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Random laser emission in Nd^{3+} doped tellurite glass

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***Abstract*— Random lasers are easier and cheaper to manufacture than regular ones, being made of several materials such as polymers, powders or dyes. Glass random lasers have been rarely studied due to their inhomogeneous broadened emission and low damage threshold. Here, we study Nd^{3+}doped TeO_2-ZnO-Al_2O_3 glasses with different concentrations of rare-earth doping (4 wt.%, 8 wt.% and 16 wt.%). Emission intensity per pump fluence and fluorescence decay time measurements showed the potential of these glasses for random laser applications in the near-infrared region.**

Keywords—random laser, glass, neodymium

I. Introduction

Random lasers are different from regular lasers since the stimulated emission is caused by scattering instead of a physical cavity, making them easier to fabricate. They were demonstrated experimentally for the first time in 1993 by Gouedard et al. [1], and since then they have been made from several materials [2-6], such as polymers [7] and dyes [8]. Glasses are promising materials to be used for random laser fabrication, since their properties can be easily manipulated with different matrices or rare-earth ions doping, which makes them able to generate a wide range of wavelengths depending on their composition. Glass random lasers, however, have been rarely studied [9-11], since they present an inhomogeneous emission band and low damage thresholds, making their stimulated emission difficult to evaluate from usual methods such as the narrowing of the emission linewidth.

Here, we study random laser emission of Nd^{3+} doped TeO_2-ZnO-Al_2O_3 glasses [12,13] with three different concentrations of Nd_2O_3 (4 wt.%, 8 wt.% and 16 wt.%). From their temporal decay measurements, it was possible to obtain the emission intensity curves, which showed a clear laser threshold for the three concentrations of Nd_2O_3, demonstrating the presence of stimulated emission for this material.

II. Materials and Methods

A. Glass and preparation

The glass was prepared with 85.0 TeO_2-12.95 ZnO-2.05 Al_2O_3 (TZA) composition, with 4 wt.%, 8 wt.% and 16 wt.% of Nd_2O_3. The melt-quenching method was used to prepare the samples. The starting reagents were melted at 835°C during 30 minutes, using a platinum crucible, and then quenched in a pre-heated brass mold, to be annealed at 300°C for two hours and then cooled down to room temperature. A more fundamental characterization of this glass matrix can be seen in [14].

The glass was turned into powder with an agate pestle, and to obtain the smallest particles, isopropyl alcohol was added and the suspension was placed within an ultrasound bath for three minutes, allowing separation of the supernatant particles (smaller) from the sunken ones (larger), giving particles of 1.5 µm average diameter, obtained through dynamic laser scattering. After drying, 100 mg of the supernatant particles were separated, and 0.1 mL of isopropyl alcohol was added to form a thick paste. This paste was placed into a (5 x 50) mm^2 well, having ~100 µm depth. The paste was left to dry, forming a film with the powder particles. Three samples were prepared, one for each Nd_2O_3 concentration

B. Temporal measurements

First, we measured the absorption spectrum (Fig. 1a) in order to verify the best wavelength for the pump. Since the 585 nm band ($^4G_{5/2} + {}^2G_{7/2}$) has higher absorption than the 806 nm ($^4F_{5/2}$) one, we chose to work with this shorter wavelength band. For the temporal measurement, we used an OPOTek model OPOlette laser system, with 10 ns pulses and repetition rate of 20 Hz, operating at 585 nm wavelength, and power varying from 0.1 mJ to 1.4 mJ. A RIGOL oscilloscope with 6 GSa/s and 350 MHz bandwidth was used to verify the decay time of the studied samples. The setup used for these measurements is shown in Fig. 1b.

III. Results and Discussion

To determine the fluorescence decay time, four measurements were taken for each pump energy to calculate an average and standard deviation for each point. For low energies, only fluorescence was observed, with a long decay time of microseconds. Increasing the pump energy led to a shorter pulse (with nanoseconds width), demonstrating the presence of stimulated emission.

Regarding the emission intensity per fluence (energy per area) measurements, by making different tests with filter combinations in front of the photodetector, we realized that the 1300 nm emission was being observed instead of the usual 1064 nm. This was verified by changing the OPO wavelength to both 1064 nm and 1300 nm, and changing the filters between a 1200 longpass filter and a 1064 nm bandpass. Afterwards, we repeated the measurement, and could see stimulated emission only for the 1300 nm line, while for the 1064 nm line only fluorescence was observed.

We verified the fluorescence decay time for each Nd_2O_3 concentration (Fig. 2), being 10.0 µs for 1064 nm and 20.4 µs

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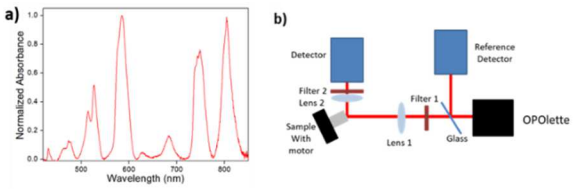

Figure 1: a) Normalized absorption spectrum for the sample with 16 wt.% of Nd_2O_3 ; b) Experimental setup used for temporal decay measurements. A glass cover slide is used to reflect 2% into a reference detector; reference detector is a Thorlabs Si photodetector; filter 1 is a longpass 550 nm filter; lens 1 has 150 mm focal length; lens 2 has 25 mm focal length; filter 2 is a 1000 nm longpass filter and either a 1200 nm longpass filter or a 1064 nm bandpass filter; detector is a Thorlabs InGaAs photodetector.

for 1300 nm for 4 wt.%; 7.2 µs for 8 wt.% for 1064 nm and 9.5 µs for 1300 nm; and for 16 wt.%, 3.7 µs for 1064 nm and 5.4 µs for 1300 nm. As expected, the decay times were shorter for higher Nd_2O_3 concentration, as reported in other works [15]. Since the 1064 nm emission band has larger emission cross-section, it was also expected that its decay times would be shorter than for the 1300 nm one. [14]

As the energy was increased, a shorter decay appeared, showing stimulated emission along with the fluorescence. By subtracting one low energy decay from the higher energy measurements we evaluated the emitted intensity of the stimulated emission; these results are presented in Fig. 2c that shows emission intensity versus fluence for the 1300 nm emission. All samples presented random laser behavior under pump power increase, with no output signal until the threshold fluence is reached, followed by a linear increase of the output intensity. We can see that the largest laser threshold occurs for the sample with 4 wt.% of Nd_2O_3, at 1.43 mJ/mm^2, and the shortest one for the sample with 16 wt.%, at 0.96 mJ/mm^2, which is approximately the same for the sample with 8 wt.%. This indicates a saturation of the laser threshold with Nd_2O_3 concentration.

Since the samples do not present the exact same thickness, it is not possible to compare their intensity values. However, it is possible to notice that the slope of the intensity is different for each Nd_2O_3 concentration; the highest slope takes place for the sample with 4 wt.% of Nd_2O_3 whereas the lowest happens for the one with 8 wt.%.

IV. Conclusions

We studied TZA samples with 4 wt.%, 8 wt.% and 16 wt.% of Nd_2O_3. The different fluorescence times for the 1064 nm and 1300 nm emission wavelengths were studied, showing that for higher concentrations, the smaller is the decay time. Moreover, the decay time for 1064 nm is always smaller than for 1300 nm. The emission intensity per fluence was studied and it was possible to verify a clear laser threshold for the three samples, with the lowest value of 0.96 mJ/mm^2 for the one with 16 wt.% of Nd_2O_3. This is, to our knowledge, the first random laser result at 1300 nm for tellurite glasses.

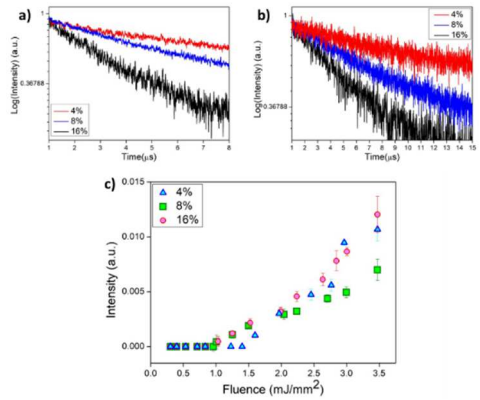

Figure 2: Decay times measured for 4 wt.% (red), 8 wt.% (blue) and 16 wt.% (black) of Nd2O3 at (a) 1064 nm and (b) 1300 nm emission. (c) Emission intensity per fluence at 1300 nm for the 4 wt.% (blue triangles), 8 wt.% (black squares) and 16 wt.% (red circles) samples.

References

[1] C. Gouedard, D. Husson, C. Sauteret, F. Auzel, and A. Migus, "Generation of spatially incoherent short pulses in laser-pumped neodymium stoichiometric crystals and powders," *Journal of the Optical Society of America B-Optical Physics,* vol. 10, no. 12, pp. 2358-2363, Dec 1993.

[2] E. Jimenez-Villar, I.F. da Silva, V. Mestre, N. U. Wetter, C. Lopez, P. C. de Oliveira *et al.*, "Random Lasing at Localization Transition in a Colloidal Suspension (TiO2@Silica)," Acs Omega, vol. 2, no. 6, pp. 2415-2421, Jun 2017.

[3] N. U. Wetter, J. M. Giehl, F. Butzbach, D. Anacleto, and E. Jimenez-Villar, "Polydispersed Powders (Nd3+:YVO4) for Ultra Efficient Random Lasers," *Particle & Particle Systems Characterization,* vol. 35, no. 4, Apr 2018, Art no. 1700335.

[4] N. U. Wetter, A. R. de Miranda, E. Pecoraro, S. J. L. Ribeiro, and E. Jimenez-Villar, "Dynamic random lasing in silica aerogel doped with rhodamine 6G," Rsc Advances, vol. 8, no. 52, pp. 29678-29685, 2018.

[5] K. C. Jorge, M. A. Alvarado, E. G. Melo, M. N. P. Carreno, M. I. Alayo, and N. U. Wetter, "Directional random laser source consisting of a HC-ARROW reservoir connected to channels for spectroscopic analysis in microfluidic devices," Applied Optics, vol. 55, no. 20, pp. 5393-5398, Jul 2016.

[6] R. J. R. Vieira, L. Gomes, J. R. Martinelli, and N. U. Wetter, "Upconversion luminescence and decay kinetics in a diode-pumped nanocrystalline Nd3+:YVO4 random laser," *Optics Express,* vol. 20, no. 11, pp. 12487-12497, May 2012.

[7] R. C. Polson, A. Chipouline, and Z. V. Vardeny, "Random lasing in pi-conjugated films and infiltrated opals," *Advanced Materials,* vol. 13, no. 10, pp. 760-764, May 2001.

[8] W. L. Sha, C. H. Liu, and R. R. Alfano, "Spectral and temporal measurements of laser action of rhodamine-640 dye in strongly scattering media," *Optics Letters,* vol. 19, no. 23, pp. 1922-1924, Dec 1994.

[9] J. G. Camara, D. M. da Silva, L. R. P. Kassab, C. B. de Araujo, A. S. L. Gomes, "Random laser emission from neodymium doped zinc tellurite glass-powder presenting luminescence concentration quenching," *Journal of Luminescence,* vol. 233, p. 117936, 2021.

[10] X. S. Wang, J. R. Qiu, J. Song, J. Xu, Y. Liao, H. Y. Sun *et al.*, "Upconversion luminescence and optical power limiting effect based on two- and three-photon absorption processes of ZnO crystal," (in English), *Optics Communications,* Article vol. 280, no. 1, pp. 197-201, Dec 2007.

[11] J. G. Camara, D. M. da Silva, L. R. P. Kassab, M. L. Silva-Neto, G. Palacios, and C. B. de Araujo, "Random lasing and replica symmetry breaking in GeO2-PbO-MgO glass-ceramics doped with neodymium," *Scientific Reports,* vol. 12, no. 1, Nov 2022, Art no. 19438.

[12] L. R. P. Kassab, M. E. Fukumoto, V. D. D. Cacho, N. U. Wetter, and N. I. Morimoto, "Spectroscopic properties of Yb3+ doped PbO-Bi2O3-Ga2O3 glasses for IR laser applications," *Optical Materials,* vol. 27, no. 10, pp. 1576-1582, Sep 2005.

[13] D. S. da Silva, N. U. Wetter, W. de Rossi, L. R. Pires Kassab, and R. E. Samad, "Production and characterization of femtosecond laser- written double line waveguides in heavy metal oxide glasses," *Optical Materials,* vol. 75, pp. 267-273, Jan 2018

[14] C. D. S. Bordon, E. S. Magalhaes, D. M. da Silva, L. R. P. Kassab, and C. B. de Araujo, "Influence of Al2O3 on the photoluminescence and optical gain performance of Nd3+ doped germanate and tellurite glasses," *Optical Materials,* vol. 109, Nov 2020, Art no. 110342.

[15] M. Sobczyk, "Temperature-dependent luminescence and temperature-stimulated NIR-to-VIS up-conversion in Nd3+-doped La2O3-Na2O-ZnO-TeO2 glasses," *Journal of Quantitative Spectroscopy & Radiative Transfer,* vol. 119, pp. 128-136, Apr 2013.