasses containing lead and bismuth were prepared, and their picosecond third-order nonlinear (NL) optical characteristics were investigated. NL refractive indices of $\approx 10^{-18} \text{ m}^2/\text{W}$ at 1064 nm were measured. Negligible NL absorption was verified and, as a consequence, the samples present a good factor-of-merit for photonic applications. © 2005 American Institute of Physics. [DOI: 10.1063/1.2137457]

Glasses with high nonlinearity have been extensively studied in the past years by many authors (Refs. 1–6, and references therein). Presently it is well known that there is a direct relation between the nonlinear (NL) refractive index, n_2 , of a glass and the polarizability of its constituents. For example, the presence of heavy-metal ions increases n_2 . From the applied point of view transparent NL glasses have many advantages compared to inorganic crystals and organic materials because of their fast response times, small linear losses, and negligible two-photon absorption coefficient, α_2 , for wavelengths in the infrared. In particular the heavy metal-oxide glasses containing lead and bismuth deserve much attention because of their large potential for photonic applications. These glasses have high refractive indices, large transmission window in the visible and near infrared, and cutoff phonon energy varying from ≈ 500 to $\approx 700 \text{ cm}^{-1}$. They are very stable and resistant to moisture.^{1–6}

This work presents measurements of the NL properties of four compositions of lead-oxide glasses at 1064 nm, using the Z-scan technique.⁷ Large values of n_2 and small values of α_2 were determined, and the results were compared with the Boiling, Glass, and Owyong (BGO) model.⁸ A good agreement is found between experimental and theoretical values.

The oxide glasses investigated in this work were obtained following the procedure described in Refs. 3–5 starting with the following composition in weight percent: 46 PbO–10 Ga₂O₃–42.6 Bi₂O₃–1.4 BaO (sample A); 46 PbO–12 Ga₂O₃–42 Bi₂O₃ (sample B); 13.3 PbO–55.5 GeO₂–31.2 Bi₂O₃ (sample C); and 35.3 PbO–48.9 PbF₂–15.8 B₂O₃ (sample D). The reagents were melted in platinum (samples A, B, and D) and alumina (sample C) crucibles, at 1000 °C, for 1 h, quenched in air (in a heated brass mold), annealed for 1 h, at 300 °C (samples A and B, considering the transition temperature of 330 °C) and at 420 °C (samples C; transition temperature ≈ 430 °C) and then cooled to room temperature, inside the furnace. Finally,

samples with dimensions $10 \times 10 \times 2 \text{ mm}^3$ and good optical quality were cut and polished for the experiments.

Optical absorption spectra of the samples were measured with a double beam spectrophotometer from 200 to 2000 nm and the linear index of refraction was measured at 632.8 nm and at 1550 nm using the prism-coupler technique. The results are shown in Figs. 1(a) and 1(b).

NL refraction and NL absorption were investigated using the Z-scan technique.⁷ Figure 2 shows the setup used. The excitation is provided by a linearly polarized mode-locked

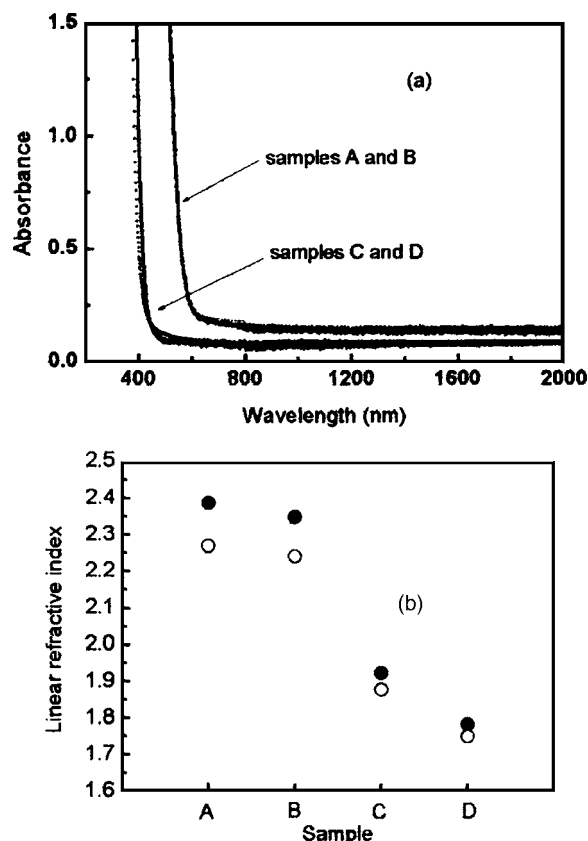


FIG. 1. (a) Absorption spectra. (b) Linear refractive indices. Full dots (632.8 nm). Open dots (1550 nm).

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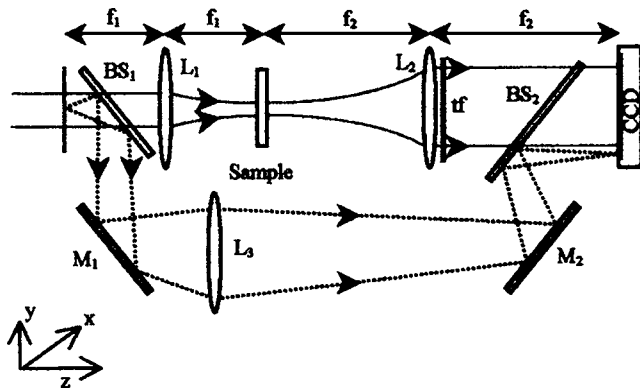


FIG. 2. Schematic of the experimental setup. The sample is located in the focal region. The labels refer to: lenses (L_1 – L_3); mirrors (M_1 , M_2); beam splitters (BS_1 , BS_2); neutral filters (tf).

Nd:yttrium-aluminum-garnet laser (1064 nm; pulse duration of 15 ps; 10 Hz). The Gaussian laser beam is focused using lens L_1 with focal distance $f_1 = 20$ mm. The waist radius at the focal plane is $w_0 = 30$ μm , corresponding to a Rayleigh length of 2.7 mm. The measurements were performed using a $4f$ system with two different incident intensities on the samples ($I_0 = 2.1$ and 4.4 GW/cm^2). The image receiver is a 1000×1018 pixels cooled camera (-30 $^\circ\text{C}$) operating with a fixed gain. It is placed at a distance equal to $f_2 = 20$ mm from lens L_2 in order to obtain the image of the focal plane into the charge coupled device. A reference beam incident in a small area of the camera allows to monitor the energy fluctuation of the laser. The sample is moved in the focus region along the beam propagation direction (Z axis). Negative values of Z correspond to locations of the sample between the focusing lens and its focal plane. Open and closed Z-scan normalized transmittance were numerically processed from the acquired images by integrating over all the pixels in the first case and over a circular numerical filter in the second one (giving a linear aperture transmittance S equal to 0.2). Lens L_2 contributes to produce the Fourier transform of the field at the exit surface of the sample, which is physically similar to the far field diffraction obtained with the original Z-scan method. The simplified Eq. (13) in Ref. 7 relating the nonlinear dephasing to ΔT_{pv} , the difference between the normalized peak and valley transmittance, remains valid for the $4f$ system. For more experimental details concerning our Z-scan procedure, see Ref. 9, and references therein.

Figure 3 shows Z-scan traces for samples A and B measured at 2.1 GW/cm^2 . The profiles are characteristic of positive nonlinearity. Samples C and D also present positive n_2 . NL absorption was not observed for all samples, which implies that the two-photon absorption coefficients are below the detection limit ($\alpha_2 < 0.1$ cm/GW). The magnitude of n_2 is obtained by comparison with the value for CS_2 , 3×10^{-18} m^2/W ,⁷ that was used as a calibration standard. Table I summarizes the results and gives the mean value of n_2 obtained at 2.1 and 4.4 GW/cm^2 .

To evaluate the potential of the present materials for all-optical switching devices, it was calculated the figure-of-merit (FOM), $n_2/\lambda\alpha_2$, and the results are in Table I. It can be seen that samples A and B present $\text{FOM} > 1$ and they are expected to have good performance in all-optical switching applications.¹⁰

Using the BGO model,⁸ it was possible to estimate values for n_2 which are in agreement with the experimental

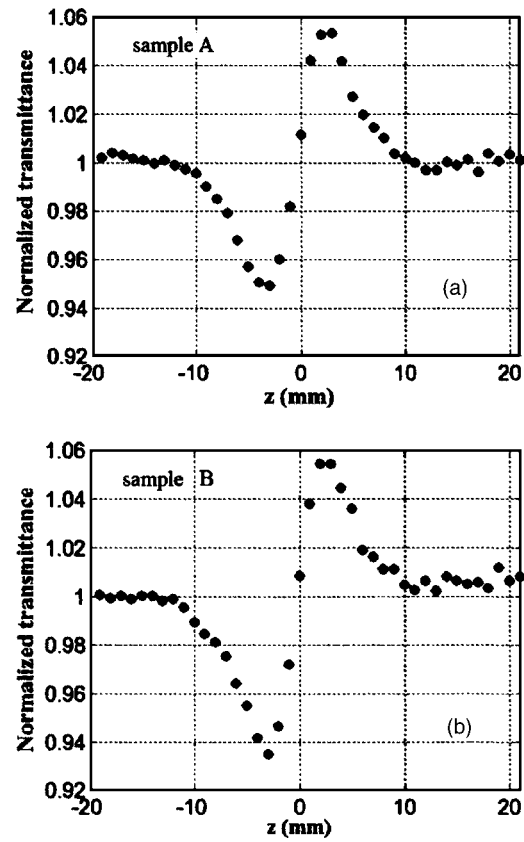


FIG. 3. Experimental Z-scan transmittances (points) for sample A (1.60 mm thick) and sample B (2.0 mm thick) for $I_0 = 2.1$ GW/cm^2 at $\lambda = 1.06$ μm .

data. In the BGO model the NL polarizability is assumed to be proportional to the linear polarizability squared and the optical dispersion of the medium is determined by only one resonance at frequency ω_0 . The incident light field of frequency ω is considered to be far from resonance ($\omega \ll \omega_0$) and n_2 , written in Gaussian units, is given by

$$n_2[\text{esu}] = \frac{[n_0(\lambda)^2 + 2]^2 [n_0(\lambda)^2 - 1]^2 (gs)}{48\pi n_0(\lambda) \hbar \omega_0 (Ns)}, \quad (1)$$

where N is the density of nonlinear oscillators, s is the effective oscillator strength, g is a dimensionless parameter given by $g = \mu s \hbar / m \omega_0$, where μ is the nonlinear coupling coefficient, $(2\pi \hbar)$ is the Planck's constant, and m is the electron mass. The linear refractive index for light wavelength λ is denoted by $n_0(\lambda)$ and satisfies the expression

$$\frac{4\pi [n_0(\lambda)^2 + 2]}{3 [n_0(\lambda)^2 - 1]} = \frac{\omega_0^2 - \omega^2}{(e^2/m)(Ns)}, \quad (2)$$

where e is the electron charge in Gaussian units.

TABLE I. Linear refractive index (n_0) at 1064 nm, NL refractive index (n_2), two-photon absorption coefficient (α_2), figure-of-merit ($n_2/\alpha_2\lambda$), and predictions of the BGO model [$n_2(\text{BGO})$].

Glass	n_0	Thickness (mm)	n_2 (10^{-18} m^2/W)	α_2 (cm/GW)	$n_2/\alpha_2\lambda$	$n_2(\text{BGO})$ (10^{-18} m^2/W)
A	2.3	1.60	1.6	≤ 0.1	> 1.5	1.4
B	2.3	2.00	1.3	≤ 0.1	> 1.2	1.2
C	1.9	1.05	0.4	≤ 0.1	> 0.4	0.33
D	1.75	3.05	0.3	≤ 0.1	> 0.3	0.21

The parameters N_s and ω_0 are obtained from the values of $n_0(\lambda=632.8\text{ nm})$ and $n_0(\lambda=1550\text{ nm})$ measured for each sample. Then, the values of N_s and ω_0 are introduced in Eq. (1) to determine n_2 . The value of $gs=3$ was used here since it is an appropriate value for oxides.^{8,11} The value of $n_0(\lambda=1064\text{ nm})$, was determined using Eq. (2) with the values of N_s and ω_0 obtained for each sample. The results calculated for n_2 at 1064 nm, in units of $10^{-18}\text{ m}^2/\text{W}$, are 1.4 (sample A), 1.2 (sample B), 0.33 (sample C), and 0.21 (sample D) which are in agreement with the measured values.

In summary, the NL refraction and NL absorption of heavy metal-oxide glasses were determined for excitation at 1064 nm. Samples with larger concentration of PbO and Bi₂O₃ present larger nonlinear refractive indices. The results indicate large potential of the samples for photonic applications.

C.B.deA. acknowledges the Université d'Angers for financial support during his stay at P.O.M.A. Financial support by the Brazilian agencies Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Fundação de Amparo à Ciência e Tecnologia do Estado de Pernambuco

(FACEPE), and Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP) is also acknowledged.

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