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Rogério F. Souza
Departamento de Eletrônica, Centro Federal de Educação Tecnológica de Alagoas, Maceió, Alagoas 57000-000, Brazil

Márcio A. R. C. Alencar[a] and Jandir M. Hickmann
Instituto de Física, Universidade Federal de Alagoas, Maceió, Alagoas 57072-970, Brazil

Renata Kobayashi
Departamento de Engenharia de Sistemas Eletrônicos, Escola Politécnica, Universidade de São Paulo, São Paulo, São Paulo 05508-900, Brazil

Luciana R. P. Kassab
Laboratório de Vidros e Datação, Faculdade de Tecnologia de São Paulo, CEETEPS/UNESP, São Paulo, São Paulo 01124-060, Brazil

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The third-order nonlinear optical properties of tellurite glasses with different compositions were investigated in the femtosecond regime at 810 nm. Using the *I*-scan technique, positive nonlinear refractive indices of $\sim 10^{-15}$ cm^2/W were measured. The authors also determined that nonlinear absorption was negligible for all studied samples. This result, added to their good chemical stability, indicates that tellurite glasses are promising materials for ultrafast photonic applications. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2364467]

In recent years, materials that present large nonlinear optical properties have been extensively studied aiming their applications to the development of several photonic devices.[1–10] Different kinds of media, such as polymers,[2] organic liquids,[3] crystals,[4] glass ceramics,[5] nanostructured materials,[6,7] and glasses,[8–12] are currently engineered to possess large nonlinear responses. For instance, it is well known that the presence of heavy-metal ions often increases the nonlinear refractive index of a medium.[8] Thus, several families of glasses containing heavy-metal ions have been developed and are promising candidates to photonic applications such as optical limiting and all-optical switchings.

Among the heavy-metal glass families, the TeO_2 based are promising materials for such applications.[10–14] These tellurite glasses are transparent in the visible, near, and middle infrared regions. In comparison with silica glasses, they present larger refraction index and smaller phonon energies (~ 700 cm^{-1}), comparable to germanate glasses. In relation to fluoride glasses, which also have various photonic applications, they offer the advantage of a better chemical durability. However, nonlinear optical properties of such glasses were not completely investigated yet.

In this letter, we report on the measurement of the nonlinear refraction and absorption in the femtosecond regime for five different compositions of tellurite glasses employing the *I*-scan technique. We observed relatively large n_2 and very small α_2 for laser excitation at 810 nm. Our results indicate that tellurite glasses are very good candidates for ultrafast photonic applications.

Figure 1 shows the linear absorption spectra, in the range from 300 to 850 nm, and the optical transmission in the medium infrared region for the studied compositions. The composition of the studied samples is presented in Table I. It should be noted that the samples present a large transparent window in the visible and near infrared regions (400–6000 nm). In the infrared region, the samples present two absorption peaks at 3.2 and 4.2 μm, associated with OH^- ions and CO_2 contaminants, respectively.

The nonlinear optical properties of the glasses were investigated employing the *I*-scan technique. It is a simple and reliable single beam technique used to determine both magnitude and sign of third-order nonlinear optical responses of a medium.[15] As well as in the *Z*-scan technique,[16] the effect of self-focusing/defocusing is exploited to evaluate the nonlinear refraction n_2 and absorption α_2 of the material under investigation. However, while in *Z*-scan the sample position with respect to the focal plane is varied, in the *I*-scan tech-

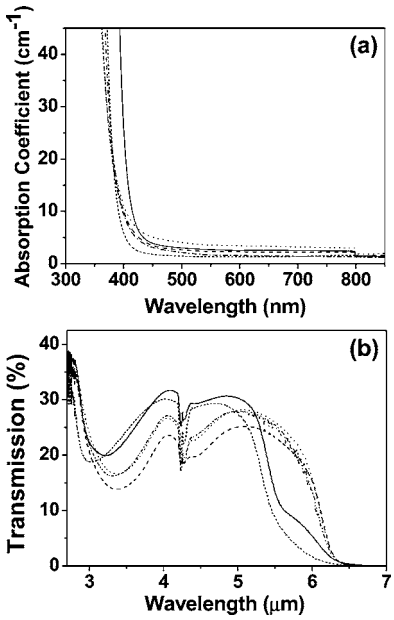

FIG. 1. UV-visible absorption (a) and middle infrared transmission spectra (b) for the glasses B3 (dashed line), B4 (solid line), Q1 (dotted line), Q2 (dash-dot curve), and Q3 (short dash curve).

[a]Electronic mail: marca@loqnl.ufal.br

TABLE I. Optical properties of the studied tellurite glasses.

Glass	Composition	n_0	n_2 (10^{-15} cm^2/W)	α_0 (cm^{-1})	α_2 (cm/GW)	W
B3	TeO_2–BaO	2.1	2.8	1.4	<0.1	0.36
B4	TeO_2–Nb_2O_5	2.1	2.7	0.9	<0.1	0.54
Q1	TeO_2–ZnO–Na_2O–PbO	2.1	1.4	0.7	<0.1	3.62
Q2	TeO_2–ZnO–Na_2O–GeO_2	2.0	2.1	1.5	<0.1	2.53
Q3	TeO_2–GeO_2–BaO–Nb_2O_5	1.9	1.1	1.4	<0.1	1.42

nique the laser intensity is changed, keeping the sample static. This technique was proposed as an alternative method to *Z*-scan in cases where the samples present surface inhomogeneity, nonparallel faces, and low damage threshold.[17] Although the studied glasses present good optical quality and high damage threshold, they have large linear refractive indices. Thus, even a small angle between the glass surfaces could lead to a large deviation in the beam propagation direction, which would be a real problem in *Z*-scan measurements. Hence, we believe that *I*-scan technique is more suitable to be employed in our case than *Z*-scan.

The *I*-scan experimental setup used in this work is depicted in Fig. 2. A mode-locked Ti:sapphire laser operating at 810 nm, producing 200 fs pulses in a 76 MHz repetition rate, was used as a light source. A pulse selector was used to reduce this rate to 1 kHz in order to avoid the cumulative effects. The laser beam was focused using a 7.5 cm focal lens preceded by a set of a wave plate followed by a polarizer (WP-P combination). This configuration allowed a continuous change in the laser intensity during the measurements. The 2 mm thick samples were mounted on a translation stage and positioned at a distance z_0 away from the focal plane by a computer controlled stepping motor. The light transmittance was measured by a closed aperture photodetector as a function of the input beam intensity. The detected signal was amplified by a lock-in amplifier and acquired by a computer. To obtain the normalized transmittance another measurement was made at a position far away from the focal plane, where the nonlinear effects are negligible. The nonlinear refractive index n_2 was then obtained by a theoretical fitting using the equation

$$T(I_0) = 1 + \frac{4kgdn_2L_{\text{eff}}}{\sqrt{2}(g^2 + 9d^2/d_0^2)d_0}I_0, \tag{1}$$

where $g=1+da/(a^2+z_0^2)$, $d_0=\pi w^2/\lambda$, I_0 is laser intensity, z_0 is the Rayleigh length, a is the sample position behind the focal plane, d is the distance between the sample and the aperture, w is the beam radius at the sample, $k=2\pi/\lambda$ is the wave vector magnitude, and $L_{\text{eff}}=(1-\exp(-\alpha_0 L))/\alpha_0$ and α_0 are the effective length and the linear absorption coefficient of the sample, respectively. A similar procedure was made to perform nonlinear absorption measurements but using an open aperture configuration. In this situation, the normalized transmittance can be expressed as

$$T(I_0) = 1 - \frac{\alpha_2(1-R)L_{\text{eff}}}{2^{3/2}}I_0, \tag{2}$$

where R is the reflection of the first face of the sample.

The geometric parameters of the *I*-scan setup are obtained performing an initial *Z*-scan measurement in a reference material. This measurement works as a calibration procedure from which the aperture factor and the sample position are selected for the *I*-scan measurements. In this way, the quality of the beam is assured from the *Z*-scan curve, and any deviation from the Gaussian model can be observed. In our work, the calibration procedure was performed using CS_2 as reference medium. In this case, it was observed that the *Z*-scan curve deviated very little from the theoretical model for Gaussian beams. Besides, the measured values for n_2 of CS_2, using *Z*-scan and *I*-scan, presented very good agreement with results of previous work.[18] These facts indicate that the geometric parameters of the *I*-scan setup were accurately chosen and that the laser beam was Gaussian.

In Fig. 3, we present typical results of the normalized *I*-scan for closed and open apertures and the corresponding theoretical fittings for sample B4. For this sample, we determined a positive nonlinear refractive index equal to 2.7 $\times 10^{-15}$ cm^2/W. Table I shows the optical properties of the investigated glasses. Moreover, the open aperture configuration did not reveal any nonlinear absorption, leading us to conclude that for all studied glasses this quantity is smaller than our system resolution, around 0.1 cm/GW. In order to assure that there was any influence of thermooptical effects on the measured n_2, we repeated the *I*-scan measurement

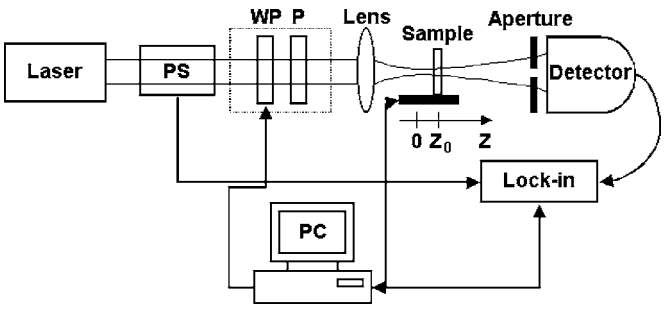

FIG. 2. Experimental setup for optical nonlinearity measurements using *I*-scan technique.

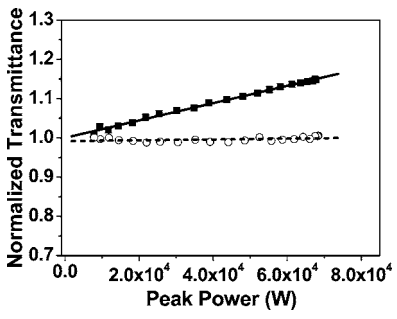

FIG. 3. *I*-scan results for sample B4, closed (solid squares) and open (open circles) aperture configurations. Solid and dashed lines correspond to the linear fitting to the closed and open aperture experiments, respectively.

using the laser with a 300 Hz repetition. Using this lower repetition rate, we obtained within the experimental error the same n_2 value as at 1 kHz. This result indicates that thermal effects contribute very little to the measured nonlinear refractive indices of the investigated tellurite glasses.

Suitable optical materials for all-optical switching devices must possess large enough n_2 to perform optical switching operations in thicknesses comparable to the absorption length. Hence, a good material for optical switching in a nonlinear Fabry-Pérot configuration should satisfy the conditions $W=\Delta n_{\max}/\lambda\alpha_0>0.27$, where $\Delta n_{\max}$ is the maximum refractive index change achievable, limited by saturation, and $T=2\alpha_2\lambda/n_2<1$.[19] As it can be observed in Table I, all samples presented very good results for W figure of merit, indicating that the tellurite glasses studied in this work are very good candidates to all-optical switching applications in the femtosecond regime.

The figure of merit T could not be accurately evaluated due to the fact that the nonlinear absorption coefficients α_2 of the studied glasses were much smaller than our system resolution.

It is worth comparing our results with a previous work that studied the optical properties of tellurite glasses in the picosecond regime.[20] Sabadel *et al.* demonstrated that the introduction of elements such as Ba or Ti changed the coordination of Te atoms in the glasses, affecting their physical properties. The authors measured the nonlinear refractive indices and absorption coefficient of four different tellurite glass compositions, using laser pulses with 250 ps, at 532 nm. Their results were about one order of magnitude larger than what we observed in our measurements.

These discrepancies can be explained as follows: firstly the compositions presented in this work are different from those investigated in Ref. 19; besides our measurements were performed with a laser tuned at 810 nm, while in Ref. 19 a laser at 532 nm was used. Hence, the detuning between the laser excitation and the glass excited state is larger in our case and consequently the n_2 values obtained in our work are expected to be smaller. We should also add that it is well known that some materials present different nonlinear optical properties when different regimes are employed for the excitation (femtosecond, picosecond, and nanosecond excitations).[21,22] This is normally due to contributions of free carriers or long lived impurity states which are more important in experiments with longer laser pulses.

In summary, we have investigated the nonlinear optical properties of five different compositions of tellurite glasses, in the femtosecond regime, employing the I-scan technique. It was observed that these glasses present positive nonlinear refractive indices of $\sim 10^{-15}$ cm^2/W, and low nonlinear absorption coefficients (<0.1 cm/GW). The figure of merit W for all-optical switching was also evaluated for these glasses, showing that they are very good candidates to the development of photonic devices in the femtosecond regime.

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