



The effect of excitation intensity variation and silver nanoparticle codoping on nonlinear optical properties of mixed tellurite and zinc oxide glass doped with Nd₂O₃ studied through ultrafast z-scan spectroscopy

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ABSTRACT

The research on Nd³⁺ doped new solid-state laser hosts with specific thermo-mechanical and optical properties is very active. Nd³⁺ doped tellurite glasses are suitable for these applications. They have high linear and nonlinear refraction index, wide transmittance range. The TeO₂-ZnO (TZO) glass considered in the present work combines all those features and the nonlinear optical properties can be used for the development of Kerr-lens mode-locked sub picosecond lasers. Recently the laser performance of Nd³⁺ doped TZO glass was reported and laser slope efficiency of 21% was observed. We investigate how the intensity variation and the silver nanoparticles codoping affects the nonlinear optical properties of Nd³⁺ doped TZO glasses. Intensity dependent nonlinear refraction indices coefficients at 750, 800 and 850 nm were observed. The nonlinear optical features were obtained through ultrafast single beam z-scan technique with excitations at 750, 800 and 850 nm and are up to two orders of magnitude higher than those reported in the literature.

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1. Introduction

Nd³⁺ doped laser materials have a wide variety of applications such as short pulse with high peak power laser systems. Thus, they are very attractive and extensively studied. Also, they have a very interesting ensemble of features, namely, easier 4-levels laser operation mode and usually higher gain cross sections if compared to Yb³⁺ doped laser materials [1]. Even though laser action of Nd³⁺ has been observed in a many solid media such as Nd:YAG systems, the research on Nd³⁺ doped new solid-state laser hosts with specific thermo-mechanical and optical properties is very active. This is the case of some Nd doped nonlinear tellurite glasses [2–8]. They have a conjunction of good thermo-mechanical properties, typical

of crystals, and broad-band spectral properties, typical of glasses. Also, a very interesting combination of large nonlinear refraction index (25 times larger than that of silica), wide transmittance range [9].

Usually, crystalline laser hosts lead to higher absorption and emission cross sections, while glasses are produced in larger volumes with optimal optical quality at lower cost. In order to minimize the non-radiative multiphonon relaxations and to optimize the quantum efficiency of the ⁴F_{3/2} → ⁴I_{11/2} emission of Nd³⁺, it is also suitable to work with Nd³⁺ doped host materials with low contents of OH impurities. In that sense, laser emission of Nd³⁺ in glasses has been reported in fluorides [10–12], chalcogenides [13], aluminosilicates [14], germanates [15], and, as just mentioned, in tellurite glasses [2–6]. Among oxi-tellurites, the TeO₂-ZnO glass which is considered here in the present article combines good mechanical stability, chemical durability, high linear refraction index together with a wide transmission window (0.4–6 μm) and a

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high rare-earth solubility [10,16,17]. The large linear refraction index (1.97) [18] of this tellurite glass imply large stimulated emission cross-sections, sometimes larger than for phosphate glasses [19]. These glasses also have high nonlinear optical properties, which can be used advantageously for the development of Kerr-lens mode-locked sub picosecond lasers.

These tellurite glasses have been also studied recently for the possibility of using thin films for the fabrication of rib waveguides [20]. The possibility of increasing the luminescent quantum yield of rare-earth ions by codoping $\text{TeO}_2\text{-ZnO}$ glasses with silver nanoparticles [21,22] showed that they are potential materials for photonic devices applications. Recently it was demonstrated the reversible memory phenomena in Au-nanoparticles-incorporated $\text{TeO}_2\text{-ZnO}$ films [23]. Also, encouraging improvements have been reported regarding the laser performance of a Nd^{3+} doped TZO ($\text{TeO}_2\text{-ZnO}$) glass [24,25]. In this work, laser slope efficiency of 21% was observed. Thermo-optical properties of tellurite glasses codoped with rare Earth ions and metallic nanoparticles have also been reported [26–28].

These results motivated the present study that reports how the incident intensity variation and the silver nanoparticles codoping affects the nonlinear optical properties of Nd^{3+} doped TZO glasses.

Measurements were carried out with the Z-scan technique. The results displayed high intensity dependent nonlinear refraction indices at the wavelength range of 750–850 nm, at 80 MHz repetition rate and 100 fs pulses.

2. Experimental details

Glasses with the composition 85% TeO_2 -15% ZnO (wt.) (TZO) were obtained with the addition of 1% Nd_2O_3 and 1% AgNO_3 (wt.) (TZO:1%Nd:1%Ag). Also a sample without AgNO_3 (TZO:1%Nd) was prepared to be used as reference. Reagents were melted at 800 °C in a platinum crucible for 20 min, quenched in a pre-heated brass mold, annealed at 325 °C for 2 h, and cooled down to room temperature during 2 h to avoid internal stresses. At the end of the fabrication process, an additional heat treatment was performed for 24 h, to the sample produced with AgNO_3 to thermally reduce the Ag^+ ions to Ag^0 and nucleate silver nanoparticles, following the procedure already reported [21,22]. Through this procedure it was possible to obtain high quality samples, with even distribution of the dopants within the glass matrix (TZO).

Absorption spectra were measured in a Perkin-Elmer LAMBDA 9 spectrophotometer in wavelength range from 350 to 1000 nm.

A 200 kV transmission electron microscope (TEM) was employed to investigate the presence of nanoparticles in the samples.

The nonlinear optical features were obtained through ultrafast single beam z-scan setup displayed in Fig. 1. The excitation beam is a Mai Tai HP, Ti:Sapphire NIR, 100 fs, 80 MHz pulsed Gaussian beam linearly polarized. A Glan-Laser linear polarizer (GL5) positioned at the laser output controls the output intensity. Just after the GL5, a lens focuses the excitation beam in the sample posed on a displacement stage. After crossing the sample the beam goes through the aperture that is set either open or partially (50%) closed depending on the experiment needs – nonlinear refraction demands closed aperture, while nonlinear absorption demands open aperture. Then, the beam crosses a lens that collimates the signal into a silicon detector connected to a computer, by which the data acquisition is performed.

3. Absorption spectra and TEM imaging

The UV-VIS-NIR absorption (absorption coefficient) spectrum of the samples, registered between 350 nm and 950 nm is shown in Fig. 2. The features corresponding to the main absorption transitions of Nd^{3+} from $4f_{9/2}$ fundamental level to excited levels $4f_{3/2}$ (890 nm), $4f_{5/2}+2h_{9/2}$ (808 nm), $4f_{7/2}+4s_{3/2}$ (750 nm), $4f_{9/2}$ (690 nm), $4g_{5/2}+2g_{7/2}$ (580 nm) and $4g_{7/2}+4g_{9/2}+2k_{13/2}$ have been identified and highlighted in the figure. As expected for a glass, the absorption features appear as broad bands. Also it is interesting to notice that the addition of silver nanoparticles (TZO:1%Nd:1%Ag) to the initial composition (TZO:1%Nd) have resulted in a reasonable enhancement of the absorption. This effect may be attributed to the

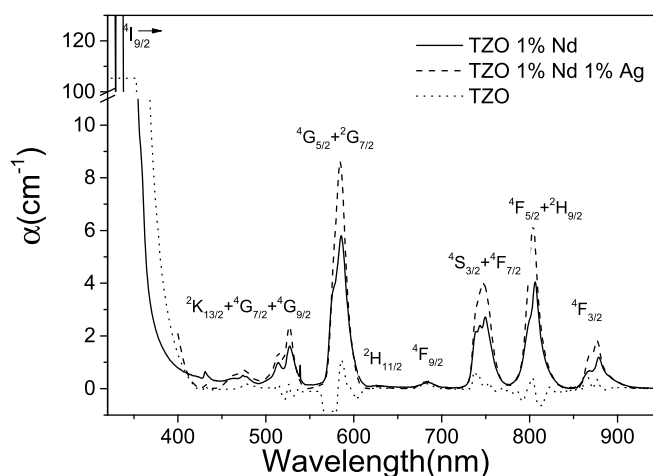


Fig. 2. UV–Vis–NIR absorption spectra for TZO doped and undoped samples.

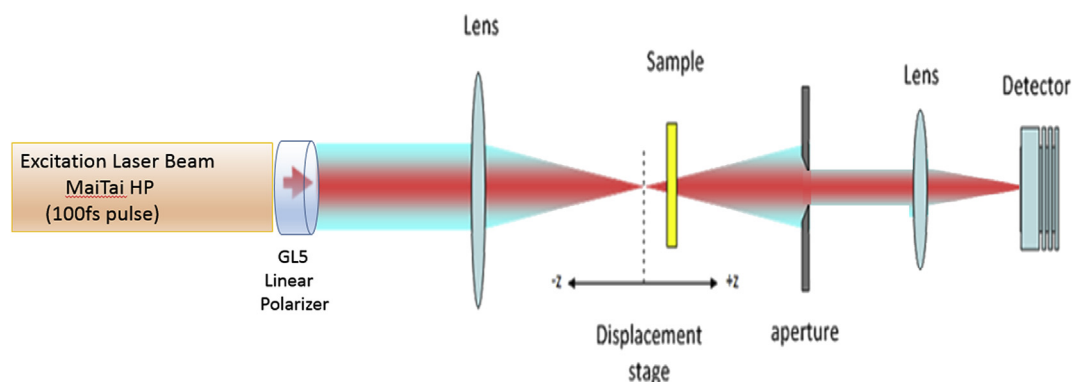


Fig. 1. Z-scan experimental setup [28].

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