Third-order nonlinear properties of a silica niobate nanocomposite

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Abstract

We have investigated the third-order nonlinear optical response of a glass-ceramic containing sodium niobate ($NaNbO_3$) nanocrystals using picosecond Z-scan and femtosecond optical Kerr gate techniques at 1064 nm and 800 nm, respectively. The results show a large dependence of the nonlinear refraction index (n_2) on the volume fraction of $NaNbO_3$ crystallites. Nonlinear absorption is negligible for all samples and the response time of the nonlinearity was shorter than 100fs demonstrating that this material is a promising candidate for ultrafast optical switching.

Introduction

The properties of glass-ceramic containing ferroelectric nanocrystals were first discussed long time ago [1]. The dielectric and optical behavior of this class of materials are different from those of the bulk materials, being a function of the crystallite size and filling factor. Not many studies of the optical properties of glass-ceramic systems were performed however, probably because of the notion that scattering losses would be large enough to prevent their use in actual devices. Presently a large variety of transparent glass-ceramic systems are known [2] and optical fibers with low intrinsic losses can be prepared based on ultra-transparent glass-ceramic. A peculiar characteristic of nanostructured glass-ceramic is that their optical nonlinearity is influenced by quantum mechanical confinement effects and local field effects, which can be exploited for specific applications.

One class of these materials currently being investigated is glass-ceramic containing ferroelectric crystallites. Photonic structures consisting of lithium niobate microcrystallites in a SiO_2 based glass were reported [4], and the nonlinear optical behavior of a glass-ceramic containing sodium niobate ($NaNbO_3$) nanocrystals (GC-SNN) has also been studied in recent experiments by our group [5,6]. We have measured the real (refractive) and imaginary (absorptive) parts of the third-order nonlinear susceptibility, $\chi^{(3)}$, of a GC-SNN in the visible (532 nm) using nanosecond power limiting [5] and picosecond Z-scan [6] techniques. It was shown that the presence of $NaNbO_3$ nanocrystals in the glass-ceramic influences the magnitude of the nonlinear (two-photon) absorption coefficient (α_2) and the nonlinear refractive index (n_2) of the material. More recently, the formation of thick GC-SNN films on lithium-silica-niobate glass substrate was reported [7], which supports the prospects for future uses of GC-SNN in integrated optics. Due to the small detuning of the laser frequency used in the previous experiments [5, 6] with respect to the energy bandgap, however, it was not clear if the measured refractive nonlinearity is dominated by the contribution of bound electrons (Kerr nonlinearity) or from free carriers (generated by two-photon absorption). If the Kerr nonlinearity dominates, the value of n_2 is expected to be independent of the excitation wavelength, and its response to the excitation should be instantaneous. Thus, it is important to evaluate the response time of the nonlinearity probing $\chi^{(3)}$ at wavelengths where nonlinear absorption contributions are negligible.

In this work we describe investigations of the third-order nonlinear properties of the GC-SNN using picosecond Z-scan and femtosecond Optical Kerr Gate (OKG) in the near-infrared with photon energies well below the energy bandgap.

Experimental Setup

The glass-ceramic samples were prepared by heat treatment of the glass with molar % composition of $35SiO_2$-$31Nb_2O_5$-$19Na_2O$-$11K_2O$-$2CdO$-$2B_2O_3$ following the procedure described in ref. [5]. Samples A, B, C and D were heat-treated at 610 °C during 141, 33, 16, and 8 hours, respectively. During the heat-treatment, sodium niobate ($NaNbO_3$) nanocrystals were formed and the volume fraction occupied by the nanocrystals were controlled by the heat-treatment time. The formation of $NaNbO_3$ crystallites was confirmed by X-ray diffraction which shows that the amplitude of the peaks due to the diffraction by the nanocrystals increases with the heat-treatment duration. The volume fraction of $NaNbO_3$ crystallites in samples A, B, C, and D was estimated from the X-ray data and density measurements and it corresponds to 0.4, 0.2, 0.08, and 0.025, respectively. The

average size of the $NaNbO_3$ crystallites does not depend on the heat-treatment time according to small-angle X-ray scattering measurements, and their typical dimensions are ~ 100 Å.

The linear absorption spectra from 200nm to 1200nm were obtained using a double beam spectrophotometer and Fig. 1 shows the results for all samples at room temperature. The samples exhibit a transparency window extending from the near-infrared to the visible and the onset of absorption occurs in the blue-green region.

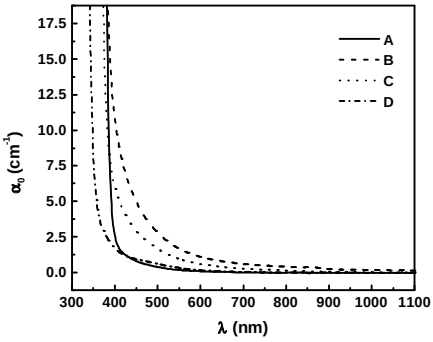


Figure 1: Absorption spectra of the glass-ceramic samples containing different volume fraction of $NaNbO_3$ nanocrystallites.

Nonlinear refraction and nonlinear absorption were investigated using the standard Z-scan technique [8]. 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 (12 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 providing intensities up to ~ 5 GW/cm^2 at the focal plane. Slow detectors connected to a boxcar integrator were used for signal analysis.

The temporal response of the GC-SNN was evaluated using an OKG setup [3]. 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 1:7 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 and the intensity at the focus was ~ 1.4 GW/cm^2. 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 photodetector and a lock-in amplifier were used for signal processing.

Results and Discussions

Figure 2 shows typical *closed – aperture Z-scan* traces for samples A and D. Each data point represents the average of 120 shots and 4 scans for $S = 0.005$. Liquid CS_2 was used as a reference sample with $n_2 = 3.3 \times 10^{-14}$ cm^2/W [8]. The prefocal (Z<0) minimum followed by the postfocal maximum corresponds to positive n_2 value**.** The separation between peak and valley in the Z-scan transmittance curve is in agreement with $\chi^{(3)}$ processes, being of $1.7Z_o$ where Z_o is the confocal parameter.

Figure 3 shows the OKG signal of sample A compared with the signal of liquid CS_2 which presents a slow decay in the picosecond range [10]. The symmetrical curves obtained with the GC-SNN samples indicate that the response time of our glasses is faster than 100 fs, which is a characteristic feature of pure electronic nonlinearities. The values found for n_2 were determined using SiO_2 as reference with $n_2 = 2.2 \times 10^{-16}$ cm^2/W [11].

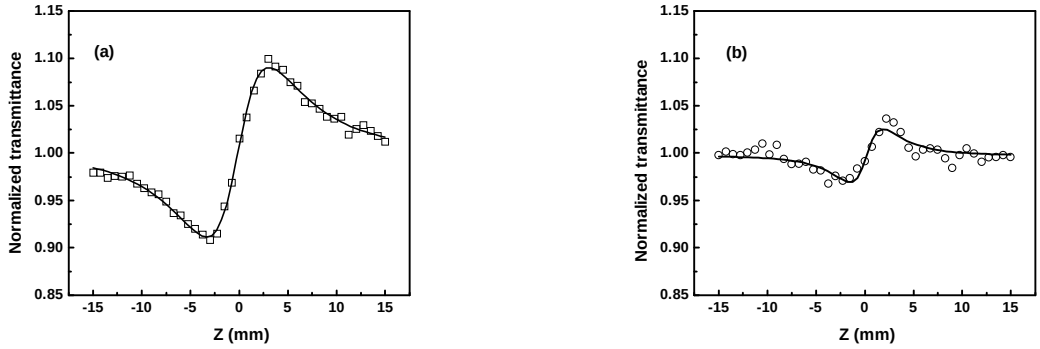


Figure 2: Normalized *closed aperture Z-scan* curves. (a) Sample A (volume fraction of nanocrystals: f=0.4); (b) Sample D (f=0.025).

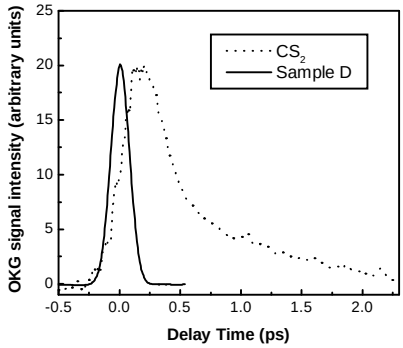
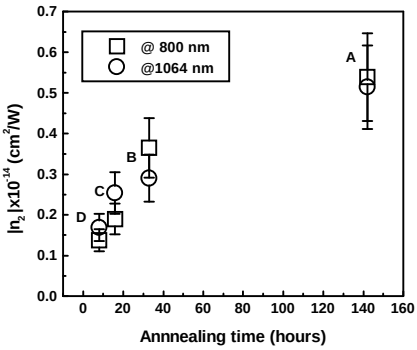


Figure 3: Optical Kerr Gate signals of glass-ceramic containing $NaNbO_3$ nanocrystallites (sample A) and liquid CS_2 .

Fig.4 displays the results for all samples as measured at 1064 nm and 800 nm with estimated errors of ± 20%. Note that n_2 is influenced by the volume fraction of $NaNbO_3$ crystallites, as already observed in refs. [5, 6] and a saturation-like behavior of n_2 is observed for increasing concentration of $NaNbO_3$ nanocrystals. The values of n_2 in the femtosecond regime at 800nm, match quite well with the values found using picosecond Z-scan at 1064 nm.



1. **Figure 4:** Dependence of n_2 as a function of the volume fraction of $NaNbO_3$ nanocrystals.

We notice that n_2 is positive in the infrared, while a negative n_2 was observed at 532 nm [6] in accordance with the predictions of a theory based upon a two-band model [9] which predicts that the sign of n_2 must change from negative to positive when the ratio between the photon energy ($h\upsilon$) and the bandgap energy (E_g) satisfies ($h\upsilon/E_g$) ~ 0.6. Two-photon absorption was not observed in the present experiments which allows to estimate α_2 as smaller than 0.02 cm/GW.

Conclusions

In conclusion, our investigation of the optical nonlinearity of a glass-ceramic containing nanocrystals of $NaNbO_3$ has shown that the magnitude of the nonlinear coefficient is influenced by the volume fraction of $NaNbO_3$. Additionally, the response time of the nonlinearity has been shown to be faster than 100 fs. The fact that the values found for n_2 are in agreement for 800nm and 1064nm, the symmetrical Z-scan curve and the ultrafast response of the samples indicate that the material response is mainly of electronic origin. These results indicate that GC-SNN may be a promising candidate for broadband all-optical switching.

Acknowledgements

We acknowledge the financial support by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Programa Nacional de Núcleos de Excelência (PRONEX), and INTAS - Russia (grant # 0818). We also thank B. V. Tatarintsev for heat treatments and B. J. P. da Silva for polishing the samples.

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