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The effects of Nd_2O_3 concentration in the laser emission of TeO_2-ZnO glasses

L.M. Moreira [a], V. Anjos [a], M.J.V. Bell [a,*], C.A.R. Ramos [b], L.R.P. Kassab [b], D.J.L. Doualan [c], P. Camy [c], R. Moncorgé [c]

[a] Laboratório de Espectroscopia de Materiais, Departamento de Física, Universidade Federal de Juiz de Fora, Juiz de Fora, MG, Brazil
[b] Faculdade de Tecnologia de São Paulo, Laboratório de Tecnologia em Materiais Fotônicos e Optoeletrônicos, CEETEPS/UNESP, São Paulo, Brazil
[c] Centre de recherche sur les Ions, les Matériaux et la Photonique (CIMAP), UMR CNRS-CEA-Ensicaen, Universite de Caen, 6 Boulevard Maréchal Juin, F-14050, Caen, France

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ABSTRACT

The present work reports the modification introduced by different Nd_2O_3 concentration on optical properties and the laser operation of Nd^{3+} doped (TeO_2-ZnO) bulk tellurite glass. The spectroscopic data are analyzed within the Judd Ofelt formalism framework and the results are compared to the fluorescence lifetime and emission measurements to derive values for the quantum efficiency and the stimulated emission cross section of the considered $^4F_{3/2} \rightarrow {}^4I_{11/2}$ infrared laser transition around 1062.5 nm. Continuous-wave laser action is achieved with this bulk tellurite glass by pumping the sample inside a standard plan-concave mirror laser cavity with different output couplers. It is possible to observe coherent emission only for the lower concentration (0.5%(wt.) of Nd_2 O_3). Also laser action could only be observed for this sample with threshold pump power of 73 mW associated with a laser slope efficiency of 8% for an output coupler transmission of 4% indicating that TeO_2-ZnO are potential materials for laser action. The results presented in this work together with those previously reported with higher concentration (1.0% (wt) of Nd_2O_3) determine the adequate Nd_2O_3 concentration for laser action and guide the correct experimental procedure for TeO_2-ZnO glasses preparation.

1. Introduction

Nd^{3+} doped laser materials are very attractive and extensively studied for a wide variety of applications due to their easier 4-levels laser operation mode and usually higher gain cross sections if compared to Yb^{3+} doped laser materials. These features show to be true even considering applications on short pulse with high peak power laser systems [1]. The search for Nd^{3+} doped new solid-state laser hosts with specific thermo-mechanical and optical properties is very active, even though laser action of Nd^{3+} has been observed in a many solid media such as Nd:YAG systems. 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 refractive index (25 times larger than that of silica), wide transmittance range, and low maximum phonon frequency which allows rare-earth ion laser emissions in a wide spectral range are observed glasses [9].

In general, crystalline laser hosts lead to higher absorption and emission cross sections, while glasses can be 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 $^4F_{3/2} \rightarrow {}^4I_{11/2}$ emission of Nd^{3+}, it is also suitable to work with Nd^{3+} doped host materials with low phonon frequencies and low contents of OH impurities. In that sense, laser emission of Nd^{3+} 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 TeO_2-ZnO glass combines good mechanical stability, chemical durability, high linear and nonlinear refractive indices, together with low phonon energies (~750 cm^{-1}), a wide transmission window (0.4–6 μm) and a high rare-earth solubility [10,16,17]. The large linear refractive index (1.97) [18] of this tellurite glass imply large stimulated emission cross-sections,

* Corresponding author.
E-mail address: mjbell@fisica.ufjf.br (M.J.V. Bell).

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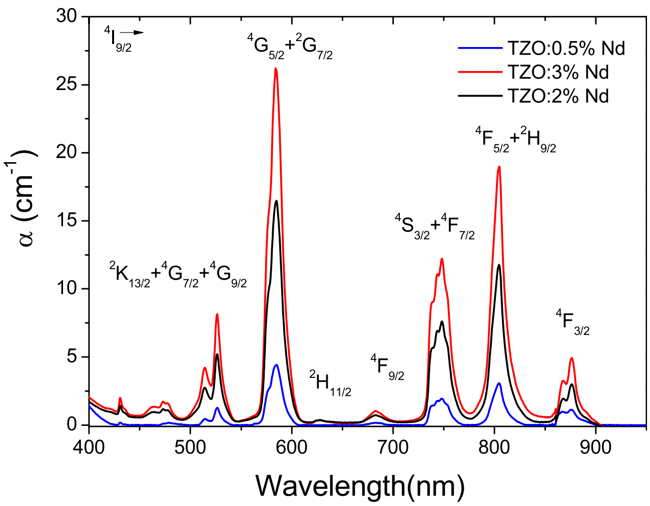

Fig. 1. UV-Vis-NIR absorption spectra for the TZO:Nd samples. The features corresponding to the main absorption transitions of Nd^{3+} from $^4I_{9/2}$ fundamental level to excited levels $^4F_{3/2}$(890 nm), $^4F_{5/2}+{}^2H_{9/2}$(808 nm), $^4F_{7/2}+{}^4S_{3/2}$(750 nm), $^4F_{9/2}$(690 nm), $^2H_{9/2}$, $^4G_{5/2}+{}^2G_{7/2}$(580 nm) and $^4G_{7/2}+{}^4G_{9/2}+{}^2K_{13/2}$ (520 nm) $^2G_{9/2}+{}^2D_{3/2}+{}^2K_{15/2}$ (480 nm) have been identified and highlighted in the figure.

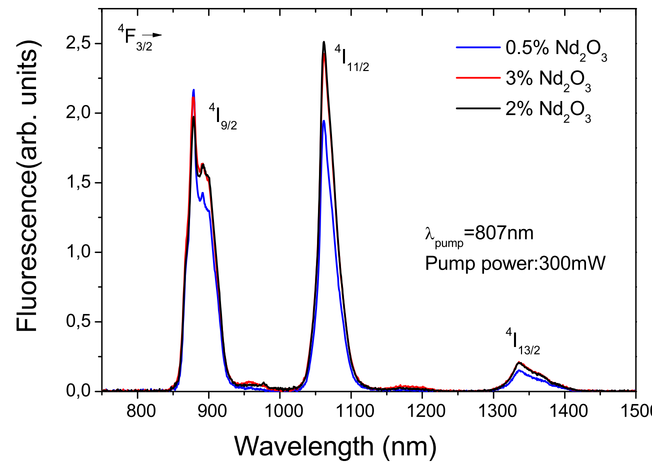

Fig. 2. NIR Fluorescence spectra of the TZO:Nd samples. They were obtained with excitation at 806 nm within the $^4I_{9/2} \rightarrow {}^4F_{5/2}+{}^2H_{9/2}$ absorption band. It consists of three broad-band emissions peaking around 882 nm, 1062 nm and 1335 nm. They are assigned to the three usual Nd^{3+} emission transitions, $^4F_{3/2} \rightarrow {}^4I_{9/2}$, $^4F_{3/2} \rightarrow {}^4I_{11/2}$, and $^4F_{3/2} \rightarrow {}^4I_{13/2}$.

sometimes larger than for phosphate glasses [19]. Their high nonlinear optical properties can be used advantageously for the development of Kerr-lens mode-locked subpicosecond lasers. The thermal properties of tellurite glasses have also been investigated and thermal conductivity and thermal diffusivity present reasonable values that encourage the development of optical devices [20,21]. Tellurite glasses have also been studied these last years for the possibility of using thin films for the fabrication of rib waveguides [22], and for the possibility of increasing the luminescent quantum yield of rare-earth ions in general with the addition of silver nanoparticles [23,24]. Recently an encouraging improvement has been reported regarding the laser performance of a TZO (TeO_2-ZnO) mixed tellurite and zinc oxide glass doped with 1% (wt.) of Nd_2O_3. In that work, a low laser threshold of 8 mW and a laser slope efficiency of 21% were observed [25]. That result motivated the present study that investigates for the first time how the Nd_2O_3 concentration influences on the optical properties as well as on the laser emission in the same TZO matrix of ref. [25]. Three different concentrations were analyzed and only at 0.5% (wt.) of Nd_2O_3 true CW laser emission could be obtained. Finally, this paper has the purpose to complement the previous results recently reported in Ref. [25] related to laser action of Nd^{3+} doped TeO_2-ZnO glasses and determine the suitable Nd_2O_3 concentration for laser action operation.

2. Experimental details

The investigated spectroscopic and laser samples were prepared by using the melting quenching technique with the following composition (in wt%): 85%TeO_2-15%ZnO (TZO). Three [3] samples were prepared with 0.5%, 2.0%, and 3.0% (wt) of Nd_2O_3 (TZO:*x*%Nd), one for each concentration. 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.

The absorption spectra were measured in a Perkin-Elmer LAMBDA 9 spectrophotometer in wavelength range from 350 to 1000 nm. The emission spectra was obtained by exciting the samples with a Titanium Safire (Ti:Sa) laser emitting at 808 nm (300 mW) and chopped at 100 Hz. The light emitted by the sample is collected with an optical fiber detector and a signal is analyzed with the aid of an Optical Spectrum Analyzer (OSA). The fluorescence lifetime measurements were achieved by exciting the sample with a pulsed optical parametric oscillator (OPO) system emitting at 808 nm (7 mJ, 5 ns pulse). The light produced by the sample is collected with a lens to the photomultiplier and finally it hits the detector that sends the signal to the oscilloscope coupled to a computer.

The laser set-up consisted in a standard plan-concave laser resonator. The flat dichroic mirror was highly reflective (R > 99.5%) around 1064 nm and with high transmittance (T > 95%) around 808 nm. Two different concave output mirrors with a radius of curvature of 100 mm and transmissions of 0.8% and 4% around 1064 nm were experimented. The laser samples were prepared in the form of a 10 × 10 mm^2 platelet of 1.65, 3.2 and 3.2 mm thickness, for the different concentrations with carefully polished and parallel end-faces but without any anti reflection coatings. They were stickled with silver paste on a copper sample holder without any particular cooling, and pumped through the dichroic input mirror by using a CW Ti: Sapphire laser tuned at 806 nm and focused with a lens of 10 cm focal length.

Table 1
Judd-Ofelt parameters for the TZO:Nd samples.

J.O. Parameters (10^{-20} cm^2)				
Sample	Ω_2	Ω_4	Ω_6	RMS
TZO:0.5%Nd	3.865	4.018	3.792	0.132
TZO:2%Nd	3.990	4.133	4.317	0.495
TZO:3%Nd	3.633	4.908	4.633	0.400

3. Spectroscopic and luminescence properties

The UV-VIS-NIR absorption (absorption coefficient) spectrum of the three samples investigated in the present work, in the range from 350 nm to 950 nm, is shown in Fig. 1. The features corresponding to the main absorption transitions of Nd^{3+} from $^4I_{9/2}$ 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), $^2H_{9/2}$, $^4G_{5/2}+{}^2G_{7/2}$ (580 nm) and $^4G_{7/2}+{}^4G_{9/2}+{}^2K_{13/2}$ (520 nm) $^2G_{9/2}+{}^2D_{3/2}+{}^2K_{15/2}$ (480 nm) have been identified and highlighted in the figure. As